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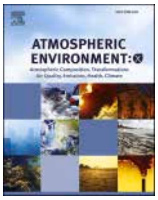

Switzerland’s PM_{10} and $PM_{2.5}$ environmental increments show the importance of non-exhaust emissions

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ABSTRACT

Atmospheric particulate matter (PM) is a priority pollutant for urban air pollution management because of its negative effects on human health and visibility. Emissions from road traffic have been a major focus of management over the past few decades, but non-exhaust emissions *i.e.*, emissions from brake, tyre, road wear, and the resuspension of dust have emerged to become a major source of unregulated PM in many locations. Here, a filter-based sampling campaign was conducted between 2018 and 2019 where a large number of PM constituents were quantified for five sites in Switzerland for both PM_{10} and $PM_{2.5}$. This had the objective of investigating urban and urban-traffic PM increments in Switzerland. The results show that PM concentrations increased as the sampling locations moved along a rural to urban-traffic gradient. However, source apportionment analysis showed that sulfate-rich, nitrate-rich, and biogenic sources were not enhanced in urban environments, but road traffic and mineral dust sources were. The total mass enhancement for PM_{10} and $PM_{2.5}$ were 2.4 $\mu g\ m^{-3}$ and 2.0 $\mu g\ m^{-3}$ for the urban environment while the corresponding urban-traffic enhancements were 5.7 $\mu g\ m^{-3}$ and 2.8 $\mu g\ m^{-3}$. Emissions from road traffic were estimated to contribute more than 75% to the urban increments and non-exhaust emissions contributed 48% (PM_{10}) and 25% ($PM_{2.5}$) to the total road traffic related increment at an urban background site and 62% (PM_{10}) and 49% ($PM_{2.5}$) at an urban-traffic site. Analysis of the composition of Switzerland’s PM showed that elements associated with non-exhaust emissions, specifically the brake wear tracers of antimony, barium, copper, and iron were the metals with the greatest urban and urban-traffic enhancements. Critically, the urban increment of these elements was enhanced for both PM_{10} and $PM_{2.5}$ by about the same magnitude as the urban-traffic increment (by 2–3 times), demonstrating non-exhaust emissions are encountered across urban areas, not just the urban-traffic environment. Therefore, non-exhaust emissions were an important contributor to the urban and urban-traffic PM_{10} and $PM_{2.5}$ increments in Switzerland’s urban areas. The relative contributions of non-exhaust emissions to the urban and urban-traffic increments could be expected to increase due to the introduction of further exhaust after-treatment technologies (such as gasoline particulate filters; GPFs) and the transition to a more electrified vehicle fleet. A management pivot will be required to control these non-exhaust emission pathways and although this work exclusively uses data from Switzerland, the conclusions are likely relevant to many other European urban areas.

1. Introduction

1.1. Background

Particulate matter (PM) is a principal atmospheric pollutant which has proven to be a challenge to control in a variety of environments across the world. In respect to public health, PM has deleterious effects and is the atmospheric pollutant which contributes the most to the global burden of disease (World Health Organization, 2016; Cohen et al., 2017). Additionally, PM perturbs the Earth’s radiation balance

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and reduces atmospheric visibility which contribute to PM being a well studied and prioritised in terms of management (Galvão et al., 2018). A key feature of PM which separates the pollutant from gases, such as oxides of nitrogen (NO_x) or ozone (O_3), is that it is composed of myriad heterogeneous liquid and solid components, all of which combine to give PM its features. Therefore, the definition of PM covers a wide range of particle sizes, shapes, surface areas, and chemical compositions and stabilities (Van Dingenen et al., 2004).

Owing to PM's diversity, it has a range of both natural and anthropogenic sources. Primary natural emission sources include: wind blown dust, volcanos, sea spray, forest fires, and bioaerosols (Sapkota et al., 2005; Samaké et al., 2019). Anthropogenic emission of PM and gaseous precursors are generally generated by combustion processes from the burning of fossil fuels or biomass, but other important sources include: industrial processes, vehicle wear, and anthropogenic dust. However, PM is also generated by secondary processes which occur in the atmosphere itself and secondary PM generation requires precursors, which are often of anthropogenic origin (Ivey et al., 2016). The oxidation of previously emitted NO_x, sulfur oxides, and gaseous ammonia produce ammonium nitrate and sulfate, and these compounds can be especially relevant PM components for many locations where secondary PM contributes a large proportion of the total PM load. The oxidation of volatile organic compounds (VOCs) can also be an important pathway for secondary organic aerosol (SOA) generation (Fine et al., 2008).

PM is usually defined by size with the PM_{10} and $PM_{2.5}$ fractions being the most common in the domain of regulation, and these are defined as PM with an aerodynamic diameter less than 10 and 2.5 μm (μm), respectively. Primarily due to the negative health effects of PM, most countries have legal limits for ambient outdoor PM_{10} and/or $PM_{2.5}$ which are to be complied with (European Commission, 2019). To ensure compliance to legal limits, PM is monitored in ambient air routinely within air quality monitoring networks.

Although PM is commonly monitored, such routine activities do not capture the very diverse nature of PM because in ambient monitoring operations, generally, only PM mass or number is reported. Therefore, there is substantial value in determining the constituents which form the PM mix in any given location (Viana et al., 2008; Zhang et al., 2014; Mukherjee and Agrawal, 2017). With such knowledge, PM can be characterised and sources inferred, and this information can be used to inform policy makers to make decisions which more effectively control PM sources and reduce the negative effects of this pollutant – a goal of all compliance-driven monitoring activities (Putaud et al., 2010; Ostro et al., 2011).

1.2. *Environmental increments*

Urban atmospheres offer interesting challenges and perspectives for air pollution which are usually burdened with complex emission, generation, chemical, and dispersion processes (Harrison, 2018). Pollutant concentrations in urban areas are subjected to heterogeneous processes because of changeable landuse and the nature of the urban environment. Strong pollutant concentration gradients are often observed in both time and space. PM is no exception to these processes.

Urban PM concentrations can be conceptualised as the result of the intersection of emission sources (and/or generation processes) resulting from different spatial scales. The first component is a regional background component which would be present, even if the urban area did not exist. The regional background component can be split into non-anthropogenically enhanced concentrations (a true "natural" component), and an enhanced amount from direct emissions and secondary generation from precursors outside the immediate urban area (Lenschow et al., 2001; Thunis et al., 2018). The urban and urban-traffic increments refer to increases in concentrations which reflect increased emissions from more intensive resource consumption in these specific environments. These increments can also be very variable and are subjected to complex atmospheric dispersion processes as well as variable emission processes, however, they also feedback, especially in the case of PM, to the regional background component (Harrison, 2018).

1.3. *Road traffic emissions*

Major sources of PM in urban atmospheres are road traffic and road vehicles (Hilker et al., 2019). Emissions from exhaust (tailpipes) have traditionally been the focus of air quality management and large reductions in this source has been achieved across the developed world due to the implementation of progressively stringent emission standards (European Environment Agency, 2020). For example, for diesel-powered light-duty vehicles (cars and vans), the Euro emission standards reduced from 0.14 to 0.004 5 g km^{-1} between 1992 and 2009 (Euro 1–5), which is a 31 fold change. The reduction of PM generated by combustion processes has not been matched by reductions in non-exhaust emissions because they have remained unregulated throughout this period (Harrison et al., 2021b) and for example, Amato et al. (2014) reported that the long-term trends of exhaust contributions to PM_{10} in Spanish cities followed a marked downward trend, but the non-exhaust contributions remained constant for the same time period.

Non-exhaust emissions refer to emissions derived from brake wear, tyre wear, road surface wear, resuspension of materials already present on the road surface, as well as other minor emissions from drive belts, clutches, and paints. Throughout Europe, the ratio between exhaust and non-exhaust emissions has altered where non-exhaust emissions are now often the dominate source of road traffic PM emissions in many locations (Beddows and Harrison, 2021). Traditionally, non-exhaust emissions were generally thought to be contained in the coarse-mode because they are emitted by abrasive processes, however it has been shown that fine PM emissions are significant, and therefore non-exhaust emissions are found in both size fractions (Vanherle et al., 2021). PM size distributions for the non-exhaust emission sources are difficult to generalise, but based on the Harrison et al. (2021a) literature review, 3 μm has been reported frequently as the mode of brake wear emissions, while the mode of tyre wear, road surface wear, and resuspension emissions distributions are more variable and have been reported between 2 and 8, 5–12, and 2–5 μm respectively. Therefore, the mass distributions of most non-exhaust emissions straddle the boundary between coarse and fine PM.

The switch of importance from exhaust and non-exhaust PM emissions will require a management pivot to address. Non-exhaust emissions are currently unregulated across the world and it is clear that even as vehicle fleets move towards becoming more and more electrified, non-exhaust emissions will remain an important emission pathway to consider after exhaust emissions are further reduced (OECD, 2020). Tyre wear has also seen an expansion of general interest because it is included in the microplastics definition by some (Hartmann et al., 2019).

1.4. *Switzerland's PM concentrations*

Switzerland's ambient PM_{10} and $PM_{2.5}$ concentrations have decreased since the commencement of widespread monitoring of PM in the late-1990s (Barmpadimos et al., 2011; Grange et al., 2018, 2020). Such reductions have been achieved in urban areas and at isolated mountainous sites and therefore, the reductions can be attributed to air quality management strategies, within Switzerland itself and across the surrounding countries which make up Europe.

