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Abstract: Exposure to outdoor air pollutants remains an important concern in Europe, as limit values for NO₂ and PM₁₀ continue to be exceeded. Few studies have addressed the long-term spatial contrasts in PM_{2.5}, PM absorbance, PM_{coarse} and especially ultrafine particles. This scarcity of data hampers the possibility to conduct epidemiological studies, assessing the health relevance of these markers of potentially harmful pollutants.

Air pollution measurements were performed in eight geographically distinct areas of the Swiss Study on Air Pollution and Lung and Heart Diseases in Adults (SAPALDIA) in Switzerland. NO₂ was measured in all eight areas at 40 sites per area, and PM_{2.5}, PM_{2.5} absorbance, PM₁₀ and ultrafine particles (particle number concentration (PNC) and lung deposited surface area (LDSA)) were measured in 4 of these areas, at a subset of 20 out of 40 sites. Each site was sampled three times during different seasons of the year, using the same equipment, sampling protocols and the same central facilities for analysis of samples. We assessed the spatial variability between areas and between individual sites, as well as pollution contrasts between the seasons and correlations between different pollutants.

Concentrations in the larger cities were generally higher than in smaller towns and rural and alpine areas. Within-area spatial variability contrasts (defined as the ratio between the 90th and 10th percentile) were highest for NO₂ (3.14), moderate for PM_{coarse} (2.19), PNC (2.00) and PM_{2.5} absorbance (1.94), and lowest for LDSA (1.63), PM_{2.5} (1.50) and PM₁₀ (1.46). Concentrations in the larger cities were generally higher than in smaller towns and rural and alpine areas, and were higher in the winter than in the summer and intermediate seasons, for all pollutants. Between-area differences accounted for more variation than within-area differences for all pollutants except NO₂ and PM_{coarse}. Despite substantial within-area contrasts for PNC and LDSA, 74.7% and 83.3% of the spatial variance was attributed to between-area variability, respectively. 65.9% and PM_{coarse} (55.1%), while for the other pollutants the majority of spatial variation occurred between areas, especially for PNC (74.7%) and LDSA (83.3%). Despite that, within area contrasts, calculated as the ratio between the 90th and 10th percentile, were lower for PM_{2.5} and PM₁₀ than for PNC and LDSA, and highest for NO₂. Coefficients of determination between long-term adjusted pollutants were high ($R^2 > 0.70$) between NO₂, PM_{2.5} absorbance, PNC and LDSA and between PM_{2.5} and PM₁₀. The measurement of spatial

Spatial and temporal variability of ultrafine particles, NO₂, PM_{2.5}, PM_{2.5} absorbance, PM₁₀ and PM_{coarse} in Swiss study areas

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Abstract

Exposure to outdoor air pollutants remains an important concern in Europe, as limit values for NO₂ and PM₁₀ continue to be exceeded. Few studies have addressed the long-term spatial contrasts in PM_{2.5}, PM absorbance, PM_{coarse} and especially ultrafine particles. This scarcity of data hampers the possibility to conduct epidemiological studies, assessing the health relevance of these markers of potentially harmful pollutants.

Air pollution measurements were performed in eight geographically distinct areas of the Swiss Study on Air Pollution and Lung and Heart Diseases in Adults (SAPALDIA) in Switzerland. NO₂ was measured in all eight areas at 40 sites per area, and PM_{2.5}, PM_{2.5} absorbance, PM₁₀ and ultrafine particles (particle number concentration (PNC) and lung deposited surface area (LDSA)) were measured in 4 of these areas, at a subset of 20 out of 40 sites. Each site was sampled three times during different seasons of the year, using the same equipment, sampling protocols and the same central facilities for analysis of samples. We assessed the spatial variability between areas and between individual sites, as well as pollution contrasts between the seasons and correlations between different pollutants.

Within-area spatial contrasts (defined as the ratio between the 90th and 10th percentile) were highest for NO₂ (3.14), moderate for PM_{coarse} (2.19), PNC (2.00) and PM_{2.5} absorbance (1.94), and lowest for LDSA (1.63), PM_{2.5} (1.50) and PM₁₀ (1.46). Concentrations in the larger cities were generally higher than in smaller towns and rural and alpine areas, and were higher in the winter than in the summer and intermediate seasons, for all pollutants. Between-area differences accounted for more variation than within-area differences for all pollutants except NO₂ and PM_{coarse}. Despite substantial within-area contrasts for PNC and LDSA, 74.7% and 83.3% of the spatial variance was attributed to between-area variability, respectively. Coefficients of determination between long-term adjusted pollutants were high ($R^2 > 0.70$) between NO₂, PM_{2.5} absorbance, PNC and LDSA and between PM_{2.5} and PM₁₀. The measurement of spatial patterns for this large range of outdoor air pollutants will contribute to a highly standardized estimation of individual long-term exposure levels for SAPALDIA cohort participants.

Highlights

We measured NO₂, PM_{2.5}, PM_{2.5} absorbance, PM₁₀ and UFP in Switzerland using standard methodology

We studied spatial and seasonal contrasts of these pollutants and PM_{coarse} across study areas

PNC and LDSA were highly correlated with each other and with NO₂ and PM_{2.5} absorbance

More spatial variance occurred within than between study areas for NO₂ and PM_{coarse}, but not for other pollutants

The air quality standard for PM_{2.5} proposed by WHO was exceeded at over 90% of measurement sites

Keywords: SAPALDIA; NO₂; particulate matter; PM_{2.5}; PM₁₀; absorbance; coarse particles; PNC; LDSA; air pollution; traffic; spatial contrast; Switzerland; seasonal contrast; exposure assessment

Abbreviations:

CV: coefficient of variation

EU: European Union

LDSA: Lung Deposited Surface Area of ultrafine particles (μm² / cm³)

LUR: Land Use Regression

PM_{2.5}: mass concentration of particles less than 2.5 μm in size

PM_{2.5} absorbance: measurement of the blackness of PM_{2.5} filters, this is a proxy for elemental carbon, which is the dominant light absorbing substance

PM₁₀: mass concentration of particles less than 10 μm in size

PM_{coarse}: mass concentration of the coarse fraction of particles between 2.5 μm and 10 μm in size

RH: relative humidity

PNC: Particle Number Concentration (units per cm³)

SAPALDIA: Swiss Cohort Study on Air Pollution and Lung and Heart Diseases in Adults

UFP: UltraFine Particles

1 Introduction

Numerous recent epidemiological studies have shown adverse health effects associated with long-term exposure to ambient air pollutants (Künzli, 2010; Rückerl et al., 2011). Nitrogen dioxide (NO₂) and the mass concentrations of all particles smaller than 2.5 µm (PM_{2.5}) or 10 µm (PM₁₀) are some of the most frequently used markers of ambient air pollution and have been associated with many long-term health endpoints. These pollutants are regulated and, thus, routinely monitored across Europe and the US. ((EEA), 2009; Cyrys et al., 2012; Eeftens et al., 2012). However, with the changes in emission control technologies and fuel formulation as well as a revival of wood combustion in some areas, those pollutants may not fully capture the health relevant aspects of air quality. This may call for the use of markers that are not yet regulated or routinely monitored. For example, black carbon particles are a marker for diesel exhaust and has been associated with morbidity and mortality.(EPA, 2012; Janssen et al., 2011) The coarse fraction of PM measures between 2.5 to 10 µm in diameter, results mostly from resuspension and abrasion and – depending on the local mix – has partly different composition and toxicity than its smaller counterparts(Brunekreef and Forsberg, 2005). The primary ultrafine particles (UFP), measuring less than 0.1 µm in diameter, are found in high concentrations in vehicle exhaust. UFP penetrate deeply into the respiratory system and may translocate directly into the blood and the brain. Because of these characteristics, UFP may exert health effects independent of larger particles, and through different pathways (Peters et al., 2006). However, so far there is a lack of epidemiological studies to quantify the exposure-response relationship for UFP (World Health Organization (WHO), 2006), and little consensus on the most health-relevant exposure metric (Wittmaack, 2007). The debate on regulating UFP is further jeopardized by the limited amount of monitoring data that is available both nationally and internationally (Heal et al., 2012; Hoek et al., 2009).

