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Air Quality Trends in Finland, 1994–2018

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To my grandchildren

Eko, Kimo and Kaïa

“Air quality is the composition of the air

in terms of how much pollution it contains.”

Collins English Dictionary

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Abstract

In this thesis, long-term, multicomponent, high-resolution (time and accuracy) air quality monitoring data from about 400 sites across Finland since 1994 are integrated into a single unified and compact view to demonstrate past air quality development and to assess the reasons behind the development at the national level.

This thesis demonstrates that internationally launched and nationally implemented regulatory controls have had an important role in improving air quality in Finland. The pollutants subject to long-term ambitious international abatement strategies (like SO_2 and persistent organic pollutants) have decreased the most. Also, NO_x emission control has been successful, but urban roadside NO_2 concentrations have not decreased as expected. The increase in diesel cars (and their potentially high primary NO_2 emissions) may have been one factor in slowing down the decline of concentrations. However, the development of emission reduction technologies together with the improved type approval test procedures have resulted in a reduction in the significance of primary NO_2 emissions in Europe. In Finland, our relatively old car fleet and the increased import of old diesel cars cause uncertainty for future development.

Due to the use of studded tyres and manifested as elevated concentrations of PM_{10}, springtime street dust is a local air pollution problem. This thesis suggests that local abatement measures (e.g., reducing traffic, changes in the car fleet, road maintenance activities) have been moving in the right direction, and the springtime street dust levels have been reduced. Although air quality standards are not exceeded today, street dust remains a persistent flaw in our otherwise good air quality.

In Finland, the ozone peak levels have been declining since 2006. Similar development has been detected in Europe and North America, and it is related to decreasing anthropogenic precursor emissions of NO_x and VOCs. For Finland, high background concentrations are more problematic, and reducing them would require international and even hemispheric cooperation.

The available long-term background data of PAH concentrations suggest that no widespread decrease in concentrations has occurred. This is not necessarily surprising as the major global sources are small-scale solid fuel combustion and wildfires. Efforts to reduce these emissions have been relatively limited or non-existent so far.

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Tekijä

Pia Anttila

Nimeke

Ilmanlaatutrendit Suomessa 1994–2018

Tiivistelmä

Tässä väitöskirjassa on koottu yhteen Suomen ilmanlaadun mittaustiedot yli parinkymmenen vuoden ajalta ja noin 400 mittausasemalta. Aineistosta arvioidaan ilmanlaadun kehitystä ja syitä havaittuun kehitykseen kansallisella tasolla.

Tämä opinnäyte osoittaa, että kansainvälisesti käynnistetyillä ja kansallisesti toteutetuilla päästöjen rajoittamistoimilla on ollut tärkeä rooli ilmanlaadun parantamisessa Suomessa. Epäpuhtaudet, joihin on kohdistunut pitkäaikaisia kunnianhimoisia kansainvälisiä päästöstrategioita (kuten SO_2 ja pysyvät orgaaniset ympäristömyrkyt), ovat vähentyneet eniten. Myös NO_x-päästöjen vähentäminen on onnistunut, mutta kaupungeissa NO_2-pitoisuudet eivät ole vähentyneet odotetusti. Dieselautojen lisääntynyt määrä (ja niiden mahdollisesti korkeat suorat NO_2-päästöt) on saattanut olla yksi tekijä, joka on hidastanut NO_2-pitoisuuksien laskua. Päästöjen vähentämistekniikoiden ja tyyppihyväksyntämenettelyjen kehittyminen on kuitenkin vähentänyt suorien NO_2-päästöjen merkitystä Euroopan tasolla. Suomessa suhteellisen vanha autokanta ja vanhojen dieselautojen lisääntynyt tuonti aiheuttavat epävarmuutta tulevaisuuden kehitykselle.

Kevään katupöly, joka johtuu nastarenkaiden käytöstä ja joka ilmenee korkeina PM_{10}-pitoisuuksina, on paikallinen ilmanlaatuongelma. Tämä opinnäyte viittaa siihen, että paikalliset vähennystoimenpiteet (esim. liikennemäärien vähentäminen, muutokset autokannassa, tienhoitotoimet) ovat olleet oikeansuuntaisia ja kevään katupölytasot ovat vähentyneet. Vaikka ilmanlaatunormeja ei tällä hetkellä ylitetä, katupöly on edelleen sitkeä ilmanlaatuhaitta muuten hyvässä ilmanlaadussamme.

Suomessa otsonin huipputasot ovat laskeneet vuodesta 2006. Vastaavaa kehitystä on havaittu Euroopassa ja Pohjois-Amerikassa, ja se on liitetty typen oksidien (NO_x) ja haihtuvien orgaanisten yhdisteiden (VOC) päästöjen vähentämiseen. Suomessa kuitenkin korkeat taustapitoisuudet ovat ongelmallisempia, ja niiden alentaminen edellyttää laajaa kansainvälistä yhteistyötä.

PAH-pitoisuuksissa ei ole tapahtunut laaja-alaista laskua. Tämä ei ole välttämättä yllättävää, koska suurimpia lähteitä maailmanlaajuisesti ovat pienpoltto ja metsä- ja maastopalot. Pyrkimykset näiden päästöjen vähentämiseksi ovat toistaiseksi olleet suhteellisen vähäisiä tai olemattomia.

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Preface

Seriously, this thesis has been underway for almost ten years. Its slow progress is not only because of my personal deficiencies as a researcher but also because of other tasks that have more strongly drawn my interest. Of those, I want to mention my participation in modernising the national air quality information system and its integration with other FMI ITC systems, the expert tasks in air quality capacity building projects in Balkan and Central Asia, and my long-lasting interest in communicating information about ambient air quality. To me, these years have been both interesting and meaningful, and I greatly appreciate the efforts, support and help I have received from my colleagues during various projects. These activities have also shaped my perception of the air quality in general and have strongly influenced how this thesis finally came to be.

I've been very fortunate to have spent most of my professional life at the Finnish Meteorological Institute, where I have had the opportunity to create such a varied career among different aspects of air quality, and for this, I express my gratitude.

I thank Academician, Academy Professor Markku Kulmala for believing that my (fragmented) research on air quality could finally make a coherent story worthy of becoming a doctoral dissertation. Of course, it helped that I have had the privilege to co-write my articles with brilliant researchers, for which I am truly grateful. I thank my pre-examiners, Professor Heikki Junninen and Associate Professor Topi Rönkkö for their supportive, sound advice. And warm thanks to my supervisor, Research Professor Hannele Hakola, who has been encouraging me towards this dissertation during these years.

And finally, special thanks are due to the Finnish air quality community, the hundreds of air quality data producers in municipalities, industries and institutions, colleagues in the reference laboratory of air quality, and the data flow and repository managers at FMI. Without your input, this work would not have been possible.

In Helsinki, Ilmala, 20 February 2020

Pia Anttila

Abbreviations

ACF	Autocorrelation function
AICC	Bias-corrected Akaike information criterion
AirBase	European air quality database maintained by the EEA
AMAP	Arctic Monitoring and Assessment Programme under the Arctic Council
Ant	Anthracene
AQM	Air quality monitoring
ARMA	Autoregressive moving-average
BaA	Benz(a)anthracene
BaP	Benzo(a)pyrene
BbF	Benzo(b)fluoranthene
BkF	Benzo(k)fluoranthene
BP	Benzo(ghi)perylene
BAPMoN	WMO Background Air Pollution Monitoring Network
CDPF	Catalytic diesel particulate filters
CEN	European Committee for Standardization
Chr	Chrysene
CLRTAP	Convention on Long-range Transboundary Air Pollution
CO	Carbon monoxide
DDD	Dichlorodiphenyldichloroethane
DDE	Dichlorodiphenyldichloroethylene
DDT	Dichlorodiphenyltrichloroethane
DhA	Dibenz(a,h)anthracene
EEA	European Environment Agency
EGAP	Group of experts on airborne pollution of the Baltic Sea area under HELCOM
EMEP	UNECE European Monitoring and Evaluation Programme
EU	European Union
FAQDMS	Finnish air quality data management system
Fl	Fluoranthene
FMI	Finnish Meteorological Institute
GAW	WMO Global Atmosphere Watch
GDP	Gross Domestic Product
GLS	Generalised least square
HCH	Hexachlorocyclohexane
HELCOM	Baltic Marine Environment Protection Commission – Helsinki Commission
HSY	Helsinki Region Environmental Services Authority (Helsingin seudun ympäristöpalvelut –kuntayhtymä)
IcP	Indeno(1,2,3-cd)pyrene
iid	independent and identically distributed
IM	UNECE Integrated Monitoring Network
INSPIRE	Directive of the Infrastructure for Spatial Information in the European Community
IPR	Directive of the reciprocal exchange of information and reporting on ambient air quality
ISO	International Organization for Standardization
IVL	Swedish Environmental Research Institute
LRT	Long-range transport

MLE	Maximum likelihood estimation
NEDC	New European Driving Cycle
NO	Nitrogen monoxide
NO_2	Nitrogen dioxide
NO_x	$NO+NO_2$
O_3	Ozone
OCP	Organochlorine pesticide
OLS	Ordinary least square
PAH	Polycyclic aromatic hydrocarbon
ppbv	parts per billion by volume
PCB	Polychlorinated biphenyl
PEMS	Portable emission measurement system
Phe	Phenanthrene
PM	Particulate matter
PM_{10}	Particles less than 10 µm in aerodynamic diameter
$PM_{2.5}$	Particles less than 2.5 µm in aerodynamic diameter
PMF	Positive matrix factorization
POP	Persistent organic pollutant
Pyr	Pyrene
RDE	Real Driving Emissions
SECA	Sulphur Emission Control Area
SIA	Secondary inorganic aerosols
SO_2	Sulphur dioxide
STD	Standard deviation
TM	Trace metals
TSP	Total suspended particles
TWC	Three-way catalytic converters
UNECE	United Nations Economic Commission for Europe
WMO	World Meteorological Organisation
WN	White noise
VOC	Volatile organic compound
WLTP	Worldwide harmonised light vehicle test procedure
WSI	Water soluble ions

gas	conversion between µg/m^3 and ppbv (293 K, 101.3 kPa)
SO_2	1 µg/m^3 = 0.376 ppb
NO_2	1 µg/m^3 = 0.523 ppb
NO	1 µg/m^3 = 0.802 ppb
O_3	1 µg/m^3 = 0.500 ppb
CO	1 µg/m^3 = 0.86 ppb

Contents

Publications of the thesis

This thesis consists of five original articles and a synthesis where these articles are reviewed in the framework of the research field in focus. The articles with their original abstracts are listed below. All measurements were either performed by the co-authors or validated data were retrieved from open-data archives. The AQM data for **Papers I** and **IV** originate from the Finnish air quality monitoring networks and were downloaded from the FMI AQ database. Data for **Paper II** were provided by the co-author at HSY. The long-term persistent organic pollutants' (POPs) data in **Paper III** were provided by the co-authors from the IVL chemical laboratory. The data for **Paper V** were received directly from the co-authors at FMI. I am responsible for the overall planning of the papers, data analysis and calculations, except in **Paper V** Lic.Sc. Sirkka Leppänen conducted the PMF calculations. Interpretation of the results was made together with the co-authors, and the co-authors also contributed to the writing of the papers.

I Anttila, P., Tuovinen, J.-P., 2010. Trends of primary and secondary pollutant concentrations in Finland in 1994–2007. Atmos. Environ., 44, 38–49.

The trends in the atmospheric concentrations of the main gaseous and particulate pollutants in urban, industrial and rural environments across Finland were estimated for the period of 1994–2007. The statistical analysis was based on generalized least-squares regression with classical decomposition and autoregressive moving-average (ARMA) errors, which were applied to monthly-averaged data. In addition, three alternative methods were tested. Altogether, 102 pollutant time series from 42 sites were analysed. During the study period, the concentrations of SO_2, CO and NO_x declined considerably and widely across Finland. The SO_2 concentrations at urban and industrial sites were approaching background levels. The reductions in NO_x and CO concentrations were comparable to those in national road traffic emissions. A downward trend was detected in half of the NO_2 time series studied, but the reductions were not as large as would be expected on the basis of emission trends, or from NO_x concentrations. For O_3, neither the mean nor the peak values showed large changes in background areas, but they were increasing in the urban data. For PM_{10}, 5 of the 12 urban time series showed decreasing mean levels. However, the highest concentrations, typically attributable to the problematic springtime street dust, did not decrease as widely. The reduction of the long-range transported major ions, mainly driven by the large-scale reduction in sulphur emissions, possibly plays a significant part in the decreases in the mean PM_{10} concentrations. It was shown that the handling of the serially correlated data with the ARMA processes improved the analysis of monthly values. The use of monthly rather than annually-averaged data helped to identify the weakest trends.

II Anttila, P., Tuovinen, J.-P., Niemi, J.V., 2011. Primary NO_2 emissions and their role in the development of NO_2 concentrations in a traffic environment. Atmos. Environ., 45, 986–992.

An assessment of the formation of NO_2 concentrations in heavily traffic-influenced environments in Helsinki, Finland was carried out. The proportion of primary NO_2 emissions from road traffic was estimated using a statistical model for the relationship between the mixing ratios of nitrogen oxides ($NO + NO_2$) and total oxidant ($O_3 + NO_2$) measured in 1994–2009. Based on this analysis, a quantitative estimate was derived for the relative importance of the primary NO_2 emissions, ambient $NO–NO_2–O_3$ equilibrium and background concentrations in the observed NO_2 concentrations. The proportion of primary NO_2 in the vehicular NO_x

emissions increased from below 10% in the 1990s to about 20% in 2009, with a more distinctive increase during the most recent years. This development was related to the changes in the proportion of diesel-powered passenger cars in Finland. Between 1994 and 2004, the photochemical NO-to-NO_2 conversion comprised on average 51% of the mean NO_2 concentration, while the primary NO_2 emissions contributed 31%. The role of the primary NO_2 emissions was limited by the steeply-decreasing total NO_x emissions. More recent data (2005–2009) yielded higher primary NO_2 emission fractions (15–21%), with a clearly increasing trend. As a result, the contribution of chemical conversion steadily decreased from 54% in 2005 to 43% in 2009, while that of the primary NO_2 emissions increased from 32 to 44%. In order not to exceed in future the annual limit of NO_2 concentration, set by the European Union, in the busiest street canyons in downtown Helsinki, the primary NO_2 emissions need to be addressed alongside the total NO_x emissions.

III Anttila, P., Brorström-Lundén, E., Hansson, K., Hakola, H., Vestenius, M., 2016. Assessment of the spatial and temporal distribution of persistent organic pollutants (POPs) in the Nordic atmosphere. Atmos. Environ., 140, 22–33.

Long-term atmospheric monitoring data (1994–2011) of persistent organic pollutants (POPs) were assembled from a rural site in southern Sweden, Råö, and a remote, sub-Arctic site in Finland, Pallas. The concentration levels, congener profiles, seasonal and temporal trends, and projections were evaluated in order to assess the status of POPs in the Scandinavian atmosphere. Our data include atmospheric concentrations of polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs), altogether comprising a selection of 27 different compounds.

The atmospheric POP levels were generally higher in the south, closer to the sources (primary emissions) of the pollutants. The levels of low-chlorinated PCBs and chlordanes were equal at the two sites, and one of the studied POPs, α-HCH, showed higher levels in the north than in the south.

Declining temporal trends in the atmospheric concentrations for the legacy POPs — PCBs (2–4% per year), HCHs (6–7% per year), chlordanes (3–4% per year) and DTTs (2–5% per year) — were identified both along Sweden's west coast and in the sub-Arctic area of northern Finland. Most of PAHs did not show any significant long-term trends.

The future projections for POP concentrations suggest that in Scandinavia, low-chlorinated PCBs and p,p′-DDE will remain in the atmospheric compartment the longest (beyond 2030). HCH's and PCB180 will be depleted from the Nordic atmosphere first, before 2020, whereas chlordanes and rest of the PCBs will be depleted between the years 2020 and 2025. PCBs tend to deplete sooner and chlordanes later from the sub-Arctic compared to the south of Sweden.

This study demonstrates that the international bans on legacy POPs have successfully reduced the concentrations of these substances in the Nordic atmosphere. However, the most long-lived compounds may continue in the atmospheric cycle for another couple of decades.

IV Anttila, P., Salmi, T., 2006. Characterizing temporal and spatial patterns of urban PM_{10} using six years of Finnish monitoring data. Boreal Env. Res., 11, 463–479.

Data from the Finnish Meteorological Institute's Air Quality Monitoring Data Management System (ILSE) for 1998–2003 were used to examine the temporal and spatial patterns of urban PM_{10} in Finland. Long term means of PM_{10} at 24 Finnish urban stations vary between 11 and 24 $\mu g/m^3$. The seasonal variation of PM_{10} at all stations was dominated by the spring maximum. A strong influence of traffic on the urban PM_{10} concentrations is shown. However, the highly synchronized day-to-day variation at a variety of sites across the country highlights the role of

large-scale weather patterns also in the formation of spring episodes. Every year, most often in August, September and October, there were also 1–5 irregular regional PM_{10} episodes, lasting from one day to six days and most likely caused by long-range transported particles. During these regional events, the PM_{10} concentrations may well reach the typical spring peak concentration levels.

V Anttila, P., Makkonen, U., Hellén, H., Pyy, K., Leppänen, S., Saari, H., Hakola, H., 2008. Impact of the open biomass fires in spring and summer of 2006 on the chemical composition of background air in south-eastern Finland, Atmos. Environ., 42, 6472–6486.

In the spring and summer of 2006, the air quality in southern Finland was affected by two major biomass fire smoke episodes. At the Virolahti background station, closest to the eastern fire areas, the episodes lasted altogether several weeks. The high point in spring was 25 April and in summer 13 August. In spring the aerosol detected at Virolahti originated at distances of even hundreds of kilometres to the south and south-east, and consequently was a mixture of material from biomass burning and from other sources (both LRT and local), all of which contributed to the detected elevation of PM_{10} concentrations. The elevated concentrations of trace elements (Cd, Pb, Zn) during the most intense biomass fire episode were associated with other anthropogenic emissions.

In contrast, during August 2006, the PM_{10} at Virolahti was quite exclusively impacted by close (ca. 50–100 km) biomass fire sources. The presumably organic component comprised, at its highest, as much as 90% of the total PM_{10}. In addition to record high PM_{10} and $PM_{2.5}$ concentrations, the concentrations of polycyclic aromatic hydrocarbons were considerably elevated, even reaching values more typical of wintertime urban environments. During the peaks of the episodes in August, the total gaseous mercury concentration in the air was more than double its background value. In general, the trace elements did not exceed their background values.

The publications are referred to in the text by their Roman numerals. The publications are reproduced with the permission of the journals concerned.

New but comparable material has also been downloaded from the FMI AQ database and is presented in this thesis.

1 Introduction

Measurements and research of outdoor, ground-level ambient air quality have been made routinely in Finland for many decades. The number of sites and pollutants measured started to expand substantially in the early 1990s following the implementation of the European Union (EU) legislation. The first major instrument was the Air Quality Framework Directive 96/62/EC and its three daughter directives. The directives established standards for pollutants, including sulphur dioxide (SO_2), nitrogen dioxide (NO_2), particulate matter (PM_{10}) and lead (Pb) in 1999; benzene (C_6H_6) and carbon monoxide (CO) in 2000; and ozone (O_3) in 2001. These were later consolidated into the Ambient Air Quality Directive 2008/50/EC (EU, 2008), which set objectives for fine particulate matter ($PM_{2.5}$). Together with Directive 2004/107/EC (EU, 2004, relating arsenic (As), cadmium (Cd), mercury (Hg), and nickel (Ni) and polycyclic aromatic hydrocarbons (PAHs)), the Ambient Air Quality Directive provides the current framework for the control of ambient concentrations of air pollution in the European Union (EU).

In addition to setting the air quality standards, the directives determined the required quantity and quality of mandatory monitoring in relation to the prevailing pollution situation and potentially exposed population as well as details of the required reporting and dissemination of information. The implementation of EU legislation in Finland strongly involved municipalities and industry in air quality monitoring in their own territories, but they also opened it up to nationwide co-operation. Nowadays, municipalities and local industry often perform the necessary monitoring tasks in economic and/or technical co-operation. The Finnish Meteorological Institute is responsible for background air quality monitoring as well as collecting, reporting, assessing and disseminating AQ information at the national level.

The objectives of air quality monitoring (AQM) are typically to establish the levels of exposure (human and/or ecosystem), to ensure compliance with legislation and to demonstrate the effectiveness of control measures. For these purposes, data are summarised as various annual statistics in accordance with the rules of the mandate in question. Such statistics serve to check compliance, but one aspect – long term trends – is not fully covered in this regulatory context.

Understanding the status and developments of air quality is crucial to supporting national air quality management and the implementation of control measures. In this work, long-term, multicomponent, high-resolution (time and accuracy) AQM data available from about 400 Finnish sites since 1994 are utilised to demonstrate past air quality development at the national level.

The focus is on Finland and Finnish monitoring results, but due to the transboundary nature of air pollution, research on a European or even a hemispheric scale is needed.

The objectives of this thesis are

- to aggregate the available long-term AQM data and process them into unified and compact information
- to answer how the air quality in Finland has developed
- to explain which factors are behind the observed air quality development.

2 Material

In Finland, systematic air quality monitoring can be said to have started in the early 1970s with the SO_2, sulphate and TSP measurements at a couple of background sites operated by FMI. In the same decade, first urban networks were also initiated in Helsinki and Oulu. The Air Protection Act of 1982 (67/1982) proclaimed that the municipalities shall see to the necessary air quality monitoring within their territories according to local conditions. The law also included a provision for the establishment of an "Air protection data register" (Ilmansuojelun tietorekisteri) to provide data for the necessary planning, control and research of air protection.