The reductions in PM concentrations has altered the relative importance of some of the primary PM components (Gianini et al., 2012b,a; Hüglin et al., 2012; Hüglin and Grange, 2021). The share of elemental carbon, nitrate, and sulfate have decreased over time, while mineral dust and organic matter has increased its share in PM_{10}, PM_{coarse}, and to a lesser extent $PM_{2.5}$. Additionally, according to the Switzerland's air pollution emission inventory (Federal Office for the Environment, 2021b,c), non-exhaust emissions (where only tyre and brake wear are reported) are one of the few PM sources increasing over time in the country (Figure S1).

1.5. Objectives

The primary objective of this work is to investigate and classify the PM urban and urban-traffic increments in Switzerland. This will be done by using data from a filter-based sampling campaign including five sites in different environments between 2018 and 2019. Receptor models using PMF (positive matrix factorisation) will be utilised to aid analysis. Urban and urban-traffic increments will be calculated and explored with an emphasis on elements which are associated with non-exhaust emissions in Switzerland.

2. Methods

2.1. Sampling sites

PM_{10} and $PM_{2.5}$ sampling was conducted in five locations in Switzerland (Table 1; Fig. 1). Four of the five monitoring sites are located on the Swiss plateau where the majority of the human population resides, however, Magadino-Cadenazzo is located south of the Alps in the southern canton of Ticino. Bern-Bollwerk is classified as an urban-traffic site (average daily traffic of 17 500 vehicles per day) and is located within a partial street canyon while the other locations are background locations in urban, suburban, or rural environments. The five sampling sites are included in Switzerland's national air pollution monitoring network (NABEL) (Federal Office for the Environment, 2021a).

2.2. Filter samples and PM mass

Daily PM_{10} and $PM_{2.5}$ filter samples were collected using Digitel DA-80H high-volume samplers with flow rates of 30 m^3 h^{-1} with appropriate PM_{10} or $PM_{2.5}$ inlets. The sampling period had a 12-month duration and ran between June 2018 and May 2019 and therefore, was not aligned with a calendar year. During the sampling period, mean air temperatures ranged from 10.6 to 13 °C and wind speed ranged from 1.7 to 2.1 m s^{-1}. Prevailing wind directions were rather variable among the five sampling sites and the wind roses for the periods are displayed in Figure S2. The quartz filters used (Pallflex Tissuquartz 2500QAT-UP) had diameters of 150 mm and the sampling duration was between midnight and midnight. Routine flow checks and other sampling parameters checks were regularly conducted in accordance to standard operating procedures for the NABEL network. Although PM_{10} samples were taken every day, punches from every fourth day were taken and delivered to laboratories and analysed with a number of analytical techniques. Unlike PM_{10}, $PM_{2.5}$ filter samples were only taken every fourth day and therefore, this analysis was conduced on every fourth day filters to allow for consistency. A very small number of filter samples were not sent for analysis due to sampling issues. A total of 899 filters were included in the analysis with 450 and 449 $p.m._{10}$ and $PM_{2.5}$ filter samples respectively. All observations, error statistics, and metadata were processed and stored in a database with a formal data model (named **pmcharacterr**) which allowed for rapid access and manipulation (Grange, 2021b).

2.3. Analytical techniques and analyses

Elemental and organic carbon (EC and OC) concentrations were determined by the thermal optical transmission (TOT) EN16909 method using the EUSAAR2 temperature protocol (European Committee for Standardization (CEN), 2017). For calculation of organic matter (OM), OC concentrations were multiplied by a factor of 1.6 to account for the mass of non-carbon atoms in OM. This multiplier has been used in the past in Switzerland (Gianini et al., 2012b), but this value is likely to be conservative for most monitoring locations (Chow et al., 2018). Additionally, when using OC for source apportionment, the variable was transformed (called reduced OC) where the sum of other organic species' carbon mass was subtracted from OC. This procedure avoided double counting of carbon mass among the many organic compounds sampled.

Water soluble inorganic ion concentrations (ammonium, calcium, chloride, magnesium, nitrate, potassium, sodium, and sulfate) were detected by ion chromatography (IC). Filter punches with 0.8 cm diameters were taken from the filters and were directly put into the vials and extracted in 4 mL of nanopure water for 4 h. The vials for cation analysis were additionally acidified using 8 μL of 0.6 mol L^{-1} nitric acid (HNO_3), allowing a more complete recovery of cations, especially for calcium. No adjustments were done to compensate for nitrate loss due to its volatility.

Additional acid digestion of 15.9 cm^2 filter punches were undertaken using a 25 mL mix of hydrofluoric, nitric, and perchloric acid (HNO_3–HF–$HClO_4$). The resulting solution was analysed for elemental concentrations by inductively coupled plasma atomic emission spectrometry (ICP-AES) and inductively coupled plasma mass spectroscopy (ICP-MS). Field blanks showed that high baselines of metals were not an issue for the sampling period and a detailed description of the applied method is given in Querol et al. (2001). Finally, sugar alcohols including arabitol, mannitol, and mannosan as well as other organics, notably levoglucosan, were determined by a high-performance liquid chromatographic method followed by pulsed amperometric detection (HPLC-PAD) following the analytical method described in Borlaza et al. (2021).

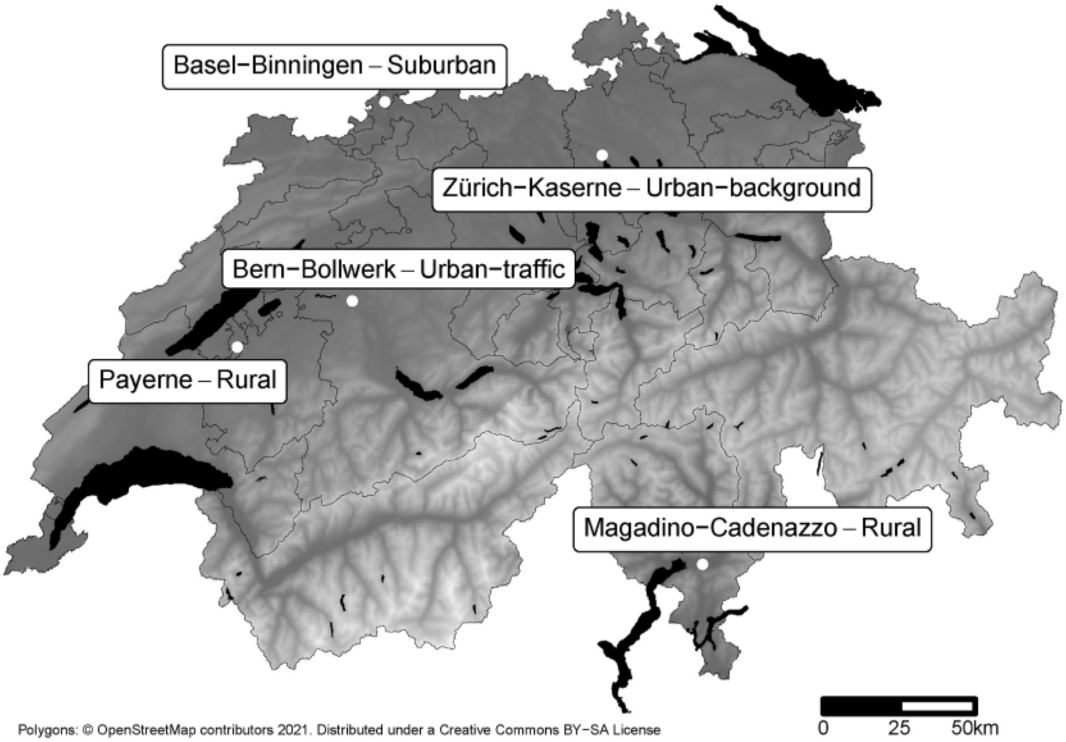
A mass closure was conducted to compare the total and reconstructed mass for validation purposes and unidentified mass ranged from 14.5 to 19.8% for PM_{10} and 8.7–15.6% for $PM_{2.5}$. Additional information about the mass closure procedure and the laboratories and analytical techniques, see Hüglin and Grange (2021).

2.4. PMF receptor modelling

Receptor modelling by positive matrix factorisation (PMF) and the use of the multilinear engine (ME-2) algorithm was conducted with the EPA PMF 5.0 software tool (Norris et al., 2014; Brown et al., 2015). The data preparation steps, PMF settings and procedure, and the application of constraints were consistent among all model runs. The harmonised procedure is documented by Favez et al. (2017); Weber et al. (2019), and is informally known as "extended PMF". All observations were included in the PMF modelling process, including two days which were "firework-influenced" (New Years Day and the day after Swiss National Day; August 2, 2018 and January 1, 2019) and two days which were clearly influenced by Saharan dust events at most sampling locations

Table 1
Basic information for the five monitoring sites in Switzerland which were used for intensive PM characterisation measurements.

Site	Site name	Local ID	Canton	Lat.	Long.	Elev. (m)	Site type
ch0002r	Payerne	PAY	Vaud	46.8	6.9	489	Rural
ch0008a	Basel-Binningen	BAS	Basel-Landschaft	47.5	7.6	316	Suburban
ch0010a	Zürich-Kaserne	ZUE	Zürich	47.4	8.5	409	Urban-background
ch0031a	Bern-Bollwerk	BER	Bern	47.0	7.4	536	Urban-traffic
ch0033a	Magadino-Cadenazzo	MAG	Ticino	46.2	8.9	203	Rural



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Elevation: Shuttle Radar Topography Mission (SRTM)

Fig. 1. The five monitoring sites in Switzerland which were used for intensive PM characterisation measurements. The shading indicates the elevation of the terrain, the lines are the cantonal boundaries, and the filled black areas show larger water bodies (lakes and reservoirs).

