

Environ Sci Pollut Res (2013) 20:6496–6508
DOI 10.1007/s11356-013-1682-1

RESEARCH ARTICLE

PM₁₀ source apportionment in a Swiss Alpine valley impacted by highway traffic

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Received: 22 December 2012 / Accepted: 26 March 2013 / Published online: 23 April 2013
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Abstract Although trans-Alpine highway traffic exhaust is one of the major sources of air pollution along the highway valleys of the Alpine regions, little is known about its contribution to residential exposure and impact on respiratory health. In this paper, source-specific contributions to particulate matter with an aerodynamic diameter < 10 μm (PM₁₀) and their spatio-temporal distribution were determined for later use in a pediatric asthma panel study in an Alpine village. PM₁₀ sources were identified by positive matrix factorization using chemical trace elements, elemental, and organic carbon from daily PM₁₀ filters collected between November 2007 and June 2009 at seven

locations within the village. Of the nine sources identified, four were directly road traffic-related: traffic exhaust, road dust, tire and brake wear, and road salt contributing 16 %, 8 %, 1 %, and 2 % to annual PM₁₀ concentrations, respectively. They showed a clear dependence with distance to highway. Additional contributions were identified from secondary particles (27 %), biomass burning (18 %), railway (11 %), and mineral dust including a local construction site (13 %). Comparing these source contributions with known source-specific biomarkers (e.g., levoglucosan, nitro-polycyclic aromatic hydrocarbons) showed high agreement with biomass burning, moderate with secondary particles (in winter), and lowest agreement with traffic exhaust.

We would like to dedicate this paper to the late Prof. Lee-Jane Sally Liu, who passed away in June 2011. We are deeply grateful for her supervision and guidance throughout the study, and she still remains a constant inspiration.

Responsible editor: Gerhard Lammel

Electronic supplementary material The online version of this article (doi:10.1007/s11356-013-1682-1) contains supplementary material, which is available to authorized users.

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Keywords Source apportionment · PMF · Highway traffic ·
PM₁₀ · Swiss Alpine valley · Levoglucosan · NitroPAH

Introduction

Inhalable particulate matter consists of a complex mixture of particles and droplets of varying size and composition. They originate from different natural and anthropogenic sources, e.g., fires, windblown dust, motor vehicles, power plants, construction activities, and contain primary and secondary particles (Seinfeld and Pandis 2006). Although there is considerable evidence that airborne particulate matter (PM) is associated with adverse respiratory and cardiovascular health effects (Davidson et al. 2005; Pope and Dockery 2006; Russell and Brunekreef 2009; US EPA 2009), only few studies have examined association of apportioned components and sources of PM with health outcomes (Stanek et al. 2011).

From a policy point of view, it is crucial to identify air pollution sources that have the greatest impact on public health in order to apply targeted emission management

strategies. For example, emission regulations are continuously tightened for traffic exhaust, one of the most investigated sources (Brugge et al. 2007; Health Effects Institute 2010). However, there is increasing evidence of health impacts from other sources such as biomass burning (Laumbach and Kipen 2012; Naeher et al. 2007) or Saharan dust events (Karanasiou et al. 2012). Therefore, application of receptor modeling has become more and more important in recent years for estimating the exposure from the different sources (Hopke 2003; Viana et al. 2008).

Due to health concerns raised in communities along the main north–south transit route crossing the Alps (Hazenkamp-von Arx et al. 2011), Swiss Federal Office for the Environment funded this pediatric asthma panel study to investigate the health impact of short-term air pollution from local sources. The primary north–south transit corridor study (Hazenkamp-von Arx et al. 2011) comprised of ten communities along the Swiss Alpine highway corridors including Erstfeld, a village located in a narrow and flat Alpine valley bordered by steep mountain slopes along the Swiss transit highway A2. The focus of the current study is the chemical composition of PM in this village. One and a half years of air pollution measurements, at several sites within the village, were used to analyze the temporal and spatial distribution of the different pollutants (Ducret-Stich et al. 2013). Traffic-related pollutants such as nitrogen dioxide (NO₂), elemental carbon (EC) and particle number concentration were highly heterogeneous in space (i.e., decreasing concentrations with increasing distance to the highway). In contrast, particulate matter with an aerodynamic diameter < 10 μm (PM₁₀) and organic carbon (OC) influenced also by sources other than traffic, showed a more homogeneous spatial distribution.

The aim of this paper is to find and apportion the main sources contributing to the total PM₁₀ in Erstfeld using positive matrix factorization (PMF). Single sources such as traffic exhaust, biomass burning, and secondary pollutants are validated by comparison with source-specific biomarkers (e.g., levoglucosan, nitro-polycyclic aromatic hydrocarbons). We describe the spatial and temporal distribution of the source-specific contributions, with special focus on highway-related sources. The modeled source-specific contribution estimates are subsequently being used to assess acute respiratory health effects in school children.

Methods

Study description

From November 2007 to June 2009, several air pollutants were measured in Erstfeld, a village with about 3,800 inhabitants located in an 800 to 900 m wide Alpine valley in

Switzerland. A detailed description of the study region, monitoring sites, and measured pollutants can be found elsewhere (Ducret-Stich et al. 2013). Daily PM₁₀ filters were concurrently collected at one fixed highway and one mobile station, which was moved each month to one of the seven locations (Fig. 1). The highway site was situated north of the village right beside the highway (22 m from the centerline). From that location, three mobile sites were additionally located at perpendicular distances of 55, 102, and 198 m to the highway (Fig. 1, sites 1–3). The other four mobile sites were chosen to represent the residential area and to capture the impact of additional sources such as the main road and railways (Fig. 1, sites 4–7). The mobile station measurements were scheduled to cover all seasons for each mobile station in the residential area during the