FMI continues to be responsible for the background air quality monitoring (see, e.g. Joffre et al., 1990; Ruoho-Airola, 2004; Makkonen, 2014). From the 1970s to the early 1990s, background monitoring was mainly driven by international research programmes (CLRTAP, EMEP, BAPMoN, EGAP, IM, AMAP, GAW), but from the 1990s onwards, the EU has also become an important actor in background monitoring by setting new components to be monitored and new measurement techniques to be used. In the mid-2000s, monitoring of trace elements, PAHs and Hg was started at three background sites as provided by Directive 2004/107/EC.

Since 2000, the national reference laboratory operated by FMI has been an important quality assurance resource to improve the reliability and comparability of the automatic analysers used in compliance monitoring (Waldén, 2009; Waldén et al., 2004; 2008; 2010; 2015; Walden and Vestenius 2018). In FMI's background network, methods based on the sampling and chemical analyses are widely used. From the beginning, chemical analyses used in the various international programmes were intercompared annually and sampling equipment a couple years later (see, e.g. Karlsson et al., 2007). Sampling and analysis methods of EU compliance monitoring are defined in the directives and are based on CEN or ISO standards. In 1997, the first accreditation of FMI's atmospheric chemistry laboratory was approved.

The air quality data register initiated in 1982 has evolved into the Finnish Air Quality Data Management System (FAQDMS) operated by FMI. This system, among other things, collects the validated air quality monitoring data (and metadata) of the FMI network and urban/industrial monitoring networks since 1973 and 1985, respectively. FMI's AQ archive aggregates Finnish data originating from long-term international research programmes, data that aim to check the compliance with air quality standards, and long-term monitoring data obliged by environmental permits.

Validated data are updated annually and processed and reported to the EU Commission, the EEA (IPR compatible) and other international organisations (e.g. EMEP). The system also processes the (near) real-time data received hourly from the stations and transfers the information to the website (https://en.ilmatieteenlaitos.fi/air-quality) and FMI's INSPIRE-compatible open data service interface (https://en.ilmatieteenlaitos.fi/open-data).

At present, around 30 independent operators (most of them municipalities) perform the compliance AQM in Finland. The biggest of these, FMI and HSY, both have a dozen fixed monitoring sites with comprehensive measurement programmes (ilmatieteenlaitos.fi/seurantamittaukset; hsy.fi/fi/asukkaalle/ilmanlaatu/). These data are also

widely used as supplementary or principal material in scientific works, while the rest of the monitoring data are not as widely exploited for scientific purposes.

The expansion in air quality monitoring is illustrated in Figure 1, which shows the steep increase in the number of continuous hourly measurements starting in the mid-1980s. The figure also reflects changes in the priorities of air quality monitoring during these decades. In 1993, sulphur dioxide levels were monitored at almost one hundred measuring stations, and since then, that number has been cut in half. Meanwhile, the number of size-selective PM mass measurements (PM_{10} or $PM_{2.5}$) has increased from fewer than 30 to over 100.

Over the years, in some cities/communities, AQM has discontinued, owing to the decline of the local smokestack (basic manufacturing/energy production) industries (e.g. Inkoo, Kaskinen, Koverhar, Valkeakoski). The focus has shifted more and more to the monitoring of pollutants generated (directly or indirectly) from vehicular emissions and other small-scale combustion, i.e. NO_x and particles. Such developments have also been enhanced by the increasing focus on the health impacts of air pollution and the role of pollutants in climate change.

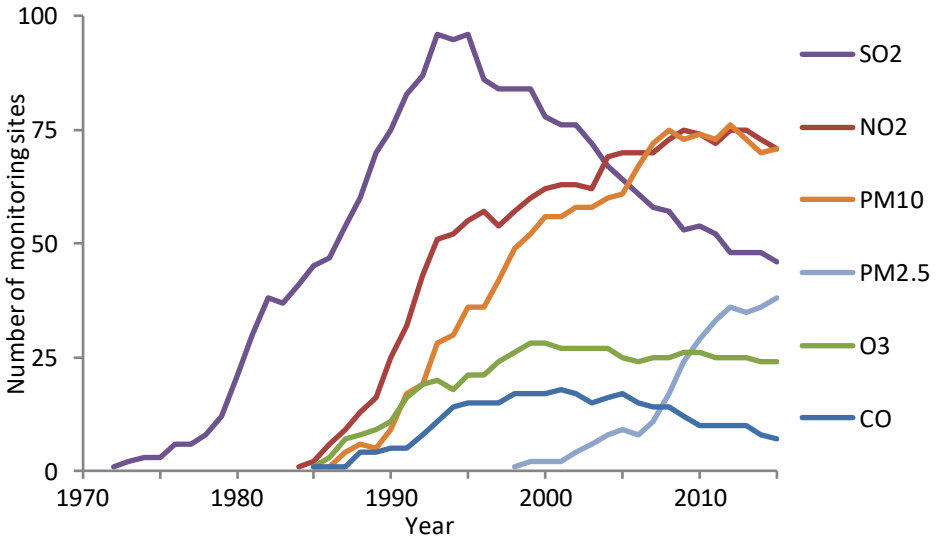

Figure 1. Number of operative air quality monitoring sites per year and per component in Finland.

Kukkonen et al. (1999) published the first review of urban air quality based on the Finnish AQM data from 1990–1993. This work focused on the comparison of concentrations to the national air quality guideline values issued in 1996. Anttila et al. (2003) compiled a summary of the AQM data from 1985–2000. By that time, it had become meaningful to make statistical analyses of air quality trends as well as comparisons to other European cities.

The data in this thesis are measured with European reference methods (or the equivalent) or with the methods determined in the international research programme in question. The set of pollutants is determined by the availability of long-term time series and is restricted to the ones studied in **Papers I**–**V**. Trends of atmospheric heavy metals at a subarctic (Pallas) site during 1996–2018 were presented in a recent paper by Kyllönen et al. (2020).

A summary of the monitoring data included in the original papers of this thesis is shown in Table 1 and the site map in Figure 2.

Table 1. List of the monitoring sites, the chemical species and PM mass size fractions included from each site in **Papers I–V**

Network/ City	Municipality/ Site	NO_2/ NO_x	O_3	SO_2	CO	SIA[1]	WSI[2]	TM[3]	PAH[4]	POP[5]	$PM_{2.5}$	PM_{10}
FMI	Ilomantsi		**I**									
	Raja-Jooseppi		**I**	**I**								
	Utö	**I**	**I**	**I**		**I**						
	Lammi Evo		**I**									
	Pallas		**I**	**I**					**III**	**III**		
	Oulanka		**I**	**I**		**I**						
	Virolahti	**I,V**	**I,V**	**I,V**		**I,V**	**V**	**V**	**V**		**V**	
	Ähtäri	**I**	**I**	**I**								
Harjavalta	Kaleva			**I**								
	Pirkkala			**I**								
	Torttila			**I**								
HSY	Espoo Leppävaara2	**IV**										**IV**
	Espoo Luukki	**I,II**	**I,II**	**I**								**IV**
	Helsinki Kallio1	**IV**										**IV**
	Helsinki Mannerheimint	**II**	**II**									
	Helsinki Töölö	**I,II**	**I,II**		**I**							**I,IV**
	Helsinki Vallila	**I**		**I**	**I**							**I,IV**
	Vantaa Tikkurila2		**I**									
	Vantaa Tikkurila3	**I**			**I**							**I,IV**
Hämeenlinna	Raatihuoneenkatu	**IV**										**IV**
Imatra	Imatra Rautionkylä[6]	**I,IV**		**I**								**I,IV**
	Imatra Teppanala											**I**
	Imatra Mansikkala	**IV**		**I**								**IV**
	Lappeenranta Keskusta	**IV**										**IV**
	Lappeenranta Lauritsala	**I**										
	Lappeenranta Tirilä			**I**								
IVL	Råö (Sweden)								**III**	**III**		
Jyväskylä	Lyseo	**I**		**I**								**I,IV**
Kajaani	Keskusta	**I**										**IV**
Kokkola	Keskusta	**IV**										**IV**
Kotka	Kirjastotalo	**I,IV**										**IV**
Kouvola	Keskusta	**I**										**I,IV**
	Valkeala Lappakoski			**I**								
Kuopio	Kasarmipuisto				**I**							
	Keskusta	**IV**										**IV**
Lahti	Kisapuisto	**I**										
	Vesku	**I**			**I**							
Lohja	Nahkurintori	**IV**										**IV**
Oulu	Keskusta	**I**			**I**							**I,IV**
	Pyyköjärvi	**I**										**I,IV**
	Nokela			**I**								
Pietarsaari	Bottenviksvägen			**I**								**IV**
Pori	Itätulli	**IV**										**IV**
	Lampaluoto			**I**								
Neste	Porvoo Mustijoki	**I**		**I**								
Rauma	Sinisaari			**I**								
Seinäjoki	Vapaudentie	**I**										

Network/ City	Municipality/ Site	NO_2/ NO_x	O_3	SO_2	CO	SIA[1]	WSI[2]	TM[3]	PAH[4]	POP[5]	$PM_{2.5}$	PM_{10}
Tampere	Lielahti[6]	**I**										
Turku	Naantali Keskusta	**IV**										**IV**
	Raisio Keskusta	**I**										**I,IV**
	Raisio Kaanaa			**I**								
	Turku Kauppatori	**I**										**I,IV**
Varkaus	Pääterveysasema	**IV**										**I,IV**

[1] Secondary inorganic aerosols: SO_4^{2-} (p), NO_3^- (g+p), $NH4^+$ (g+p)
[2] Water-Soluble Inorganic Ions: Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-
[3] Trace Metals: Hg (g), Al, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, V, Zn
[4] Polycyclic aromatic hydrocarbons (g+p): Phe, Ant, Fl, Pyr, BaA, Chr, BbF, BkF, BaP, DhA, BP, IcP
[5] Persistent organic pollutants (g+p): polychlorinated biphenyls (seven PCB congeners), organochlorine pesticides (α- and γ-HCH, chlordanes, DDT, DDE, DDD)
[6] no NO_x

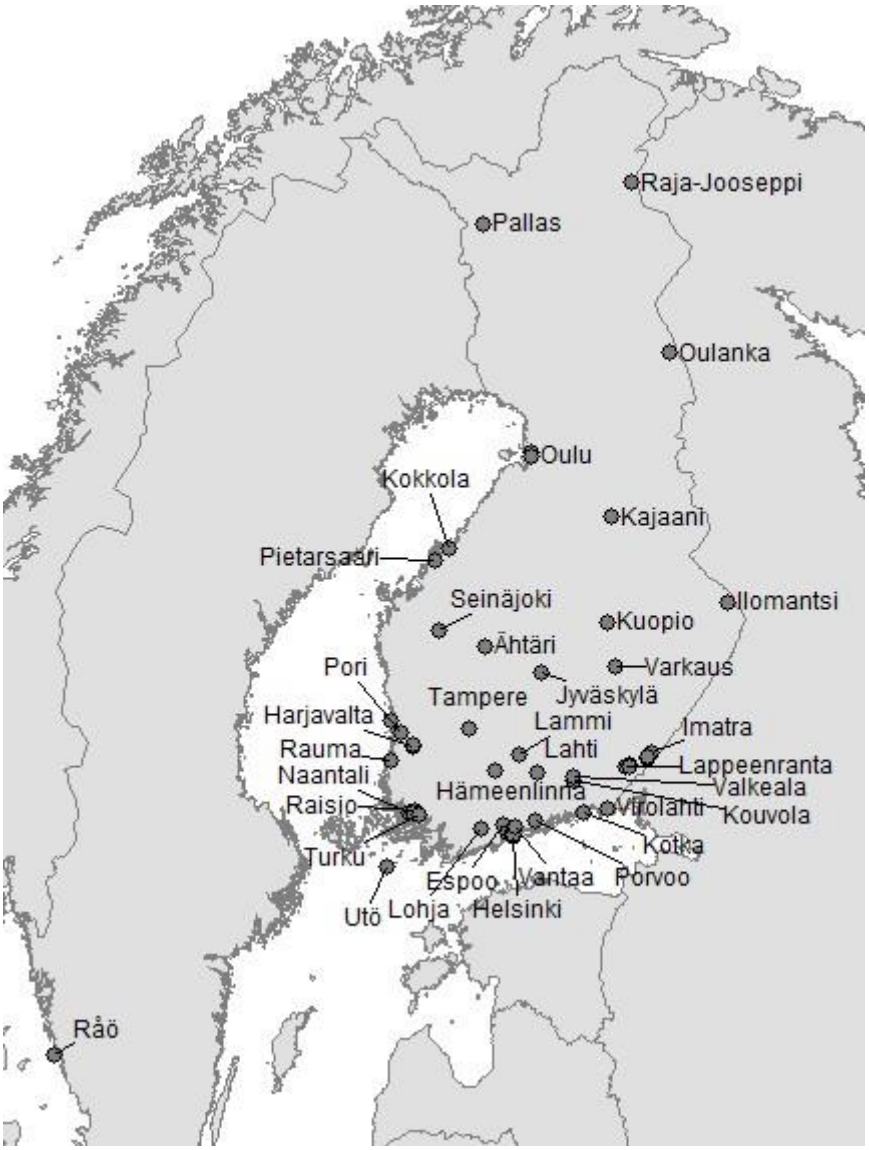

Figure 2. Locations of the air quality monitoring sites in **Papers I**–**V**.

3 Data analysis method

3.1 Generalised least squares (GLS) regression with ARMA errors

The generalised least squares regression with ARMA errors (GLS-ARMA) method is essential time series analysis method in **Papers I** and **III**.. A light methodological background and practical implementation of the method is presented here. Chapters 3.1.1 and 3.1.2 are based on the general time series analysis framework presented, e.g. by Hamilton (1994) and Brockwell and Davies (2002). The application of the method is demonstrated in chapter 3.1.3.

3.1.1 Generalisation of ordinary least squares regression

In standard linear regression, the errors (or residuals of the fit) are assumed to be independent and identically distributed (iid) (Hamilton, 1994). In an air quality time series, this assumption is typically violated due to cyclic dependencies (e.g. diurnal, seasonal) in the observed data. Generalised least squares (GLS) regression with ARMA errors extends the ordinary least squares (OLS) estimation of the linear model by providing for possible correlations between different residuals and for possibly unequal residual variances (Brockwell and Davis, 2002; Hamilton, 1994).

Let us first consider the ordinary least squares (OLS) regression model in matrix form,

$$y = X\beta + \varepsilon \quad , \qquad \text{(Eq. 1)}$$

where y is the vector of n observations of some random variable Y at times $t = 1,\ldots, n$, (y is a nx1 matrix). X is the design matrix of the explanatory variables (non-stochastic); its columns can be, e.g. any function of time t. β is the vector of regression coefficients and ε is the vector of random errors (nx1). The goal is to obtain the OLS estimators $\hat{\beta}_{\mathrm{OLS}}$ of vector β, so that

$$y = X\hat{\beta}_{\mathrm{OLS}} + e \ , \qquad \text{(Eq. 2)}$$

where e is the residual vector (nx1) of the OLS fit.

The OLS estimator $\hat{\beta}_{\mathrm{OLS}}$ minimises the sum of squared residuals (prime denotes the transpose)

$$\sum e_t^2 = e'e = (y - X\hat{\beta}_{\mathrm{OLS}})'(y - X\hat{\beta}_{\mathrm{OLS}}) \ . \qquad \text{(Eq. 3)}$$

Expanding this out, differentiating with respect to $\hat{\beta}_{\mathrm{OLS}}$ and setting to zero, we find that

$$\hat{\beta}_{\mathrm{OLS}} = (X'X)^{-1}X'y, \qquad \text{(Eq. 4)}$$

i.e. the OLS coefficients can be calculated as a function of data matrix X and observation vector y.

To find the "significance" of the trend, we need to determine the standard error (SE) of the slope coefficient, i.e. one of the terms of the OLS estimator $\hat{\beta}_{\mathrm{OLS}}$. For this, we need to make use of the iid assumptions of the residuals of this OLS fit; $e_t \sim \mathrm{N}(0, \sigma^2 I)$, where σ^2 is the true error variance and I is the identity matrix.

The variance of any random variable is the expectation of the squared deviation of its expected value. For the random vector $\hat{\beta}_{\mathrm{OLS}}$,

$$\mathrm{Var}\,\hat{\beta}_{\mathrm{OLS}} = \mathrm{E}\left[\left(\hat{\beta}_{\mathrm{OLS}} - \mathrm{E}(\hat{\beta}_{\mathrm{OLS}})\right)\left(\hat{\beta}_{\mathrm{OLS}} - \mathrm{E}(\hat{\beta}_{\mathrm{OLS}})\right)'\right] \quad , \qquad \text{(Eq. 5)}$$

where E stands for the expected value.

After some matrix calculus (not shown), we get

$$\mathrm{Var}\,\hat{\beta}_{\mathrm{OLS}} = (X'X)^{-1}X'\mathrm{E}(ee')X(X'X)^{-1} \quad . \qquad \text{(Eq. 6)}$$

The term $\mathrm{E}(ee')$ is the covariance matrix of e which, under OLS assumptions ($e_t \sim \mathrm{N}(0, \sigma^2 I)$ is $\mathrm{E}(ee')=\sigma^2 I$ and (Eq. 6) reduces to

$$\mathrm{Var}\,\hat{\beta}_{\mathrm{OLS}} = \sigma^2(X'X)^{-1} \text{ and SE } \hat{\beta}_{\mathrm{OLS}} = \sigma\sqrt{(X'X)^{-1}} \quad . \qquad \text{(Eq. 7)}$$

The true error variance σ^2 is unknown, but it is estimated based on the regression residuals (mean squared error)

$$\hat{\sigma}^2 \sim \frac{1}{n-2}\sum_{t=1}^{n} e_t^2 \quad . \qquad \text{(Eq. 8)}$$

This is where the OLS method goes: statistical packages give the regression parameters from (Eq. 4) and estimates of the standard errors from (Eq. 7) and (Eq. 8).

If residual errors are not independent and identically distributed (iid), we need a specification of how the dependence varies with time. This dependence can be parameterised in the variance–covariance matrix and can be fitted by generalised least squares (GLS). In this case, the residual covariance matrix term in (Eq. 6) is $\mathrm{E}(ee') = \mathrm{Cov}(e) = \Gamma$, where Γ is any matrix so that $e_t \sim \mathrm{N}(0, \Gamma)$. Different diagonal entries in Γ correspond to non-constant error variances, while nonzero off-diagonal entries correspond to correlated errors.

The GLS solution lies in transforming the original linear regression model to $y^* = X^*\beta + e^*$ so that we can then estimate the $\hat{\beta}$ matrix by OLS on the transformed variables, i.e.

$$\mathrm{Cov}(e^*) = \mathrm{E}(e^* e^{*\prime}) = \sigma^2 I \quad . \qquad \text{(Eq. 9)}$$

For this, we define matrix T so that

$$T'T = \sigma^2 \Gamma^{-1} \text{ and hence (not shown) } T\Gamma T' = \sigma^2 I \; . \qquad \text{(Eq. 10)}$$

If we now multiply (Eq. 2) by matrix T, we get

$$Ty = TX\beta + Te \quad , \qquad \text{(Eq. 11)}$$

a regression equation with coefficient vector β, data vector Ty, design matrix TX, and error vector Te. The covariance matrix of the transformed error term will be

$$\begin{aligned} \text{Cov}\,(Te) &= \text{E}(Te(Te)') \\ &= T\text{E}(ee')T' \\ &= T\Gamma T' = \sigma^2 I\,, \end{aligned} \qquad \text{(Eq. 12)}$$

so the transformed model (Eq. 11) has uncorrelated, zero mean errors, each with variance σ^2. The estimator of β in terms of Ty can be obtained by applying an OLS estimation to the transformed regression equation (Eq. 11). This gives the generalised least squares (GLS) estimator $\hat{\beta}_{\text{GLS}}$

$$\hat{\beta}_{\text{GLS}} = \left(X'\Gamma^{-1}X\right)^{-1}X'\Gamma^{-1}y \qquad \text{(Eq. 13)}$$

with the covariance matrix

$$\text{Cov}\,\hat{\beta}_{\text{GLS}} = \left(X'\Gamma^{-1}X\right)^{-1} \quad . \qquad \text{(Eq. 14)}$$

The error covariance matrix Γ is not known, and it must be estimated from the data along the regression coefficients, e.g. by the maximum likelihood method; for this, further restrictions are needed due to too many elements in Γ ($n(n+1)/2$).

In our case (and more generally, in time series data), after transformations, the error e_t terms are already stationary. But they are serially correlated, and the covariance of two errors depends only upon their separation in time: voilà, this is exactly what ARMA processes can provide.

3.1.2 Autoregressive moving-average (ARMA) models

Autoregressive moving-average (ARMA) models (e.g. Hamilton, 1994; Brockwell and Davies, 2002) provide a commonly used description of a stationary time series e_t in terms of two polynomials: one describes the value of the time series as a function of its lagged values (this is the so-called autoregressive (AR) part), and the second describes the effect of lagged random terms (the moving average (MA) part).

The ARMA(p,q) process is defined as

$$e_t = \phi_1 e_{t-1} + \ldots + \phi_p e_{t-p} + Z_t + \theta_1 Z_{t-1} + \ldots + \theta_q Z_{t-q} \quad , \qquad \text{(Eq. 15)}$$

where ϕ_i, i=1,…, p and θ_j, j=1,..., q are constants and $Z_t \sim \text{WN}(0, \sigma^2)$; p is the number of autoregressive parameters, and q is the number of moving-average parameters (Brockwell and Davis, 2002).