(February 20, 2019 and April 23, 2019).

The construction of the observation errors (s) was done by using Equation (1) where, DL was the detection limit and was calculated by two times the standard deviation of the blanks of each species, CV was the coefficient of variation (also known as the relative standard deviation; μ/σ), x was the concentration of the species, and finally, α was an additional error term to include other, miscellaneous sources of uncertainty. α was varied for the different analytical techniques and the values used are shown in Table S1.

$$s = \sqrt{DL^2 + (CV \times x)^2 + (\alpha \times x)^2} \quad (1)$$

Concentrations which were below the species' DL were replaced by half of the DL, and the corresponding errors were replaced with 5/6 of the DL. Missing values were replaced by the species median and their errors set to four times the concentration's median. Signal-to-noise (S/N) for all variables (Table S1) were calculated (Equation (2)), and for those variables with S/N $\leq$ 0.2 were excluded from the model, while variables which had S/N $\leq$ 2.0 were set as a "weak" variable in the EPA PMF software which results in such species having additional uncertainty applied. Finally, PM mass was set as the "total variable" in the EPA PMF tool which also results in this species gaining additional uncertainty.

$$S\Big/N = \sum\nolimits_{i=1}^{n} \frac{\left(\frac{x}{s}\right)}{n} \quad (2)$$

For each PM fraction, the five sites' observations and error tables were combined together and exposed to PMF with the use of the ID/site column functionality. Using the EPA PMF tool in such a pooled fashion allowed the factors' characteristics to be conserved among the sites which aided model stability and interpretation. The PMF models were sequentially run with a range of factor numbers (4–12), and primarily based on bootstrap stability, the best models were selected. However, other metrics such as the Qtrue/Qrobust ratio (Q-ratio) and Fpeak values were investigated too (Norris et al., 2014). The factors were decoded into PM sources by evaluating the factors' time series and factor profiles (Figure S3; Figure S4; Figure S5).

In a final step to optimise the PMF models, constraints were sparingly used on some of the factors. This constraining process followed those outlined in the SOURCES methodology documentation (Favez et al., 2017) and are displayed in Table S2. The use of such constraints had a very small influence on the total mass contributions of sources, but they did increase the stability of the solutions (bootstrap mapping was usually in the very high-nineties), thus the use of constraints offered an additional quality control step.

The PMF modelling process was challenging with this dataset due to 91 filter samples being available for each site and PM size fraction which is fewer samples than the generally recommended minimum of 100 for PMF (Norris et al., 2014). Despite the many validation steps conducted, the models had some limitations and therefore, caution was required when interpreting the results. These limitations were considered and are further discussed in Section 3.1.

The various data files generated by the EPA PMF tool were read, processed, analysed, and visualised by the **pmfr** R package (Grange, 2021c). The PMF input data, PMF outputs, and complementary data files are stored in a persistent data repository for readers who wish to investigate all the specific steps and settings which were applied for the PMF analysis (Grange, 2021a).

2.5. *Environmental increment calculations*

The urban and urban-traffic increments were unable to be calculated within Switzerland's urban areas because the sampling campaign did not include multiple sampling locations within the same urban areas of interest. Therefore, Payerne, Zürich-Kaserne, and Bern-Bollwerk were used as representative rural, urban-background, and urban-traffic sites for this work (Table 1; Fig. 1). The increments were thought of being representative of such environments across Switzerland. Although there could be some weakness associated with such an assumption, previous work such as Pandolfi et al. (2020) has demonstrated that using sampling sites in this way offers useful and valid comparisons. Payerne was chosen instead of Magadino-Cadenazzo as the rural site because Payerne is like Zurich-Kaserne and Bern-Bollwerk situated on the Swiss plateau north of the Alps and the three sites are in closer proximity (Fig. 1) and share similar climatic properties. Magadino-Cadenazzo is exposed to high levels of wood smoke in the winter which results in the site not being representative of most of Switzerland's rural locations.

The urban increment was defined as the urban-background concentration minus the rural concentrations (Zürich-Kaserne − Payerne) while the urban-traffic increment was calculated by urban-traffic minus urban-background (Bern-Bollwerk − Zürich-Kaserne). Urban to rural and urban-traffic to urban ratios were also calculated by dividing the two sites' concentrations, *i.e.*: Zürich-Kaserne ÷ Payerne and Bern-Bollwerk ÷ Zürich-Kaserne respectively. Urban and urban-traffic increments were calculated for measured components and elements in PM_{10} and $PM_{2.5}$ as well as the source contributions as obtained from the PMF models. For the later, corrections were applied to the road traffic factor because of the limitations of the PMF procedure, specifically, the presence of impure factors due to imperfect separation of contributions among the identified sources.

3. Results and discussion

3.1. *Spatial patterns of Switzerland's PM components*

Mean PM_{10} and $PM_{2.5}$ concentrations calculated from the filter-based every forth-day sampling regime between June 2018 and May 2019 showed that mean PM concentrations were variable among the five sampling sites across Switzerland (Fig. 2). Clear, and statistically significant increases in PM concentrations were observed as the sampling sites' classifications became increasingly urban (pairwise Wilcoxon signed-rank tests) which is typical for most atmospheric pollutants, with the exception of O_3 (Harrison, 2018). Although there was an increase in PM_{10} and $PM_{2.5}$ concentrations when moving from rural to urban locations, the urban-traffic site, Bern-Bollwerk clearly had the highest PM concentrations for the sampling period with PM_{10} and $PM_{2.5}$ means of 21.4 and 14 µg m^{-3}. Bern-Bollwerk's PM_{10} and $PM_{2.5}$ concentrations were significantly higher than the other four sampling sites. The pairwise Wilcoxon signed-rank tests' results for the five sites and two particulate fractions are displayed in Table S3. These data indicate that the increased PM concentrations are associated with urban and urban-traffic environments in Switzerland.

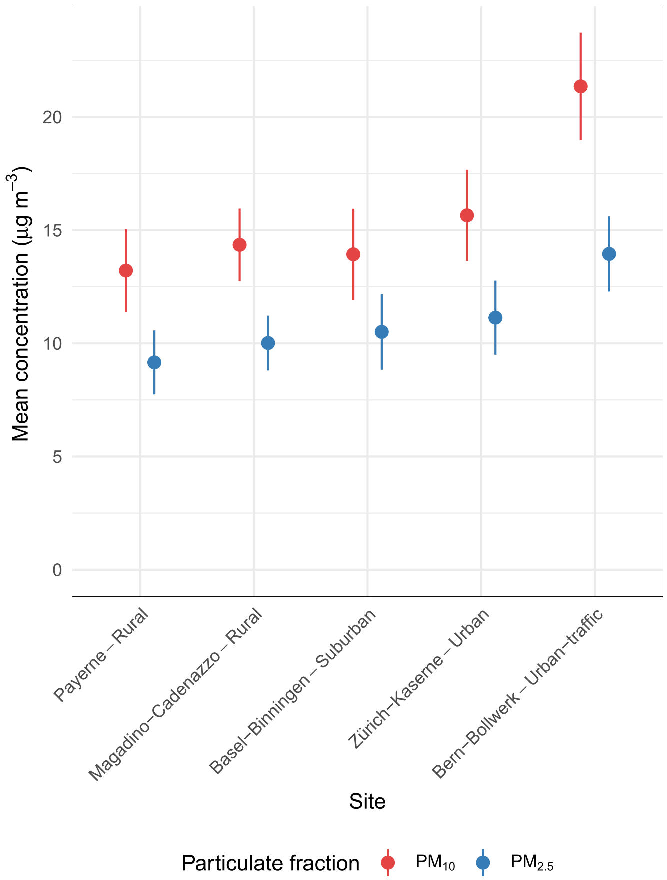

Fig. 2. Mean PM_{10} and $PM_{2.5}$ concentrations for the filter-based every forth-day sampling regime for five Swiss monitoring sites between June 2018 and May 2019.

To investigate what sources compose Switzerland's PM and what sources were responsible for the enhanced urban and urban-traffic concentrations shown in Fig. 2, source apportionment with PMF was applied to both the PM_{10} and $PM_{2.5}$ size fractions for the five sampling sites. The PMF models identified eight distinct sources for PM_{10} and five sources for $PM_{2.5}$ (Fig. 3; Fig. 4).

The largely secondary sulfate-rich, nitrate-rich, and the long range aged sea salt sources (only for PM_{10} for the latter) were a major part of the PM load across Switzerland (Fig. 4). On average (of all sites), these two or three sources contributed 48% and 47% to the PM_{10} and $PM_{2.5}$ mass respectively. The next most prominent sources were road traffic, mineral dust, and wood combustion, all contributing 12–14% of PM_{10} and 9–23% of $PM_{2.5}$. In the case of $PM_{2.5}$, the constituents usually associated with mineral dust (for example magnesium, aluminium, and titanium) were contained within the road traffic source (Figure S3) which indicated a linkage between these two sources. The two biogenic sources (primary and secondary) contributed the least to the PM_{10} mass on average, but these two sources displayed a distinct seasonal cycle with higher contributions during summer and autumn (Fig. 3). The contribution of secondary biogenic source to $PM_{2.5}$ was higher than that to PM_{10}. However, the first included important components that composed the primary biogenic source for PM_{10}. Therefore, this might be considered a more 'mixed biogenic' source in the case of $PM_{2.5}$, but the secondary biogenic label has been used because of the secondary PM is expected to prevail due to its fine size mode.