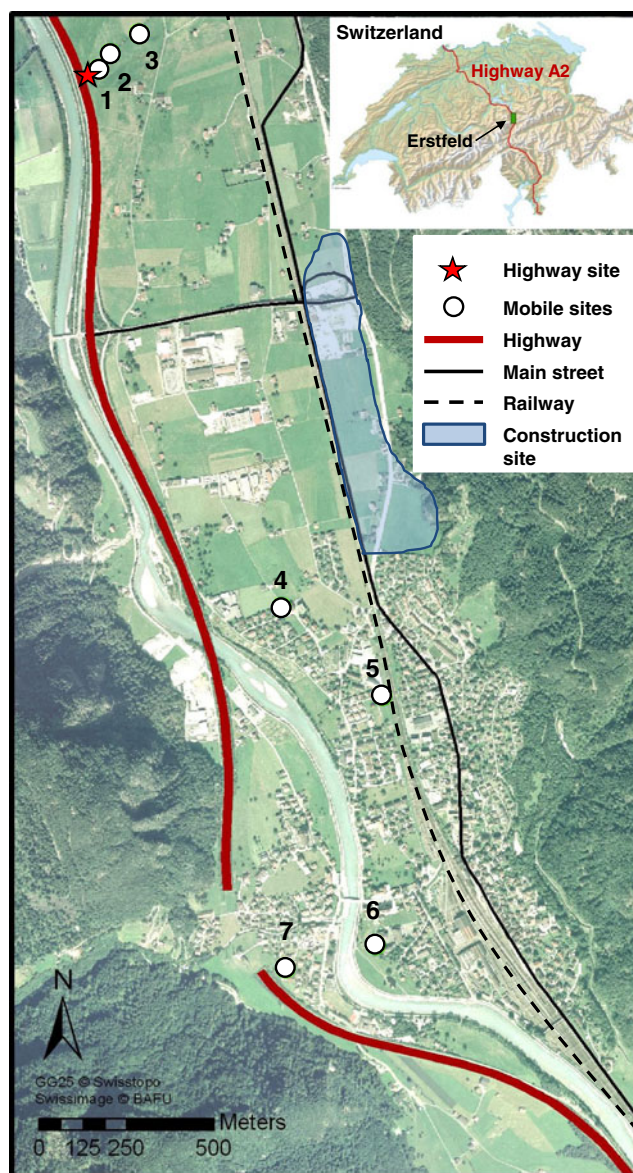


Fig. 1 Map of Erstfeld with measurement sites

study. During the whole study period there was a major construction site north-east of site 4 (Fig. 1), where the largest train tunnel in the world, the base tunnel through the Gotthard mountain (Alp Transit), is still being built.

Sampling methods

At the highway and mobile sites, high volume samplers (Digitel DHA-80) with a flow rate of 30 m³/h were used to collect concurrent 24-h PM₁₀ on 150 mm quartz fiber filters (Pallflex Tissuquartz 2500QAT-UP, Pall AG, Switzerland). At the mobile sites, 24-h PM₁₀ was additionally collected on 47 mm Teflon filters (PTFE 2.0 μm pore size, Blanc-Labo S.A., Switzerland) using a Partisol low volume sampler (Partisol®-Plus Modell 2025 Sequential Air Sampler, Thermo Fisher Scientific) with a flow rate of 1 m³/h. All filters were pre-weighted and pre-conditioned (48 h at 22 °C and 50 % RH), and their mass concentrations were determined by standard gravimetric methods. The limit of detection, determined as three times the standard deviation of the field blanks, was 2.3 μg/m³ (*N*=57) and 2.4 μg/m³ (*N*=30) for the quartz- and Teflon-filters, respectively. The two measurement methods (high volume versus Partisol) agreed very well with an *R*² of 0.93 (slope=0.96; intercept=-1.78 μg/m³) and a mean difference of 1.12±2.86 μg/m³ (*N*=521).

Chemical analysis

All of the quartz fiber filters were analyzed for EC and OC using a thermal-optical transmission method (TOT, OCEC Analyzer Sunset Laboratory Inc.). Due to a protocol change in 2009, the filters were analyzed with two different temperature profiles. Filters from 2007/2008 were analyzed with the NIOSH700⁺ protocol, while filters from 2009 were analyzed with the EUSAAR2 protocol, details of the protocols are described separately (Ducret-Stich et al. 2013). Since the two protocols provided different EC–OC fractions, it was not possible to use the fractionated EC–OC data without losing a substantial portion of the data (>30 % corresponding to 2009 measurements). Thus, we used total EC and OC and adjusted the NIOSH700⁺ concentrations to levels of the EUSAAR2 protocol with the following factors derived from orthogonal regression analysis of 102 ambient filters analyzed with both protocols (Ducret-Stich et al. 2013):

$$EC_{\text{EUSAAR2}} = 1.10 \times EC_{\text{NIOSH700+}} \quad (1)$$

$$OC_{\text{EUSAAR2}} = 0.92 \times OC_{\text{NIOSH700+}} \quad (2)$$

The Teflon filters at the mobile sites were used to determine concentrations of 48 chemical elements (Table 1 and

Table 1 Summary statistics, method detection limit (MDL), % of data below MDL, signal-to-noise ratio (S/N), and PMF category of all species which were used for PMF analysis (*N*=510)

Species	Mean (ng/m ³)	SD (ng/m ³)	MDL (ng/m ³)	% below MDL	S/N	PMF category
PM ₁₀	15,490	9,732	2,400	0	0.8	Weak
MM ^a	3,030	3,944			0.7	Weak
EC	1,112	706	51	1	3.0	Strong
OC	2,761	1,587	476	2	2.1	Strong
Al	132	232	1.63	5	25.9	Strong
Ba	5.98	7.21	0.77	25	2.4	Weak
Br	1.77	1.07	0.29	4	8.3	Weak
Ca	340	319	0.48	0	26.3	Strong
Cl	85	172	1.73	44	25.5	Strong
Cr	3.86	3.76	0.34	3	11.9	Strong
Cu	12	10	0.43	0	20.3	Weak
Fe	975	1,024	0.48	0	26.6	Strong
K	352	281	1.10	0	26.4	Strong
Mg	32	51	2.02	22	10.4	Strong
Mn	13	11	0.43	0	20.9	Strong
Na	24	55	7.82	75	5.6	Weak
Ni	0.65	0.74	0.34	45	2.8	Strong
Pb	4.75	4.65	0.48	9	12.3	Weak
Rb	1.04	0.79	0.34	19	3.8	Strong
S	606	457	0.53	0	27.1	Strong
Si	442	625	1.20	0	26.9	Strong
Ti	14	21	0.29	3	19.1	Strong
V	0.60	0.69	0.29	42	1.4	Weak
Zn	21	15	0.58	0	22.6	Strong

For summary of all other species see Online Resource Table S1

^aMissing mass

Online Resource Table S1) by energy-dispersive X-ray fluorescence (XRF) (Cooper Environmental Services LLC, Portland, OR, USA). After the non-destructive XRF analysis, PM was extracted from the filters using Dichloromethane (CH₂Cl₂) (Miller-Schulze et al. 2010) and combined into 1- and 2-week composites for analysis of three nitro-polycyclic aromatic hydrocarbons (NPAHs) and levoglucosan (LG), respectively. NPAHs were analyzed with two-dimensional high-performance liquid chromatography tandem mass spectrometry (Miller-Schulze et al. 2010) and included 1-nitropyrene (1-NP), a diesel marker (Albinet et al. 2007; Ringuet et al. 2012b; Scheepers et al. 1995), and 2-nitropyrene (2-NP) and 2-nitrofluoranthene (2-NFI), two markers for secondary pollution (Arey et al. 1967; Ringuet et al. 2012a, b; Sweetman et al. 1967). The wood smoke marker LG (Jordan et al. 2006; Simoneit et al. 1999) was analyzed by gas chromatography/mass spectrometry as described in Simpson et al. (2004).