The next step is to find the most satisfactory ARMA(p, q) model to represent the residual series e_t in order to eliminate the remaining dependencies and finally get the white noise error terms. The parameters ϕ_i, i=1,…, p and θ_j j=1,..., q are estimated with the maximum likelihood method. The selection of the parameters p, q is based on the bias-corrected Akaike information criterion (AICC) (Akaike, 1973; Hurvich and Tsai, 1989). All combinations of p and q (within the pre-set ranges) are walked through, and the combination of p and q values that returns the maximum likelihood model with the smallest AICC value is chosen.

So, we build the error covariance matrix with the ARMA processes and estimate the unknown parameters ($\hat{\phi}_i$, $\hat{\theta}_j$, $\hat{\beta}_{\mathrm{GLS}}$) using the maximum likelihood method.

3.1.3 Application of GLS-ARMA in AQ trend analysis

The purpose of trend testing is to determine whether the values of a random variable generally increase (or decrease) over some period in statistical terms. The concentrations of airborne pollutants are our random variables C_t, and the observed concentrations c_t are the realisation of these variables. The time resolution of our original data (**Papers I** and **III**) varied from one hour to one week, but all data have been averaged to monthly means, resulting in a time series with consecutive measurements taken at equally spaced time intervals; the independent variable, time t, gets the values from 1 to n. The Windows-based computer package ITSM2000 (B and D Enterprises, Inc. 7.3 (Professional) Oct 1, 2005) was used in the computations.

The application of the method is demonstrated with the help of diagnostic plots from an example: the monthly time series of NO_2 concentrations at Oulu Keskusta station in 1994–2007 (**Paper I**) with the extension of new data up to the end of 2015 (n = 168 + 96 monthly values) (Figure 3).

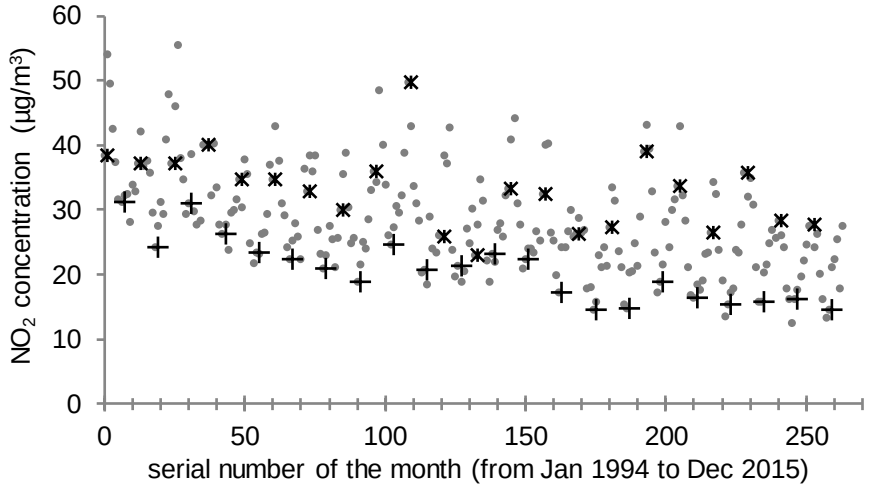

Figure 3. Scatterplot of the monthly mean concentrations of NO_2 at Oulu Keskusta station in 1994–2015. January and July values are denoted with stars and crosses, respectively.

The trend analysis begins with the examination of the detected time series (c_t) plot. The plot (Figure 3) reveals that there are no obvious exponential changes or heteroscedasticity (strongly increasing or decreasing variance), which suggests that a linear model could be appropriate. We

also see seasonal variation (concentrations are systematically higher in winter months) and trend (decreasing mean) patterns (Figure 3).

Before introducing the GLS-ARMA, we do a preliminary transformation of the observed time series c_t; seasonal adjustment is needed as most of the ambient air pollutants have a strong seasonal variation. This is done here by applying a moving average filter to c_t, calculating the monthly indices and subtracting them from the original time series c_t (see details in **Paper I**).

Now, the deseasonalised time series c_t^{ds} becomes our dependent variable, and we fit an OLS line, $c_t^{\mathrm{ds}} = \hat{\beta}_1 + \hat{\beta}_2 t + e_t$, where $\hat{\beta}_1$ and $\hat{\beta}_2$ are the regression parameters and e_t are the residuals of this fit to this remaining time series (Figure 4a). Now, we have the OLS approximation of the regression parameters (Eq. 4) and their standard errors (in parentheses) (Eq. 7 and (Eq. 8),

$$\hat{\beta}_1 = 34.88(0.5164) \text{ and } \hat{\beta}_2 = \text{-}0.05175(0.003378) \quad . \qquad \text{(Eq. 16)}$$

It is time to study the residuals e_t of this fit. In a valid regression model, the residuals need to be independent and identically distributed (iid), i.e. normally distributed with a zero mean and constant variance and not serially autocorrelated. So, the residuals must be investigated for the normality, homoscedasticity and autocorrelation to ensure the appropriateness of our linear regression model. This can be done using descriptive plots and formal statistical tests.

Figure 4b suggests that residuals e_t now have ~zero mean and constant variance; both Breusch–Pagan and White tests suggest homoscedasticity with *p*-values 0.084 and 0.225, respectively. Figure 4c shows that residuals are moderately close to normal distribution; the Kolmogorov–Smirnov test does not reject normal distribution, while the Jarque–Bera and Shapiro–Wilk tests reject normality assumption. We conclude that the deviation from normality is so small that we continue with the selected model.

However, the sample autocorrelation function in Figure 4d displays high autocorrelations of the residuals e_t up to a lag of five months. Hence, our residual sequence e_t is not (yet) iid, and we must introduce additional terms – ARMA processes – into our model to account for the autocorrelation.

We can then use the ARMA processes to model the remaining dependences (autocorrelation) of the residuals and finally use them (ARMA processes) in conjunction with the trend to re-estimate the parameters (and standard errors) of the original model.

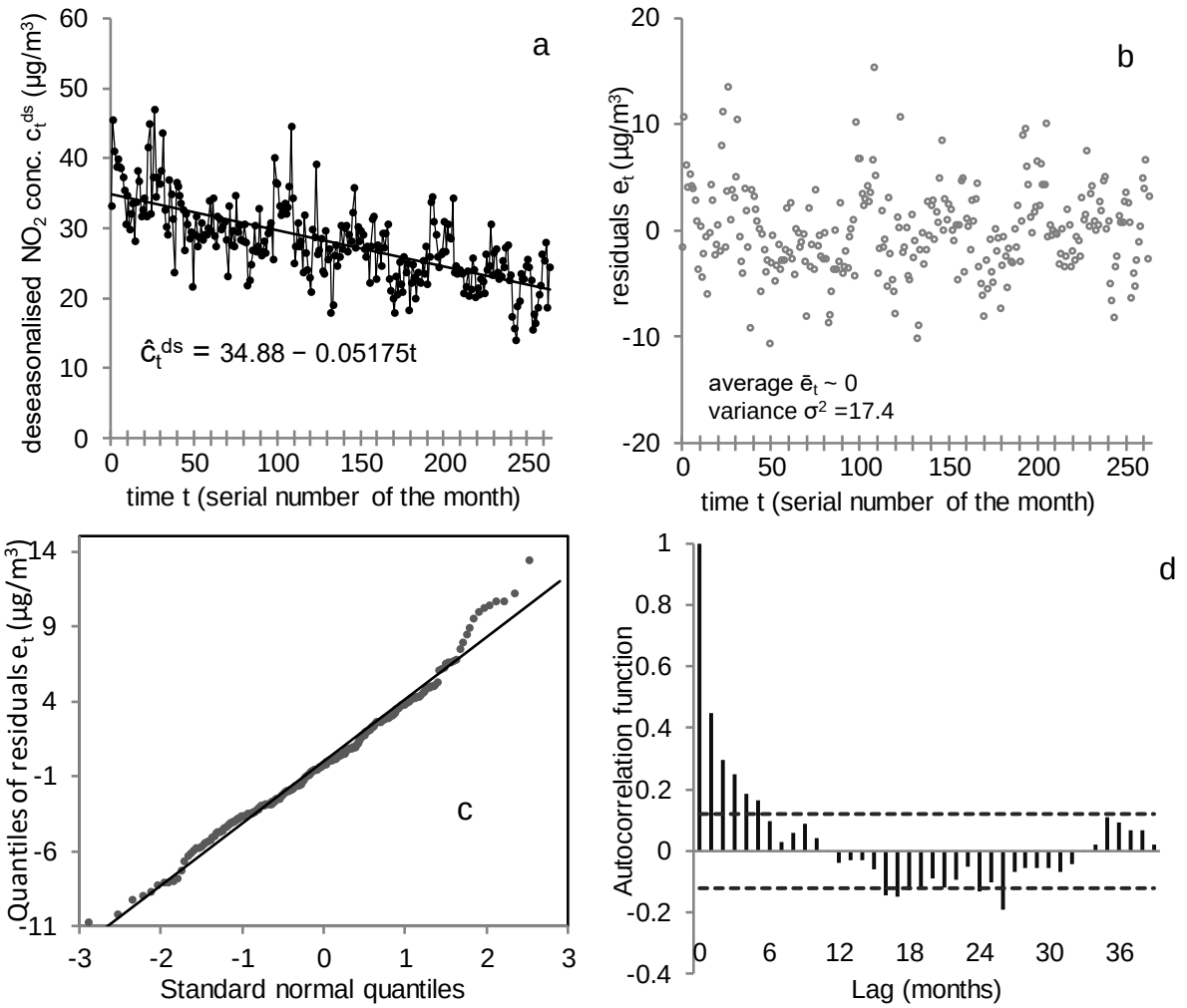

Figure 4. (a) seasonally adjusted c_t^{ds} together with the fitted OLS line $\hat{c}_t^{\text{ds}}$, (b) residual e_t time series, (c) a quantile-quantile plot of a the residuals e_t versus the standard normal quantiles with an OLS line as a reference, and (d) the autocorrelation plot of the residuals e_t (the horizontal lines are the 95% confidence level bounds).

Next, we fit the maximum likelihood ARMA(p, q) values for all p and q (in a specified range) and select from these the model with the smallest AICC value. In ITSM software, the maximum likelihood estimation of the ARMA(p, q) model is based on the innovation algorithm (see Brockwell and Davies, 2002).

In our example, the minimum AICC model for the OLS regression residuals e_t turns out to be an ARMA(1,1) model with $\hat{\phi}_1 = 0.7331$, $\hat{\theta}_1 = -0.3747$. Thus, our new model for c_t^{ds} is

$$\begin{aligned} c_t^{\text{ds}} &= 34.88(0.5164) - 0.05175(0.003378)t \\ &+ 0.7331(0.09079)e_{t-1} + Z_t - 0.3747(0.1279)Z_{t-1} \end{aligned}, \qquad \text{(Eq. 17)}$$

where $Z_t \sim \text{WN}(0, 13.564996)$.

Next, both the ARMA parameters ($\hat{\phi}$ and $\hat{\theta}$) and the regression coefficients ($\hat{\beta}$) are reestimated with the maximum likelihood estimation. MLE is repeated until all parameters are

stabilised. After several iterations, we arrive at the final model with the following maximum likelihood estimators (the standard errors in parentheses):

$$c_t^{ds} = 34.92(1.044) - 0.05177(0.006803)t + 0.7332(0.09076)e_{t-1} + Z_t - 0.3748(0.1279)Z_{t-1} \quad \text{(Eq. 18)}$$

and $Z_t \sim \text{WN}(0, 13.5648)$.

The residuals of this GLS-ARMA model are (Brockwell and Davis, 2002)

$$\hat{e}_t^{\text{GLS-ARMA}} = (e_t - \hat{e}_t)/\sqrt{r_{t-1}} \ , \quad \text{(Eq. 19)}$$

where $\hat{e}_t$ is a one-step predictor of e_t, $r_{t-1} = \text{E}(e_t - \hat{e}_t)^2/\sigma^2$ and σ^2 is the white noise variance of the fitted model.

The next step is to check the model for goodness of fit. The statistical confidence intervals for the zero autocorrelation (null hypothesis) are estimated as follows: For iid noise ~N(0, σ^2), the sample autocorrelations are approximately iid N(0,1/*n*) for large *n* (see, e.g. Brockwell and Davis, 2002). Hence, approximately 95% of the autocorrelations should fall between the bounds $\pm 1.96/\sqrt{n} = \pm 0.12$ (since 1.96 is the 0.975 quantile of the standard normal distribution, and *n* is 264).

While the OLS residuals e_t failed to meet the iid assumptions (due to autocorrelation; see Figure 4d), the ACF of the GLS-ARMA residuals $\hat{e}_t^{\text{GLS-ARMA}}$ shows that all but one of the 40 sample autocorrelations calculated fall between the 95% bounds, and there is no cause to reject the fitted model based on the autocorrelations (see Figure 5). (Other tests of randomness were comparable to previously presented OLS residual tests.)

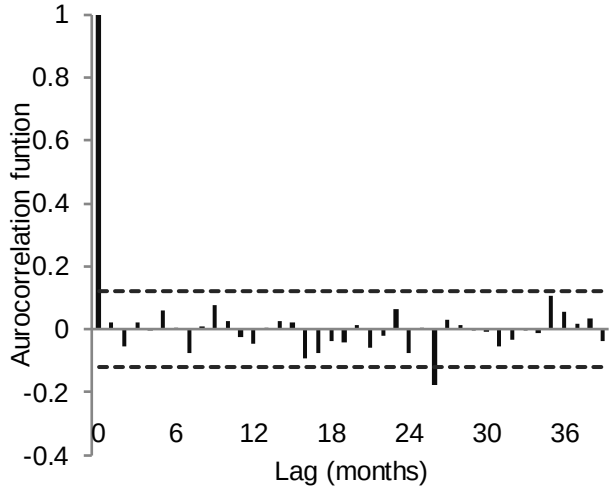

Figure 5. ACF plot of the residuals $\hat{e}_t^{\text{GLS-ARMA}}$. The horizontal lines are the 95% confidence level bounds.

Next, we decide whether there is a statistically significant trend. The null hypothesis H0 (no trend, i.e. slope is zero) is tested against the alternative hypothesis H1 where there is a trend (slope $\neq 0$).

Estimated 95% confidence bounds for the slope using this GLS-ARMA estimate are slope ± 1.96 x standard error = −0.0518 ± 1.96 x 0.0068 = (−0.0651, −0.0384). Zero does not belong to this interval, and we reject the null hypothesis and conclude that there is a significant decreasing trend in our time series at a 95% confidence level.

The final trend equation, with standard errors, is

$$\hat{c}_t^{\mathrm{ds}} = ((34.9 \pm 1.0) - 0.0518 \pm 0.0068)t \quad . \qquad \text{(Eq. 20)}$$

If we had settled for the OLS (Eq. 16) instead of GLS in this example, the standard error estimates of the slope and intercept would have been about half of the resulting GLS estimates (Eq. 20). So, ignoring the autocorrelation of the residuals would have led to an underestimation of the standard errors – and an overestimation of the significance. But on the other hand, both methods would have suggested a decreasing trend even at a 99.9% confidence level.

In **Paper I**, the slopes and significance of the trends calculated with the GLS-ARMA method were compared with three other trend analysis methods, i.e. deseasonalised monthly OLS regression, annual OLS regression and annual non-parametric Sen's slope and the Mann-Kendall significance test (Salmi et al., 2002).

4 Results

In **Paper I**, the atmospheric concentration time series of the main inorganic gases, secondary inorganic ions and PM_{10} mass from 42 Finnish rural, urban or industrial AQM sites (Table 1) between 1994 and 2007 were analysed with GLS-ARMA for potential trends. Analogously, in **Paper III**, the long-term temporal trends of 27 persistent organic pollutant species from 1996 to 2011 from two Scandinavian rural background sites (Råö and Pallas) were investigated (Table 1). **Paper II** accompanies **Paper I** by focusing on the urban NO_x anomaly in more detail. **Papers IV** and **V** are case studies that provide supportive material for the AQ trend analysis.

4.1 Sulphur dioxide (SO_2) and sulphate (SO_4^{2-}) trends

In Europe, the abatement of long-range transboundary air pollution started under the United Nations Economic Commission for Europe (UNECE) in 1979 with the signature of the Convention on Long-range Transboundary Air Pollution (CLRTAP) by 32 governments and the European Community. The first sulphur protocol under the CLRTAP was adopted in 1985 (UNECE, 1985). The acidification of ecosystems was the main driving force for SO_2 emission abatement in Europe in the 1980s (see, e.g. Kauppi et al., 1990). The timing and implementation of emission reductions varied between countries, but the major reduction measures generally included the shift to cleaner, low-sulphur fuels, the adoption of flue gas desulphurisation and advances in technologies and industrial processes (Crippa et al., 2016; Vestreng et al., 2007). Rafaj et al. (2014) emphasise the role of reduced energy intensity (energy/GDP) in SO_2 emission reduction. In Finland, SO_2 emission reduction was enhanced by a comprehensive legislative package approved by the government in 1987, which limited the maximum allowed sulphur content of light and diesel oil and coal as well as SO_2 emissions in heavy oil and coal-fuelled power plants, the pulp industry, sulphuric acid plants and oil refineries (e.g. Huutoniemi et al., 2006).

As a result, in Finland as in the whole of Europe, SO_2 emissions have been declining for over 40 years now (Vestreng et al., 2007; Crippa et al., 2016; EEA, 2018). These reductions of SO_2 emissions were reflected in the decreasing ambient SO_2 (-3% to -8%/yr) concentrations at 15 sites during the study period, 1994–2007 (**Paper I**). Comparable downward trends have been reported in both urban and rural environments in Europe (Jones and Harrison, 2011; Henschel et al., 2013; Guerreiro et al., 2014; Crippa et al., 2016; Tørseth et al., 2012; Colette et al., 2016) and in Finland (Ruoho-Airola et al., 2015; Riuttanen et al., 2013; Nieminen et al., 2014). Aas et al. (2019) report that an average reduction in SO_2 concentrations was -5.03%/yr (STD=2.04; *n*=20) in European background areas between 1980 and 1990 and -7.56%/yr (1.81; 43) and -4.23%/yr (3.17; 51) in 1990−2000 and 2000−2010, respectively.

Figure 6 summarises all the Finnish SO_2 monitoring data available at the FAQDMS since 1987. After removing years with data captures below 70%, the data set contained 1406 individual annual values from 201 separate sites. Caution is required when averaging data from a monitoring network containing multiple time series of variable durations. Movement, opening and closing of monitoring sites may introduce biases into the average trend. However, an aggregate view of the overall changes in concentrations offers an interesting perspective.

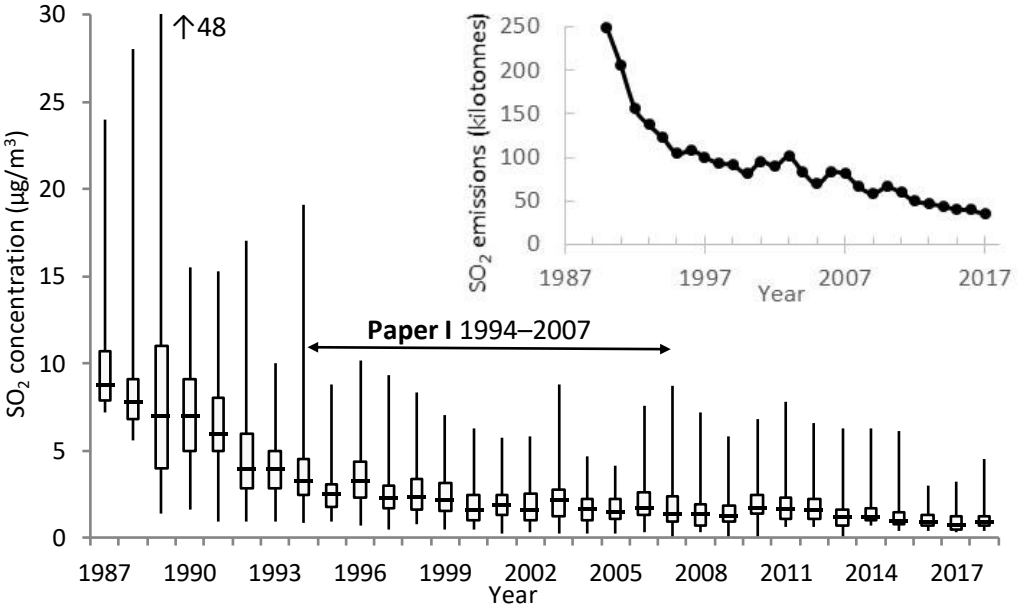

Figure 6. Distribution of annual mean concentrations of SO_2 measured at the Finnish monitoring sites in 1987−2018. At least a 70% annual data capture was required for each site and year. For each year, the lowest, highest and median values together with the 25th and 75th percentile values (box) are shown. The number of sites included varies from 6 in 1987 to 76 in 1993, and after that, there is a gradual decrease to 36 sites in 2018. A total of 1406 annual values were included. The subplot shows the development of the annual SO_2 emissions in Finland (SYKE, 2019a).