For PM, a recent study confirms for Europe that mortality rates are still affected at exposure levels well below the EU limits (Beelen et al., 2013), adding support for both the science based WHO guideline values to protect public health and the need for continued research not only in highly polluted areas but also in regions with low to moderate pollution to guide future global policy. The Swiss Cohort Study on Air Pollution and Lung and Heart Diseases in Adults (SAPALDIA), initiated in 1991, is one of the landmark studies investigating the long-term effects of air pollution. Its second follow-up (since baseline) finished in 2011.

This air pollution assessment was carried out within the framework of the third follow-up of SAPALDIA. The purpose of this paper is to describe and discuss the spatial and seasonal contrasts for outdoor NO₂, PM_{2.5}, PM_{2.5} absorbance, PM₁₀, PM_{coarse} and UFP within and between the SAPALDIA study areas. For the first time, we report these spatial and seasonal variability for both the particle number concentration (PNC) as well as the lung deposited surface area of these ultrafine particles (LDSA). Long-term effects of UFP exposure have not been studied extensively, partly because of a lack of spatially resolved concentration data.(Hoek et al., 2010), and it is important to investigate how long-term spatial contrasts in UFP are related to those of other, more traditional pollutants. From a toxicological point of view, the surface area may be a highly attractive marker for the health relevance of ambient particles (Duffin et al., 2002; Wittmaack, 2007). However, this metric has not been used in epidemiological studies on long-term health effects so far. To better understand to what extent single pollutants may be markers for other pollutants, we explored the seasonal variation of the pollutants, and the spatial correlations between them. Given the focus of SAPALDIA on the long-term health effects of air pollution, we derived for all pollutants an estimate of the long-

term mean, which provides the primary base for the development of long-term spatial models to estimate air pollution exposure for SAPALDIA participants. Results for daily and hourly UFP variation, time resolved short-term correlations with other pollutants and the comparison between data from the novel device used in our study (MiniDisc) and routine monitoring data are reported in a separate paper.(Meier et al., 2015)

2 Methods

2.1 Study area and design of the sampling campaign

Air pollution measurements were taken in the eight geographically diverse study areas of the SAPALDIA cohort, which include major cities (Basel, Geneva), medium-size cities (Aarau, Lugano), rural areas (Payerne, Wald) and alpine areas (Davos, Montana) (Figure 1). NO₂ was measured in all eight study areas at 40 sites per area. In Basel, Geneva, Lugano and Wald, the additional pollutants PM_{2.5}, PM₁₀, and UFP were measured in 20 out of 40 sites. Measurements in all areas were conducted by the same team, using identical equipment, sampling protocols and criteria for the selection of sampling sites. Samples were analysed centrally at Swiss Tropical and Public Health Institute in Basel (PM mass and absorbance) and Passam laboratory (NO₂).

Figure 1: Placement of the air pollution monitors in the study areas of the SAPALDIA 3 air pollution sampling campaign.

2.2 Selection of sampling sites

Forty sampling sites were selected in each of the study areas. In Aarau, Davos, Montana and Payerne, where only NO₂ was measured, street level 40 sites were chosen to: 1) cover the geographic extent of the cohort within each study area, and 2) reflect a range of pollution levels by ensuring a large range in determinants, such as population density, nearby traffic intensity, altitude and proximity to industry. In Basel, Geneva, Lugano and Wald, the 20 sites where only NO₂ was measured were selected in the same manner. The 20 sites where additional pollutants PM_{2.5}, PM₁₀ and PNC were measured were chosen at the homes of SAPALDIA cohort participants. Like the NO₂-only sites, these homes were chosen to include a wide range of pollution levels. Our site selection approach generally follows that used in various other sampling campaigns which form the basis for land-use regression modelling (Cyrus et al., 2012; Eeftens et al., 2012; Hoek et al., 2002; Lewné et al., 2004). Regional background sites were located outside of the towns and away from traffic sources, while urban background sites were situated in the towns, but away from major roads. Street sites were selected at sites and homes which were located on major streets with different intensity and street configuration. Since road traffic is known to be a major source of spatial contrast, we chose to over-represent street sites. However, in this paper, we do not focus on site classification, because of the geographic dissimilarity of the study areas (e.g. some areas contained no roads with traffic intensity higher than 5,000 vehicles/24 hours) and the fact that the traffic intensity is distributed on a continuous scale. Rather than setting hard criteria for site classification, we selected typical major streets, representative for each of the study areas, choosing city ring roads and major arteries into or through the town centre. Online Supplement A shows the traffic intensity around the sites within a 100m buffer, and distance to the nearest major road, by study area.

In each study area, we selected a centrally located routine monitoring site to conduct additional reference measurements for NO₂, PM_{2.5}, PM₁₀ and UFP. This reference site was located away from traffic influences, and served two main purposes: 1) To evaluate compliance of our instruments with the reference methods, and 2) to adjust for seasonal variation of concentrations between the measurement rounds to ultimately estimate the annual mean. In four study areas, a second suitable routine monitoring site was available. This site was used for co-location of samplers, and for validation of the seasonal adjustment procedure.

2.3 Sampling schedule

Repeated air pollution measurements were conducted at the selected sites during three seasons (winter, summer and spring). In Aarau, Davos, Montana and Payerne, where only NO₂ was measured, 14-day samples were taken simultaneously at all 40 sites. In Basel, Geneva, Lugano and Wald, 20 sites were measured simultaneously, 10 of them for NO₂-only and 10 for NO₂ and PM. Both NO₂ and PM measurements ran for 14 days, after which equipment was redeployed to the remaining 20 locations. For PNC and LDSA, only 5 sites could be measured simultaneously, typically for one week each, due to the limited availability of monitors. In addition, all pollutants (NO₂, PM_{2.5}, PM₁₀, PNC and LDSA) were measured at area-specific reference stations during all sampling periods. A schematic overview of the sampling periods is given in Online Supplement B.

2.4 Sampling equipment and methodology

NO₂ was measured using passive diffusion samplers (Passam AG, Männedorf, Switzerland) and analysed in the Passam AG laboratory, where the collected NO₂ was determined photospectrometrically using the Saltzmann method.

PM_{2.5} and PM₁₀ were collected on filters using Harvard Impactors, designed to collect particles at a flow rate of 4.0 l min⁻¹. One MEDO pump per site (VP0125, MEDO USA, Roselle, IL, USA) was operated on two separate timers, which consecutively routed power to the PM_{2.5} and PM₁₀ inlet for 10 minutes each hour, thereby effectively sampling an air volume of 13.5 m³ over 14 days for each size fraction. Teflon filters (Pall Corporation, Port Washington, NY, USA) were pre- and post-weighed on a micro-balance (Mettler Toledo, Greifensee, Switzerland), following procedures prescribed by the US Environmental Protection Agency (Agency, 1998). Specifications of the weighing facility have been published previously (Allen et al., 2001). Filters were conditioned to the temperature (20-23°C ± 2°C) and relative humidity (30-40%±5%) of the weighing room for at least 24 hours prior to weighing. Filters were excluded from further analysis if: serious handling problems were reported (14 filters), if the average flow deviated >10% from the target of 4.0 l min⁻¹ (2), the sampling duration was less than one week (4), the total runtime was less than 25.2 hours (equivalent to 90% of a one-week sample) (2), or if the PM₁₀ mass was less than the PM_{2.5} mass (2). The coarse fraction was determined by subtracting the PM_{2.5} mass per m³ concentration from the PM₁₀ concentration at each occasion where both were available.