The aged sea salt factor found in PM_{10} was also not strictly a pure source. This source contained the classic sodium, magnesium, and chloride markers with some contribution of nitrate and sulfate (Figure S3; Figure S4; Figure S5). However, the source also contained small contributions from metals including iron, copper, zinc, and tin – typically tracers for traffic emissions. This indicated that despite the source containing the necessary markers to classify the source, there was some contamination from other emission sources which the PMF model was unable to fully separate. Additionally, the aged sea salt factor demonstrated an inverse gradient where the mean contribution decreased as the site become increasingly urban (Fig. 5) which may be a result of a compensation process from the differing traffic source profiles among the different sampling sites. This highlights that although the aged sea salt label is often used in source apportionment studies, it can often be an imperfect description.

The representative rural, urban, and urban-traffic sites' PMF source means are shown in Fig. 5. As it could be expected, it was observed that the sulfate-rich, nitrate-rich, aged sea salt, and the two biogenic sources had no urban or urban-traffic enhancement across Switzerland during the sampling period. However, the road traffic source (identified by the presence of iron, cooper, zinc, tin, antimony, barium, and EC) was very clearly enhanced with urban and urban-traffic increments of 2.2 µg m^{-3} and 4.5 µg m^{-3} for PM_{10} and 1.7 µg m^{-3} and 2.5 µg m^{-3} for $PM_{2.5}$. The PM_{10} mineral dust source was also progressively enhanced along the Payerne, Zürich-Kaserne, and Bern-Bollwerk site gradient (enhancements of 0.7 µg m^{-3} and 1.6 µg m^{-3} for the urban and urban-traffic

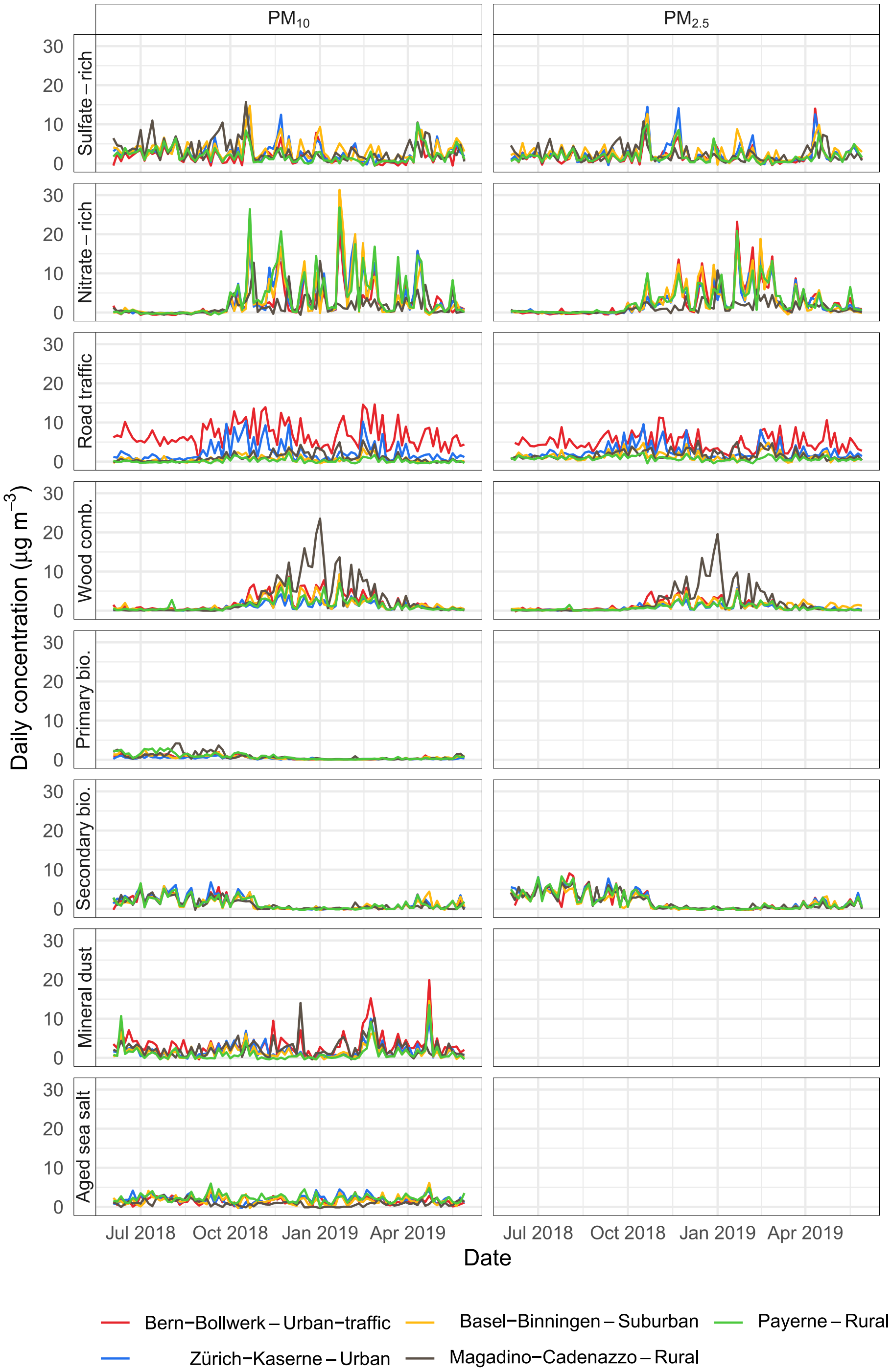

Fig. 3. Time series of the eight PMF-identified sources for PM_{10} and $PM_{2.5}$ at five sampling sites in Switzerland between June 2018 and May 2019. Three of the eight sources were not identified for $PM_{2.5}$ and therefore, are not present.

environments were determined). Magadino-Cadenazzo's mineral dust contributions were however greater than expected (Fig. 4), and this was most likely due to the differing geology south of the Alps compared to the Swiss plateau (Gianini et al., 2012b). Wood combustion did display an urban-traffic increment, but this was most likely an overestimation of mass to this source in the street canyon environment by the PMF models. Interestingly, an overestimation of wood burning sourced PM at this site has been reported by other data analysis methods unrelated to PMF too (Grange et al., 2020).

Inconsistencies were identified between the PM_{10} and $PM_{2.5}$ road traffic sources at the rural and suburban sites (Payerne and Basel-Binningen) where the $PM_{2.5}$ source, was on average, greater than PM_{10} (Fig. 5). This result cannot reflect reality and suggests that this source was underestimated for PM_{10} because the mean mass of this source was very low. The apparent underestimation of the PM_{10} fraction may represent bias in the $PM_{2.5}$ models where fewer sources were identified. However, this feature may be a result of modelling all sites' time series together with PMF. Careful examination of the source profiles shows that the aged sea salt source contains components commonly associated with road traffic (Figure S3) and therefore, is likely overestimated in respect to mean concentrations. The road traffic source profiles are likely to be at least somewhat different when considering rural sites and urban-traffic sites due to their different proximities to the emission source. This outlines a compromise or a limitation with the