Figure 6 demonstrates the steepest decline in SO_2 concentrations in the late 1980s and early 1990s and a moderate decrease in 1994–2007 (**Paper I**). Overall, SO_2 concentration levels are lower now than 10 years ago, but the decline has not been as straightforward as anticipated in **Paper I**. The high end of the SO_2 concentration distribution nowadays (2016–2018) is occupied by industrial monitoring sites in Harjavalta, Imatra, Kokkola, Lappeenranta, Porvoo and Raahe. The annual means of SO_2 have varied (2016–2018) between 2–5 µg/m^3, and hourly values have occasionally reached maximums of 1000 µg/m^3. European air quality standards are not exceeded anywhere.

In Finnish background areas, the SO_2 concentration level is around 0.5–1 µg/m^3. Nowadays (2016–2018) in Finnish background areas, the lowest SO_2 concentrations (hourly, daily, annual) are observed at Utö, and the highest are in northernmost Lapland (Raja-Jooseppi and Kevo) under the influence of the ongoing and still partly obscure emissions in the Kola Peninsula (e.g. Prank et al., 2010). On the other hand, the worst SO_2 episode of the last decade in Lapland was due to the volcanic plume coming from the Icelandic Holuhraun fissure eruption in September 2014 (Ialongo et al., 2015). The highest SO_2 hourly values were 188 µg/m^3, 172 µg/m^3 and 88 µg/m^3 at Pallas/Sammaltunturi, Oulanka and Raja-Jooseppi, respectively.

As of 1 January, 2015, the stricter Sulphur Emission Control Area (SECA) regulations (IMO, 2008; EU, 2016) came into force in the North Sea and the Baltic Sea, reducing the maximum sulphur content allowed in marine fuels from 1.0% to 0.1%. By means of three chemical transport models, Karl et al. (2019) estimated that in Finnish southern coastal areas in the pre-SECA conditions (2012), the contribution of ship emissions to the SO_2 concentrations was around 0.2–0.5 µg/m^3. Indeed, at Utö, the difference between the observed daily SO_2 means in the pre-SECA period, 2012–2014 (0.72 ±0.70 µg/m^3), and the post-SECA period, 2015–2017

(0.4 ±0.4 µg/m^3), was -0.3 µg/m^3 ($p<0.0001$). A similar stepwise reduction is also clearly visible in the SO_4^{2-} concentrations (Figure 7).

In the atmosphere, gaseous SO_2 becomes oxidised to form particulate sulphate (SO_4^{2-}). Figure 7 assembles the monthly time series of SO_4^{2-} concentrations at FMI's three monitoring stations between 1994 and 2018. At the beginning of 2007, the change of the main ions' sampling method was introduced (Ruoho-Airola et al., 2015), so the demonstrative OLS lines are drawn in two parts (Figure 7). The decrease in concentrations has also continued after 2006, the end of the study period in **Paper I**.

For SO_4^{2-}, Aas et al. (2019) gave a European average trend of -2.67%/yr (2.03; 36) for the 2000–2015 period, which is in line with the Finnish observations (-2% to -3%/yr in 1994–2006) reported in **Paper I**.

Since the late 1970s, the European-wide co-operation to tackle acid rain and its impacts on ecosystems has been followed by a number of policies (both under the EU and the UN) that either directly or indirectly act to reduce SO_2 emissions. Through these actions, SO_2 emissions, acid rain and ecosystem impacts have decreased (see, e.g. Grennfelt et al., 2019). Simultaneously, marked sulphur-related air quality improvements have been achieved, and the protection of human health from air pollution has substantially benefitted.

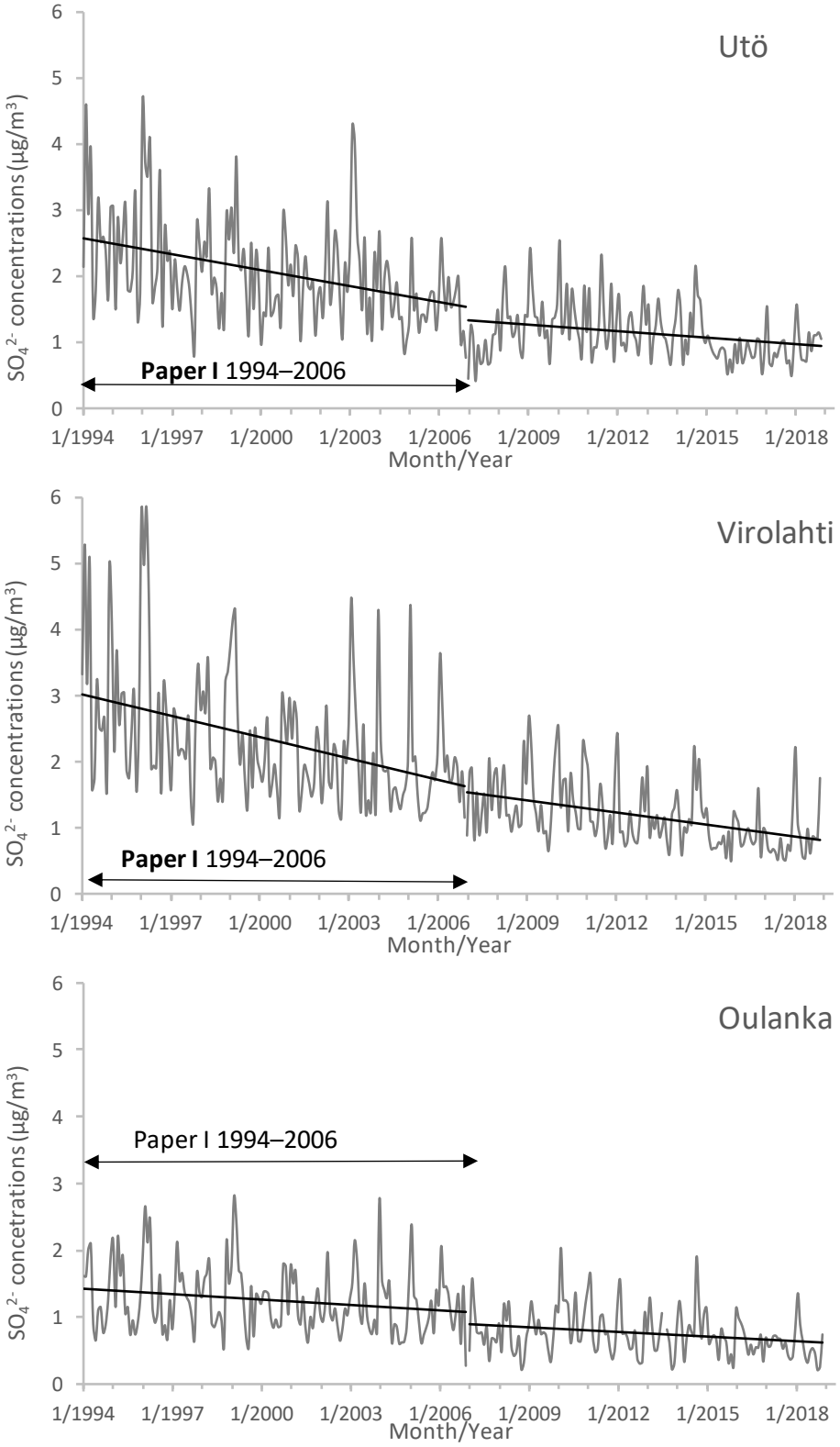

Figure 7. Monthly time series of SO_4^{2-} concentrations at Utö, Virolahti and Oulanka in 1994–2018. OLS lines are drawn for visualisation.

4.2 Nitrogen oxides (NO_x) trends

As in the case of SO_2, the control of NO_x ($NO+NO_2$) emissions was agreed under the UNECE in 1988 (UNECE, 1988); only stabilisation was mandated. In Europe, a major step

forward in limiting road transport emissions was the introduction of the first of the so-called Euro emission standards, Euro 1, in 1992. In practice, then, a three-way catalytic converter (TWC) became mandatory in all new petrol vehicles. TWCs reduce the emission of the three primary pollutants NO_x, CO and VOCs. The introduction of TWCs to petrol cars has reduced these emissions significantly in Finland (Table 2) and in Europe (Guerreiro et al., 2014; EEA, 2018).

Table 2. Annual anthropogenic emissions of NO_x, CO and VOC in Finland in 1994 and 2017 (SYKE, 2019b).

	Finnish anthropogenic emissions (1000 tonnes)				Reduction	
	1994		2017		%	
	total	traffic	total	traffic	total	traffic
NO_x	293	179	130	58	56	68
CO	700	490	379	141	46	71
VOC	208	101	89	15	57	85

In **Paper I,** the concentration time series of NO_2 and NO_x from 22 and 20 monitoring sites during 1994–2007 (Table 1) were studied. Half of the NO_2 time series displayed a statistically significant decreasing trend, while as much as 70% of the studied NO_x time series were decreasing. The typical annual reductions were 1–2% and 3–5% for NO_2 and NO_x, respectively, typically occuring in traffic-influenced environments. During the same period, the national total NO_x emissions and road traffic emissions were reduced by 2% and 5%, respectively, so the decrease in NO_x concentrations was generally consistent with the national NO_x emission data (**Paper I**).

Figure 8 summarises the Finnish NO_2 and NO monitoring results up to 2018. Visually, the downward trends of NO_2 have continued throughout the 1994–2018 period. Statistically, the trends were decreasing (OLS, *t*-test, $p<0.001$) for all percentiles except the time series consisting of the highest annual means. Also, the post-2007 trends were decreasing ($p<0.05$) apart from the minimum time series. Analogously, NO concentrations (Figure 8) decreased throughout the whole period ($p<0.01$ for all studied percentiles), while the post-2007 trends were decreasing but were not statistically significant ($p>0.05$).

The air quality directive (EU, 2008) sets two limit values for NO_2 for the protection of human health: the NO_2 hourly mean value may not exceed 200 µg/m^3 more than 18 times in a year, and the NO_2 annual mean value may not exceed 40 µg/m^3; both are to be met from 2010 onwards. In Finland, the hourly limit value of NO_2 is not critical, but the annual limit value was exceeded beyond 2010 in the busiest street canyons in Helsinki. Since 2016, the annual limit value of NO_2 has not been exceeded at any monitoring station.

During 1994–2017, Finnish total and traffic NO_x emissions decreased by 56% and 68%, respectively (Table 2). At the same time, NO concentrations decreased by 80% (calculated from the medium values in Figure 8) and NO_2 concentrations by 50%. In the past twenty years or so, the NO_2 concentrations have been decreasing, albeit slower than NO_x emissions and NO concentrations, and nowadays, the air quality standards for the protection of human health are not exceeded (since 2016).

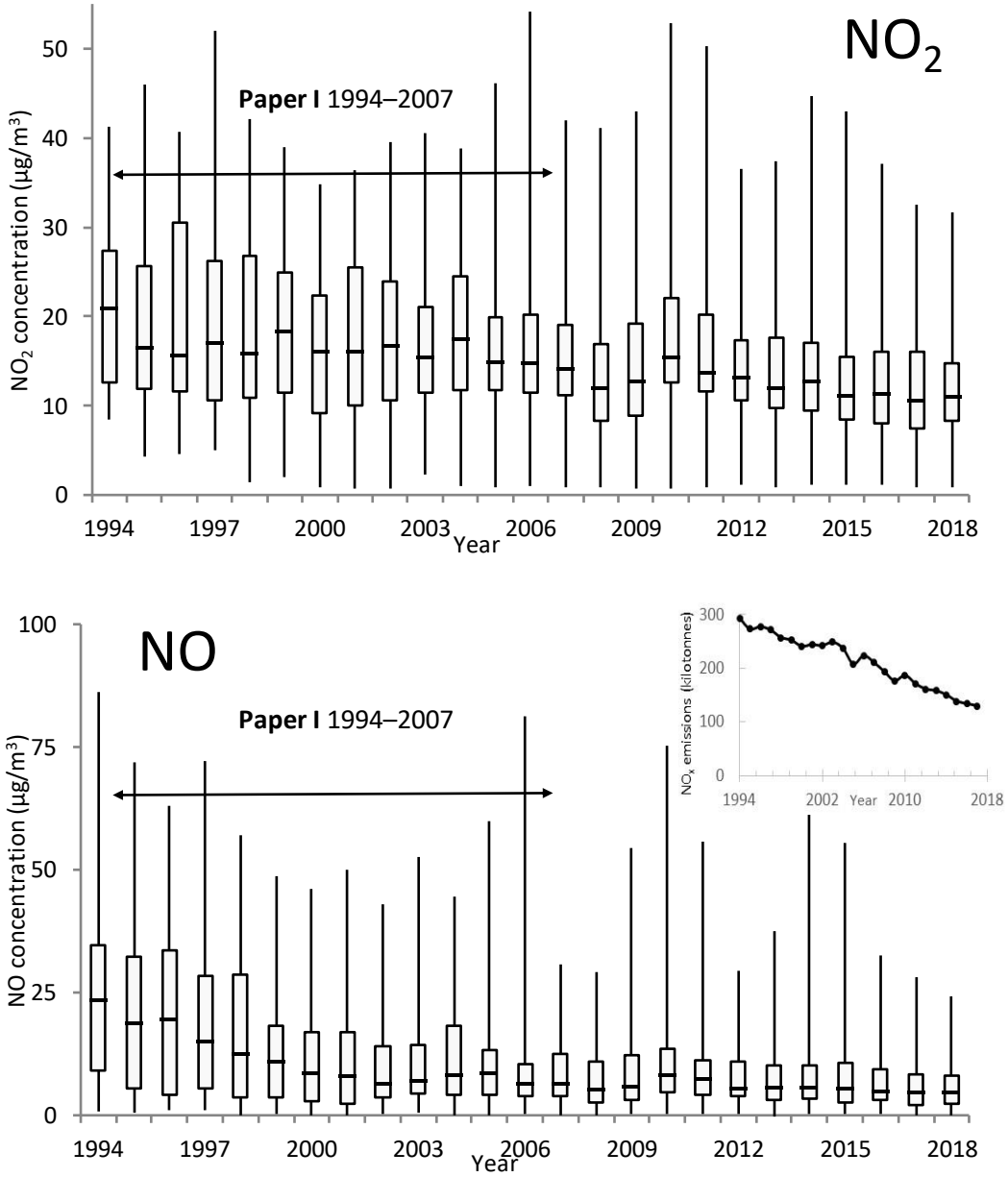

Figure 8. Distribution of annual mean concentrations of NO_2 and NO (lower) measured at the Finnish monitoring sites in 1994−2018. At least a 75% annual data capture was required for each site and year. For each year, the lowest, highest and median values together with the 25th and 75th percentile values (box) are shown. The number of sites included varies from 26 (NO 26) in 1994 to 59 (55) in 2018, the total number of sites being 209 (205). A total of 1326 (1271) annual values were included. The subplot shows the development of the annual NO_x emissions in Finland (SYKE, 2019b).

Primary NO_2 emission trend

Despite the considerable NO_x emission reductions, in many larger European cities, the urban traffic/roadside NO_2 concentrations have not decreased as expected or have stabilised or even increased, and NO_2 air quality standards are widely exceeded (e.g. Guerreiro et al., 2014; Henschel et al., 2015; Grange et al., 2017). This discrepancy has been attributed largely to the increasing usage of diesel vehicles in Europe and the increase in directly emitted tailpipe NO_2.

Already in the early 2000s, Carslaw and Beevers (2004) and Carslaw (2005) suggested that the detected increase in the ambient NO_2/NO_x concentration ratio in London is related to the increase in the NO_2/NO_x ratio in vehicular exhaust, especially in this London case, due to the increasing number of buses and cars fitted with catalytic diesel particulate filters (CDPF). These devices oxidise carbon monoxide and hydrocarbons and generate NO_2 to aid in the emissions control of diesel particulate. Since then, the increase in the urban NO_2/NO_x concentration ratio and the underlying causes of the phenomenon have been widely studied (Keuken et al., 2009; **Paper II**; Carslaw et al., 2011; Mavroidis and Chaloulakou, 2011; Keuken et al., 2012; Kurtenbach et al., 2012; Melkonyan and Kuttler, 2012; Carslaw and Rhys-Tyler, 2013; O'Driscoll et al., 2016; Carslaw et al., 2016; Degraeuwe et al., 2017; Wild et al., 2017; Olstrup et al., 2018; Casquero-Vera et al., 2019). Nowadays, it is well-established that the proportion of primary NO_2 (i.e. the NO_2/NO_x ratio) in diesel vehicle exhaust has increased over the past decade because of exhaust gas after-treatment technologies (CDPF and others).

Additionally, it has long been known (and addressed in, e.g. emission inventories; e.g. Ntziachristos et al., 2016) that the on-road real-driving NO_x emissions of vehicles – especially those of diesel vehicles – differ from the standardised approval emission tests. This discrepancy rose to the spotlight in 2015 when Volkswagen's "diesel defeat devices" were exposed (BMVI, 2016). In certain vehicles (belonging to the emission classes Euro 5 or Euro 6), the manufacturer had installed a specific engine control software that recognises if the vehicle follows a type approval test cycle in lab conditions (NEDC) and accordingly switches to a low-NO_x emission strategy. In real-life road operations, these vehicles run in a different mode, resulting in higher NO_x emissions. However, it quickly emerged that the problem was not limited just to Volkswagen, but most manufacturers had been "optimising" the performance of their vehicles to pass the type approval tests. Permissive testing procedures at the EU level and defective emissions control strategies (so-called "lawful defeat devices") were found to legally allow higher NO_x emissions on the road than in a laboratory setting (BMVI, 2016).

It should be noted that, by now, the EU has already adopted several measures to improve the type approval emission testing; e.g. real driving emissions (RDE) measurements of NO_x (but not NO_2) and particle number (PN) by portable emission measuring systems (PEMS) gradually became compulsory for new car models, starting from September 2017; also, an improved laboratory test cycle (worldwide harmonised light vehicle test procedure – WLTP) replaced the NEDC as of 2017.

Also, in Finland, the number of diesel passenger cars began to grow rapidly in the mid-2000s due to the new CO_2 emission–based tax for passenger cars. In **Paper I**, we showed that the ambient NO_2/NO_x ratio was increasing at urban and suburban sites in the mid-1990s and early 2000s. To see the further development, the observed NO_2/NO_x concentration ratios are reproduced here, but they now include all Finnish monitoring stations up to the year 2018 (Figure 9). All five calculated percentiles of the annual NO_2/NO_x ratios displayed a statistically significant increasing trend (OLS, *t*-test; $p<0.05$) for the whole 1994–2018 period, but of the latter years (2007–2018), only the 25th percentile displayed an increasing trend ($p=0.04$). The summary of all monitoring data suggests that after 2007, the NO_2/NO_x ratio continued to increase at roadsides (with heavy traffic and high NO emission), albeit at a clearly slower pace (Figure 9).

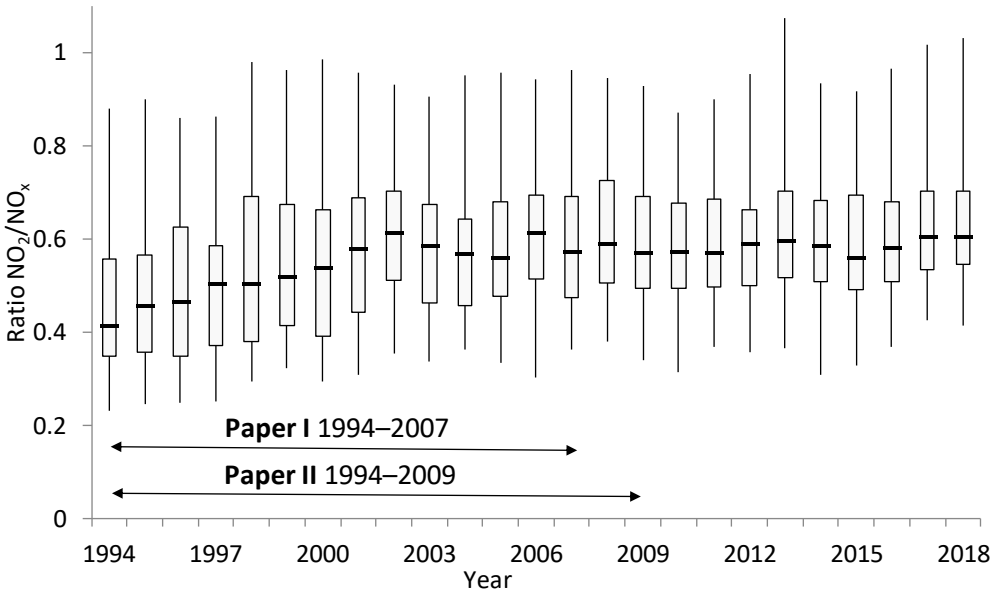

Figure 9. Distribution of annual NO_2/NO_x ratios calculated from NO_2 (in ppbv) and NO (in ppbv) concentrations measured at the Finnish monitoring sites in 1994−2018. At least a 75% annual data capture was required for each site and year. For each year, the lowest, highest and median values together with the 25th and 75th percentile values (box) are shown. The number of sites varies from 24 in 1994 to 54 in 2018, the total number of sites being 203. A total of 1257 annual ratios were included.

The steepest growth of the NO_2/NO_x ratio occurred in the first decade of the whole study period (1994–2004). During this period, the passenger car fleet in Finland was dominated by petrol-fuelled vehicles (92% and 88% of the car fleet in 1994 and 2004, respectively) and by the increasing fraction of cars with three-way catalysts (TWCs). Petrol cars emitted/emit very little, if any, tailpipe NO_2 (less than 5% of total NO_x) and TWC controlled NO (and HC and CO) very efficiently. So, vehicles' NO emissions declined systematically, while secondary NO_2 (formation from $NO+O_3$) and regional background concentrations declined less or not at all, resulting in an increasing NO_2/NO_x concentration ratio in urban areas. In general, the situation was comparable to the Töölö case presented in **Paper II**.