Reflectance was measured on PM_{2.5} filters using a smoke stain reflectometer (EEL Model 43D, Diffusion Systems Ltd., London, U.K.). All filters were measured twice, and the average was transformed into absorbance using Equation 1, following ISO, 1993. ((ISO), 1993)

$$PM_{2.5} \text{ absorbance} = \frac{A}{2V} \times \ln \left(\frac{R_B}{R_S} \right)$$

Equation 1: Calculation of PM_{2.5} absorbance, where A is the deposition area on the filter (determined to be 6.39*10⁻⁴ m²), V is the sampling volume in m³, R_B and R_S are the average reflectance of the blank filters (set at 100%) and sample filters, respectively.

Measurements of ultrafine particles were conducted with Miniature Diffusion Size Classifier (MiniDiSC) (Fachhochschule Nordwestschweiz, Switzerland) (Fierz et al., 2011), measuring particles between 10 and 300 nm, with a flow rate of 1.0 l/min. This is a larger size range than the general definition of UFP, namely particles smaller than 0.1 µm. However, this smaller fraction dominates the total particle number count (Sioutas et al., 2005). Raw data collected at 1-second logging intervals were processed with the provided software and averaged over one minute. Individual minute-values were excluded if: 1) the average particle size was smaller than 10 or larger than 300 nm, corresponding to the calibrated measurement range of the device (Meier et al., 2012), 2) the flow deviated more than 10% or 3) there was an error related to the functioning of the instrument (negative values in the diffusion or filter stage, low corona voltage, dirt on the counter-electrode, low battery charge or high offset on diffusion or filter zero). Minute-values were further averaged into hourly means, which were excluded if more than 20% of minute-values were missing. Entire measurements were excluded if the total sampling duration was less than 5.6 days (80% of 7 days).

2.5 Quality assurance/quality control

For NO₂, PM_{2.5}, PM_{2.5} absorbance and PM₁₀, blanks and duplicates for time-integrated pollutants were taken in each area in each season. Equipment was also co-located with at least one routine monitoring station in the study area.

Because of the limited availability of miniDiSCs, simultaneous test measurements with all devices were run in the laboratory, with all devices running simultaneously. This was done 12 times throughout the 2-year measurement campaign. In addition, MiniDisc samplers were co-located with condensation particle counters at Basel St Johann (CPC3022A, TSI Inc., MN, USA, particle size range 7nm-1000nm) and Lugano University (CPC3775, TSI Inc., MN, USA, particle size range 4nm-1000nm).

2.6 Adjustment for temporal variability

In Switzerland, air pollution concentrations follow typical patterns of seasonal variation. Our measurements at each site cover only part of sampling period of 2011-2012. NO₂, PM_{2.5}, PM_{2.5} absorbance, PM₁₀ and PM_{coarse} measurements covered a total of 6 weeks per site, while PNC and LDSA measurements typically covered 3 weeks per site. Yet, for each site, we aimed to derive a reliable long-term mean, representative of the entire two-year period using continuous data from a reference site. In addition, we derived season-specific long-term means, representative of the summer season (27 May to 26 September), the intermediate seasons (27 September – 12 November and 13 March – 26 May) and the winter season (13 November – 12 March). Taking into account the temporal variation enabled a direct comparison of bi-annual and season-specific average concentrations both within and between study areas. Typically, the seasonal patterns are a regional phenomenon, and therefore the continuous measurements taken at one reference station are assumed to reflect the seasonal variability for the entire study area.

Temporally corrected concentrations for site *i* were calculated by multiplying the uncorrected concentration at site *i* in period *j* by the ratio between the long-term concentration measured at the reference site, and the concentration measured at that same reference site during period *j* (typically two weeks for NO₂, PM_{2.5}, PM_{2.5} absorbance, PM₁₀ and PM_{coarse} and one week for PNC and LDSA).

$$\text{Corrected concentration}_{i,j} = \frac{\text{longterm refsite concentration}}{\text{refsite concentration}_j} \times \text{uncorrected concentration}_{i,j}$$

Measurements in Basel, Davos, Geneva and Payerne were conducted in 2011 and in Aarau, Lugano, Montana and Wald in 2012. For NO₂, PM_{2.5}, PM_{2.5} absorbance, PM₁₀ and PM_{coarse}, long-term averages were calculated over the entire two-year period (2011-2012). To derive bi-annual mean concentrations for each site, the long-term refsite concentration was equal to the bi-annual mean at the reference site. Each of the three measurements was then adjusted, and subsequently averaged to one bi-annual average per site. To derive season-specific mean concentrations for each site, the long-term refsite concentration was equal to the summer, intermediate or winter mean, and three seasonally adjusted measurements were derived for each site. We used routine monitoring data for the temporal adjustment, because no measurements were available from the dedicated campaign between the sampling rounds. For PM_{2.5}, PM_{2.5} absorbance and PM_{coarse}, which were not routinely measured at all reference sites, reference site data were estimated from other pollutants using area-specific linear regression with the most correlated pollutant (details provided in Online Supplement C). To validate the use of routine monitoring measurements for the purpose of seasonal adjustment, we compared if temporal adjustments were different if we used a different routine monitoring

station, or if we used difference adjustment instead of ratio-adjustment. Details are provided in Online Supplement C.

For PNC and LDSA, no routine monitoring data were available which covered all areas and seasons. Therefore, the average long-term concentration at the reference site was calculated over the 12-week measurement period, which was available for each area and assumed to be representative for the entire 2011-2012 period. Long-term summer, intermediate and winter average concentrations were calculated over the 4-week periods within each season and assumed to be representative for each season, respectively.

2.7 Statistical analyses

In all analyses, we used temporally adjusted concentrations from all sampling sites where at least two out of three measurements were available. All analyses were performed in SAS 9.3. Averages and ranges for each pollutant were calculated by study area. Pollutant contrast were calculated as the P90/P10 ratio for each area separately and by pooling all areas together. The mean of area-specific P90/P10 contrasts was also calculated. The apportionment of variance was done using PROC MIXED of SAS using the REML method in two ways: 1) on the bi-annual average concentrations, considering only between and within-area spatial variability while ignoring seasonal variation and 2) on the season-adjusted concentrations, thereby considering three components of variance: between areas, within areas (between sites) and within sites (between seasons). We evaluated the relationships between pollutants as R^2 . Median ratios between pollutants were determined by first calculating the ratio between long-term adjusted concentrations for each site individually, and then taking the median.

3 Results

3.1 Quality assurance/quality control

Blanks for the time-integrated pollutants were generally very low resulting in low detection limits for NO_2 ($<0.8 \mu\text{g}/\text{m}^3$ for most areas), PM mass ($<1.2 \mu\text{g}/\text{m}^3$) and PM absorbance ($<0.017 \cdot 10^{-5} \text{ m}^{-1}$), and few observations under the detection limit (Online Supplement C, Table C.1). Duplicates agreed well with one another for NO_2 (coefficient of variation (CV)=4.5%) and PM absorbance (5.1%), but the agreement was lower for PM mass (28.6%) (Online Supplement C, Table C.2). However, for PM absorbance and PM mass, only three valid duplicate pairs were available. Our measurements showed good to excellent agreement with routine monitors for NO_2 ($R^2 > 0.88$, maximum average deviance $6.8 \mu\text{g}/\text{m}^3$), $\text{PM}_{2.5}$ ($R^2 > 0.99$, maximum average deviance $1.3 \mu\text{g}/\text{m}^3$) and PM_{10} ($R^2 > 0.90$, maximum average deviance $5.2 \mu\text{g}/\text{m}^3$). PNC was also highly comparable to co-located CPC monitors in Basel and Lugano, with generally high correlations between hourly (R^2 0.70-0.83), daily (R^2 0.87-0.94) and seasonal ($R^2=1.00$) average values (Online Supplement C, Table C.3, Figures C.2, C.3, C.4). The CPC3022A at Basel St Johann (particle size range 7nm-1000nm) measured slightly fewer particles than the MiniDiSC (10nm-300nm) despite its larger particle size range. The CPC3775 at Lugano University samples an even wider particle size range (4nm-1000nm) and did measure slightly more particles than the MiniDiSC (Online Supplement C, Table C.3).