Mass closure

A mass closure analysis was performed on the data by comparing the reconstructed mass (RCM) with the measured mass. To calculate the RCM, the following estimates were used:

$$OM = 1.6[OC] \tag{3}$$

$$\text{Sulfate} = 4.125[S] \tag{4}$$

$$\begin{aligned} \text{Mineral dust} = & 2.14[Si] + 1.89[Al] + 1.66[Mg] \\ & + 1.40[Ca_{\text{geo}}] + 1.21[K_{\text{geo}}] \\ & + 1.43[Fe_{\text{geo}}] \end{aligned} \tag{5}$$

$$\begin{aligned} \text{RCM} = & [EC] + OM + \text{Sulfate} + \text{Mineral dust} \\ & + \sum \text{anthropogenic part of Ca, K, Fe} \\ & + \sum \text{all other species} \end{aligned} \tag{6}$$

where [] represent the concentration of the specific element. Organic matter (OM) (Eq. 3) is accounting for the mass of oxygen and other unmeasured elements in non-carbon organic matter (Turpin and Lim 2001). Although Turpin and Lim recommended a multiplication factor larger than 1.6 for non-urban areas, we used 1.6 because of the impact from the highway traffic on the air pollution in this valley (Ducret-Stich et al. 2013). In Eq. 4, the ratio of the atomic weights of ammonium sulfate ((NH₄)₂SO₄) and Sulfur (S) is used assuming all the sulfate is fully neutralized and occurs on the Teflon filter as ammonium sulfate. For mineral dust (Eq. 5), the elements are assumed to be present as oxides (Chow et al. 1994). However, as anthropogenic sources also emit calcium (Ca), potassium (K), and iron (Fe), only the geogenic part of these elements were used, i.e., Ca_{geo}=1.15[Al], K_{geo}=0.61[Al], and Fe_{geo}=1.13[Al] (Gianini et al. 2012b).

RCM, as defined in Eq. 6, explained on average 76 % (SD=12 %) of the measured mass (Online Resource Fig. S2). As our OM estimate (Eq. 3) was rather conservative (Turpin and Lim 2001), the missing mass (MM), defined as measured mass minus RCM, also contained the organic mass not accounted for in Eq. 6. In addition, RCM did not include nitrates, which were not measured in this study. Because MM showed higher concentrations in winter, MM was assumed to represent nitrates, which are mostly present as ammonium nitrate (NH₄NO₃) and readily form and persist in cold temperature environments (Seinfeld and Pandis 2006). MM, with an average of 24 % PM₁₀ mass, was also comparable to secondary nitrate contributions to PM₁₀ in other Swiss

regions (22–32 %) (Gianini et al. 2012a). Therefore, we included the MM as an additional “species” representing nitrates and additional organic matter in the source apportionment analysis.

Receptor modeling

To find and apportion the different sources of PM₁₀, we used the PMF software from the U.S. Environmental Protection Agency (EPA PMF 3.0). PMF is a receptor model, which solves a positively constrained bilinear mass balance model based on a weighted least-squares fit (Paatero 1997; Paatero and Tapper 1994).

Database construction

Chemical species with a large fraction (>50 %) of concentrations below the method detection limit (MDL) were excluded from the analysis (Online Resource Table S1), except sodium (Na), which was included as a tracer for road salt. Arsenic (As) was excluded due to weak signals where only 31 % of the concentrations were above three times the MDL. Most species with an atomic number above 40 (zirconium (Zr)) except for lead (Pb), and barium (Ba) were excluded because of the difficulties in XRF analysis. Phosphorus (P) was also excluded as it is generally a poor variable in XRF analysis due to the influence of the large Sulfur (S) peak that is adjacent in the X-ray spectrum. The final data included 24 species (Table 1).

Concentrations and uncertainties of the chemical species data were processed according to Polissar et al. (1998) for the PMF analysis. Values below the MDL were replaced by half of the MDL values, and their uncertainties were set to 5/6 of the MDL values. There were no missing values in our data set. For EC and OC, the uncertainties were set to the sum of ½ of the minimum measured value of all samples (EC, 0.01 µg/m³; OC, 0.12 µg/m³) plus instrument detection limit obtained from the manufacturer (0.04 µg/m³) plus MDL, defined as three times the standard deviation of the field blanks calculated for each set of blanks (2007/2008 data (NIOSH700+) and 2009 data (EUSAAR2)) (e-CFR, 2012). The uncertainty in the MM was calculated as the square root of the sum of the variances of all measured species and measured mass (Wu et al. 2007). PM₁₀ was additionally included as an independent variable into the PMF model to directly obtain the source contributions to the daily PM₁₀ mass concentrations. It is assigned a high uncertainty of four times the value to decrease its weight in the model fit (Kim et al. 2003b). Uncertainty of species with a signal-to-noise ratio (S/N)<2 (PM₁₀, MM, vanadium (V)) were increased by a factor of three by categorizing them as “weak” in the PMF analysis (Norris et al. 2008; Paatero and Hopke 2003). Also Na, Ba, and Pb were set to weak to

account for their poor data quality described above. In addition, bromine (Br) and copper (Cu) were categorized as weak to reduce the number of scaled residuals $>\pm 3$ (Norris et al. 2008; Paatero and Hopke 2003).

Unusual events can distort the results of source profiles and contributions (Norris et al. 2008); therefore, 19 of 529 samples were excluded from our analysis due to: fireworks (high Ba and K, $N=3$); two Saharan dust events in May and October 2008 (high PM_{10} , silicon (Si), magnesium (Mg), aluminum (Al), titanium (Ti), $N=6$); possible local events from the construction site (high Si, Mg, Al, Ti, $N=6$); local road salt spreading (very high Na and chlorine (Cl), $N=1$); and three unexplained local events with high Pb, zinc (Zn); high Ba, K, molybdenum (Mo); and high P, respectively. Summary statistics of the final 510 samples included in the PMF analysis are provided in Table 1.

Model execution

PMF was run in the robust mode for seven to ten sources using all 510 samples together. To determine the final number of sources, the distribution of the scaled residuals was examined to ensure the data were properly fitted. The extracted profiles were compared with profiles from the literature. This combination of tests was used to identify the most physically reasonable solution. The best results were found for nine sources. For up to eight sources, the residuals were positively skewed and with ten sources the tire and brake wear factor split into two unrecognizable factors. The model uncertainty was tested with bootstrapping (100 runs) (Norris et al. 2008). All factors were mapped to a base factor in every run, indicating a stable result.

PMF does not generally produce a unique solution due to rotational ambiguity (Paatero et al. 2002). Therefore, PMF solutions were systematically explored by varying the F_{peak} parameter between -1 and $+1$ to find F_{peak} -intervals for which the penalty function $Q(E)$ values remained relatively constant (Norris et al. 2008; Paatero et al. 2002). For these intervals, PMF solutions were analyzed using *G space plotting* to investigate statistical dependence between source contributions caused by unrealistic rotations (Paatero et al. 2005). An F_{peak} value of -0.2 was found to give the best results after comparing the *G space* plots and source profiles. For $F_{peak}=0$ several profiles displayed unrealistic values as shown in Online Resource Fig. S3.