Instead, the data from Mannerheimintie (**Paper II**) span over the period of the most intense diesel penetration, and thus, the total oxidant model calculation is supplemented here with new data up to 2015, when O_3 monitoring at Mannerheimintie ended. Table 3 and Figure 10 summarise the updated results of the primary NO_2 emission fraction calculated with the total oxidant model (see **Paper II**).

Table 3. Annual regression results of $[O_x]_{local} = c[NO_x]_{local} + d$ (see **Paper II**) at the Mannerheimintie site in 2005–2015 (SE = standard error)

							diesel share %	
year	c	SE.	d	SE.	r	n	passenger car fleet	1st registrations
2005	0.151	0.002	2	0.1	0.78	4162	12.5	17
2006	0.173	0.003	0.3	0.2	0.72	2953	13.3	20.3
2007	0.169	0.002	2.3	0.1	0.78	4216	14.5	28.5
2008	0.206	0.002	1.4	0.2	0.81	4212	17.1	49.6
2009	0.21	0.002	2.1	0.1	0.85	4045	18.7	46.3
2010	0.153	0.003	3.5	0.2	0.66	4060	20.3	41.8
2011	0.161	0.002	2.1	0.1	0.73	4083	21.8	42.1
2012	0.134	0.002	2.1	0.1	0.71	3947	23.1	38.7
2013	0.153	0.003	1.4	0.2	0.65	4333	24.1	37.3
2014	0.154	0.003	1.1	0.2	0.63	3918	25.1	39
2015	0.187	0.003	3.2	0.1	0.68	4213	26	35.7

It turns out that the rise in the primary NO_2 emission fraction – calculated with the total oxidant model – was already reversed in 2010, and since then, it has varied between 15% and 19% at the Mannerheimintie site (Table 3). Correspondingly, since 2010, the contribution of the conversion from NO to NO_2 has been larger (51–56% of the total NO_2 concentration) than the primary NO_2 fraction (28–34%). The contribution of the regional background to this urban NO_2 concentration was 14–19% (Figure 10). So, the anticipated future development, that the primary NO_2 emissions will make an increasingly important contribution to the ambient NO_2 concentrations (**Paper II**), has not realised during these six years at this study site.

On the other hand, the number of new registrations of diesel passenger cars has become a steady decrease (24% in 2018) since the peak year 2008 (50%) (Table 3). The share of diesel cars of the total car fleet is still steadily growing; diesel engines last longer than petrol engines, and almost 60% of the imported used cars (total of 45 000) were diesel cars in 2018 (AuT, 2018).

Obviously, the magnitude and time development of the primary NO_2 emission and its contribution to the ambient NO_2 concentrations at roadsides is highly dependent on the country/city and road-specific vehicle fleets. However, based on this updated data, it seems that in Finland, the development of air quality in terms of NO_2 has remained positive despite the (temporarily) accelerated diesel penetration. Ambient NO_2 concentrations are decreasing (Figure 8), and the NO_2/NO_x ratio has stabilised (Figure 9).

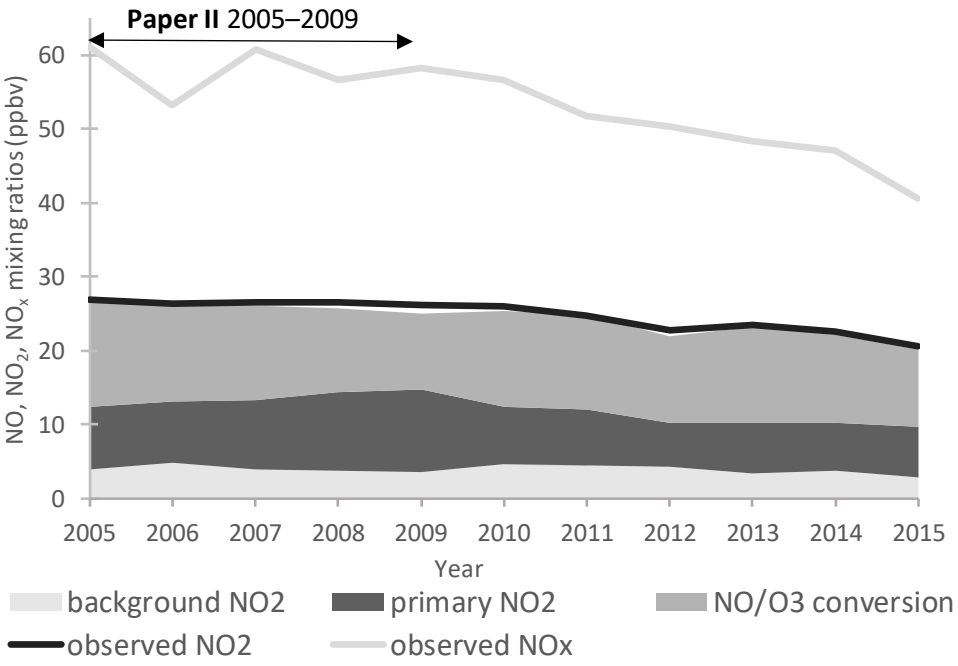

Figure 10. Annual NO_2 budgets during weekday hours between 5:00 and 21:00 at Mannerheimintie, 2005–2015.

Similar results come from the rest of Europe, too. Grange et al. (2017) show that from around 2010, the NO_2/NO_x ratio at most roadside locations across Europe have stabilised. Degraeuwe et al. (2017) conclude that the absolute level of traffic NO_x emissions is more relevant for the NO_2 concentration in urban street canyons than the NO_2 fraction in the total NO_x emissions. Carslaw et al. (2019) found that the primary and absolute NO_2 emissions of diesel cars vary a lot depending on the year of the manufacture (Euro status), used emission control techniques, the vehicle "family" manufacturer and mileage. Carslaw et al. (2019) conclude that the importance of primary NO_2 from vehicles has been decreasing in recent years.

4.3 Carbon monoxide (CO) trends

A reduction of CO emissions and concentrations has not encountered problems similar to NO_2. The strong and uniform ambient air concentration reductions of CO (-3% to -7%/yr) at the six studied urban monitoring sites can be related to the realised reductions in road traffic emissions. In Finland, the urban CO concentration levels are nowadays below 300 µg/m^3, and CO is no longer monitored in cities (Figure 11).

Similar temporal behaviour, i.e. marked declines of CO, were also detected in Europe in 2002−2011 (Guerreiro et al., 2014). Nowadays, CO is only a very localised and infrequent air quality problem in Europe. This can be considered another example of the successful implementation of air pollution control

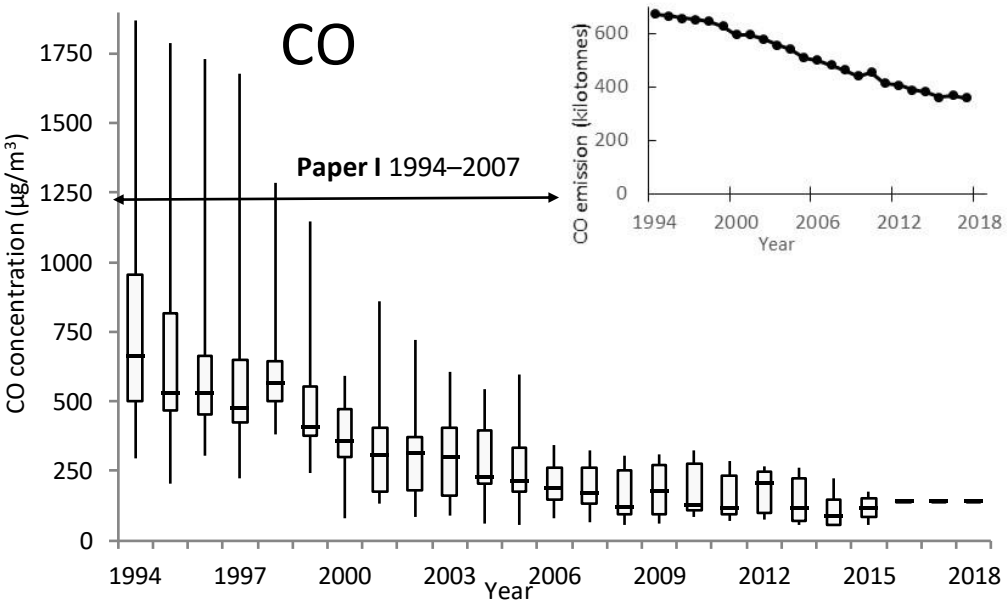

Figure 11. Distribution of annual mean concentrations of CO registered at the Finnish monitoring sites in 1994−2018. At least a 75% annual data capture was required for each site and year. For each year, the lowest, highest and median values together with the 25th and 75th percentile values (box) are shown. The number of sites included varies from 9 in 1994 to 17 in 2002, and after that, it follows a gradual decrease to 4 sites in 2015 and one site (Pallas) since then. A total of 261 annual values were included. The subplot shows the development of the annual CO emissions in Finland (SYKE, 2019a).

Long-term rural background measurements of CO were not available, but international studies combining emission inventories, surface observations, satellite data and modelling suggest widespread decreasing trends in the 2000s across Western Europe, eastern USA (Yoon and Pozzer, 2014) and East Asia (Zheng at al., 2018), mostly due to the changes in anthropogenic emissions.

4.4 Ozone (O_3) trends

Surface-level O_3 concentrations, especially in urban areas, are closely linked to the NO_x emissions and concentrations. Once released into the atmosphere, both NO and NO_2 join in complicated atmospheric photooxidation reaction chains. The simplest of them, the one producing O_3, the photostationary state of the NO_x–O_3 cycle, is fast and important, especially in urban NO_x-rich environments. More generally, O_3 production takes place via the O_3–VOC–NO_x system, which also involves CO and inorganic radicals like hydroxyl, hydroperoxyl and nitrate radicals (e.g. Seinfeld and Pandis 1998; Monks et al., 2015). These chains of reactions can be long, complex and non-linear. Meanwhile, the ozone and its precursors can be transported throughout the lower atmosphere on regional, intercontinental and hemispheric scales (Monks et al., 2015). As a result, an emission reduction of one component is not necessarily reflected as a parallel reduction of ozone ambient concentrations.

Anthropogenic emissions of the major ozone precursors (i.e. NO_x, NMVOCs and CO) have decreased during the past 20 years in Finland as well as in Europe and North America (e.g. Monks et al., 2015). On the other hand, global emission trends vary greatly from region to

region, and while emissions are declining in Europe and North America, the global trend may be stagnant (NO_x) or even rising (NMVOCs) (Miyazaki et al., 2017; Huang et al., 2017).

Ozone concentration trends remain challenging for air pollution control. In Europe, for example, both positive and negative (generally weak) trends have been documented. Guerreiro et al. (2014) presented an overview and an analysis of ozone trends in Europe (AirBase data) in 2002–2011. They found that 80% of the sites (both urban and rural) did not have a statistically significant trend for the studied metric (93.15th percentile of a maximum daily 8 hour mean); 18% of the stations registered a statistically significant decreasing trend, while 2% registered a significant increasing trend.

Yan et al. (2018) report significant ozone enhancements (0.20–0.59 µg/m^3/yr) for the annual means over the European suburban and urban stations (AirBase data) during 1995–2012. For European background sites (EMEP), Yan at al. (2018) report a decreasing trend (-0.9 µg/m^3/yr) in the 95th percentile ozone concentrations (especially at noon), while the 5th percentile ozone concentrations increased with a trend of 0.3 µg/m^3/yr during 1995–2014. Klingsberg et al. (2017) found decreasing summer daytime trends (slopes for the 75th–98th percentiles ranging from -0.2 to -1 µg/m^3/yr) and increasing summer nighttime and winter daytime and nighttime ozone trends at northern European background sites (EMEP) during 1990–2015. Olstrup et al. (2018) found increasing O_3 concentrations at urban background sites in Stockholm, Gothenburg and Malmö during 1990–2015. Hellén et al. (2015) found no ozone trend at Pallas/ Sammaltunturi during 1994–2013. Of the nine studied NMVOCs, only one (ethyne) had a statistically significant decreasing trend.

Fleming et al. (2018) conclude that Europe and North America in particular (both urban and non-urban) are characterised by reduced exposure to peak levels of ozone related to photochemical episodes over the 2000–2014 period. The short-term mid-high and long-term high ozone metrics exhibited more mixed trends. Furthermore, Fleming et al. (2018) conclude that in Europe, the trend results for non-urban and urban stations are broadly similar.

Similar miscellaneous ozone trends were reported in **Paper I**, too. The concentration time series of O_3 were studied from 9 rural background sites and two urban/suburban traffic sites. Increasing and decreasing trends of ozone mean levels were equally common, although the only three statistically significant trends were increasing (Helsinki Töölö, Vantaa Tikkurila and Espoo Luukki). On the other hand, the predominantly increasing trends of the monthly calculated 95th percentiles (**Paper I**) were detected, four being statistically significant, i.e. Raja-Jooseppi, Pallas/Sammaltunturi, Helsinki Töölö and Vantaa Tikkurila.

Figure 12 displays the 95th and 5th percentiles of summertime (March–August) ozone concentrations from all Finnish ozone monitoring sites calculated on an annual basis from 1994 to 2018. Trends, if any, are vague, but some patterns can be extracted.

The increase in summertime ozone peak levels documented in **Paper I** have not continued since 2007, but rather, Figure 12 indicates decreasing peak ozone concentrations for most sites – both urban and non-urban – since the top year 2006. In spring and summer 2006, Finland was affected by two major LRT episodes lasting several days (see **Paper V**), which are clearly reflected in these peak ozone concentrations. For example, the all-time (since 1987) hourly

ozone record (195 µg/m^3) of Finnish monitoring stations was registered on 5 May 2006, at the Virolahti site.

The decrease of episodic ozone peak levels in rural areas of Europe have been attributed to regional pollution emission controls applied to the VOC and NO_x and the following reductions in photochemical ozone formation (Monks et al., 2015 and references therein). On the other hand, in the high-NO_x environments of major European cities, the ozone levels are increasing due to locally decreasing NO emissions and following reductions in ozone scavenging by NO (Monks et al., 2015 and references therein). This latter development is no longer evident in this Finnish data; peak levels (as well as mean levels) of ozone increased at the sites next to the busiest roads in the 1990s (see the red lines in Figure 12) but have since been rather stable and have quite closely followed the interannual variation found in the surrounding urban/suburban and rural areas. The increase in lower-level concentrations (Yan et al., 2018) is not as apparent (Figure 12) in this Finnish material.

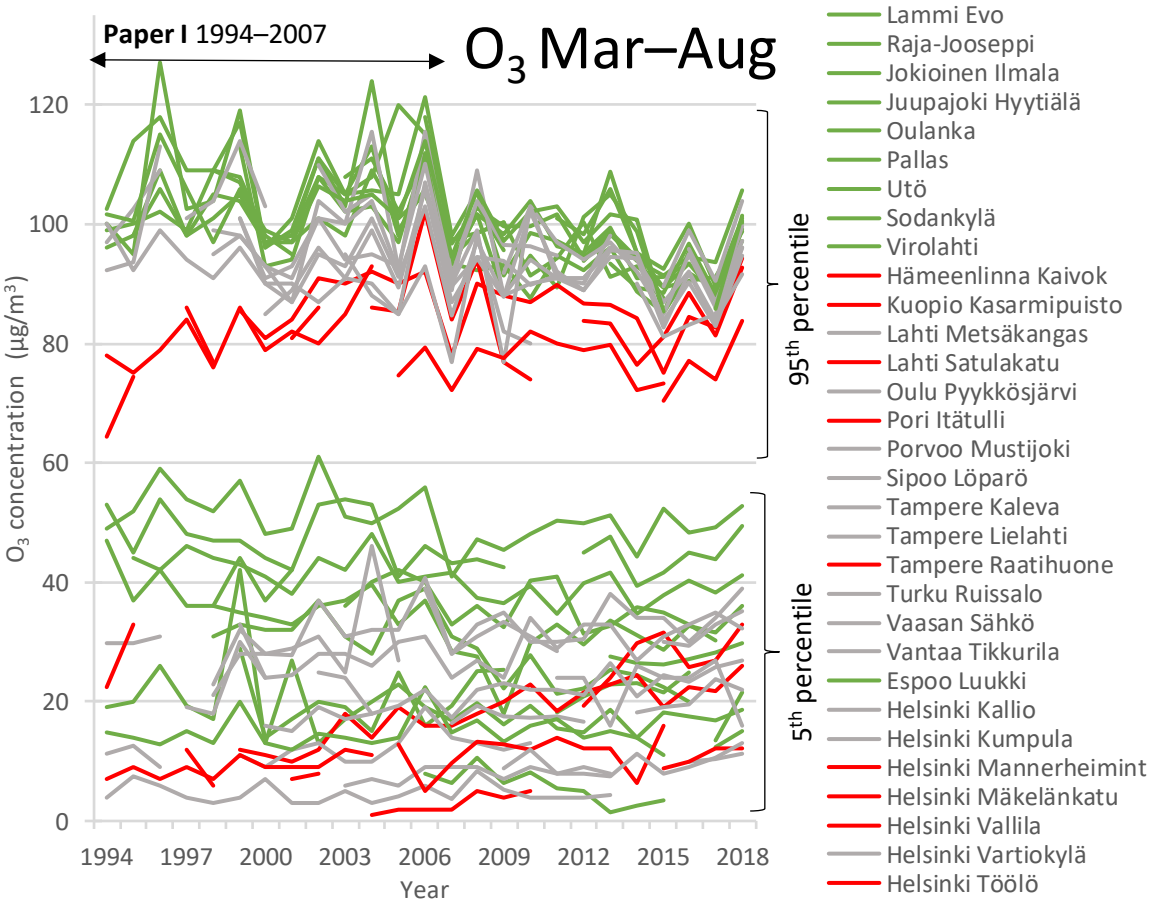

Figure 12. The summer period's (March–August) 5th and 95th percentiles of hourly O_3 concentrations at Finnish monitoring sites 1994–2018. The green lines are rural stations; the grey lines are urban, suburban or industrial stations and red roadside sites.

4.5 PM_{10} particle mass trends

In Finland, elevated PM_{10} concentrations are typically detected near busy roads in springtime (Kukkonen et al., 1999; Pohjola et al., 2002; **Paper IV**; Teinilä et al., 2019). In

Paper IV, it was shown that during the 1998–2003 study period in 20 Finnish cities, the annual peak concentrations occurred very concurrently. Further, it was shown that PM_{10} concentrations started to increase in the first half of March (a week later in northern cities) concurrently with the snowmelt and the low rainfall period. The highest concentrations occurred in urban areas with high traffic volume.

In Finnish studies, urban (road) dust has been identified to be mainly mineral matter in the coarse mode (aerodynamic diameter between 2.5 and 10 µm) (Hosiokangas et al., 1999; Pakkanen et al., 2001; Kupiainen and Tervahattu 2004; Sillanpää et al., 2006). The source of road dust and factors influencing the source strength have been studied both in laboratory tests (Räisänen et al., 2003; Kupiainen et al., 2003; Kupiainen et al., 2005; Räisänen et al., 2005; Tervahattu et al., 2006), near-road measurements (Kupiainen and Tervahattu 2002; Kupiainen et al., 2016), on-road/mobile measurements (Pirjola et al. 2009; Kupiainen and Pirjola 2011; Kupiainen et al., 2016) and by modelling (e.g. Stojiljkovic et al., 2019). Generally, these studies showed that the interactions between studded tyres, traction sanding and pavement aggregate are the major underlying factors behind the production of road dust emissions. Recent study (Kupiainen et al., 2016) suggests that the pavement wear associated with the use of studded tyres contributes most to high ambient PM_{10} concentrations, while the use of traction sanding has a lesser impact. The resulting ambient PM_{10} concentrations depend, obviously, on the traffic amounts, vehicle fleet, road maintenance activities and meteorological factors.

In addition to the spring dust period, **Paper IV** highlighted the PM_{10} episodes that were irregular in time but spatially wide-ranging and concluded that they are related to long-range transport rather than to local sources. Niemi et al. (2009) summarised the frequency and origin of fine particle episodes in Southern Finland; episodes occur several times per year, and they most often originate in Eastern Europe. Starting from the early 2000s, the characteristics and/or sources of the major LRT PM episodes in Finland have become identified in detailed case studies. In September 2001, the dust plumes from the Kazakhstan Ryn-Peski desert mixed with Estonian and Russian oil-shale industry emissions and spread over coastal areas of the Baltic Sea (Sofiev et al., 2003; Hongisto and Sofiev, 2004; Tervahattu et al., 2004). The most serious LRT episodes, though, are related to biomass burning events in 2002 (Niemi et al., 2004; Sillanpää et al., 2005; Niemi et al., 2005; Aarnio et al., 2008), in 2006 (Saarikoski et al., 2007; Saarnio et al., 2010; **Paper V**; Makkonen et al., 2010) and in 2010 (Portin et al., 2012; Mielonen et al., 2012; Leino et al., 2014). In addition to these, wildfire episodes were reported in 2004 (Niemi et al., 2006; Saarikoski et al., 2008), in 2007 (Niemi et al., 2009), in 2008 (Hyvärinen et al., 2011) and in 2009 (Timonen et al., 2010).