3.2 Adjustment for long-term and seasonal variability

Site-specific averages of bi-annually adjusted concentrations correlated highly with the averages taken over the unadjusted measurements for NO_2 , and moderately to highly for $\text{PM}_{2.5}$ absorbance (Online Supplement D, Table D.3). For $\text{PM}_{2.5}$, PM_{10} , $\text{PM}_{\text{coarse}}$, PNC and LDSA, which are known to exhibit more temporal variation, we found low to moderate correlations between unadjusted and long-term adjusted average concentrations. There were no large differences depending on which reference station in the study area was used for the adjustment (Online Supplement D, Table D.3) or whether we used ratio-adjustment or difference-adjustment (data not shown). All data shown in this paper are temporally adjusted and are representative of the bi-annual (2011-2012) or seasonal (summer, intermediate, winter) concentration.

3.3 Spatial and seasonal contrasts within and between study areas

Figure 2 shows the spatial and seasonal variability between and within all study areas for the different pollutants. Within-area contrasts and seasonal deviations from the long-term mean are quantified in Table 1.

For NO_2 , the highest concentrations were found in Geneva and Lugano, and lowest in the alpine and rural areas Montana and Payerne (Figure 2). Generally, concentrations were highest in the larger cities (particularly Geneva and Lugano), and lowest in the alpine and rural areas Montana and Payerne. Despite the modest absolute concentrations, within-area relative contrasts were highest in Davos and Wald (Table 1). Within-area NO_2 contrasts were relatively high, compared to other pollutants, and were somewhat lower in winter than in the other seasons (Table 1). NO_2 concentrations were generally similar over the course of a year in all areas, and only Lugano showed substantially higher concentrations in summer and lower concentrations in winter (Table 1).

$\text{PM}_{2.5}$ and PM_{10} concentrations were generally highest in Lugano and lower in the other areas (Figure 2). Within-area contrasts were low compared to other pollutants and were similar between study areas (Table 1). In most areas, both $\text{PM}_{2.5}$ and PM_{10} concentrations were substantially higher in winter than during the other seasons (Table 1). These seasonal contrasts were less strong for PM_{10} . In the rural area Wald, wintertime $\text{PM}_{2.5}$ and PM_{10} concentrations were elevated more than in the other areas, likely because of woodburning.

Long-term average $\text{PM}_{2.5}$ absorbance concentrations were substantially higher in Lugano and Geneva than in Basel, and much higher than in Wald (Figure 2). Within area contrasts were moderate, though lower than for NO_2 (Table 1). Seasonal variability was low, but concentrations were generally lowest in the intermediate season and highest in winter (Table 1).

Like PM_{10} and $\text{PM}_{2.5}$, $\text{PM}_{\text{coarse}}$ showed the highest concentrations in Lugano and the lowest in Wald (Figure 2), but in contrast to $\text{PM}_{2.5}$ and PM_{10} , within-area contrasts were higher for the coarse fraction (Table 1). Seasonal variability of $\text{PM}_{\text{coarse}}$ was not strong, but contrary to other pollutants, we found somewhat lower concentrations in the wintertime (Table 1).

The highest PNC and LDSA values were found in Geneva and Lugano, and the lowest were measured in Wald (Figure 2). Within-area spatial contrasts for PNC and LDSA were moderate on average, and slightly lower for LDSA than for PNC, but showed substantial differences between areas (Table 1). Seasonal variability for both PNC and LDSA was larger than for the other pollutants, and levels increased notably in the winter period (Table 1).

3.4 Variance apportionment

Table 2 shows the apportionment of variance between areas and sites for all pollutants, based on bi-annual means (2011-2012) means and seasonal means.

If we focussed on spatial variability in bi-annual means, within-area variability was 65.9% and 55.1% of the total for NO_2 and $\text{PM}_{\text{coarse}}$, respectively, accounting for the majority of spatial variability. For the other pollutants, the majority of spatial variability was observed between study areas, with especially high between-area variability for PNC and LDSA, despite the presence of substantial within-area contrasts, which were actually higher than for $\text{PM}_{2.5}$ or PM_{10} (Table 1).

When considering spatial as well as seasonal variability, seasonal variation became the most important variance factor for $\text{PM}_{2.5}$, PM_{10} , $\text{PM}_{\text{coarse}}$, PNC and LDSA. In comparison to the large seasonal variability, the within-area variance became (nearly) negligible for $\text{PM}_{2.5}$, PM_{10} , PNC and LDSA, while within-area contrasts contributed substantially to spatial variability on the long term. For NO_2 , within-area variation remained the most important source of variance, while for $\text{PM}_{2.5}$ absorbance, most variance was attributed to differences between areas.

Figure 2: Within-area distribution of temporally adjusted concentrations reflecting the biannual mean (2011-2012) (grey), summer mean (red), intermediate season mean (yellow) and winter mean (blue) for NO_2 , $\text{PM}_{2.5}$, $\text{PM}_{2.5}$ absorbance, PM_{10} , $\text{PM}_{\text{coarse}}$, PNC and LDSA. Median, 25th and 75th percentiles are shown by the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points. Pooled data for 8 areas (Aarau, Basel, Davos, Geneva, Lugano, Montana, Payerne and Wald) and 4 areas (Basel, Geneva, Lugano and Wald) are shown on the right side of each graph.

Table 1: Within-area contrasts (ratio of 90th/10th percentile) and differences of seasonal average concentration (95% confidence interval) to the long-term average by area (all areas combined) for NO₂, PM_{2.5}, PM_{2.5} absorbance, PM₁₀, PM_{coarse}, PNC and LDSA in the SAPALDIA 3 study areas.