Validation of the final PMF solution was done by regressing the predicted PM_{10} (sum of all source contributions from PMF) with the measured PM_{10} which resulted in an R^2 of 0.87 (Fig. 2). However, an increasing dispersion around the regression line was observed for higher mass values measured mainly in winter (Fig. 2). This might be caused by the underestimation of the organic matter in winter and/or some loss of semi-volatile material on the filters due to temperature changes before weighing. The

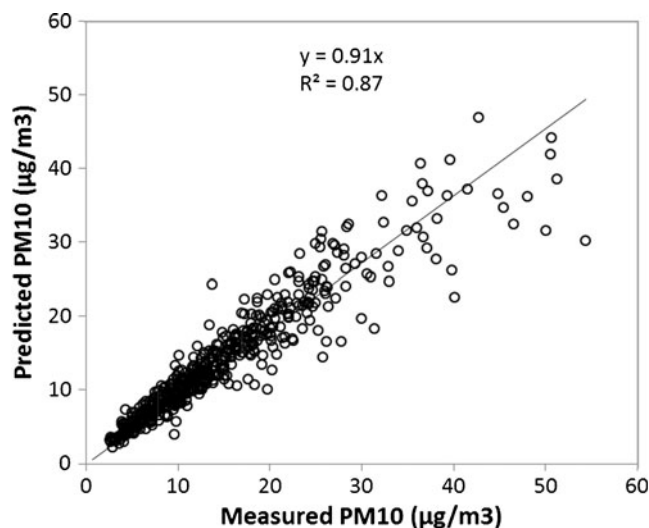


Fig. 2 Comparison of measured PM_{10} concentrations versus PMF predictions

PMF solution was additionally validated using measurements of the biomarkers levoglucosan and NPAHs to further compare source-specific contributions from biomass burning, traffic exhaust, and secondary pollution.

Results and discussion

Source profiles

We identified nine sources of which four were directly traffic-related, namely, traffic exhaust, road dust, tire and brake wear, and road salt (Fig. 3). Traffic was also indirectly related to secondary particles by producing some of their precursors. Other sources were biomass burning, railway, and two mineral-related sources, mineral dust, and minerals from the construction site (Fig. 4).

Traffic exhaust

The traffic exhaust profile was dominated by EC and OC with some contributions of abrasion-related metals such as Zn, Ba, and Cu (Schauer et al. 2006) (Fig. 3a). Source contributions were higher on weekdays than on weekends. This result corresponded to the higher weekday traffic counts of heavy duty trucks (with lower counts on weekends due to a Sunday truck traffic ban) than to the total traffic, which showed higher counts on weekends (Ducret-Stich et al. 2013) (Online Resource Fig. S4b). Although this relationship seemed to point to a strong correlation between attributed traffic exhaust PM_{10} and diesel-powered heavy duty trucks, comparison with the diesel marker, 1-NP, showed only an R^2 of 0.13 (Fig. 5a). A higher correlation was obtained with the more general traffic marker NO_2 ($R^2=0.50$) (Fig. 5b). In contrast to daily

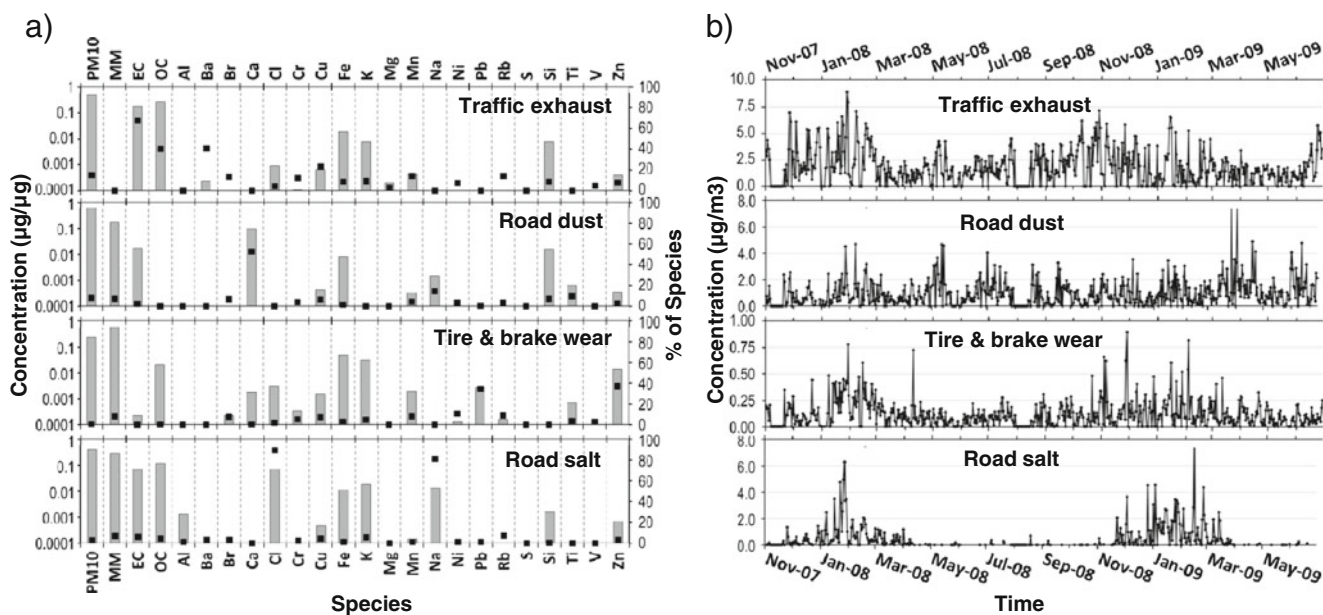


Fig. 3 Attributed sources directly related to traffic: **a** PM₁₀ source profiles (solid bars, left axis) with explained variation (squares, right axis) and **b** daily PM₁₀ source contributions. Left axis in source profiles

describes the normed concentration such that the sum of all elemental contribution within a factor equals 1

NO₂ measurements, 1-NP was only available as weekly average concentrations. Moreover, filter reaction artifacts during sampling may have resulted in overestimation of 1-NP and thus to the low correlation with traffic exhaust PM₁₀. Reported EC/OC ratios for gasoline and diesel vehicle emissions

typically range between 0.1 to 0.5 and 1.0 to 4.0, respectively (Liu et al. 2006; Lough et al. 2007). The EC/OC ratio of 0.7 in our study reflected the mixed traffic fleet with about 14 % of heavy duty vehicles (diesel) and 86 % passenger cars of which about 18 % used diesel.