These wildfire episodes typically occurred in spring (March–May) and late summer (July–September), and they originated from Eastern/Central Europe and the western part of Russia. The episodes were strongest in Southern Finland, but some of them were detected in Northern Finland, too (e.g. Leino et al., 2014), and further in the High Artic (Stohl et al., 2007). In episode cases, the first alarm typically came from the elevated concentrations of PM mass detected with the continuous monitors. During LRT episodes, the particle mass size distribution was dominated by fine particles (<2.5 µm) rather than by the coarse fraction. In some PM episode cases, however, PM_{10} served as a feasible LRT episode indicator, too (e.g. **Paper V**, Makra et al., 2011). Globally, wildfires release huge amounts of primary organic aerosol (POA) but also photochemically active organic gases (VOCs) and black/elemental carbon (e.g. Andreae, 2019), all contributing to (aged) PM mass. Depending on the trajectory of the polluted air mass, the

aerosol could be a mixture of biomass burning (agricultural/forest) and fossil/industrial emissions like trace metals, resuspended persistent organic pollutants, gaseous mercury and radionuclides (e.g. **Paper V**; **Paper III**; Paatero et al., 2009).

Somewhat surprisingly, the latest reported episodic long-range transport of particles was different from those described above. In Helsinki on 25 May 2011, the ash plume from the eruption in Grimsvötn, Iceland, enhanced PM_{10} mass concentrations up to several tens of $\mu g/m^3$ (Kerminen et al., 2011).

PM mass recorded at any monitoring site is a mixture of primary or secondary, natural or anthropogenic, fossil or biogenic, and local or far away emission/source, which makes PM pollution particularly difficult to control. So, it's not surprising that PM_{10} concentration trends in Finland and elsewhere in Europe are not decreasing in any systematic way.

In **Paper I**, 12 PM_{10} time series of monthly means and monthly 95^{th} percentiles were investigated for potential trends. No nationwide convergent development was found, but instead, both increasing and decreasing trends were identified. This result is not a surprise as all studied time series were from urban/suburban/industrial monitoring sites and given the pronounced contribution of local street dust as a source of ambient PM_{10} particles. Similar partly mixed trend results of PM have been reported elsewhere in Europe, too (Guerreiro et al., 2014; Colette et al., 2011).

In Finland, the springtime street dust raises a lot of public concern and has been identified as one of our major air quality problems; it was also addressed in the National Air Pollution Control Programme (NAPCP, 2019). In this thesis, an attempt is made to extract the spring dust trends by summarising the recent seasonal PM_{10} monitoring results in Finland (Figure 13). Years were divided to three seasons: Feb–May (representing the spring dust period), Jun–Sept (wildfires) and Oct–Jan (other).

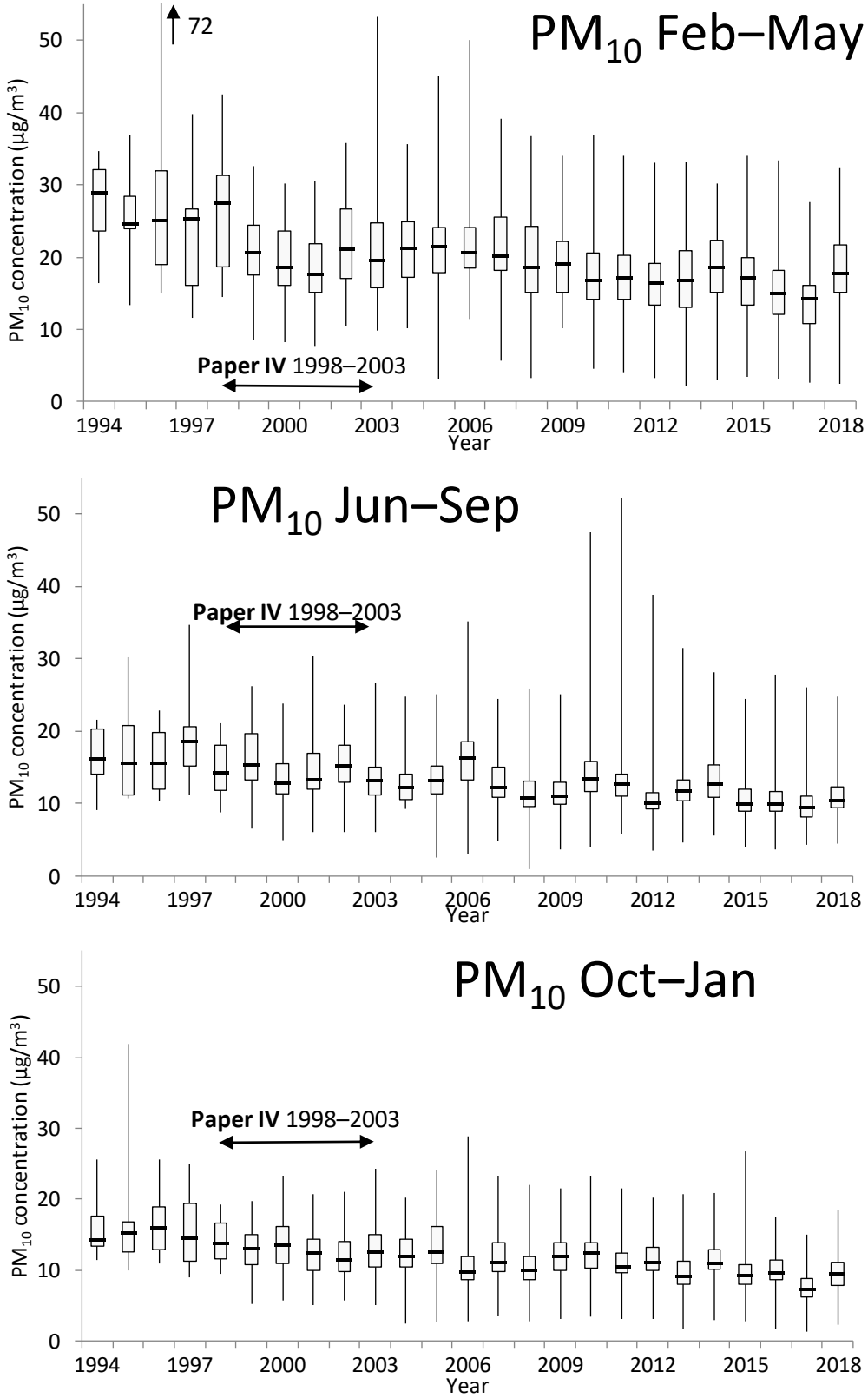

Figure 13. Distribution of seasonal mean concentrations of PM_{10} measured at the Finnish monitoring sites in 1994−2018. At least a 75% seasonal data capture was required for each site and year. For each year, the lowest, highest and median values together with the 25th and 75th percentile values (box) are shown. The number of sites included increased from 9 in 1994 to 60 in 2007, and after that, it has been steady. A total of 1115, 1120 and 1194 seasonal values were included for Feb–May, Jun–Sep and Oct–Jan, respectively. The total number of sites was 171.

It should be noted that in 2017, the updated correction factors for PM continuous monitors (see Waldén et al., 2017) were released and recommended for use. Of around 60 instruments in use in 2017, 32 adopted a correction factor which reduced the measurement result compared to the one used previously. At the median level, the reduction percents were between -30% and -1%. Nine instruments were corrected with a factor that increased the result (8–26%), and the coefficients for twenty devices were not changed or the information was not provided. So, PM results for 2017 onwards are not necessarily comparable to previous years, and they are not included in the following trend analysis.

A quick look at Figure 13 suggests that PM_{10} concentrations have generally been decreasing and that the decrease was steeper at the beginning of the study period. During the first 10 years, the monitoring network expanded strongly (from 10 to 60 sites). At the beginning, the new sites were typically opened at the most polluted roadside locations, which provide highly localised data, especially when it comes to street dust. At the lowest tail of the concentration distribution, the start of PM_{10} monitoring at the remote sites at Pallas and Raja-Jooseppi in 2007 is clearly visible. These kinds of changes in the network may cause some bias in the aggregated data. Since 2007, the network has been roughly stable, and the trend estimates more representative.

For an exact picture of the trends, linear fits to the time series of each percentile were performed over the whole 1994–2016 period and separately over the last ten years, 2007–2016. Table 4 summarises the trend statistics.

Table 4. Trend results of the seasonal mean concentrations of PM_{10} (slopes and their *p*-values; two-tailed t-test, percent change); significance level of at least 5% is bolded

		1994–2016			2007–2016		
		slope	*p*-value	%	slope	*p*-value	%
Feb–May	highest	-0.485	0.103		**-0.615**	**0.014**	**-36**
	75th percentile	**-0.497**	**0.000**	**-37**	**-0.577**	**0.010**	**-53**
	median	**-0.459**	**0.000**	**-40**	**-0.358**	**0.017**	**-41**
	25th percentile	**-0.354**	**0.000**	**-38**	**-0.438**	**0.007**	**-59**
	lowest	**-0.564**	**0.000**	**-88**	-0.397	0.115	
June–Sept	highest	+0.421	0.093		-0.160	0.896	
	75th percentile	**-0.381**	**0.000**	**-42**	-0.209	0.243	
	median	**-0.265**	**0.000**	**-36**	-0.143	0.333	
	25th percentile	**-0.171**	**0.000**	**-28**	-0.134	0.225	
	lowest	**-0.297**	**0.000**	**-71**	+0.130	0.422	
Oct–Jan	highest	-0.283	0.062		-0.194	0.499	
	75th percentile	**-0.308**	**0.000**	**-38**	**-0.259**	**0.034**	**-42**
	median	**-0.249**	**0.000**	**-37**	-0.197	0.100	
	25th percentile	**-0.178**	**0.000**	**-32**	-0.133	0.199	
	lowest	**-0.391**	**0.000**	**-93**	**-0.136**	**0.043**	**-87**

Indeed, when the whole study period is considered for all seasons, all concentration levels except the highest exhibit a s.s. decreasing trend. In contrast, in the latter part of the study period, only the spring season exhibits systematically decreasing trends (all concentration levels except the lowest).

This suggests that, to some extent, the local abatement measures (e.g. reducing traffic amounts, changes in the car fleet, road maintenance activities) have been successful, and the springtime street dust levels have been reduced. However, the springtime concentration levels of PM_{10} are still one and a half times higher than in other seasons on average. In spring, the daily mean of 50 µg/m^3 is exceeded frequently, not only in the biggest cities but also in small towns of 20 000 people. At most, there are about 20–30 exceedances/site/year, so the European PM_{10} norm (daily mean should not exceed 50 µg/m^3 more than 35 five times per year) is not exceeded. Although the European air quality norm is not being violated, street dust remains a persistent flaw in our otherwise good air quality.

4.6 Persistent organic pollutants (POPs) trends

In **Paper III**, long-term atmospheric monitoring data of persistent organic pollutants (POPs) were assembled from a rural site in Southern Sweden, Råö, and the Pallas/Matorova site in Northern Finland. The concentration levels, congener profiles and seasonal and temporal trends (1994–2011) were evaluated to assess the status of POPs in the Scandinavian background atmosphere.

Polycyclic aromatic hydrocarbons (PAHs)

In 2007, globally, the biggest sources of PAHs (i.e. PAH-16) were residential/commercial biomass burning (60.5%), transportation (14.8%) and open-field biomass burning (agricultural waste burning, deforestation and wildfire) (13.6%) (Shen et al., 2013). About 80% of the transportation emissions were in the form of light molecular weight naphthalene, while residential fuel consumption, industrial emissions and nonorganised waste burning contributed more high molecular weight carcinogenic PAHs (6% of total PAH emissions, eight congeners starting from benz(a)anthracene) (Shen et al., 2013). South and East Asia contributed half of the total global PAH emissions. From 1995 to 2008, the global PAH emissions decreased by 15%, and significant emission reductions are expected to continue until 2030 (Shen et al., 2013).

Considerable differences in PAH source profiles and the time development of emissions varied a lot due to energy structure, development status and vegetation cover. According to Shen et al. (2013), in developed countries (like Europe), the PAH emissions peaked in the early 1970s and decreased gradually since, primarily due to the introduction of emission mitigation technologies and the subsequent decline in the PAH emissions from on-road motor vehicles. Meanwhile, only slow decreases in PAH emissions occurred in the residential and commercial sources.

Finnish PAH-4 emissions (the four indicator compounds benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene and indeno(1,2,3cd)pyrene reported under the UNECE LRTAP) increased from 7 tonnes to 11 tonnes between 1990 and 2010. Since then, the PAH-4 emissions have fluctuated around 10 tonnes/year with no clear trend (SYKE, 2019b).

In Finland, the fuel wood consumption in the residential sector has increased by 23% between 1993 and 2017 (LUKE, 2019). Residential wood combustion has been identified as the major emission source of (primary) fine particles and black carbon (e.g. Savolahti et al.,

2016; NAPCP, 2019). And finally, residential wood combustion has been shown to be a significant source of airborne fine particles ($PM_{2.5}$) (Saarnio et al., 2012) and PAHs (Hellén et al., 2008; 2017) in urban and suburban areas, especially in wintertime.

In light of the considerations above, the trends of atmospheric background PAH concentrations in **Paper III** are understandable. Our data comprised 12 PAH congeners consisting of those with three to six aromatic rings. The low molecular weight congeners with two aromatic rings, including naphthalene, were not included in the chemical speciation due to methodological reasons. Therefore, it is not expected that the reduction in European/Finnish traffic emissions will be reflected in this trend analysis, but rather, they will bring out the evolution of the use of biomass fuel in the residential sector and/or wild/agricultural fires.

Of the 12 studied PAH compounds, only two at Råö and three at Pallas exhibited s.s. decreasing trends. At Råö, the three- and four-ring anthracene and benz(a)anthracene decreased, while at Pallas, three-, five- and six-ring fluoranthene, benzo(b)fluoranthene and indeno(123cd)pyrene decreased. Most PAH time series had negative regression slopes, but trends were not statistically significant. Reasonably, these weak or non-existent decreasing trends of PAHs can be linked to the only slowly decreasing regional and long-range transported emissions of biomass burning in the residential sector and/or wild/agricultural fires.

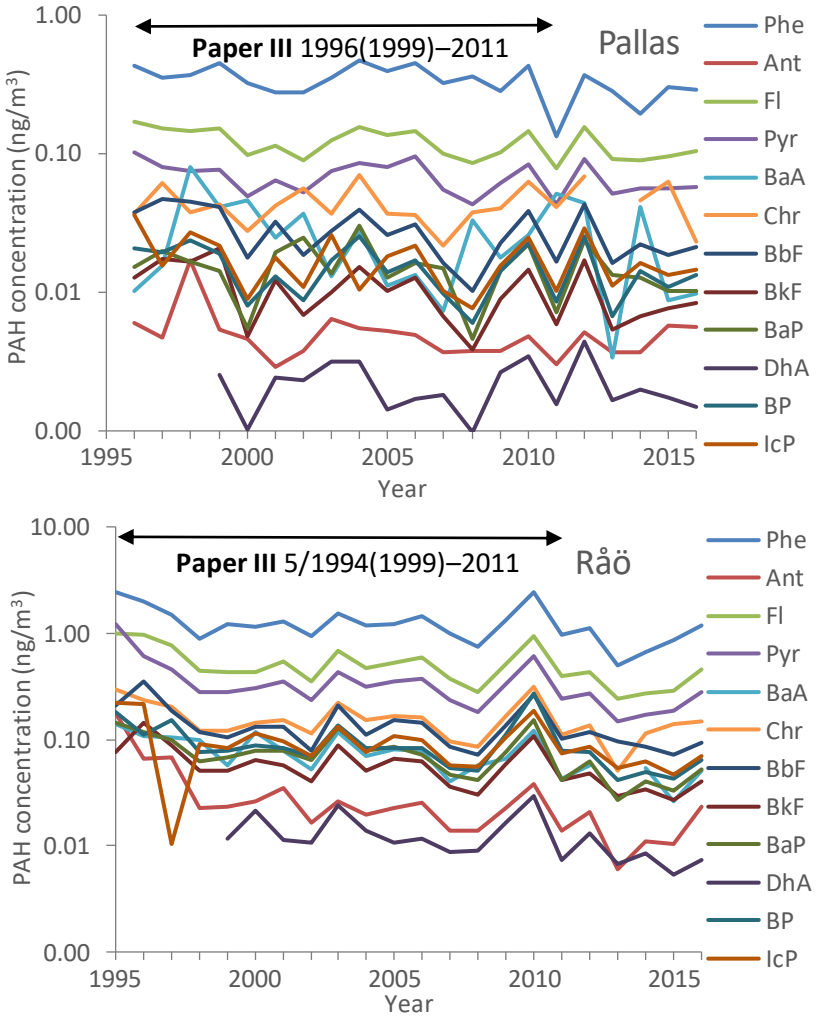

Figure 14. Annual means of 12 PAH compounds at Pallas and Råö in 1995–2016.

In a recent study, Yu at al. (2018) analysed the long-term trends (1992–2015) of selected PAHs (phenanthrene, pyrene and benzo(a)pyrene) at three arctic/subarctic sites, i.e. Alert, Canada; Zeppelin, Svalbard and Pallas, Finland. In line with the findings of **Paper III**, a significant decline in these PAHs was not observed. Forest fires were identified as an important contributing source.

At Råö, closer to the European and Scandinavian emissions sources, the concentrations of PAHs were 3–9 times higher than at the remote Pallas site. Yu et al. (2018) also found that the decrease in PAH concentrations continued when going farther north from Pallas (Pallas > Zeppelin > Alert).

Polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs)

Paper III reported declining trends of atmospheric concentrations for the PCBs, HCHs, chlordanes and DDTs at Råö and Pallas. These declines were a very prominent feature in this material; of the studied 28 time series, all but one showed a s.s. decreasing trend. In a recent paper, Kalina et al. (2019) published long-term trends (1996–2016/2018) of PCBs and HCHs based on the very same but extended data from Råö and Pallas. For PCBs and p,p'-DDE, the trends (annual changes per year) were in good agreement with the findings of **Paper III**. For α-HCH, annual changes were -10% and -9% and for γ-HCH, they were -11% and -12% at Råö and Pallas, respectively (Kalina et al., 2019). These rates of changes are almost twice as high as given in **Paper III**. The differences are most likely related to the different transformations applied to the original data. Kalina et al. (2019) fitted the linear trend to log-transformed data accompanied by the Theil–Sen estimator and the Mann-Kendall test.

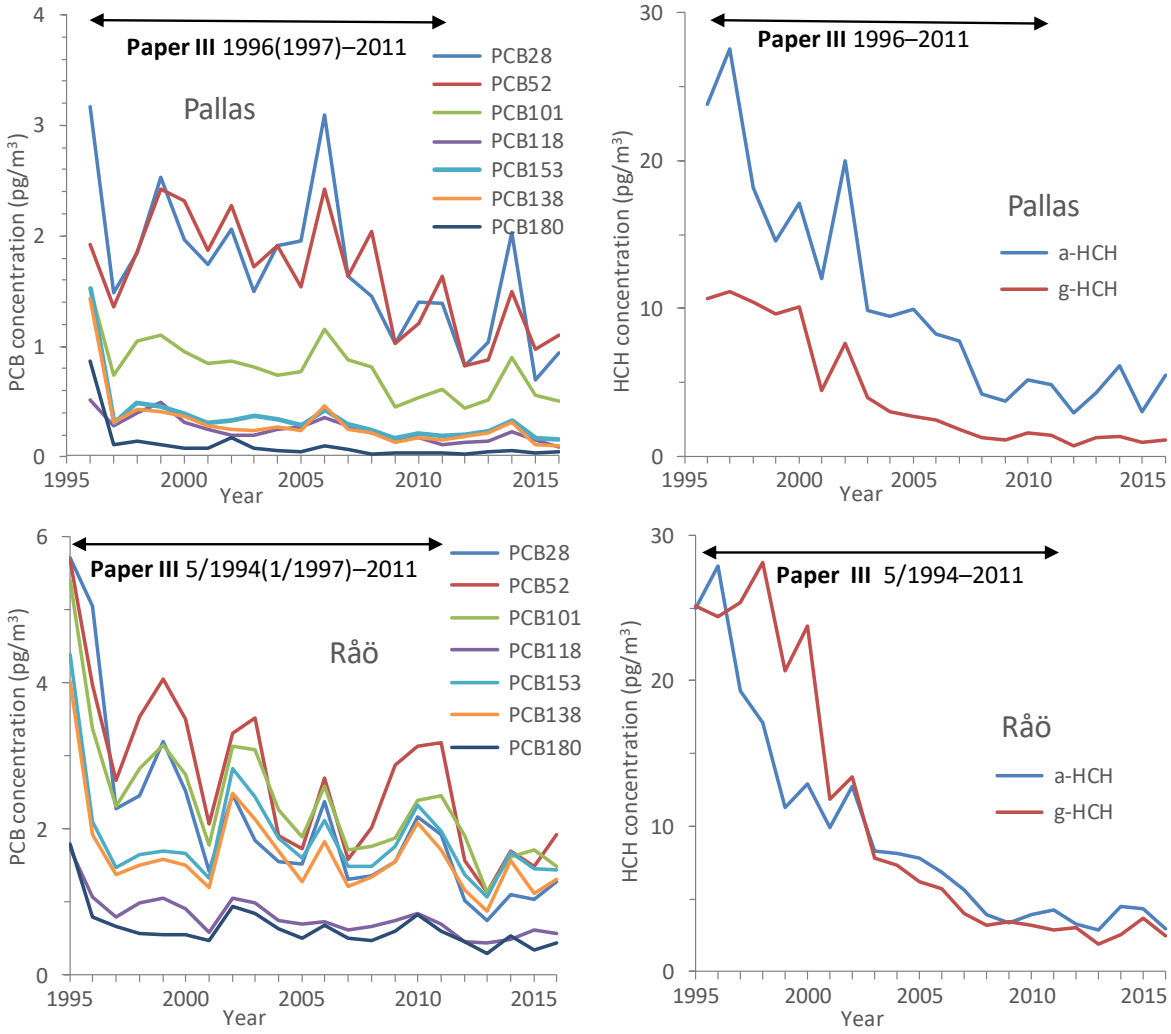

Figure 15. Annual means of PCBs and HCHs at Pallas and Råö in 1995/1996–2016.