Pollutant	Area	n	Within-area contrast (ratio of 90 th /10 th percentile)				Differences of seasonal average concentration from the annual mean		
			Whole year	Summer	Intermediate	Winter	Summer	Intermediate	Winter
NO ₂ (µg/m ³)	Aarau	40	2.5	3.8	2.7	1.8	-1.1 (-4.4, 3.0)	0.1 (-2.9, 3.5)	0.7 (-1.9, 3.6)
	Basel	40	2.3	3.1	2.3	1.7	0.2 (-3.1, 4.0)	-0.7 (-3.5, 2.5)	0.4 (-2.0, 3.1)
	Davos	38	5.2	4.8	5.8	3.5	2.7 (-2.1, 8.8)	0.2 (-4.2, 6.0)	-3.5 (-6.7, 0.6)
	Geneva	38	2.8	2.8	3.2	2.4	-2.4 (-6.3, 2.2)	1.5 (-3.2, 7.2)	0.0 (-3.9, 4.6)
	Lugano	37	2.3	3.2	2.2	1.6	6.0 (0.3, 12.6)	-2.1 (-6.2, 2.6)	-4.5 (-7.4, -1.2)
	Montana	40	3.0	2.9	3.8	2.5	-0.9 (-3.2, 1.8)	0.8 (-1.9, 4.1)	-0.1 (-2.3, 2.5)
	Payerne	40	2.2	2.8	2.3	1.5	1.9 (-0.3, 4.6)	1.1 (-0.8, 3.4)	-3.3 (-4.4, -2.1)
	Wald	39	4.8	4.7	5.9	3.3	0.3 (-3.1, 4.8)	-1.4 (-4.4, 2.4)	0.5 (-2.5, 4.1)
	Average 8 areas ^{a,c}	312	3.1	3.5	3.5	2.3			
	Average 4 areas ^{a,d}	154	3.0	3.4	3.4	2.2			
	Pooled 8 areas ^{b,c}	312	3.2	4.2	3.7	3.0	0.8 (-2.8, 5.2)	-0.1 (-3.4, 4.0)	-1.2 (-3.9, 1.9)
PM _{2.5} (µg/m ³)	Basel	20	1.5	1.9	1.8	1.4	-0.78 (-2.17, 0.8)	-1.6 (-3.0, 0.1)	2.7 (1.2, 4.3)
	Geneva	18	1.3	1.4	1.4	1.5	-2.2 (-3.0, -1.4)	0.4 (-0.75, 1.7)	1.8 (0.5, 3.2)
	Lugano	17	1.4	1.6	1.7	1.5	2.7 (0.6, 5.1)	-1.1 (-3.1, 1.1)	-2.1 (-4.3, 0.5)
	Wald	19	1.8	1.3	2.1	1.8	-4.2 (-4.9, -3.3)	-4.1 (-5.1, -2.9)	8.7 (5.5, 12.5)
	Average 4 areas ^{a,d}	74	1.5	1.5	1.8	1.6			
	Pooled 4 areas ^{b,d}	74	1.7	2.7	2.5	1.6	-1.1 (-2.4, 0.3)	-1.6 (-3.0, 0.0)	2.8 (0.7, 5.1)
PM _{2.5} absorbance (10 ⁻⁵ m ⁻¹)	Basel	20	1.5	1.9	1.9	1.3	-0.11 (-0.20, 0.01)	-0.08 (-0.16, 0.02)	0.21 (0.12, 0.31)
	Geneva	18	1.9	1.8	2.1	2.4	-0.09 (-0.25, 0.10)	-0.10 (-0.28, 0.11)	0.10 (-0.12, 0.38)
	Lugano	17	1.8	2.4	1.7	1.3	0.37 (0.10, 0.69)	-0.32 (-0.45, -0.17)	0.02 (-0.13, 0.20)
	Wald	19	2.6	2.1	3.4	3.2	0.07 (-0.04, 0.20)	-0.12 (-0.20, -0.01)	0.02 (-0.10, 0.17)
	Average 4 areas ^{a,d}	74	1.9	2.1	2.3	2.0			
	Pooled 4 areas ^{b,d}	74	3.5	4.3	3.8	3.4	0.06 (-0.10, 0.25)	-0.15 (-0.27, -0.01)	0.09 (-0.06, 0.26)
PM ₁₀ (µg/m ³)	Basel	20	1.3	1.5	1.4	1.2	-0.6 (-1.9, 0.7)	-1.3 (-2.8, 0.4)	2.2 (1.1, 3.3)
	Geneva	18	1.4	1.6	1.5	1.3	-0.8 (-2.5, 1.0)	1.6 (-0.7, 4.2)	-0.8 (-2.2, 0.8)
	Lugano	17	1.5	1.3	1.5	1.7	0.9 (-1.6, 3.6)	-0.4 (-2.4, 1.8)	-0.8 (-3.9, 2.9)
	Wald	19	1.7	1.7	1.3	1.9	-4.4 (-5.6, -3.2)	-2.8 (-4.6, -0.7)	7.6 (4.8, 10.8)
	Average 4 areas ^{a,d}	74	1.5	1.6	1.4	1.5			
	Pooled 4 areas ^{b,d}	74	1.7	2.3	2.1	1.5	-1.3 (-2.9, 0.5)	-0.7 (-2.6, 1.4)	2.1 (-0.1, 4.4)
PM _{coarse} (µg/m ³)	Basel	20	2.2	4.7	11.7	3.1	-1.0 (-2.7, 1.9)	-0.4 (-1.8, 1.6)	-0.6 (-1.5, 0.6)
	Geneva	18	1.8	1.7	2.0	3.0	1.2 (0.1, 2.5)	1.0 (-0.3, 2.5)	-2.4 (-3.2, -1.4)
	Lugano	17	2.4	2.3	3.4	2.1	0.2 (-1.2, 1.8)	-0.3 (-1.8, 1.7)	-0.2 (-1.6, 1.6)
	Wald	19	2.4	1.8	3.1	5.1	-0.4 (-1.1, 0.4)	0.7 (-0.6, 2.3)	-1.3 (-2.1, -0.3)
	Average 4 areas ^{a,d}	74	2.2	2.6	5.1	3.4			
	Pooled 4 areas ^{b,d}	74	2.5	2.7	2.8	3.6	0.0 (-1.2, 1.6)	0.2 (-1.1, 2.0)	-1.1 (-2.1, 0.1)
PNC (particles / cm ³)	Basel	17	1.4	1.5	1.3	1.3	-2010 (-2797, -1145)	511 (-320, 1410)	645 (-219, 1580)
	Geneva	16	2.1	1.6	1.8	2.4	-1989 (-4221, 694)	-3141 (-5309, -501)	5071 (1023, 10124)
	Lugano	16	1.7	1.5	1.8	1.8	-6425 (-7685, -4981)	-4916 (-6401, -3208)	8971 (5234, 13356)

	Wald	18	2.9	2.0	2.5	2.8	-1251 (-2003 , -338)	100 (-1051 , 1548)	1171 (-461 , 3330)
	Average 4 areas ^{a,d}	67	2.0	1.7	1.9	2.1			
	Pooled 4 areas ^{b,d}	67					-2698 (-3983 , -1170)	-1448 (-2839 , 187)	3128 (656 , 6135)
LDSA ($\mu\text{m}^2/\text{cm}^3$)	Basel	17	1.2	1.4	1.2	1.2	-7.3 (-9.4 , -4.9)	1.1 (-0.9 , 3.2)	3.8 (1.9 , 5.7)
	Geneva	16	1.5	1.4	1.5	1.7	-4.1 (-8.0 , 0.2)	-6.7 (-10.3 , -2.6)	11.8 (5.9 , 18.5)
	Lugano	16	1.4	1.3	1.5	1.4	-17.1 (-20.4 , -13.5)	-16 (-18.9 , -12.8)	30.2 (23.1 , 38.1)
	Wald	18	2.3	1.7	2.0	2.3	-1.5 (-3.5 , 0.9)	-2.2 (-4.7 , 0.7)	3.6 (-0.2 , 8.1)
	Average 4 areas ^{a,d}	67	1.6	1.5	1.5	1.7			
	Pooled 4 areas ^{b,d}	67	3.0	2.3	2.8	4.2	-6.4 (-9.2 , -3.1)	-5.3 (-8.4 , -1.7)	9.5 (3.6 , 16.3)

Bold = significant seasonal differences at $p = 0.05$; ^a Average of P90/P10 ratios as calculated for all areas; ^b P90/P10 ratio calculated from pooled data from all areas; ^c All 8 areas: Aarau, Basel, Davos, Geneva, Lugano, Montana, Payerne, Wald; ^d All 4 NO₂+PM+UFP areas: Basel, Geneva, Lugano, Wald

Table 2: Variance apportionment based on adjusted biannual means (2011-2012) and adjusted seasonal means.