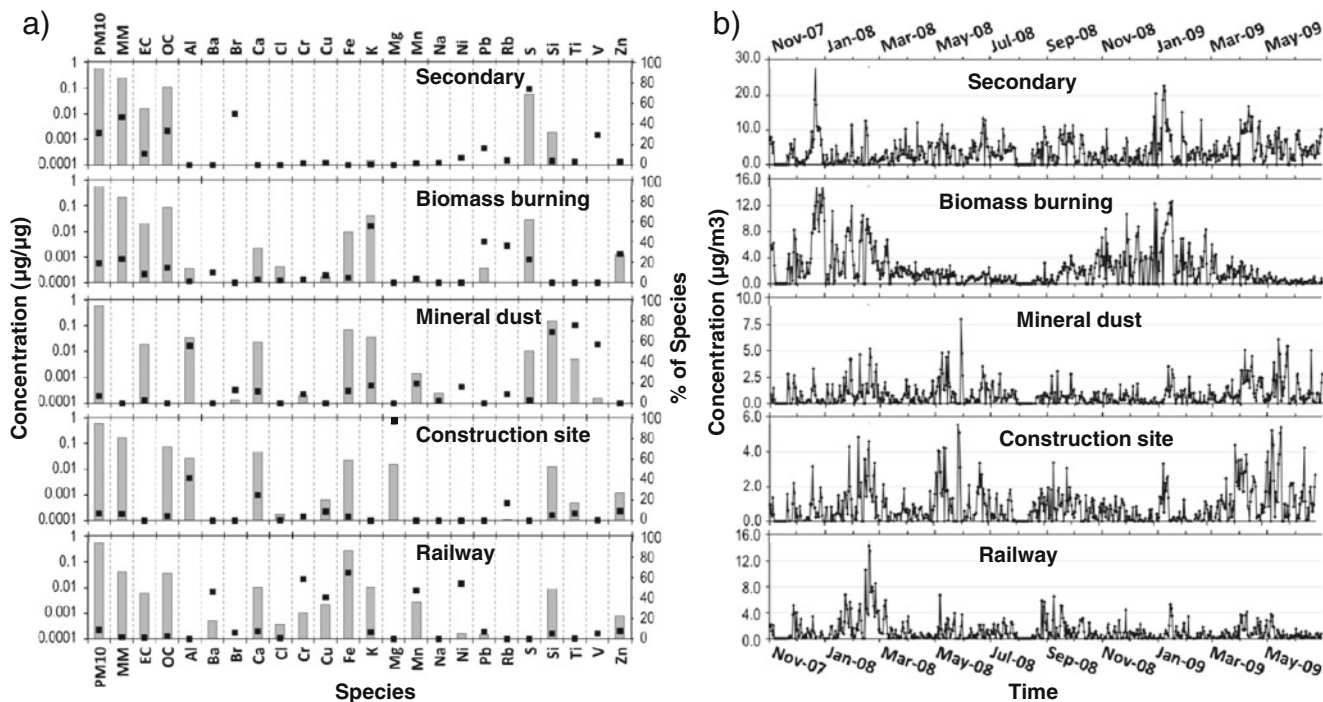


Fig. 4 Attributed sources not directly related to traffic: **a** PM₁₀ source profiles (solid bars, left axis) with explained variation (squares, right axis) and **b** daily PM₁₀ source contributions. Left axis in source profiles

describes the normed concentration such that the sum of all elemental contribution within a factor equals 1

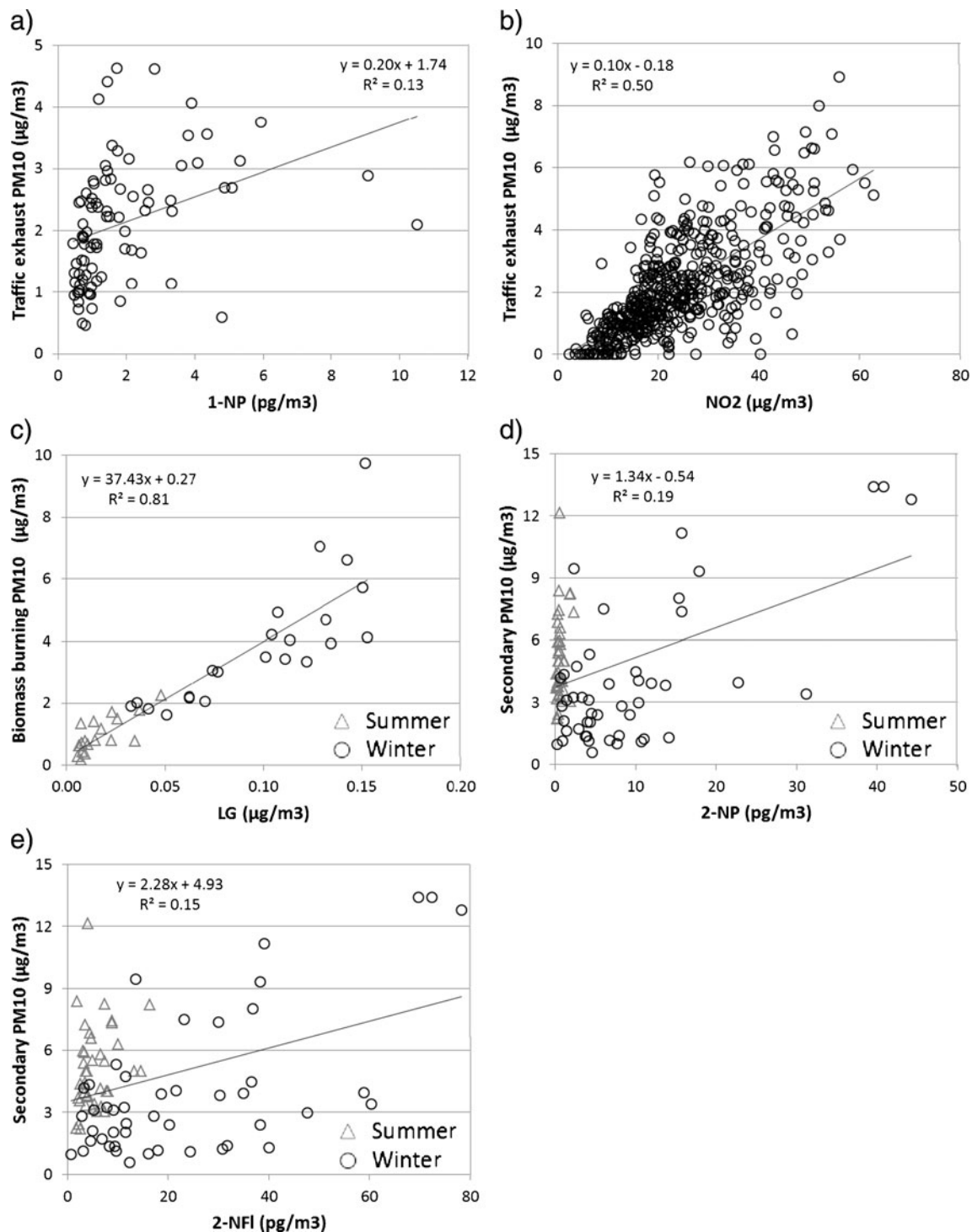


Fig. 5 Comparison of source contributions with biomarkers: traffic exhaust PM₁₀ versus **a** 1-NP and **b** NO₂; **c** biomass burning PM₁₀ versus levoglucosan; secondary particles versus **d** 2-NP and **e** 2-NFI. The individual regression equations correspond to all data combined

The seasonal differences with higher concentrations in winter than summer (Fig. 3b) were not related to traffic counts, which were higher in summer (Ducret-Stich et al. 2013) but rather to the meteorology. Air pollution levels were generally higher in winter than in summer due to lower wind speeds and lower mixing heights including frequent

inversions in this valley (Ducret-Stich et al. 2013). In Fig. 6, exhaust and non-exhaust (i.e., resuspended road dust, tire, and brake wear) traffic source contributions were compared between sites at different distances from the highway. For traffic exhaust, a clear gradient was observed with the higher contributions being closer to the highway.