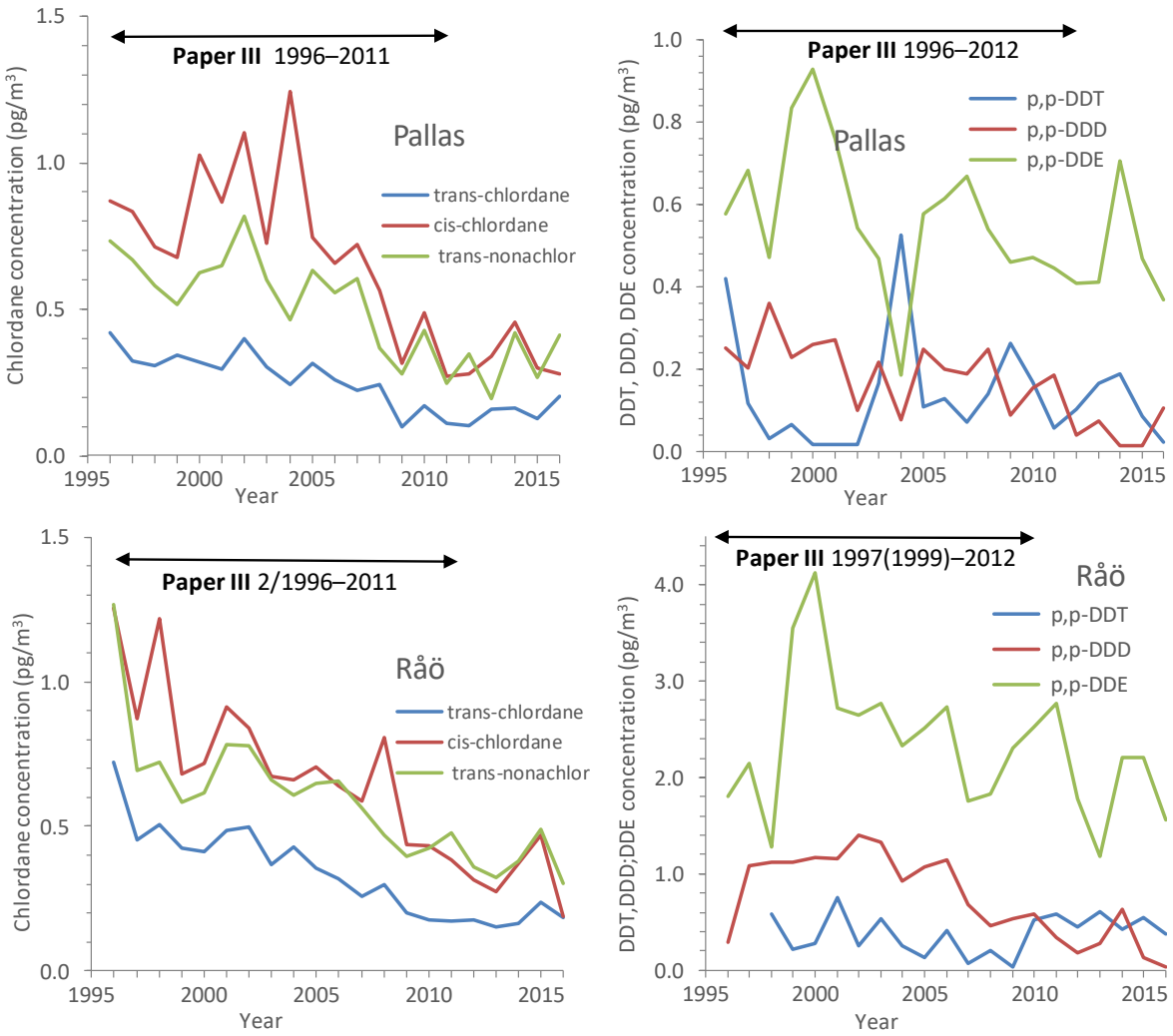

Figure 16. Annual means of chlordanes and DDT and its residues at Pallas and Råö in 1996–2016.

Nevertheless, these studies (**Paper III** and the references therein; Kalina et al., 2019) show large decreases in the atmospheric concentrations of PCB and OCPs in the Nordic atmosphere. PCBs, DDT and chlordanes belonged to the first set and HCHs to the second set of the controlled POPs by the UN Stockholm Convention in 2001 and 2009, respectively. This global treaty and the preceding national and regional regulations (since the late 1970s) banning intentional production, use, import and export have thus been successful in achieving improvements to the atmospheric environment. However, the projections suggest that the most long-lived compounds will persist in the atmospheric cycle beyond 2030 (**Paper III**).

5 Conclusions

Of the 176 analysed long-term time series in the original papers, 51% showed a statistically significant downward trend (**Paper I;** years 1994–2007 and **Paper III,** years 1994–2011). The decrease in concentrations was most common for CO, SO_4^{2-}, POPs (other than PAHs), NO_x, SO_2 and NO_2, i.e. 100%, 100%, 96%, 70%, 68% and 50% of the studied time series decreased, respectively. PM_{10} yielded both decreasing (38%) and increasing (21%) trends. O_3 exhibited only increasing trends (32%), and PAHs, NO_3^- and NH_4^+ were rather characterised by the lack of any uniform trends.

The assessment of the extended material presented in this thesis largely confirmed the above summarised development. The decrease of SO_2 and SO_4^{2-} concentrations have continued, and nowadays, elevated SO_2 concentrations are detected at a handful of industrial monitoring sites, while in Finnish background areas, the highest SO_2 concentrations are detected in northeastern Lapland under the influence of the ongoing and still partly obscure emissions in the Kola Peninsula. Urban CO concentrations have declined to such an extent that monitoring has been discontinued. Atmospheric concentrations of PCBs, HCHs and chlordanes have substantially decreased. NO concentrations have declined, and so have NO_2 concentrations, albeit slower than could be expected based on national NO_x emissions or parallel NO concentrations. There are indications that springtime street dust (PM_{10}) levels have declined over the last 10 years. Since 2006, the peak ozone concentrations have declined. Contrary to these predominantly decreasing trends, the limited available long-term data of PAHs suggest that no widespread decrease in concentrations has occurred.

This thesis demonstrates that internationally launched and nationally implemented regulatory controls have had an important role in improving air quality in Finland. The pollutants subject to long-term ambitious international abatement strategies (like SO_2, PCBs and OCPs) have decreased the most. Also, NO_x emission control has been succesful, but the outcome is a bit more complex. The emission regulations (both stationary and mobile sources) target the total NO_x emissions (which have been reduced successfully), while NO_2 is targeted in the ambient air standards.

The increasing penetration of diesel cars with relatively high (or unknown) primary NO_2 emissions may have been one contributing factor for why urban roadside NO_2 concentrations have not decreased as expected. However, the recent studies from European roadsides suggest that the still-decreasing absolute total NO_x emissions are compensating for the relative increase in primary NO_2 emissions, and the importance of primary NO_2 from vehicles has been decreasing in recent years. The newly adopted improved type approval tests may further reduce emissions – or at least clarify the situation.

In Finland, in terms of EU air quality norms for NO_2, the only problematic areas have been certain busy street canyon sites in Helsinki, which have been below the limit value since 2016. However, our relatively old car fleet and the increased import of old diesel cars cause uncertainty for future development.

A very different type of traffic-related air pollution is springtime street dust due to the use of studded tyres and manifested as elevated concentrations of PM_{10}. This thesis suggests that

springtime PM_{10} concentrations are on the decline. This could mean that the local abatement measures (e.g. reducing traffic amounts, changes in the car fleet, road maintenance activities) have been in the right direction, and the springtime street dust levels have been reduced. However, every spring, strong (even on a European scale) dust episodes occur, not only in the biggest cities but also in the small towns. So even though the European air quality norm is not being violated, street dust remains a persistent flaw in our otherwise good air quality.

In Finland, the ozone peak levels have been declining since 2006. A similar development has been detected in Europe and North America, and it is related to decreasing anthropogenic precursor emissions of NO_x and VOCs. For Finland, high background concentrations are more problematic, the reduction of which would require international and even hemispheric cooperation.

The available long-term background data of PAH concentrations suggest that no widespread decrease in concentrations has occurred. This is not necessarily surprising as the major global sources are small-scale solid fuel combustion and wildfires. Efforts to reduce these emissions have been relatively limited or non-existent so far.

Increasing wood burning and an increasing number of diesel cars are examples of cases when policies or economic controls to address one environmental issue (climate change) have had unintended consequences on another (air pollution).

References

Aarnio, P., Martikainen, J., Hussein, T., Vehkamäki, H., Sogacheva, L., Kulmala, M., 2008. Analysis and evaluation of selected PM pollution episodes in the Helsinki Metropolitan Area in 2002. Atmos. Environ., 42, 3992–4005.

Aas, W., Mortier, A., Bowersox, V., Cherian, R., Faluvegi, G., Fagerli, H., Hand, J., Klimont, Z., Galy-Lacaux, C., Lehmann, C.M.B., Myhre Lund, C., Myhre, G., Olivié, D., Sato, K., Quaas, J., Rao, P.S.P., Schulz, M., Shindell, D., Skeie, R.B., Stein, A., Takemura, T., Tsyro, S., Vet, R., Xu, X., 2019. Global and regional trends of atmospheric sulfur. Scientific Reports 9:953.

Akaike, H., 1973. Information theory as an extension of the maximum likelihood principle. In: Petrov, B.N., Csaki, F. (Eds.), Second International Symposium on Information Theory, Akademiai Kiado, Budapest pp. 267–281.

Andreae, M.O., 2019. Emission of trace gases and aerosols from biomass burning – an updated assessment, Atmos. Chem. Phys., 19, 8523–8546.

Anttila, P., Alaviippola, B., Salmi, T., 2003. Air quality in Finland–monitoring results in relation to the guideline and limit values and comparisons with European concentration levels. Publications on Air quality No. 33. Finnish Meteorological Institute. (In Finnish with an English abstract)

AuT, 2018. Share of diesel cars in new registrations and car fleet. The Finnish Information Centre of Automobile Sector, viewed 28 August 2018, <http://www.aut.fi/en/statistics/long-term_statistics/share_of_diesel_cars>.

BMVI, 2016. Report by the "Volkswagen" Commission of Inquiry. Investigations and administrative measures with regard to Volkswagen, results of the field investigation of the Federal Motor Transport Authority into unlawful defeat devices in diesel vehicles, and conclusions. Federal Ministry of Transport and Digital Infrastructure (BMVI), viewed 1 October 2019, <https://www.bmvi.de/SharedDocs/EN/publications/bericht-untersuchungskommission-volkswagen.pdf?__blob=publicationFile>.

Brockwell, P.J., Davis, R.A., 2002. Introduction to Time Series and Forecasting, Second Edition, Springer-Verlag, New York, Inc.

Carslaw, D., 2005. Evidence of an increasing NO_2/NO_x emissions ratio from road traffic emissions. Atmos. Environ., 39, 4793–4802.

Carslaw, D.C., Beevers, S.D., 2004. Investigating the potential importance of primary NO_2 emissions in a street canyon. Atmos. Environ., 38, 3585–3594.

Carslaw, D.C., Beevers, S.D., Tate, J.E., Westmoreland, E.J., Williams, M.L., 2011. Recent evidence concerning higher NOx emissions from passenger cars and light duty vehicles. Atmos. Environ., 45 (39), 7053v7063.

Carslaw, D.C., Farren, N.J., Vaughan, A.R., Drysdale, W.S., Young, S., Lee, J.D., 2019. The diminishing importance of nitrogen dioxide emissions from road vehicle exhaust. Atmos. Environ., X 1, 100002.

Carslaw, D.C., Murrels, T.P., Andersson, J., Keenan, M., 2016. Have vehicle emissions of primary NO_2 peaked? Faraday Discussions, 189, 439–454.

Carslaw, D.C., Rhys-Tyler, G., 2013. New insights from comprehensive on-road measurements of NO_x, NO_2 and NH_3 from vehicle emission remote sensing in London, UK. Atmos. Environ., 81, 339–347.

Casquero-Vera, J.A., Lyamani, H., Titos, G. Borrás, E., Olmo, J.F., Alados-Arboledas, L., 2019. Impact of primary NO_2 emissions at different urban sites exceeding the European NO_2 standard limit. Sci. Total Environ., 646, 1117–1125.

Colette, A., Aas, W., Banin, L., Braban, C. F., Ferm, M., González Ortiz, A., Ilyin, I., Mar, K., Pandolfi, M., Putaud, J.-P., Shatalov, V., Solberg, S., Spindler, G., Tarasova, O., Vana, M., Adani, M., Almodovar, P., Berton, E., Bessagnet, B., Bohlin-Nizzetto, P., Boruvkova, J., Breivik, K., Briganti, G., Cappelletti, A., Cuvelier, K., Derwent, R., D'Isidoro, M., Fagerli, H., Funk, C., Garcia Vivanco, M., Haeuber, R., Hueglin, C., Jenkins, S., Kerr, J., de Leeuw, F., Lynch, J., Manders, A., Mircea, M., Pay, M.T., Pritula, D., Querol, X., Raffort, V., Reiss, I., Roustan, Y., Sauvage, S., Scavo, K., Simpson, D., Smith, R.I., Tang, Y.S., Theobald, M.,

Tørseth, K., Tsyro, S., van Pul, A., Vidic, S., Wallasch, M., Wind, P., 2016. Air pollution trends in the EMEP region between 1990 and 2012. EMEP/CCC-Report 1/2016.

Colette, A., Granier, C., Hodnebrog, Ø., Jakobs, H., Maurizi, A., Nyiri, A., Bessagnet, B., D'Angiola, A., D'Isidoro, M., Gauss, M., Meleux, F., Memmesheimer, M., Mieville, A., Rouïl, L., Russo, F., Solberg, S., Stordal, F., Tampieri, F., 2011. Air quality trends in Europe over the past decade: a first multi-model assessment, Atmos. Chem. Phys., 11, 11657–11678.

Crippa, M., Janssens-Maenhout, G., Dentener, F., Guizzardi, D., Sindelarova, K., Muntean, M., Van Dingenen, R., Granier, C., 2016. Forty years of improvements in European air quality: regional policy-industry interactions with global impacts. Atmos. Chem. Phys., 16, 3825–3841.

EU, 2004. Directive 2004/107/EC of the European Parliament and of the Council of 15 December 2004 relating to arsenic, cadmium, mercury, nickel and polycyclic aromatic hydrocarbons in ambient air.

EU, 2008. Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe.

EU, 2016. Directive 2016/802 of the European Parliament and of the Council of 11 May 2016 relating to a reduction in the sulphur content of certain liquid fuels.

Degraeuwe, B., Thunis, P., Clappier, A., Weiss, M., Lefebvre, W., Janssen, S., Vranck, S., 2017. Impact of passenger car NO_x emissions on urban NO_2 pollution – Scenario analysis for 8 European cities. Atmos. Environ., 171, 330–337.

EEA, 2018. Air quality in Europe −2018 report. EEA Report 12/2018. 83pp.

Fleming, Z.L., Doherty, R.M., von Schneidemesser, E., Malley, C.S., Cooper, O.R., Pinto, J.P., Colette, A., Xu, X., Simpson, D., Schultz, M.G., Lefohn, A.S., Hamad, S., Moolla, R., Solberg, S., Feng, Z., 2018. Tropospheric Ozone Assessment Report: Present-day ozone distribution and trends relevant to human health. Elementa: Science of the Anthropocene, 6(1), p.12.

Grange, S.K., Lewis, A.C., Moller, S.J., Carslaw, D.C., 2017. Lower vehicular primary emissions of NO_2 in Europe than assumed in policy projections. Nature Geoscience 10, 914–918.

Grennfelt, P., Engleryd, A., Forsius, M., Hov, Ø., Rohde, H., Cowling, E., 2020. Acid rain and air pollution: 50 years of progress in environmental science and policy. Ambio. 49, 849–864.

Guerreiro, C.B.B., Foltescu, V., de Leeuw, F., 2014. Air quality status and trends in Europe. Atmos. Environ., 98, 376–384.

Hamilton, J.D., 1994. Time Series Analysis. Princeton University Press, New Jersey.

Hellén, H., Hakola, H., Haaparanta, S., Pietarila, H., Kauhaniemi, M., 2008. Influence of residential wood burning on local air quality. Sci. Total Environ., 393, 283–290.

Hellén, H., Kouznetsov, R., Anttila, P., Hakola, H., 2015. Increasing influence of easterly air masses on NMHC concentrations at the Pallas-Sodankylä GAW station. Boreal Env. Res., 20, 542–552.

Hellén, H., Kangas, L., Kousa, A., Vestenius, M., Teinilä, K., Karppinen, A., Kukkonen, J., Niemi, J.V., 2017. 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.

Henschel, S., Querol, X., Atkinson, R., Pandolfi, M., Zeka, A., Le Tertre, A., Analitis, A., Katsouyanni, K., Chanel, O., Pascal, M., Bouland, C., Haluza, D., Medina, S., Goodman, P.G., 2013. Ambient air SO_2 patterns in 6 European cities. Atmos. Environ., 79, 236–247.

Henschel, S., Le Tertre, A., Atkinson, R.W., Querol, X., Pandolfi, M., Zeka, A., Haluza, D., Analitis, A., Katsouyanni, K., Bouland, C., Pascal, M., Medina, S., Goodman, P.G., 2015. Trends of nitrogen oxides in ambient air in nine European cities between 1999 and 2010 Atmos. Environ., 117, 234–241.

Hongisto, M., Sofiev, M., 2004. Long-range transport of dust to the Baltic Sea Region. Int. J. Environ. Pollut., 22(1–2), 72–86.

Hosiokangas, J., Ruuskanen, J., Pekkanen, J., 1999. Effects of soil dust episodes and mixed fuel sources on source apportionment of PM_{10} particles in Kuopio, Finland. Atmos. Environ., 33, 3821–3829.

Huang, G., Brook, R., Crippa, M., Janssens-Maenhout, G., Schieberle, C., Dore, C., Guizzardi, D., Muntean, M., Schaaf, E., Friedrich, R., 2017. Speciation of anthropogenic emissions of non-methane volatile organic compounds: a global gridded data set for 1970–2012. Atmos. Chem. Phys., 17, 7683–7701.

Hurvich, C.M., Tsai, C.L., 1989. Regression and time series model selection in small samples. Biometrika, 76, 297–307.

Huutoniemi, K., Estlander, A., Hahkala, M., Hämekoski, K., Kulmala, A., Lahdes, R., Laukkanen, T., (Eds.). 2006. Savuntarkastajista päästökauppiasiin. Suomen ilmansuojelun historiaa. Ilmansuojeluyhdistys ry. Gummerus, Jyväskylä.

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., Lihavainen, H., 2011. Aerosol black carbon at five background measurement sites over Finland, a gateway to the Arctic. Atmos. Environ., 45, 4042–4050.

Ialongo, I., Hakkarainen, J., Kivi, R., Anttila, P., Krotkov, N.A., Yang, K., Li, C., Tukiainen, S., Hassinen, S., Tamminen, J., 2015. Validation of satellite SO_2 observations in northern Finland during the Icelandic Holuhraun fissure eruption. Atmos. Meas. Tech. Discuss., 8, 599–621.

IMO, 2008. Revised MARPOL Annex VI of the International Convention for the Prevention of Air Pollution from Ships. International Maritime Organization, viewed 9 September 2020 <http://www.imo.org/en/KnowledgeCentre/IndexofIMOResolutions/Marine-Environment-Protection-Committee-(MEPC)/Documents/MEPC.176(58).pdf>

Joffre, S.M., Laurila, T., Hakola, H., Lindfors, V., Konttinen, S., Taalas, P., 1990. On the effects of meteorological factors on air pollution concentrations and depositions in Finland. In: Kauppi, P., Anttila, P., Kenttämies, K., (eds.). Acidification in Finland, Springer-Verlag, Berlin, pp.43–94.

Jones, A.M., Harrison, R.M., 2011. Temporal trends in sulphate concentrations at European sites and relationships to sulphur dioxide. Atmos. Environ., 45, 873–882.

Kalina, J., White, K.B., Scheringer, M., Přibylová, P., Kukučka, P., Audy, O., Klánová, J., 2019. Comparability of long-term temporal trends of POPs from co-located active and passive air monitoring networks in Europe. Environmental Science, Processes and Impacts, 21, 1132.

Karl, M., Jonson, J. E., Uppstu, A., Aulinger, A., Prank, M., Sofiev, M., Jalkanen, J.-P., Johansson, L., Quante, M., and Matthias, V.: Effects of ship emissions on air quality in the Baltic Sea region simulated with three different chemistry transport models, Atmos. Chem. Phys., 19, 7019–7053.

Karlsson, V., Pyy, K., Saari, H., 2007. Measurement uncertainty of sulphur and nitrogen containing inorganic compounds by 1-stage and 2-stage filter-pack methods. Water Air Soil Pollut., 182, 395–405.

Karlsson, P.E., Klingbeg, J., Engardt, M., Andersson, C., Langner, J., Pihl Karlsson, G., Pleijel, H., 2017. Past, present and future concentrations of ground-level ozone and potential impacts on ecosystems and human health in northern Europe. Sci. Total Environ., 576, 22–35.