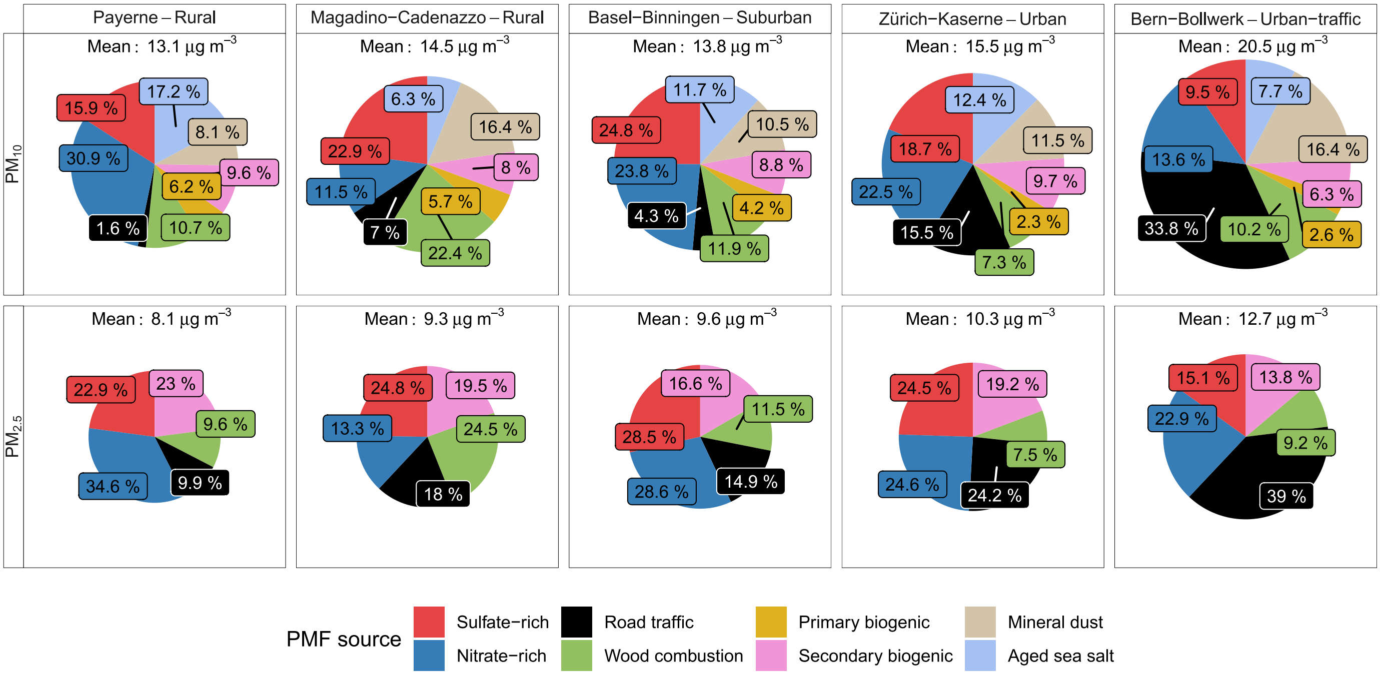

Fig. 4. PMF identified PM_{10} and $PM_{2.5}$ sources for five sampling sites in Switzerland between June 2018 and May 2019. The pie charts' areas are scaled by the sampling period's mean concentration.

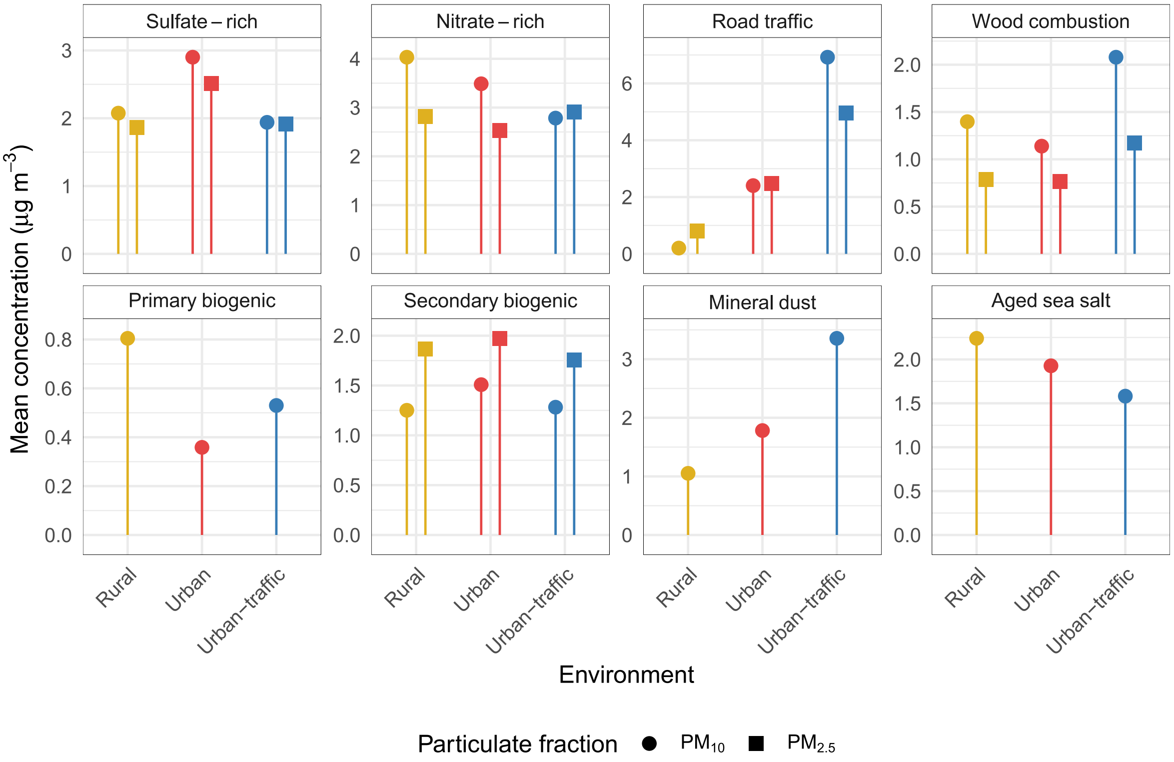

Fig. 5. PMF identified PM_{10} and $PM_{2.5}$ sources' means at three environments in Switzerland between 2018 and 2019.

different usage-modes of PMF – a user can run individual models and classify potentially heterogeneous factors with the same source label or, a user can pool all observations to get consistent factor profiles (which has been done here) and potentially miss subtle differences in the sources among the different sites included in the analysis. The 2 p.m. size fractions available in this analysis allows for this limitation to be outlined.

The secondary biogenic source for $PM_{2.5}$ shows this bias too, but this can be explained due to the $PM_{2.5}$ source containing significant components which were found in the primary biogenic PM_{10} source, thus increasing the mean mass of the source. Despite the points related to PMF limitations discussed above, the source apportionment results clearly indicate the road traffic and mineral dust sources were the two sources which were enhanced when moving along the rural, urban, and urban-traffic gradient (Fig. 4; Fig. 5).

3.2. Road traffic emission contributions

The PM_{10} and $PM_{2.5}$ concentration gradient across the different site types and the identified PMF sources indicated that emissions from road traffic and mineral dust were primarily responsible for the urban and urban-traffic PM increments observed in Switzerland (Fig. 5). Using the detailed compositional data gained from filter measurements and various laboratory analyses, an exploration of what processes drive the enhanced concentrations was conducted. Road traffic emissions can be sub-classified into three processes: exhaust emissions, non-exhaust emissions, and secondary generation from gaseous precursor emissions (Harrison et al., 2021b). Although secondary generation from road traffic emissions can be an important source of PM in some locations, especially when considering SOA (Platt et al., 2014), secondary PM components show a spatially low variability and did not contribute to the urban and urban-traffic increments. Therefore, the focus here is on primary components. As discussed in Section 1.3, non-exhaust emissions have become an increasingly important component to consider in developed countries because of the dramatic improvements of controlling combustion-sourced products emitted from light- and heavy-duty vehicles' exhausts.

Tracers of brake, tyre, road wear, and resuspended road dust are somewhat difficult to determine because of the variation of vehicles, vehicle components, regulation of materials, and environments experienced in different urban areas (Amato et al., 2011; Lawrence et al., 2013). However, antimony, copper, and barium have been identified as appropriate tracers for brake wear due to their use in brake pad and shoe fillers (Luhana et al., 2004; Gietl et al., 2010; Varrica et al., 2013; Charron et al., 2019; Harrison et al., 2021a). Barium has been commonly used a major tracer of brake wear because it lacks other common sources, with the exception of intermittent firework emissions (Gietl et al., 2010). Iron is also used as a brake wear indicator, but not of brake pads or shoes, but of discs or rotors. However, iron is also a significant component of crustal matter which indicates road wear and resuspended road dust too, so iron is usually given the definition of both a brake and road wear tracer (Gietl et al., 2010). Zinc has been used as a tracer for tyre wear because zinc oxide is used as a vulcaniser (hardener) for road vehicle tyres and zinc oxide forms approximately 1–2% of tyre tread content (Luhana et al., 2004). Although zinc does have other sources, it has been used successfully as a tyre wear tracer by some (Harrison et al., 2012). Cadmium has also been associated with tyre wear particles in some urban areas (Hjortenkrans et al., 2007; Adamiec et al., 2016).