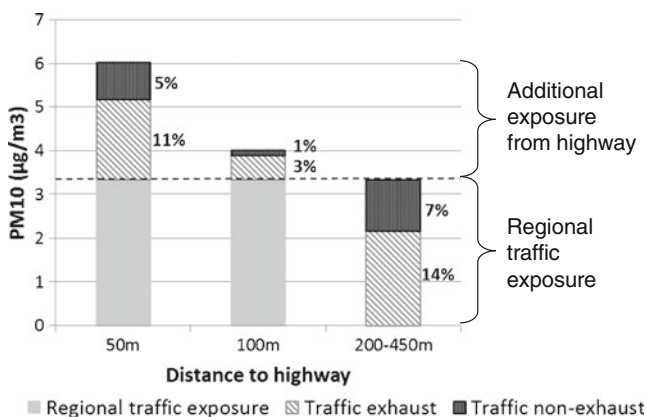


Fig. 6 Spatial distribution of traffic source contributions for the year 2008. Concentrations are normalized by the ratio of the concurrently measured PM₁₀ and the annual average PM₁₀ measured at the highway site. Data were averaged as follows: site 1 (50 m), site 2 (100 m), average of sites 4–6 (200–450 m). Exclusions: site 3 (only nine samples) and site 7 (protected by tunnel and noise wall)

Some recent studies have used EC and OC fractions to apportion traffic exhaust into diesel and gasoline sources (Kim and Hopke 2005; Kim and Hopke 2004; Liu et al. 2006). However, in our attempt to use EC and OC fractions in the PMF analysis, the traffic exhaust factor was not split. It must be mentioned that for the analysis with fractions, only two thirds of the data were available due to different procedures in the analysis of EC and OC (see “Chemical analysis”).

Road dust

Road dust profiles are often difficult to identify due to similar elements seen in mineral dust. Our road dust profile included Si and Fe but differed from mineral dust with a high contribution of Ca (Hueglin et al. 2005; Schauer et al. 2006; Thorpe and Harrison 2008) (Fig. 3a). Contributions of this factor were higher in spring and summer (Fig. 3b), probably due to dryer weather conditions enhancing resuspension. They were also higher on weekdays, probably due to the heavy duty traffic (Online Resource Fig. S4c), which causes more resuspension of road dust than passenger cars (Bukowiecki et al. 2010; Gehrig et al. 2004).

Tire and brake wear

Tire and brake wear contributed to higher concentrations of Cu, Fe, Zn, and Pb (Bukowiecki et al. 2010; Schauer et al. 2006; Thorpe and Harrison 2008) (Fig. 3a). Concentration levels were a little elevated in winter (Fig. 3b) probably caused by higher abrasion of winter tires and increased braking due to the weather conditions. Higher concentrations were again observed on weekdays (Online Resource Fig. S4d) likely the result of heavy duty truck traffic (Bukowiecki et al. 2010).

Road salt

The de-icing road salt factor had a very clear signature with more than 80 % of Na and Cl apportioned to it (Gianini et al. 2012a) (Fig. 3a). This factor was only seen in winter (Fig. 3b) and showed no differences with respect to day of the week.

Biomass

The biomass burning profile was characterized by high concentrations of K, EC, and OC (Gianini et al. 2012b; Khalil and Rasmussen 2003; Reid et al. 2005) and other typical elements for wood smoke (Rb, S, Fe, Ca) (Godoy et al. 2005; Reid et al. 2005) (Fig. 4a). The EC/OC ratio was 0.24, which was on the upper level compared with other studies in Switzerland (Gianini et al. 2012b; Szidat et al. 2006). This somewhat higher ratio might be explained by higher contributions from EC combined with substantial amounts of Pb and Zn observed in the profile (Fig. 4a) pointing to a mixture of biomass burning with some traffic. This was supported by factor contributions in summer, when almost no biomass burning should have been present (Fig. 4b). Nevertheless, PM₁₀ contribution from this biomass profile was highly correlated with the known wood biomarker levoglucosan ($R^2=0.81$, Fig. 5c). In winter, OC also showed a good correlation with biomass burning ($R^2=0.72$), whereas EC primarily originated from traffic ($R^2=0.86$) (Online Resource Fig. S5).

Secondary particles

The secondary particle factor included about 75 % of MM, 50 % of S, and about 30 % of OC (Fig. 4a). MM was assumed to represent nitrates and part of the organic matter (see “Mass closure”) while S associated with OC represented sulfates in the secondary aerosol (Gianini et al. 2012a; Kim et al. 2003a). Except during two inversion episodes in December 2007 and January 2009, this factor showed generally higher contributions in spring and summer (Fig. 4b), when increased photochemical activity is favoring the formation of secondary particles (Kim et al. 2003a; Seinfeld and Pandis 2006). This result was also shown by the measured ratio between 2-nitrofluoranthene and 2-nitropyrene (2-NFI/2-NP) of 5.7 (9.0 in summer, 2.6 in winter) indicating that secondary particles were mainly formed by reaction with daytime OH⁻ rather than NO₃-radicals (Arey et al. 1986). Nevertheless, the correlations between secondary PM₁₀ and 2-NFI or 2-NP were small with an R^2 of 0.15 and 0.19, respectively (Fig. 5d, e). These results were heavily influenced by the very low concentrations of 2-NFI and 2-NP in summer showing almost no correlation with secondary PM₁₀. Correlations in winter were stronger with 0.40 and 0.49 for 2-NFI and 2-NP, respectively. In contrast to daily secondary particle contributions, nitro-PAHs were

only available as weekly concentrations leading to generally low correlations. Also, some of the nitro-PAHs may be present as a result of sampling artifacts (Schauer et al. 2003) and thereby lower the observed correlations.

Mineral factors

Contributions from mineral sources were split into two profiles: mineral dust and construction site emissions (railway tunnel “Alp Transit”) (Fig. 4a). The crustal elements Si, Al, Ca, and Fe were shared by both profiles. While most of the Si was apportioned to mineral dust, almost all Mg was present in the construction site factor. Mg as an additive to the concrete, which was prepared with recycled excavated material and used on site, might have caused the PMF to apportion Mg unevenly between the two sources. The construction site factor also contained substantial amounts of Ca and Zn. These elements might be explained by the extensive use of lubricating oil for drilling machines in the tunnel as well as conveyor belts and diesel locomotives outside. Ratios of Ca/Al, K/Al, and Fe/Al for the mineral dust factor (0.64, 1.03, and 2.02, respectively) and for the construction site factor (1.80, 0.0, and 0.85, respectively) were different compared with the geogenic ratios (1.15, 0.61, and 1.13,

respectively) in northern Switzerland (Gianini et al. 2012b). However, the ratios of the two source profiles combined were again similar to the geogenic ratios with Ca/Al=1.14, K/Al=0.59, and Fe/Al=1.52. PMF was not able to clearly split these two sources, which was also reflected in the persistently observed correlations in the *G space* plots between these two factors. If additional information were available on the composition of the material from either source, then a clearer separation such as that obtained by Amato et al. (2009) might have been possible. Both mineral factors showed similar weekday patterns with peaks on Tuesdays and Thursdays (Online Resource Fig. S4e). These peaks might be driven by some activity patterns on the construction site, because all high mineral factor concentrations were measured at site 5, which was closest to the construction site. Due to the similarities of the two factors, we combined them to one total mineral dust factor for the description of contributions in “Source contributions.”