Pollutant	Number of area(s)	Based on biannual seasonally-adjusted means		Based on seasonal means, considering seasonal variability		
		Between area variance (%)	Within area / Between site variance (%)	Between area variance (%)	Within area / Between site variance (%)	Within site (seasonal) variance (%)
NO ₂	8 ^a	34.1	65.9	29.1	48.9	22.0
NO ₂	4 ^b	32.2	67.7	27.2	49.2	23.7
PM _{2.5}	4	55.4	44.6	21.4	0	78.6
PM _{2.5} abs	4	72.5	27.5	57.3	9.8	32.9
PM ₁₀	4	65.1	34.9	39.5	0.6	59.9
PM _{coarse}	4	44.9	55.1	27.8	6.4	65.8
PNC	4	74.7	25.3	43.0	0	57.0
LDSA	4	83.3	16.7	40.8	0	59.2

^a All areas: Aarau, Basel, Davos, Geneva, Lugano, Montana, Payerne, Wald; ^b All NO₂+PM+UFP areas: Basel, Geneva, Lugano, Wald

3.5 Correlations between pollutants

Coefficients of determination between the bi-annually adjusted concentrations were variable across the different study areas (Table 3). Generally, high coefficients of determination ($R^2 > 0.70$) were found between NO₂ and PM_{2.5} absorbance, NO₂ and PNC, NO₂ and LDSA, PM_{2.5} and PM₁₀, PM_{2.5} absorbance and PNC, PM_{2.5} absorbance and LDSA, PM₁₀ and LDSA and between PNC and LDSA. Moderate overall coefficients of determination ($R^2 < 0.50$) were found between NO₂ and PM_{2.5}, PM_{2.5} and PNC and between PM_{coarse} and all other pollutants except PM₁₀. Coefficients of determination varied markedly between the study areas, as a result of the more limited concentration range within any single area and the reduced number of comparisons.

Table 3: Coefficients of determination, expressed as R^2 between adjusted biannual average concentrations for all pollutants, corrected for temporal variation.

Coefficient of determination, R^2 (n)																					
Area	NO ₂						PM _{2.5}					PM _{2.5} abs				PM ₁₀			PM _{coarse}		PNC
	PM _{2.5}	PM _{2.5} abs	PM ₁₀	PM _{coarse}	PNC	LDSA	PM _{2.5} abs	PM ₁₀	PM _{coarse}	PNC	LDSA	PM ₁₀	PM _{coarse}	PNC	LDSA	PM _{coarse}	PNC	LDSA	PNC	LDSA	LDSA
Basel	0.21	0.35	0.18	0.03	0.47	0.32	0.63	0.49	0.32	0.43	0.50	0.25	0.25	0.35	0.36	0.01	0.71	0.59	0.08	0.19	0.63
Geneva	0.21	0.44	0.39	0.32	0.60	0.64	0.18	0.63	0.24	0.17	0.33	0.22	0.13	0.07	0.19	0.83	0.10	0.17	0.02	0.03	0.92
Lugano	0.11	0.82	0.25	0.13	0.74	0.61	0.16	0.75	0.16	0.10	0.38	0.28	0.14	0.61	0.48	0.51	0.27	0.50	0.17	0.12	0.75
Wald	0.35	0.90	0.62	0.37	0.82	0.80	0.51	0.45	0.20	0.34	0.30	0.71	0.44	0.86	0.84	0.30	0.69	0.72	0.33	0.34	0.98
All areas	0.42	0.80	0.63	0.41	0.81	0.80	0.51	0.74	0.21	0.40	0.58	0.68	0.41	0.74	0.79	0.59	0.62	0.76	0.42	0.40	0.90

For PM_{2.5}, PM_{2.5} absorbance, PM₁₀ and PM_{coarse} the number of sites is 20, 18, 17, 19 and 74 respectively for Basel, Geneva, Lugano, Wald and all areas together. For PNC and LDSA, the number of sites is 17, 16, 16, 18 and 67 for Basel, Geneva, Lugano, Wald and all areas.

4 Discussion

We measured a wide range of air pollutants, across a dense monitoring network covering the eight focus areas of the ongoing Swiss cohort study SAPALDIA. This extensive purpose-designed network comprised a total of 312 sites for NO₂, 74 sites for PM_{2.5}, PM_{2.5} absorbance, PM₁₀ and PM_{coarse} and 67

sites for PNC and LDSA. We found substantial concentration differences for all pollutants within and between the eight study areas. Especially for NO₂, but also for PM_{2.5} absorbance, PM_{coarse} and PNC, we saw strong within-area contrasts, indicating that within-area spatial variability in air pollution results from various local sources, including tailpipe and non-tailpipe emissions. The modest contrast found for PNC and LDSA, compared to some of the other pollutants, is. In the long term, variance was predominantly attributed to between-area contrasts for most pollutants, except NO₂ and PM_{coarse}. However, when considering also seasonal variability, the seasonal variation made up the majority of the total, and so the fraction attributed to within area variability was substantially reduced for many pollutants. Pollutants generally correlated well with one another when areas were pooled, though within-area correlations varied considerably. The spatial patterns observed for the two metrics of UFP (PNC and LDSA) were highly correlated. 4.1 Spatial and temporal variation of NO₂, PM_{2.5}, PM_{2.5} absorbance, PM₁₀ and PM_{coarse}

Although absolute concentrations for the 2011-2012 period are somewhat lower than observed for 2003, the between-area ranking for NO₂ between the eight areas is comparable to the situation observed in the previous monitoring campaign in 2003 (Liu et al., 2012). PM_{2.5}, PM₁₀ and PM_{coarse} levels and contrasts in Lugano are virtually identical to those observed in the same area for the ESCAPE study (2009), but PM_{2.5} absorbance concentrations observed in the current study are lower (Eeftens et al., 2012). This could be due to lower traffic intensity on the street sites selected for this campaign, compared to the ESCAPE campaign, which required a minimum traffic intensity of 10,000 vehicles / 24 hours for the “street” type sites. NO₂ levels and contrasts in Geneva are comparable to those observed in the ESCAPE study, but while contrasts were similar, concentrations were observed to be somewhat lower in Basel, and higher in Lugano for the current study (Cyrus et al., 2012). This could be due to variations between years, as well as differences in site selection.

4.2 Spatial and temporal variation of PNC and LDSA

As previously noted, fewer studies are available on long-term exposure to PNC and we know of no previous studies investigating long-term spatial contrasts in LDSA. A study conducted in Helsinki, Athens, Amsterdam and Birmingham, also performed week-long measurements of ultrafine particles, and found median levels of PNC in Athens, Amsterdam and Birmingham that were comparable to the levels we observed in Geneva and Lugano (Puustinen et al., 2007). However, it is important to note that there is currently no standard definition of ultrafine particles and different instruments can have great differences in particle counts. Two other studies found that PNC winter concentrations in Helsinki, Barcelona, Rome, Augsburg and Stockholm were 2 to 5 times higher than summer concentrations (Aalto et al., 2005), and winter-spring contrasts were lower in Augsburg (Cyrus et al., 2008). In Geneva and Lugano, we observed similarly high winter-summer and winter-spring contrasts as were observed in Helsinki, Augsburg and Stockholm, but we found lower seasonal contrasts in Basel and Wald. Higher seasonal differences were also previously reported for Basel by Ragettli et al. (2014), but were less apparent in the current study. This may be a result of specific weather conditions on the particular dates and times of day, during which the short-term 20-minute measurements were made, since seasonal variability in UFP is affected by photochemical activity in summer and by a lower mixing height and lower temperature in winter. Concentrations and spatial variability in this study were slightly lower than those recently published in a short-term study from Basel, which also used the MiniDiSC. (Ragettli et al., 2014) This is likely due to differences in site selection, as Ragettli et al included 70% sites that were situated directly on major roads, and measured on sidewalks rather than home facades. Ragettli et al (2014) included 60 monitoring sites

were measured for 20 minutes each, on work days between the hours of 9:30 and 16:00, thereby avoiding peak traffic hours. Nevertheless, concentrations during the measurement times were likely higher than the typical 24-hour daily average. Ragettli et al. (2014) also found that when simultaneous measurements were taken on the sidewalk and on the balcony of SAPALDIA homes, concentrations at the sidewalk were 20% higher than those at the homes, and showed more variability. Our current study is exclusively focussed on exposure at the home from a health perspective, and does not make any inferences about spatial contrasts closer to the tailpipe, which are likely larger, but less relevant for long-term health. Indeed, we showed a sharp decrease in PNC concentration, with increasing distance to major roads in a previous publication, focussed at short-term PNC concentrations. (Meier et al., 2015) This study also showed dramatic increases in PNC concentration during the morning and evening rush hours, indicating that road traffic is a major source of PNC. (Meier et al., 2015) Light-duty diesel vehicles are relatively common in Switzerland and most other European countries: the percentage of diesel vehicles is estimated at 26 to 50% in the study areas of this project (Online Supplement E). Heavy-duty diesel vehicles (buses and trucks) emit relatively high amounts of NO₂, and retrofitting them with particle filters which also capture UFP is an ongoing process. Reduced UFP emissions due to particle filters are a possible explanation for the modest intra-area contrasts of UFP metrics, compared to NO₂, but also the positioning of monitors at homes rather than sidewalks likely plays an important role here.