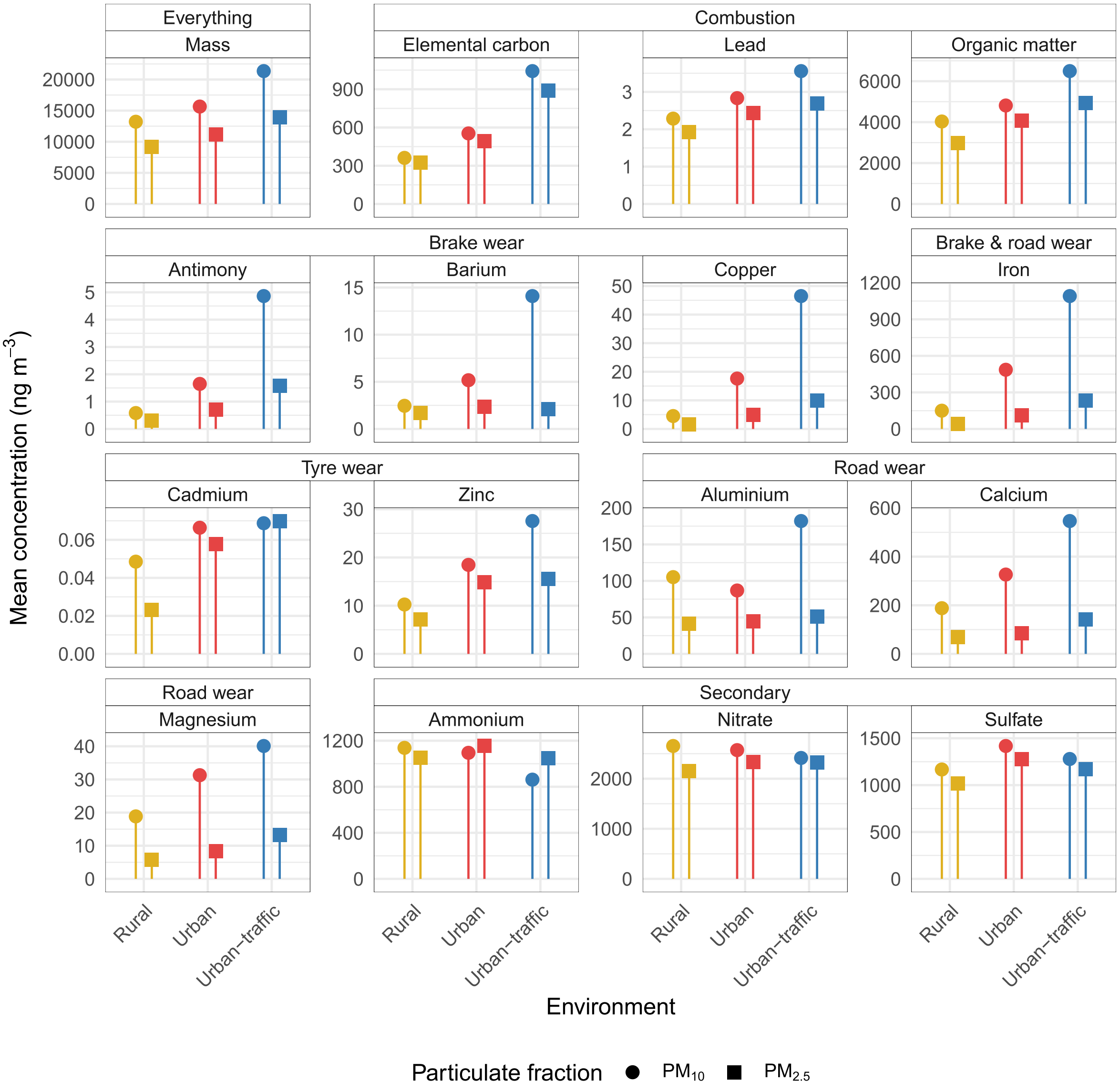

Fig. 6. Mean concentrations of PM_{10} and $PM_{2.5}$ constituents in three environments across Switzerland for selected groups of components between 2018 and 2019.

Notably, in a parallel sampling campaign complementing this work, Rausch et al. (2022) reported tyre and road wear particles (TRWP) being prominent at Zürich-Kaserne and Bern-Bollwerk when PM was sampled and analysed by passive samplers and scanning electron microscopy and energy dispersive X-ray spectroscopy (SEM/EDX).

All components sourced from combustion, brake, tyre, and road wear processes show increased concentrations when considering their urban and urban-traffic PM_{10} increments across Switzerland (Fig. 6; for box-plots see Figure S6). The brake wear elements, specifically antimony, barium, copper, and iron showed the strongest enhancements across the rural-urban-urban-traffic gradient. Tyre wear tracers, especially zinc showed substantial enhancement, as did the crustal elements, calcium and magnesium, and to a lesser extent aluminium, all of which are indicative of road surface wear or road dust resuspension. Combustion sourced products, EC and OM also demonstrated clear enhancements. The inclusion of lead as a combustion-sourced element can be debated, but it is included for completeness and showed urban and urban-traffic increments, despite its use in fuels being extremely limited after 2000. The persistence of lead in road transport corridors is the likely cause of this lead enhancement (Resongles et al., 2021), and although the national emission inventory indicates most contemporary lead emissions are from heavy industrial processes (Federal Office for the Environment, 2021c), it is unlikely that the sampling sites in this analysis will be influenced by such activities due to their locations. In contrast, the secondary generated ions, ammonium, nitrate, and sulfate showed no urban or urban-traffic increments indicating that these ions along with their products (primarily ammonium nitrate and sulfate), were not enhanced within Switzerland's urban areas which is consistent to the PMF analysis presented in Section 3.1. Curiously, Bern-Bollwerk and Zürich-Kaserne's mean ammonium concentrations were slightly higher in $PM_{2.5}$ than PM_{10}. Even after a careful exportation of the analytical process, the reasons for this inconstancy is unknown.

When the urban-rural and urban-traffic-urban ratios are plotted with the same source groupings as Fig. 6, the brake wear elements are clearly the most enhanced components in Switzerland's urban areas (Fig. 7). For example, antimony, an element used in brake pad fillers had concentrations in PM_{10} that were ≈ 3 times higher in urban areas compared to rural environments, while the urban-traffic environment was also enhanced by ≈ 3 times when considering PM_{10}.

It is important to note that the urban enhancement was similar for many specific non-exhaust emission tracers to the urban-traffic enhancement. For example, antimony, barium, copper, and iron all had over twice the concentrations in urban environments compared to rural areas (Fig. 7). This demonstrates that despite urban-background site not being directly influenced by road traffic, non-exhaust particles significantly contaminated the urban atmosphere, even when away from

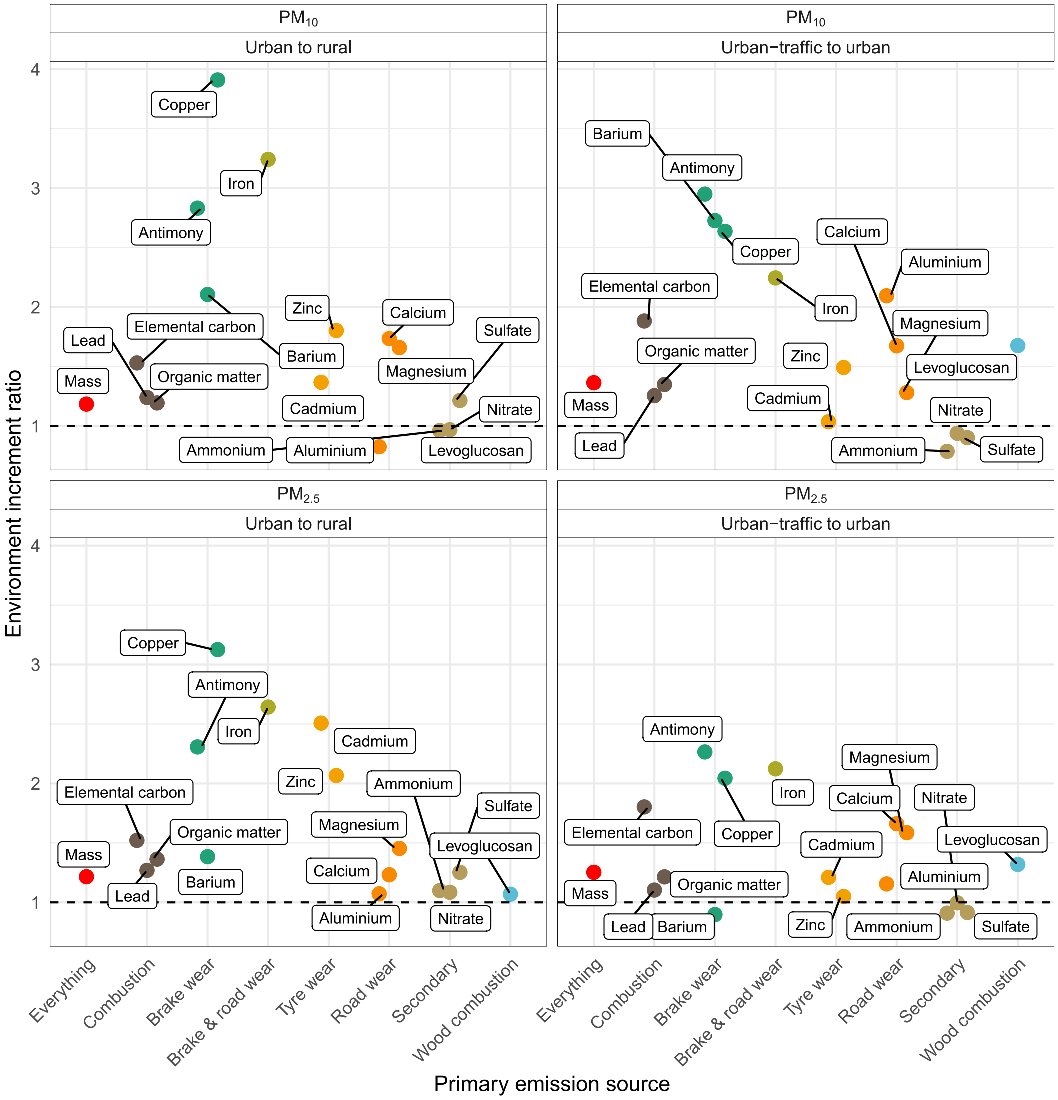

Fig. 7. Switzerland's urban to rural and urban-traffic to urban increment ratios for a selected number of species grouped by their emission source.

the immediate area surrounding road networks.

The $PM_{2.5}$ plots (lower panels of Fig. 7) also demonstrate that although the non-exhaust emissions are generated by abrasive processes, fine PM was also emitted. This resulted in enhanced concentrations of many metals in the $PM_{2.5}$ fraction. Like PM_{10}, the most enhanced tracers for $PM_{2.5}$ were those belonging to the brake wear group, but tyre and road wear tracers were also enhanced to a lesser extent. These observations reinforce the importance of fine-mode non-exhaust emissions and their importance for $PM_{2.5}$ concentrations (Harrison et al., 2021a; Vanherle et al., 2021). The primarily combustion-sourced EC and OM species had very similar enhancements in both PM_{10} and $PM_{2.5}$ demonstrating that the carbonaceous species were almost entirely in the fine PM fraction.