Railway

The railway profile showed typical abrasion elements such as Fe, Cu, manganese (Mn), chromium (Cr), and nickel (Ni) (Bukowiecki et al. 2007) (Fig. 4a). However, Cu, Mn, and

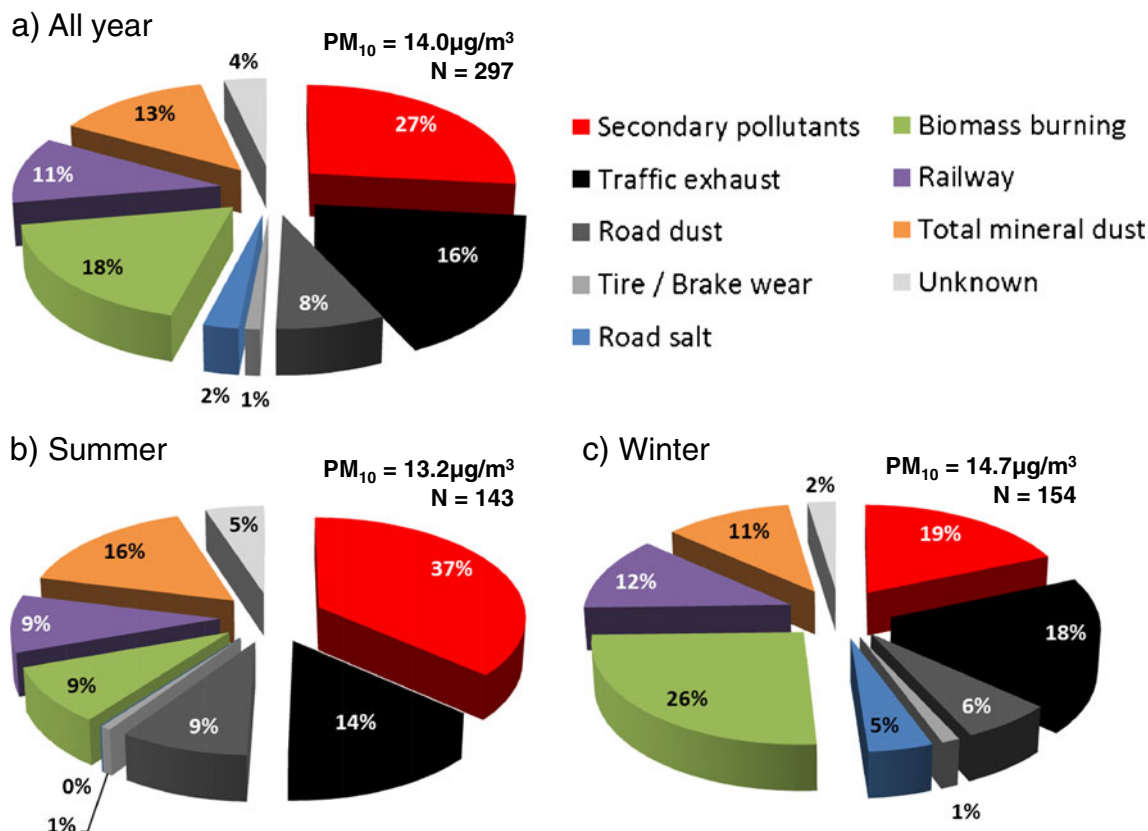


Fig. 7 Average source contributions for the year 2008: **a** the whole year, **b** in summer (Apr–Sep), and **c** in winter (Oct–Mar). Average daily PM₁₀ concentrations with number of measurements for the respective measurement periods are shown adjacent to the pie charts

Cr were emitted in very low quantities compared with Fe (Cu/Fe=0.01, Mn/Fe=0.01, Cr/Fe<0.005) comparable to a study in Zürich (Gehrig et al. 2007). In Switzerland, trains nearly exclusively operate with electric locomotives. However, due to the absence of power lines, diesel locomotives were used for the construction site. This presence of diesel engines might explain the additional contributions of Ba, Zn, EC, and OC in the source profile (Fig. 4a). Contributions of this factor were always highest at the site 5, which was closest to the railway tracks (Fig. 8).

Source contributions

In Fig. 7, source contributions are summarized by season for all sites in Erstfeld. To prevent over-representation of the winter months, only data from the year 2008 were used. To compare the contributions among the sites and seasons (Fig. 8), the sites closest to the highway (sites 1-3, Fig. 1) were excluded because of limited and unbalanced data in 2008 for these sites (i.e., only summer data for site 1 (N=9), only winter data for site 2 (N=32) and site 3 (N=9)). Overall, secondary pollution was the main contributor to PM₁₀ in summer, while traffic and biomass burning were

more important in winter (Fig. 7). In summer, greater photochemical activity boosts the formation of secondary pollutants, while stable air masses and frequent inversions trap the pollutants in the valley in winter, increasing the importance of local sources such as traffic and biomass burning. The importance of secondary pollution in Erstfeld was also reflected in the ratio of 2-nitrofluoranthene and 1-nitropyrene (2-NF/1-NP), which was >5 at all sites except site 1, indicating a domination of nitro-PAHs by atmospheric reactions rather than primary emissions (Miller-Schulze et al. 2010).

Although total PM₁₀ concentrations were spatially homogeneously distributed (Ducret-Stich et al. 2013), large differences were observed for local traffic, railway, and mineral contributions to PM₁₀ (Fig. 8). In both seasons, relative traffic source contributions were higher close to the highway (Figs. 6 and 8). At site 5, which was only 22 m away from the railway tracks and the closest measuring station to the construction site, we observed the highest contribution from the railway and mineral sources (Fig. 8).

The average PM₁₀ concentration of 14.0 µg/m³ in this study was lower compared with other sites from the Swiss national air pollution monitoring network (NABEL) (Gianini