4.3 Correlation between pollutants

We found that the spatial contrasts for the different pollutants were moderately to highly correlated, both within areas, as well as overall. These high correlations imply that it remains difficult to disentangle the health effects associated with each pollutant separately. High correlations between NO₂ and PM_{2.5} absorbance (Eeftens et al., 2012; Lewné et al., 2004) and between PM_{2.5} and PM₁₀ (Eeftens et al., 2012) were also reported in previous studies. Low correlations between PM_{coarse} and other pollutants were also previously reported, (Eeftens et al., 2012) and are likely a consequence of higher uncertainty surrounding PM_{coarse} data, which are derived by subtracting PM_{2.5} from PM₁₀ mass and are therefore affected by measurement error from both of these measurements. In comparison with Geneva and Lugano, the PM_{coarse} concentrations are lower in Basel and Wald and have a smaller range in those areas. Therefore, the ranking and correlation with other pollutants in Basel and Wald are more affected by the uncertainty that results from the subtraction, than other areas. UFP measures PNC and LDSA were correlated highly with each other and with NO₂ and PM_{2.5} absorbance. Both NO₂ and PM_{2.5} absorbance characterizes local motor vehicle exhaust emissions, more from diesel than from gasoline engines. UFP, which are emitted from all tailpipes, though with different size distribution and composition, is also elevated in the direct vicinity of “general” traffic, and therefore both, PNC and LDSA, are highly correlated with NO₂ and PM_{2.5} absorbance in this study.

4.4 Air quality guidelines

Switzerland has set annual average limit values for both NO₂ (30 µg/m³) and PM₁₀ (20 µg/m³), whereas PM_{2.5} is not yet regulated. The Swiss Federal Commission for Air Hygiene recently advised the government to additionally introduce an annual average limit value for PM_{2.5}, following the guideline value of 10 µg/m³, as recommended by WHO (WHO, 2006). This study shows that at many sites, the corrected bi-annual averages for both NO₂ and PM₁₀ still exceed these standards in Switzerland, predominantly in Geneva and Lugano (Figure 2). The corrected bi-annual average exceeds the proposed standard of 10 µg/m³ for PM_{2.5} at over 90% of measurement sites in this study

(Figure 2). While we did not measure at all sites over an entire year, we documented exceedances of the above-mentioned standard in all areas, and in all 3 seasons. The temporal correction was carried out to make our measurements directly comparable to one another, and representative for a bi-annual average. We validated that the temporal variability at the reference site was representative for the entire area and documented that the use of a different reference site did not affect the results (Table C.3). As recently confirmed (Beelen et al., 2013), $PM_{2.5}$ exposure, also affects health at these relatively low levels. Therefore, benefits of further improvements of air quality continue to be substantial (Henschel and Chan, 2013).

4.5 Strengths and limitations

PNC and LDSA were temporally adjusted using long-term data derived for the 12-week period during which miniDiSCs were operated at the reference sites. The other pollutants were adjusted to concentrations representing the average concentration over 2011 and 2012. When assessing the relationships (correlation and ratio) between the pollutants, we therefore assumed that the corrected PNC and LDSA concentrations were also representative for the 2011-2012 period. We could only verify this assumption for Basel and Lugano, where long-term station monitoring data were available for some of the seasons (Online Supplement D).

In the biannually adjusted concentrations, most of the spatial contrasts for UFP (both PNC and LDSA) were attributed to differences between study areas (Figure 2). However, considerable temporal contrasts become apparent when we look at the seasonal variation. Future research on UFP and LDSA will need to investigate the role of seasonal, day-to-day and diurnal temporal as well as spatial variability. In this study, we have limited our scope to long-term and seasonal patterns in space. But also shorter-term variations also pose a challenge in the assessment of exposure for epidemiological studies as people move through space, experiencing vastly varying UFP concentrations, e.g. during their commute.

5 Conclusion

We found substantial spatial contrasts between and within study areas for NO_2 , $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10} , PM_{coarse} , PNC and LDSA. For most pollutants, except NO_2 and PM_{coarse} , the majority of spatial variance was attributable to differences between the study areas, but substantial contrast was also found within areas for all pollutants. Correlations between the pollutants were generally moderate to high, and PNC and LDSA correlated very highly with each other and with NO_2 and $PM_{2.5}$ absorbance. Through land-use regression (LUR) modelling, these standardized measurements will greatly aid in characterizing the long-term exposure of the SAPALDIA cohort to these pollutants.

6 Competing interest

The authors declare that they have no competing financial interest

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Supplementary data

Supplementary data related to this article can be found at [\[\[put website information\]\]](#).