3.2.1. *Non-exhaust contributions*

To evaluate the non-exhaust contribution to the urban and urban-traffic increments, the sum of carbonaceous species, namely EC and OM (OC × 1.6) was used as a proxy for combustion emissions. Exhaust emissions from vehicles are overwhelmingly carbonaceous (Ruellan and Cachier, 2001; Kupiainen and Klimont, 2007), but Switzerland's urban areas have a significant loading from woodburning activities too (Fig. 4) (Hüglin and Grange, 2021) and therefore, the carbonaceous species cannot be considered an exclusive vehicular emission source in this analysis. The contribution of such emissions to the urban and urban-traffic increments was however small (Fig. 5) as also indicated by the minor urban and urban-traffic increment for levoglucosan, a key wood smoke tracer (Fig. 7).

In the case of the urban increment, the PM_{10} mass enhancement was 2.4 $\mu g\ m^{-3}$ while the combustion enhancement was 1.0 $\mu g\ m^{-3}$ (Table 2). Therefore, the proportion of the mass enhancement by combustion processes was 40%, while the remaining non-combustion enhancement was 60% of the urban PM_{10} increment. The urban-traffic increment was 5.7 $\mu g\ m^{-3}$ with a combustion-derived enhancement of 2.1 $\mu g\ m^{-3}$ and this forms a combustion to non-combustion split of 38 and 62% – a very similar split to the urban increment. The complementary $PM_{2.5}$ splits were more variable where the urban and urban-traffic combustion to non-combustion ratios were 63 to 37% and 45–55% respectively. The different ratios in $PM_{2.5}$ can be explained by the bias of combustion-sourced carbonaceous species being primarily contained in the fine fraction, while non-combustion emissions straddle both the course and fine fractions, and therefore $PM_{2.5}$ will not capture all non-combustion emission features. Table 2 presents the numbers discussed above in tabular form.

The use of carbonaceous and non-carbonaceous species in this way gives an upper bound of non-exhaust emissions because the non-carbonaceous mass includes everything apart from EC and OM and therefore, may contain additional components from other emission pathways not associated with non-exhaust emissions. To verify the above ratios, the ability to explore and interpret the PMF source profiles was leveraged. However, the limitations of the source separation ability of the PMF models required a series of corrections to make the best possible urban and urban-traffic increment estimates.

In the case of PM_{10}, the contamination of road traffic tracers in the aged sea salt factor as discussed in Section 3.1 was estimated from the progressively decreasing aged sea salt factor concentrations along the rural to urban-traffic gradient (Fig. 5), assuming that these concentration differences were caused by contributions from road traffic. Consequently, 0.6 $\mu g\ m^{-3}$ and 0.3 $\mu g\ m^{-3}$ were added to the PMF derived road traffic contribution at the rural site Payerne and the urban site Zurich-Kaserne respectively, leading to a road traffic contribution to the urban PM_{10} increment of 1.9 $\mu g\ m^{-3}$ (Table 3). Following the same approach, we assumed that the higher mineral dust contribution at the urban-traffic site Bern-Bollwerk compared to the urban site Zurich-Kaserne was due to road wear and road dust resuspension. The concentration difference of 1.6 $\mu g\ m^{-3}$ was therefore, added to the road traffic contribution at the urban-traffic site. This resulted in a road traffic contribution to the urban-traffic PM_{10} increment of 5.8 $\mu g\ m^{-3}$.

In the case of $PM_{2.5}$, the mineral dust factor was not detected by the PMF models and elements that are specific for mineral dust (for example, aluminium and titanium) were largely explained by the road traffic factor (Figure S3). Therefore, it was assumed that the road traffic factor was contaminated by geogenic mineral dust and the geogenic mineral dust concentration in $PM_{2.5}$ was calculated based on the compositional data as described in (Hüglin and Grange, 2021). This led to a subtraction of 0.3 $\mu g\ m^{-3}$ from the PMF derived road traffic contributions at the rural, urban and urban-background site, leaving the calculated road traffic contribution to the urban and urban-traffic $PM_{2.5}$ increment unchanged at 1.7 $\mu g\ m^{-3}$ and 2.5 $\mu g\ m^{-3}$, respectively (Table 3).

The above calculations suggested that road traffic emissions contributed more than 75% to the total mass increments for both environments and PM size fractions (Table 3). For the urban site, non-exhaust emissions were estimated to contribute 48% and 25% to the PM_{10} and $PM_{2.5}$ increments, while for the urban-traffic increment, the estimated non-exhaust contributions were 62% and 49%. Admittedly, these calculations and applied corrections are based on assumptions and therefore have significant uncertainties. However, they complement and agree with the compositional analysis and show that non-exhaust emissions contribute about half of the mass to the urban and urban-traffic PM_{10} increments in Switzerland and a smaller extent to $PM_{2.5}$. These calculations show that non-exhaust emissions are a very important component contributing to Switzerland's urban and urban-traffic PM_{10} and $PM_{2.5}$ increments.

3.3. *Changes in concentrations over time*

The components presented by emission source in Figs. 6 and 7 are also available for two other time periods for the same sampling sites where intensive sampling and PM characterisation was conducted (1998–1999 and 2008–2009; only PM_{10} is available for the later period) (Hueglin et al., 2005; Gianini et al., 2012a). Although the measurement programmes were not identical, they were very similar and the results are able to be compared (Hüglin and Grange, 2021). The mean concentrations of these periods, grouped by primary source are shown in Fig. 8.

Fig. 8 demonstrates that most components which are displayed decreased at the five sampling locations between 1998 and 2019. The highest concentrations of the combustion and non-exhaust sourced products (almost always at Bern-Bollwerk) show progressive decreases during the three measurement periods. Although this analysis and

Table 2

Estimates of the combustion and non-combustion contributions to the urban and urban-traffic increments based on compositional analysis. The percentages have been rounded to whole numbers.

Environment/ increment	PM size fraction	Total increment ($\mu g\ m^{-3}$)	Combustion increment ($\mu g\ m^{-3}$)	Non-combustion increment ($\mu g\ m^{-3}$)	Combustion contribution(%)	Non-combustion contribution (%)
Urban	PM_{10}	2.4	1.0	1.5	40	60
Urban-traffic	PM_{10}	5.7	2.1	3.5	38	62
Urban	$PM_{2.5}$	2.0	1.2	0.7	63	37
Urban-traffic	$PM_{2.5}$	2.8	1.3	1.6	45	55

Table 3

Estimates of the exhaust and non-exhaust contributions to the urban and urban-traffic increments based on the PMF analyses. The PMF estimates have been adjusted and the details are discussed in text. The total mass increments can be found in Table 2 and the percentages have been rounded to whole numbers.

Environment/ increment	PM size fraction	Road traffic increment (μg m^{-3})	Combustion increment (μg m^{-3})	Non-exhaust increment (μg m^{-3})	Combustion contribution (%)	Non-exhaust contribution (%)	Road traffic contribution to total (%)	Combustion contribution to total (%)	Non-exhaust contribution to total (%)
Urban	PM_{10}	1.9	1.0	0.9	52	48	78	40	37
Urban-traffic	PM_{10}	5.8	2.2	3.6	38	62	101	38	63
Urban	$PM_{2.5}$	1.7	1.2	0.4	75	25	85	63	21
Urban-traffic	$PM_{2.5}$	2.5	1.3	1.2	51	49	88	45	43