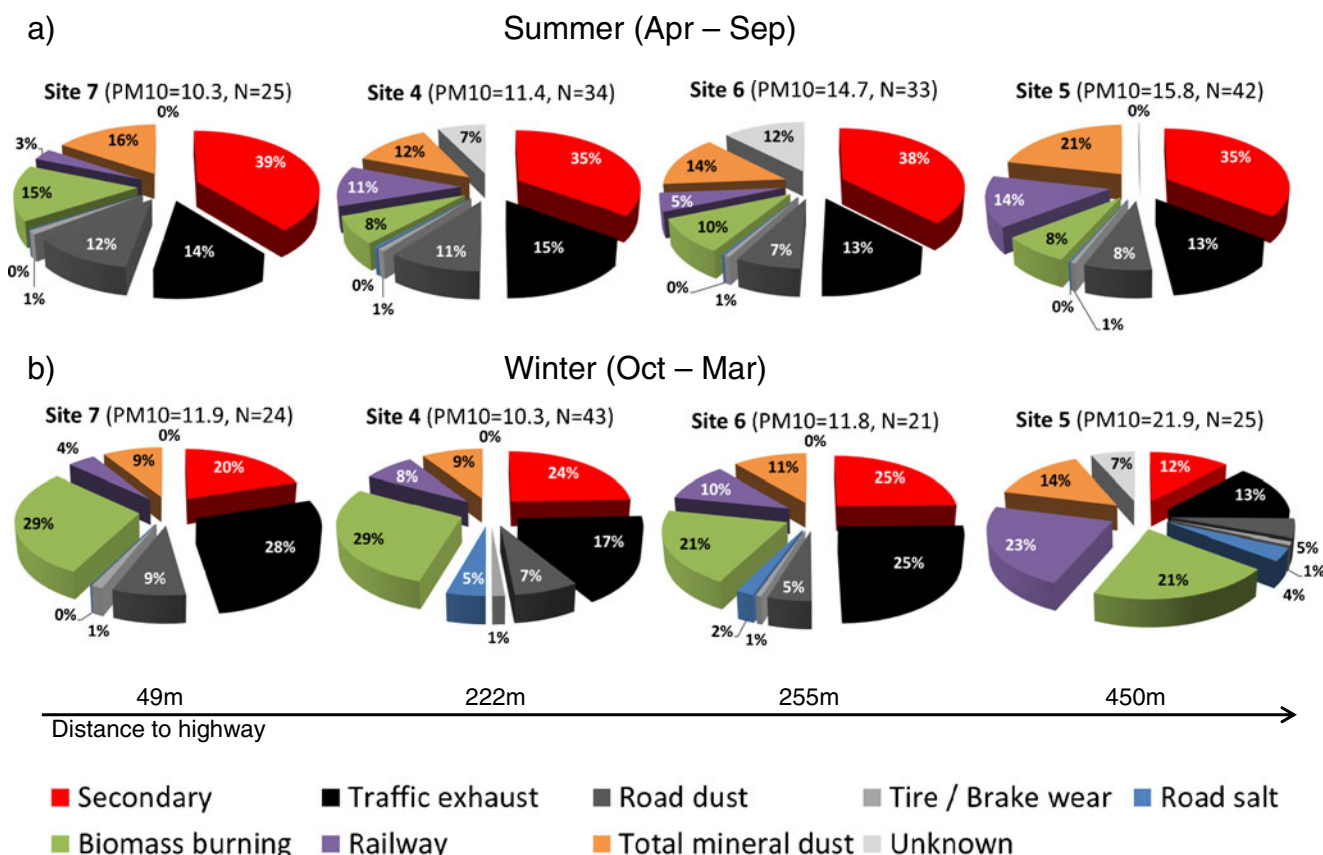


Fig. 8 Estimated source contributions by season for the four sites in the residential area for the year 2008. Site 7 is protected by a tunnel and a noise barrier from the direct influence of the highway; site 5 is closest

to the railway and the construction site. Average daily PM₁₀ concentrations [micrograms per cubic meter] together with number of measurements at various sites are presented in brackets

et al. 2012b) and comparable to other European rural background sites (Putaud et al. 2010). However, the relative contribution to PM₁₀ of total traffic (i.e., exhaust, road dust, tire and brake wear) was about 25 %, which was higher than in Zürich (18 %, urban background), almost as high as in Bern (30 %; urban roadside), and comparable to the rural site Magadino (24 %) south of the Alps, which is also influenced by the same highway as in our study (Gianini et al. 2012a). These results emphasize the large influence of highway traffic on the composition of particulate matter.

Secondary pollution contributions (27 %) were again comparable to Magadino (29 %) and lower than at all other NABEL sites (37–52 %) (Gianini et al. 2012a). In the European context, secondary aerosol contributions were comparable to urban sites in Central Europe (Putaud et al. 2010). This lower contribution might be a result of less photochemical activity due to the decreased sunlight in the narrow mountain valley of Erstfeld. Also, PMF might underestimate the contribution of this factor due to the lack of measurements of sulfate, nitrate, and ammonium in this study.

Another important source was biomass burning (primarily wood burning in winter). With a relative contribution of 18 %, biomass PM₁₀ was higher in our study than at NABEL sites (11–14 %), except for Magadino, which had a very high contribution of 31 % (Gianini et al. 2012a). In other studies, close to Magadino, it has been shown that wood burning is a major source in this region (Sandradewi et al. 2008; Szidat et al. 2007). The combination of the mountain–valley terrain, strong inversion conditions in winter, and extensive use of wood as a home heating fuel led to these high contributions to PM₁₀.

In Erstfeld, the construction site was probably the reason for the higher total mineral dust (i.e., mineral dust and construction site) contribution of 13 % compared with other rural sites in Central Europe (9 %) (Putaud et al. 2010) and the NABEL sites in Switzerland (8–13 %) (Gianini et al. 2012a).

As the result of having an extensive data set of daily measurements from seven different monitoring sites within the valley over one and a half years, we were able to separate different sources spatially and seasonally even though our study region was rather small. Also, additionally measured biomarkers (i.e., levoglucosan, NPAHs) allowed us to independently compare and validate some of the source contributions. However, as measurements were not concurrently collected at the different sites and no representative background measurements were available, it was not possible to compare daily contributions between the sites. The aim to distinguish between diesel and gasoline traffic sources from the highway could also not be achieved. Because of the method change in the EC and OC analysis, comparable fractionated data were only available for part of the data set. Using only the subsets, PMF was not able to give stable results.

Conclusions

In general, traffic, secondary particles from precursors of various sources—including traffic—and biomass burning were the major contributors to PM₁₀ in the alpine village of Erstfeld in 2008 accounting for 27 %, 27 %, and 18 % of the total PM₁₀ concentrations, respectively. PM₁₀ exposure attributed to traffic was more comparable to urban sites than those observed in rural sites in Switzerland. Also, a clear spatial dependency from the nearby highway was observed, showing increasing contributions of traffic exhaust and non-exhaust PM₁₀ with decreasing distance to the highway. While secondary particle contributions were lower, the biomass burning contributions were higher in Erstfeld compared with other northern Swiss villages, due to the combination of topography, winter inversions, and the importance of wood as an energy source in the Alps. However, contributions were much lower than in Magadino in the southern Alps, where biomass burning is known to be the major source of PM. The local influences of railway traffic and a major construction site were also observed. These receptor modeling results are being used to investigate the source-specific impact of air pollution exposures on short-term respiratory health outcomes in children in the village.

Acknowledgments This work was funded by the Federal Office for the Environment FOEN (No. 050303 / G161-1955) of the Swiss Government. Its contents are solely the responsibility of the authors and do not necessarily represent the official views of the FOEN. The authors declare they have no competing financial interests. We would like to thank Peter Boehler and Raffael Kaenzig from inNET Monitoring AG (Altdorf, Switzerland) for conducting the extensive air pollution measurements. We are also grateful to Christoph Hüglin and Matthias Gianini at Empa (Dübendorf, Switzerland) and to Chang-Fu Wu at National Taiwan University for their helpful inputs to the PMF analysis.

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