Kauppi, P., Anttila, P., Kenttämies, K., (Eds.), 1990. Acidification in Finland. Springer Verlag, 1237 p.

Kerminen, V.-M., Niemi, J.V., Timonen, H., Aurela, M., Frey, A., Carbone, S., Saarikoski, S., Teinilä, K., Hakkarainen, J., Tamminen, J., Vira, J., Prank, M., Sofiev, M., Hillamo, R., 2011. Characterization of a volcanic ash episode in southern Finland caused by the Grimsvötn eruption in Iceland in May 2011. Atmos. Chem. Phys., 11, 12227–12239.

Keuken, M., Roemer, M., van den Elshout, S., 2009. Trend analysis of urban NO_2 concentrations and the importance of direct NO_2 emissions versus ozone/NO_x equilibrium. Atmos. Environ., 43, 4780–4783.

Keuken, M.P., Roemer, M.G.M., Zandveld, P., Verbeek, R.P., Velders, G.J.M., 2012. Trends in primary NO_2 and exhaust PM emissions from road traffic for the period 2000–2020 and

implications for air quality and health in the Netherlands. Atmos. Environ., 54, 313–319.

Kukkonen, J., Salmi, T., Saari, H., Konttinen, M., Kartastenpää, R., 1999. Review of urban air quality in Finland. Boreal Env. Res., 4, 55–65.

Kupiainen, K., Pirjola, P., 2011. Vehicle non-exhaust emissions from the tyre-road interface – effect of stud properties, traction sanding and resuspension. Atmos. Environ., 45, 4141–4146.

Kupiainen, K., Ritola, R., Stojiljkovic, A., Pirjola, L., Malinen, A., Niemi, J., 2016. Contribution of mineral dust sources to street side ambient and suspension PM_{10} samples. Atmos. Environ., 147, 178–189.

Kupiainen, K., Tervahattu, H., Räisänen, M., 2003. Experimental studies about the impact of traction sand on urban road dust composition. The Sci. Total Environ., 308, 175–184.

Kupiainen, K., Tervahattu, H., 2004. The effect of traction sanding on urban suspended particles in Finland. Environ. Monit. Assess., 93, 287–300.

Kupiainen K.J., Tervahattu H., Räisänen M., Mäkelä T., Aurela M., Hillamo R., 2005. Size and composition of airborne particles from pavement wear, tires, and traction sanding. Environ. Sci. Technol., 39, 699–706.

Kurtenbach, R., Kleffmann, J., Niedojadlo, A., Wiesen, P., 2012. Primary NO_2 emissions and their impact on air quality in traffic environments in Germany. Environmental Sciences Europe, 24:21.

Kyllönen, K., Vestenius, M., Anttila, P., Makkonen, U., Aurela, M., Wängberg, I., Nerentorp Mastromonaco, M., Hakola, H., 2020. Trends and source apportionment of atmospheric heavy metals at a subarctic site during 1996–2018. Atmos. Environ., 236, (2020) 117644, <doi.org/10.1016/j.atmosenv.2020.117644>.

Kyrö, E.-M., Väänänen, R., Kerminen, V.-M., Virkkula, A., Petäjä, T., Asmi, A., Dal Maso, M., Nieminen, T., Juhola, S., Shcherbinin, A., Riipinen, I., Lehtipalo, K., Keronen, P., Aalto, P.P., Hari, P., Kulmala, M., 2014. Trends in new particle formation in eastern Lapland, Finland: effect of decreasing sulfur emissions from Kola Peninsula. Atmos. Chem. Phys., 14, 4383–4396.

Leino, K., Riuttanen, L., Nieminen, T., Dal Maso, M., Väänänen, R., Pohja, T., Keronen, P., Järvi, L., Aalto, P.P., Virkkula, A., Kerminen, V.-M., Petäjä, T., Kulmala, M., 2014. Biomass-burning smoke episodes in Finland from Eastern European wildfires. Boreal Env. Res., 19 (suppl. B), 275–292.

LUKE, 2019. Residential firewood consumption, Natural Resources Institute Finland, viewed 3 January 2020, <http://statdb.luke.fi/PXWeb/pxweb/fi/LUKE>.

Makra, L., Matyasovszky, I., Guba, Z., Karatzas, K., Anttila, P., 2011. Monitoring the long-range transport effects on urban PM_{10} levels using 3D clusters of backward trajectories. Atmos. Environ., 45, 2630–2641.

Miyazaki, K., Eskes, H., Sudo, K., Boersma, K.F., Bowman, K., Kanaya, Y., 2017. Decadal changes in global surface NO_x emissions from multi-constituent satellite data assimilation, Atmos. Chem. Phys., 17, 807–837.

Monks, P.S., Archibald, A.T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, C., Law, K.S., Mills, G.E., Stevenson, D.S., Tarasova, O., Thouret, V., von Schneidemesser, E., Sommariva, R., Wild, O., Williams, M.L., 2015. Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer. Atmos. Chem. Phys., 15, 8889–8973.

Makkonen, U., 2014. Measurements of inorganic ions and their precursor gases in ambient air. Finnish Meteorological Institute Contributions, 110, 59 pp.

Makkonen, U., Hellén, H., Anttila, P., Ferm, M., 2010. Size distribution and chemical composition of airborne particles in south-eastern Finland during different seasons and wildfire episodes in 2006. Sci. Total Environ., 408, 644–651.

Mavroidis, I., Chaloulakou, A., 2011. Long-term trends of primary and secondary NO_2 production in the Athens area. Variation of the NO_2/NO_x ratio. Atmos. Environ., 45, 38, 6872–6879.

Melkonyan, A., Kuttler, W., 2012. Long-term analysis of NO, NO_2 and O_3 concentrations in North Rhine-Westphalia, Germany. Atmos. Environ., 60, 316–326.

Mielonen, T., Portin, H., Komppula, M., Leskinen, A., Tamminen, J., Ialongo, I., Hakkarainen, J., Lehtinen, K.E.J., Arola, A., 2012. Biomass burning aerosols observed in eastern Finland during the Russian wildfires in summer 2010 — Part 2: Remote sensing. Atmos. Environ., 47, 279–287.

NAPCP, 2019. National Air Pollution Control Programme 2030. Publications of the Ministry of the Environment, 14, 96 p.

Niemi, J.V., Tervahattu, H., Vehkamäki, H., Kulmala, M., Koskentalo, T., Sillanpää, M., Rantamäki, M., 2004. Characterization and source identification of a fine particulate episode in Finland. Atmos. Environ., 38, 5003–5012.

Niemi, J.V., Tervahattu, H., Vehkamäki, H., Martikainen, J., Laakso, L., Kulmala, M., Aarnio, P., Koskentalo, T., Sillanpää, M., Makkonen, U., 2005. Characterization of aerosol particle episodes in Finland caused by wildfires in Eastern Europe. Atmos. Chem. Phys., 5, 2299–2310.

Niemi, J.V., Saarikoski, S., Tervahattu, H., Mäkelä, T., Hillamo, R., Vehkamäki, H., Sogacheva, L., Kulmala, M., 2006. Changes in background aerosol composition in Finland during polluted and clean periods studied by TEM/EDX individual particle analysis. Atmos. Chem. Phys., 6, 5049–5066.

Niemi, J.V., Saarikoski, S., Aurela, M., Tervahattu, H., Hillamo, R., Westphal, D.L., Aarnio, P., Koskentalo, T., Makkonen, U., Vehkamäki, H., Kulmala, M., 2009. Long-range transport episodes of fine particles in southern Finland during 1999–2007. Atmos. Environ., 43, 1255–1264.

Nieminen, T., Asmi, A., Dal Maso, M., Aalto, P., Keronen, P., Petäjä, T., Kulmala, M., Kerminen, V.-M., 2014. Trends in atmospheric new-particle formation: 16 years of observations in a boreal-forest environment. Boreal Env. Res., 19 (suppl. B), 191–214.

O'Driscoll, R., ApSimon, H.M., Oxley, T., Molden, N., Stettler, M.E.J., Thiyagarajah, A., 2016. A Portable Emissions Measurement System (PEMS) study of NO_x and primary NO_2 emissions from Euro 6 diesel passenger cars and comparison with COPERT emission factors. Atmos. Environ., 145, 81–91.

Olstrup, H., Forsberg, B., Orru, H., Nguyen, H., Molnár, P., Johansson, C., 2018. Trends in air pollutants and health impacts in three Swedish cities over the past three decades. Atmos. Chem. Phys., 18, 15705–15723.

Paatero, J., Vesterbacka, K., Makkonen, U., Kyllönen, K., Hellén, H., Hatakka, J., Anttila, P., 2009. Resuspension of radionuclides into the atmosphere due to forest fires. Journal of Radioanalytical and Nuclear Chemistry, 282, 473–476.

Pakkanen, T., Loukkola, K., Korhonen, C., Aurela, M., Mäkelä, T., Hillamo, R., Aarnio, P., Koskentalo, T., Kousa, A., Maenhaut, W., 2001. Sources and chemical composition of atmospheric fine and coarse particles in the Helsinki area. Atmos. Environ., 35, 5381–5391.

Pirjola, L., Kupiainen, K.J., Perhoniemi, P., Tervahattu, H., Vesala, H., 2009. Non-exhaust emission measurement system of the mobile laboratory SNIFFER. Atmos. Environ., 43, 4703−4713.

Pohjola, M., Kousa, A., Kukkonen, J., Härkönen, J., Karppinen, A., Aarnio, P., Koskentalo, T., 2002. The spatial and temporal variation of measured urban PM_{10} and $PM_{2.5}$ in the Helsinki metropolitan area. Water, Air and Soil Pollution: Focus 2(5–6), 189–201.

Portin, H., Mielonen, T., Leskinen, A., Arola, A., Pärjälä E., Romakkaniemi, S., Laaksonen, A., Lehtinen, K.E.J., Komppula, M., 2012. Biomass burning aerosols observed in Eastern Finland during the Russian wildfires in summer 2010 – Part 1: In-situ aerosol characterization. Atmos. Environ., 47, 269–278.

Prank, M., Sofiev, M., Denier van der Gon, H.A.C., Kaasik, M., Ruuskanen, T.M., Kukkonen, J., 2010. A refinement of the emission data for Kola Peninsula based on inverse dispersion modelling, Atmos. Chem. Phys., 10, 10849–10865.

Rafaj, P., Amann, M., Siri, J., Wuester, H., 2014. Changes in European greenhouse gas and air pollutant emissions 1960–2010: decomposition of determining factors. Climatic Change 124, 477–504.

Riuttanen, L., Hulkkonen, M., Dal Maso, M., Junninen, H., Kulmala, M., 2013. Trajectory analysis of atmospheric transport of fine particles, SO_2, NO_x and O_3 to the SMEAR II station in Finland in 1996–2008. Atmos. Chem. Phys., 13, 2153–2164.

Ruoho-Airola, T., 2004. Temporal and regional patterns of atmospheric components affecting acidification in Finland. Finnish Meteorological Institute, Contributions 44.

Ruoho-Airola, T., Anttila, P., Hakola, H., Ryyppö, T., Tuovinen, J.-P., 2015. Trends in the bulk deposition and atmospheric concentration of air pollutants in the Finnish Integrated Monitoring catchment Pallas during 1992–2012. Boreal Env. Res., 20, 553–569.

Räisänen, M., Kupiainen, K., Tervahattu, H., 2003. The effect of mineralogy, texture and mechanical properties of anti-skid and asphalt aggregates on urban dust. Bulletin of Engineering, Geology and the Environment 62, 359–368.

Räisänen, M., Kupiainen, K., Tervahattu, H., 2005. The Effect of mineralogy, texture and mechanical properties of anti-skid and asphalt aggregates on urban dust, Stages II and III. Bulletin of Engineering Geology and the Environment 64, 247–256.

Saarnio, K., Aurela, M., Timonen, H., Saarikoski, S., Teinilä, K., Mäkelä, T., Sofiev, M., Koskinen, J., Aalto, P.P., Kulmala, M., Kukkonen, J., Hillamo, R. 2010. Chemical composition of fine particles in fresh smoke plumes from boreal wild-land fires in Europe. Sci. Total Environ., 408, 2527–2542.

Saarnio, K., Niemi, J.V., Saarikoski, S., Aurela, M., Timonen, H., Teinilä, K., Myllynen, M., Frey, A., Lamberg, H., Jokiniemi, J., Hillamo, R., 2012. Using monosaccharide anhydrides to estimate the impact of wood combustion on fine particles in the Helsinki Metropolitan Area. Boreal Env. Res., 17, 163–183.

Saarikoski, S., Sillanpää, M., Saarnio, K., Hillamo, R., Pennanen, A., Salonen R., 2008. Impact of biomass combustion on urban fine particulate matter in Central and Northern Europe. Water Air Soil Pollut., 191, 265–77.

Saarikoski S., Sillanpää M., Sofiev M., Timonen H., Saarnio K., Teinilä K., Karppinen A., Kukkonen J., Hillamo R., 2007. Chemical composition of aerosols during a major biomass burning episode over Northern Europe in spring 2006: Experimental and modeling assessment. Atmos. Environ., 41, 3577–3589.

Salmi, T., Määttä, A., Anttila, P., Ruoho-Airola, T., Amnell, T., 2002. Detecting trends of annual values of atmospheric pollutants by the Mann-Kendall test and Sen's slope estimates – the Excel template application MAKESENS. Finnish Meteorological Institute, Publications on Air Quality No. 31.

Savolahti, M., Karvosenoja, N., Tissari, J., Kupiainen, K., Sippula, O., Jokiniemi, J., 2016. 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.

Seinfeld, J.H., Pandis, S.N., 1998. Atmospheric chemistry and physics. From air pollution to climate change. John Wiley and Sons. New York. 1326 p.

Shen, H., Huang, Y., Wang, R., Zhu, D., Li, W., Shen, G., Wang, B., Zhang, Y., Chen, Y., Lu, Y., Chen, H., Li, T., Sun, K., Li, B., Liu, W., Liu, J., Tao, S., 2013. Global atmospheric emissions of polycyclic aromatic hydrocarbons from 1960 to 2008 and future predictions. Environ. Sci. Technol., 47, 6415– 6424.

Sillanpää, M., Saarikoski, S., Hillamo, R., Pennanen, A., Makkonen, U., Spolnik, Z., Van Grieken, R., Koskentalo, T., Salonen, R.O., 2005. Chemical composition, mass size distribution and source analysis of long-range transported wildfire smokes in Helsinki. Sci. Total Environ., 350, 119–135.

Sillanpää, M., Hillamo, R., Saarikoski, S., Frey, A., Pennanen, A., Makkonen, U., Spolnik, Z., Van Grieken, R., Braniš, M., Brunekreef, B., Chalbot, M.-C., Kuhlbusch, T., Sunyer, J., Kerminen, V.-M., Kulmala, M. & Salonen, R.O., 2006. Chemical composition and mass closure of particulate matter at six urban sites in Europe. Atmos. Environ., 40S2, 212–223.

Sofiev, M., Kaasik, M., Hongisto, M., 2003. Model simulations of the alkaline dust distribution from Estonian sources over the Baltic Sea Basin. Water Air Soil Pollut., 146, 211–223.

Stohl, A., Berg, T., Burkhart, J.F., Fjæraa, A.M., Forster, C., Herber, A., Hov, Ø., Lunder, C., McMillan, W.W., Oltmans, S., Shiobara, M., Simpson, D., Solberg, S., Stebel, K., Ström J., Tørseth, K., Treffeisen, R., Virkkunen, K., Yttri, K.E., 2007. Arctic smoke – record high air pollution levels in the European Arctic due to agricultural fires in Eastern Europe. Atmos. Chem. Phys., 7, 511–534.

Stojiljkovic, A., Kauhaniemi, M., Kukkonen, J., Kupiainen, K., Karppinen, A., Denby, B.R., Kousa, A., Niemi, J.V., Ketzel, M., 2019. The impact of measures to reduce ambient air PM_{10} concentrations originating from road dust, evaluated for a street canyon in Helsinki. Atmos. Chem. Phys., 19, 11199–11212.

SYKE, 2019a. Emissions of air pollutants in Finland (in Finnish). Finnish Environment Institute, viewed 23 March 2019,

<https://www.ymparisto.fi/fi-FI/Kartat_ja_tilastot/Ilman_epapuhtauksien_paastot>.

SYKE, 2019b. Emissions of air pollutants in Finland (in Finnish). Finnish Environment Institute, viewed 16 September 2019,

<https://www.ymparisto.fi/fi-FI/Kartat_ja_tilastot/Ilman_epapuhtauksien_paastot>.

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

Tervahattu, H., Hongisto, M., Aarnio, P., Kupiainen, K., Sillanpää, M., 2004. Composition and origins of aerosol during a high PM_{10} episode in Finland. Boreal Env. Res., 9, 335–345.

Tervahattu, H., Kupiainen, K.J., Räisänen, M., Mäkelä, T., Hillamo, R., 2006. Generation of urban road dust from anti-skid and asphalt concrete aggregates. Journal of Hazardous Materials, 132, 39–46.

Timonen, H., Aurela, M., Carbone, S., Saarnio, K., Saarikoski, S., Mäkelä, T., Kulmala, M., Kerminen, V.-M., Worsnop, D. R., Hillamo, R., 2010. High time-resolution chemical characterization of the water-soluble fraction of ambient aerosols with PILS-TOC-IC and AMS. Atmos. Meas. Tech., 3, 1063–1074.

Tørseth, K., Aas, W., Breivik, K., Fjæraa, A.M., Fiebig, M., Hjellbrekke, A.G., Lund Myhre, C., Solberg, S., Yttri, K.E., 2012. Introduction to the European Monitoring and Evaluation Programme (EMEP) and observed atmospheric composition change during 1972–2009, Atmos. Chem. Phys., 12, 5447–5481.

UNECE, 1985. Protocol to the 1979 Convention on Long-Range Transboundary Air Pollution on the reduction of sulphur emissions or their transboundary fluxes by at least 30 per cent. United Nations Economic Commission for Europe, Helsinki.

UNECE, 1988. Protocol to the 1979 convention on long-range transboundary air pollution concerning the control of emissions of nitrogen oxides or their transboundary fluxes. United Nations Economic Commission for Europe, Sofia.

Vestreng, V., Myhre, G., Fagerli, H., Reis, S., Tarrasón, L., 2007. Twenty-five years of continuous sulphur dioxide emission reduction in Europe. Atmos. Chem. Phys., 7, 3663–3681.

Waldén, J., 2009. Metrology of gaseous air pollutants. Finnish Meteorological Institute, Contributions 79.

Waldén, J., Bergius, J., Pohjola, V., Laurila, S., Kuronen, P., Wemberg, A., 2008. Field comparison of CO, SO_2, NO, H_2S and O_3 ambient air measurements and field audit 2006 (in Finnish). Finnish Meteorological Institute, Studies No. 2.

Waldén, J., Hillamo, R., Aurela, M., Mäkelä, T., Laurila, S., 2010. Demonstration of the equivalence of $PM_{2.5}$ and PM_{10} measurement methods in Helsinki 2007–2008 (in Finnish). Finnish Meteorological Institute, Studies No. 3.

Waldén, J., Laurila, S., Lusa, K., Kuronen, P., Waldén, T., Anttila, T., 2015. Interlaboratory comparison exercise and field audit 2011 – measurements of CO, SO_2, NO, and O_3 (in Finnish). Finnish Meteorological Institute, Reports 2015:2.

Waldén, J., Talka, M., Pohjola, V., Häkkinen, T., Lusa, K., Sassi, M.-K., Laurila, S., 2004. The field comparison of carbon monoxide, sulphur dioxide and nitrogen monoxide ambient air measurements and a field audit 2002–2003 (in Finnish). Finnish Meteorological Institute, Publications on Air Quality, No. 35.

Waldén, J., Waldén, T., Laurila, S., Hakola, H., 2017. Demonstration of the equivalence of $PM_{2.5}$ and PM_{10} measurement methods in Kuopio 2014–2015. Finnish Meteorological Institute, Reports 2017:1.

Waldén, J., Vestenius, M., 2018.Verification of PM-analyzers for PM_{10} and $PM_{2.5}$ with the PM reference method. Finnish Meteorological Institute, Reports 2018:2.

Wild, R.J., Dubé, W.P., Aikin, K.C., Eilerman, S.J., Neuman, J.A., Peischl, J., Ryerson T.B., Brown, S.S., 2017. On-road measurements of vehicle NO_2/NO_x emission ratios in Denver, Colorado, USA. Atmos. Environ., 148, 182−189.

Yan, Y., Pozzer, A., Ojha, N., Lin, J., Lelieveld, J., 2018. Analysis of European ozone trends in the period 1995–2014. Atmos. Chem. Phys., 18, 5589–5605.

Yoon, J., Pozzer, A., 2014. Model-simulated trend of surface carbon monoxide for the 2001–2010 decade. Atmos. Chem. Phys., 14, 10465–10482.

Yu, Y., Katsoyiannis, A., Bohlin-Nizzetto, P., Brorström-Lundén, E., Ma, J., Zhao, Y., Wu, Z., Tych, W., Mindham, D., Sverko, E., Barresi, E., Dryfhout-Clark, H., Fellin, P., Hung, H., 2019. Polycyclic aromatic hydrocarbons not declining in Arctic air despite global emission reduction. Environ. Sci. Technol., 53, 2375–2382.

Zheng, B., Chevallier, F., Ciais, P., Yin, Y., Deeter, M.N., Worden, H.M., Wang, Y., Zhang, Q., He, K., 2018. Rapid decline in carbon monoxide emissions and export from East Asia between years 2005 and 2016. Environ. Res. Lett., 13, 044007.