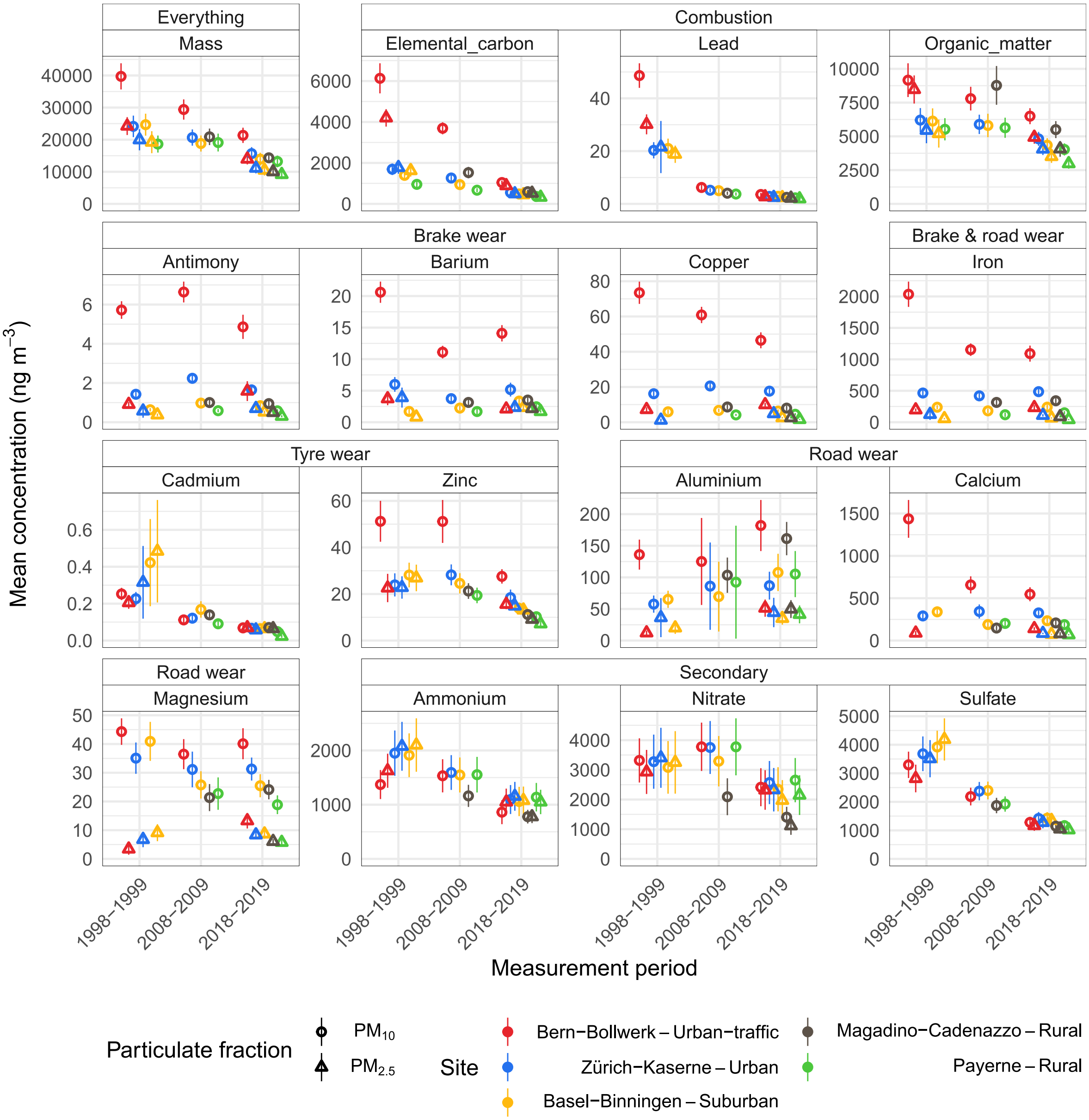

Fig. 8. Three measurement period means for selected PM_{10} and $PM_{2.5}$ constituents grouped by primary source. $PM_{2.5}$ was not sampled in the 2008–2009 measurement period and therefore, has not been included.

Switzerland's national emission inventory suggest that emissions of non-exhaust emissions have increased in the last two decades, the decreases in absolute concentrations can be explained by very significant reductions in traffic flows at Bern-Bollwerk. Between 2000 and 2019, mean daily traffic counts reduced from 28 000 to 17 500 (Federal Office for the Environment, 2017) which is a −37.8% change in the number of vehicles during these two contrasting years.

During the 1998–1999 and 2018–2019 sampling periods, EC, a prominent combustion sourced product, decreased by 80% at Bern-Bollwerk in both PM size fractions (Fig. 8). Because this decrease was over twice as large as the reduction in traffic counts, individual vehicle emission factors for EC must have strongly reduced during the two measurement periods. The decrease in emission factors can be explained with the successful control of exhaust emissions by the implementation of progressively more stringent Euro emission standards during this period. However, other constituents such as the brake wear tracers saw less of a reduction at Bern-Bollwerk compared to EC. There was a reduction in these tracers between 1998–1999 and 2019–2019 in PM_{10}, but the results for $PM_{2.5}$ were more variable. The road wear tracers are difficult to interpret at Bern-Bollwerk because the road was resurfaced in late-2007 and early 2008 which altered the make-up of the road immediately adjacent to the Bern-Bollwerk sampling site. For the other four Swiss sampling sites, changes in concentrations of the brake, tyre, and road wear tracers between 1998–1999 and 2018–2019 were rather mixed and clear patterns are difficult to resolve.

4. Conclusions and implications

The filter-based PM sampling campaign between 2018 and 2019 showed that road traffic was the emission source largely responsible for the urban and urban-traffic enhancement of PM_{10} and $PM_{2.5}$ concentrations in Switzerland. In the case of PM_{10}, non-exhaust vehicle emissions contributed ≈50% to the urban and urban-traffic increments and this emission source is currently unregulated. Additionally, the Swiss emissions inventory (Federal Office for the Environment, 2021b,c) indicates that non-exhaust emissions are one of the few PM sources which are increasing over time (Figure S1). Past observations (in 1998–1999 and 2008–2009) clearly indicate a disconnect between the change in concentrations for combustion sourced and non-exhaust tracers during this period too.

The reasons for this absolute increase in non-exhaust emissions are likely diverse. The average weight of light-duty vehicles is increasing across Europe due to consumer preference for sport utility vehicles (SUVs) and crossovers over saloons and station wagons/estate cars (Diaz et al., 2020). The progressive penetration of electric vehicles which, due to their batteries, are ≈20% heavier than their internal combustion counterparts (Beddows and Harrison, 2021) might also play a role. At a fleet or country level, such increases in weight could result in greater emissions from brake, tyre, and road surface wear processes and shows the fallacy of referring to electric vehicles as zero-emission vehicles (Timmers and Achten, 2016; Timmers et al., 2018). There are however outstanding questions regarding non-exhaust emission factors from electric vehicles, especially around the use of regenerative braking technologies. In principal, regenerative brakes could reduce friction brakes emissions to near-zero in some usage situations, but the quantification of these effects have yet to be resolved accurately (Timmers and Achten, 2016; Beddows and Harrison, 2021). Whether electric vehicles will have greater non-exhaust emissions than their internal combustion counterparts is yet to be fully determined, however, it is clear that non-exhaust emissions will remain an important process to consider, even if vehicle fleets move towards being 100% electric in the near future.

Diesel particulate filters (DPFs) have been required for light-duty vehicles since 2009 to achieve the Euro 5 emission standards across Europe. Gasoline direct injection (GDI) technology has seen rapid uptake across Europe, mostly motivated by fuel efficiency and CO_2 concerns, and this technology increases PM emissions when compared to the previous generation of port injection systems (Awad et al., 2020). Thus, GDI vehicles also require gasoline particle filters (GPF) to meet current PM emission standards. The use of PM filters for both diesel and gasoline engines are likely to further reduce exhaust PM emissions as more vehicles equipped with such a technology are put in-service and older, more polluting vehicles drop out of the in-service fleet.

All of these progressive changes to the vehicle fleet create a situation where combustion products (which are generally carbonaceous) can be expected to reduce in the near future while non-exhaust emissions will remain more stable. The estimate of ≈50% to the PM_{10} urban and urban-traffic increments by non-exhaust emissions will likely grow over the next decade. Although the results of $PM_{2.5}$ were more variable than those for PM_{10}, non-exhaust emissions, especially the tracers used for brake wear were clearly enhanced in $PM_{2.5}$. This demonstrates that non-exhaust emissions can significantly enhance urban and urban-traffic $PM_{2.5}$ concentrations, at least in some situations and environments.

The importance of non-exhaust emissions for urban and urban-traffic PM concentrations offers a challenge for regulatory bodies to address because such emission pathways are currently unregulated, but there is a need for these process to be controlled to ensure the reductions of future ambient PM concentrations in urban areas (Wu et al., 2021). The elements which were most enhanced – copper, iron, antimony, and barium have also been shown to be potent in respect to drive oxidative stress (Calas et al., 2019; Daellenbach et al., 2020). Therefore, these metals may be very important to consider when evaluating the oxidative potential of urban PM. Additional work investigating such questions using pieces of the dataset presented here is in preparation (Grange et al. In preparation). In a related point, there is a tendency of regulatory bodies to replace PM_{10} standards with those for $PM_{2.5}$. However, in respect to non-exhaust emissions, the dominating fraction is coarse and this emphasises that there remains significant value in continuing PM_{10} monitoring and regulation to understand and control this important emission pathway (European Commission, 2004; Querol et al., 2004).

Finally, the filter based sampling campaign was done in a single country in Europe, namely Switzerland. However, the conclusions of non-exhaust emissions being a large component of urban and urban-traffic PM increments is likely applicable to many similar countries. Indeed, similar conclusions have been reported elsewhere (Amato et al., 2014; Achten et al., 2018; Charron et al., 2019). We recommend regulatory bodies to evaluate the importance of non-exhaust emissions in their jurisdictions and start introducing interventions to reduce non-exhaust emissions to ensure PM concentrations continue to decrease in the near future. Secondary PM components also contribute a large portion of total PM_{10} and $PM_{2.5}$ mass, and further policy efforts at regional and transboundary scales will be required to achieve background reductions in PM mass in Switzerland and across Europe.

Data availability

The data sources used in this work are described and some data sets are publicly accessible in a persistent data repository (Grange, 2021a, https://doi.org/10.5281/zenodo.4668158). Additional data and information are available from the authors on reasonable request.

CRediT authorship contribution statement

Stuart K. Grange: Conceptualization, Methodology, Software, Formal analysis, Data curation, Writing – original draft, Writing – review & editing. **Andrea Fischer:** Data curation, Writing – review & editing. **Claudia Zellweger:** Data curation. **Andrés Alastuey:** Data curation, Writing – review & editing. **Xavier Querol:** Data curation, Writing – review & editing. **Jean-Luc Jaffrezo:** Data curation, Writing – review & editing. **Samuël Weber:** Software, Formal analysis. **Gaëlle Uzu:** Data curation, Writing – review & editing. **Christoph Hueglin:** Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.aeaoa.2021.100145.

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