8 References

- Aalto, P., Hämeri, K., Paatero, P., Kulmala, M., Bellander, T., Berglind, N., Bouso, L., Castaño-Vinyals, G., Sunyer, J., Cattani, G., Marconi, A., Cyrys, J., Klot, S.v., Peters, A., Zetzsche, K., Lanki, T., Pekkanen, J., Nyberg, F., Sjövall, B., Forastiere, F., 2005. Aerosol Particle Number Concentration Measurements in Five European Cities Using TSI-3022 Condensation Particle Counter over a Three-Year Period during Health Effects of Air Pollution on Susceptible Subpopulations. *Journal of the Air & Waste Management Association* 55, 1064-1076.
- Allen, R., Box, M., Liu, L.-J.S., Larson, T.V., 2001. A Cost-Effective Weighing Chamber for Particulate Matter Filters. *Journal of the Air & Waste Management Association* 51, 1650-1653.
- Beelen, R., Raaschou-Nielsen, O., Stafoggia, M., Andersen, Z.J., Weinmayr, G., Hoffmann, B., Wolf, K., Samoli, E., Fischer, P., Nieuwenhuijsen, M., 2013. Effects of long-term exposure to air pollution on natural-cause mortality: an analysis of 22 European cohorts within the multicentre ESCAPE project. *The Lancet* 383, 785-795.
- Brunekreef, B., Forsberg, B., 2005. Epidemiological evidence of effects of coarse airborne particles on health. *European Respiratory Journal* 26, 309-318.
- Cyrys, J., Eeftens, M., Heinrich, J., Ampe, C., Armengaud, A., Beelen, R., Bellander, T., Beregszaszi, T., Birk, M., Cesaroni, G., Cirach, M., de Hoogh, K., De Nazelle, A., de Vocht, F., Declercq, C., Dèdelè, A., Dimakopoulou, K., Eriksen, K., Galassi, C., Gražulevičienė, R., Grivas, G., Gruzieva, O., Gustafsson, A.H., Hoffmann, B., Iakovides, M., Ineichen, A., Krämer, U., Lanki, T., Lozano, P., Madsen, C., Meliefste, K., Modig, L., Mölter, A., Mosler, G., Nieuwenhuijsen, M., Nonnemacher, M., Oldenwening, M., Peters, A., Pontet, S., Probst-Hensch, N., Quass, U., Raaschou-Nielsen, O., Ranzi, A., Sugiri, D., Stephanou, E.G., Taimisto, P., Tsai, M.-Y., Vaskövi, É., Villani, S., Wang, M., Brunekreef, B., Hoek, G., 2012. Variation of NO₂ and NO_x concentrations between and within 36 European study areas: Results from the ESCAPE study. *Atmospheric Environment* 62, 374-390.
- Cyrys, J., Pitz, M., Heinrich, J., Wichmann, H.E., Peters, A., 2008. Spatial and temporal variation of particle number concentration in Augsburg, Germany. *Science of The Total Environment* 401, 168-175.
- Duffin, R., Tran, C., Clouter, A., Brown, D., MacNee, W., Stone, V., Donaldson, K., 2002. The importance of surface area and specific reactivity in the acute pulmonary inflammatory response to particles. *Annals of Occupational Hygiene* 46, 242-245.
- Eeftens, M., Tsai, M.-Y., Ampe, C., Anwender, B., Beelen, R., Bellander, T., Cesaroni, G., Cirach, M., Cyrys, J., de Hoogh, K., De Nazelle, A., de Vocht, F., Declercq, C., Dèdelè, A., Eriksen, K., Galassi, C., Gražulevičienė, R., Grivas, G., Heinrich, J., Hoffmann, B., Iakovides, M., Ineichen, A., Katsouyanni, K., Korek, M., Krämer, U., Kuhlbusch, T., Lanki, T., Madsen, C., Meliefste, K., Mölter, A., Mosler, G., Nieuwenhuijsen, M., Oldenwening, M., Pennanen, A., Probst-Hensch, N., Quass, U., Raaschou-Nielsen, O., Ranzi, A., Stephanou, E., Sugiri, D., Udvardy, O., Vaskövi, É., Weinmayr, G., Brunekreef, B., Hoek, G., 2012. Spatial variation of PM_{2.5}, PM₁₀, PM_{2.5} absorbance and PM_{coarse} concentrations between and within 20 European study areas and the relationship with NO₂ – Results of the ESCAPE project. *Atmospheric Environment* 62, 303-317.
- European Environment Agency (EEA), 2009. Spatial Assessment of PM₁₀ and Ozone Concentrations in Europe (2005).
- Fierz, M., Houle, C., Steigmeier, P., Burtcher, H., 2011. Design, calibration, and field performance of a miniature diffusion size classifier. *Aerosol Science and Technology* 45, 1-10.
- Heal, M.R., Kumar, P., Harrison, R.M., 2012. Particles, air quality, policy and health. *Chemical Society Reviews* 41, 6606-6630.
- Henschel, S., Chan, G., 2013. Health risks of air pollution in Europe—HRAPIE project.
- Hoek, G., Beelen, R., Kos, G., Dijkema, M., Zee, S.C.v.d., Fischer, P.H., Brunekreef, B., 2010. Land use regression model for ultrafine particles in Amsterdam. *Environmental Science & Technology* 45, 622-628.
- Hoek, G., Boogaard, H., Knol, A., de Hartog, J., Slottje, P., Ayres, J.G., Borm, P., Brunekreef, B., Donaldson, K., Forastiere, F., Holgate, S., Kreyling, W.G., Nemery, B., Pekkanen, J., Stone, V., Wichmann, H.E., van der Sluijs, J., 2009. Concentration Response Functions for Ultrafine Particles and All-Cause Mortality and

- Hospital Admissions: Results of a European Expert Panel Elicitation. *Environmental Science & Technology* 44, 476-482.
- Hoek, G., Meliefste, K., Cyrys, J., Lewné, M., Bellander, T., Brauer, M., Fischer, P., Gehring, U., Heinrich, J., van Vliet, P., 2002. Spatial variability of fine particle concentrations in three European areas. *Atmospheric Environment* 36, 4077-4088.
- International Standardization Organization (ISO), 1993. ISO 9835 Ambient Air: Determination of a Black Smoke Index.
- Janssen, N., Hoek, G., Simic-Lawson, M., Fischer, P., van Bree, L., ten Brink, H., Keuken, M., Atkinson, R.W., Anderson, H.R., Brunekreef, B., 2011. Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM₁₀ and PM_{2.5}. *Environmental Health Perspectives* 119, 1691-1699.
- Künzli, N.P., Laura; Rapp, Regula, 2010. Air Quality and Health - European Respiratory Society.
- Lewné, M., Cyrys, J., Meliefste, K., Hoek, G., Brauer, M., Fischer, P., Gehring, U., Heinrich, J., Brunekreef, B., Bellander, T., 2004. Spatial variation in nitrogen dioxide in three European areas. *Science of The Total Environment* 332, 217-230.
- Liu, S., Tsai, M.-Y., Keidel, D., Gemperli, A., Ineichen, A., Hazenkamp-von Arx, M., Bayer-Oglesby, L., Rochat, T., Künzli, N., Ackermann-Liebrich, U., Straehl, P., Schwartz, J., Schindler, C., 2012. Long-term exposure models for traffic related NO₂ across geographically diverse areas over separate years. *Atmospheric Environment* 46, 460-471.
- Meier, R., Clark, K., Riediker, M., 2012. Comparative Testing of a Miniature Diffusion Size Classifier to Assess Airborne Ultrafine Particles Under Field Conditions. *Aerosol Science and Technology* 47, 22-28.
- Meier, R., Eeftens, M., Aguilera, I., Phuleria, H.C., Ineichen, A., Davey, M., Ragettli, M.S., Fierz, M., Schindler, C., Probst-Hensch, N., Tsai, M.-Y., Künzli, N., 2015. Ambient Ultrafine Particle Levels at Residential and Reference Sites in Urban and Rural Switzerland. *Environmental Science & Technology* 49, 2709-2715.
- Peters, A., Veronesi, B., Calderón-Garcidueñas, L., Gehr, P., Chen, L.C., Geiser, M., Reed, W., Rothen-Rutishauser, B., Schurch, S., Schulz, H., 2006. Translocation and potential neurological effects of fine and ultrafine particles a critical update. *Particle and Fibre Toxicology* 3, 1-13.
- Puustinen, A., Hämeri, K., Pekkanen, J., Kulmala, M., de Hartog, J., Meliefste, K., ten Brink, H., Kos, G., Katsouyanni, K., Karakatsani, A., Kotronarou, A., Kavouras, I., Meddings, C., Thomas, S., Harrison, R., Ayres, J.G., van der Zee, S., Hoek, G., 2007. Spatial variation of particle number and mass over four European cities. *Atmospheric Environment* 41, 6622-6636.
- Ragettli, M.S., Ducret-Stich, R.E., Foraster, M., Morelli, X., Aguilera, I., Basagaña, X., Corradi, E., Ineichen, A., Tsai, M.-Y., Probst-Hensch, N., 2014. Spatio-temporal variation of urban ultrafine particle number concentrations. *Atmospheric Environment* 96, 275-283.
- Rückerl, R., Schneider, A., Breitner, S., Cyrys, J., Peters, A., 2011. Health effects of particulate air pollution: a review of epidemiological evidence. *Inhalation Toxicology* 23, 555-592.
- Sioutas, C., Delfino, R.J., Singh, M., 2005. Exposure assessment for atmospheric ultrafine particles (UFPs) and implications in epidemiologic research. *Environmental Health Perspectives*, 113, 947-955.
- United States Environmental Protection Agency (US EPA), 1998. Summary of Guidance; Filter Conditioning and Weighing Facilities and Procedures for PM_{2.5} Reference and Class I Equivalent Methods; US EPA: Research Triangle Park, NC, USA.
- United States Environmental Protection Agency (US EPA), 2012. Report to congress on black carbon.
- World Health Organization (WHO), 2006. Air Quality Guidelines Global Update 2005.
- Wittmaack, K., 2007. In search of the most relevant parameter for quantifying lung inflammatory response to nanoparticle exposure: particle number, surface area, or what? *Environmental health perspectives*, 115, 187-194.

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