



Dominant impact of residential wood burning on particulate matter in Alpine valleys during winter

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[1] Within the project AEROWOOD (Aerosols from wood burning versus other sources), particulate matter was collected at two Swiss Alpine valleys during winter. Apportionment of aerosols from transit traffic and residential wood burning was performed using radiocarbon (¹⁴C) determinations of the organic carbon (OC) and elemental carbon (EC) fractions. On daily average, 88% and 65% of the total carbonaceous matter (including all other atoms than carbon) originated from non-fossil sources inside and outside of the village, respectively, which is mainly attributed to wood burning. These proportions were generally higher during the evening compared to morning hours, when traffic intensity waned and heating activities increased. ¹⁴C measurements of EC indicated episodes of both nearly pure traffic and wood-burning sources, respectively, of which the latter extreme has not been observed for EC under ambient conditions before. Results show that regulatory measures for the improvement of air quality must consider residential wood burning as an important source even in industrialized countries.

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1. Introduction

[2] Carbonaceous aerosols as a major component of the total fine particulate matter (PM) burden comprise a huge variety of compounds [Penner, 1995] and may cause asthmatic attacks, cardio-pulmonary diseases, and lung cancer incidents [Jerrett *et al.*, 2005]. As most of these compounds cannot be isolated and, therefore, are still unknown, sub-fractions of total carbon (TC) with similar chemical or physical behavior are defined, namely organic carbon (OC) and elemental carbon (EC). The latter fraction is also designated as black carbon (BC) and consists of refractory and optically absorptive combustion polymers [Bond *et al.*, 2004]. As EC from both diesel and wood smoke is carcinogenic [Knaapen *et al.*, 2004], it is of special interest for regulatory air quality measures.

[3] Anthropogenic activities like fossil-fuel usage and biomass burning as well as natural processes like plant abrasion and secondary particle formation by atmospheric oxidation of biogenic precursors are identified as the major sources of the carbonaceous PM [Penner, 1995; Turpin and Lim, 2001; Bond *et al.*, 2004]. The importance of these different sources, however, still can not be estimated accurately. Emission inventory modeling, on the one hand, is based on the estimation of all possible contributions for a certain aerosol component. This allows the distinction of many different sources, but their sum often substantially underestimates ambient concentrations of this component, e.g. for EC [Bond *et al.*, 2004]. The analysis of many elemental and organic tracer compounds from ambient PM, on the other hand, enables a mass balance of the sources (e.g. levoglucosan traces wood burning emissions) assuming a constant composition fingerprint for every emission type, which remains unchanged during atmospheric transport and processing [Schauer *et al.*, 1996; Zheng *et al.*, 2006]. Whereas this method may be valid for special emission conditions and close to the sources, when the effect of processing can be neglected, it bears considerable uncertainties for ambient environmental sampling. In contrast to these methods, radiocarbon (¹⁴C, T_{1/2} = 5730 a) measurements of the carbonaceous aerosol offer the opportunity of a direct distinction of fossil and non-fossil sources. Due to its age, ¹⁴C has completely disintegrated in fossil substances, whereas modern plant material is on the contemporary radiocarbon level [Currie *et al.*, 1994; Szidat *et al.*, 2004b, 2006]. This intrinsic isotopic information characterizes the sources of ambient PM independent of its history regarding emission conditions or atmospheric transport.

[4] Only little attention was paid to the anthropogenic contribution of biomass burning in industrialized countries [Schauer *et al.*, 1996; Zdráhal *et al.*, 2002]. Recent studies have shown that especially residential wood burning for heating purposes may be important during winter, because the efficiency of aerosol production can be very high depending on the burning conditions [Szidat *et al.*, 2006; Ward *et al.*, 2006; Aymoz *et al.*, 2007]. Although wood burning contributes only marginally to the total energy production in Switzerland compared to fossil-fuel combustion, it became relevant for Zurich air quality during an exceptional inversion episode in February 2003 [Szidat *et al.*, 2006]. That study showed that wood burning accounted for ~25% of EC and ~41% of OC during this episode, which was 4 times more compared to summer 2002. In this work, we measured ¹⁴C in OC and EC at two sites in southern Switzerland for apportionment of the carbonaceous PM sources within the project AEROWOOD (Aero-

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Table 1. Sampling Details

	Campaign I	Campaign II	Campaign III
Time	January 2005	February 2005	March 2005
Site	Roveredo	Moleno	Roveredo
Position	46°14'18"N, 9°07'45"E, 298 m a.s.l.	46°16'46"N, 8°59'49"E, 254 m a.s.l.	See Campaign I
Location	Inside the village, directly next to motorway and railway	800 m outside the village, at motorway parking lot	See Campaign I
Valley	Mesolcina Valley	Reviera Valley	See Campaign I
Valley direction	Northeast - Southwest	North - South	See Campaign I
Transit route	A13: San Bernardino Route	A2: Gotthard Route	See Campaign I
Traffic density ^a	7900	19,700 ^b	9700
Sampling inlet	PM10	PM10	PM1
Daytime of sampling	Morning: 6 am – 2 pm Evening: 6 pm – 2 am	See Campaign I	See Campaign I

^aMean traffic density during the month of sampling given as vehicles per day [Swiss Federal Roads Authority, 2006].

^bDetermination performed on motorway A2 at Biasca, 10 km north of the sampling site.

sols from wood burning versus other sources). Both sites are situated in Alpine valleys which are typical for this area with often stable inversion weather conditions during winter. Local emissions derive from wood burning, which is a traditionally popular method for residential heating in this region, and from the transit highways through these valleys as well as some local traffic.

2. Methods

2.1. Aerosol Sampling

[5] During three campaigns in January–March 2005, aerosols were collected on pre-heated quartz fiber filters (QF20, Schleicher & Schuell) with high-volume samplers (DA80, Digitel) at two Alpine valley sites in southern Switzerland. Table 1 presents details of these campaigns. Morning (6 am–2 pm) and evening (6 pm–2 am) sampling periods were separated by two simultaneously running samplers in order to probe episodes of higher transit-traffic and wood-burning contributions, respectively. Each filter was used during the same period of day (morning or evening) on two consecutive days, resulting in an air throughput of 480 m³ during 16 hours for each sample. After sampling, filters were wrapped in aluminum foil, packed into air-tight plastic bags, and stored at –20°C until analysis. PM10 concentrations were determined with a betameter, which was regularly calibrated with weighted filters from a high-volume sampler.

[6] Both stations represent typical Alpine valley sites for central and southern Switzerland. The valley floors have an extension of <2 km and the surrounding mountains tower above ground by >1.5 km. Human activities in the villages, on roads and cultivated fields are concentrated at the valley floor. Campaigns I and III were conducted in the center of the village of Roveredo in the Mesolcina Valley (Table 1). Due to the topography of this valley, the sun does not reach the village during December and January. This fact gives rise to strong inversion conditions with boundary layer heights of several tens to several hundreds of meters and midday temperatures below freezing. Campaign II was carried out at a parking lot near the motorway A2, 800 m northeast of the small village of Moleno (Table 1). Traffic emissions are more important at this site compared to Roveredo due to the higher traffic density and the larger distance to the next populated area. In contrast to Roveredo, sunlight reaches the valley floor at the Moleno site in

February at midday enabling maximum temperatures of ~10°C and reducing inversion conditions. At Roveredo, the sampling site was separated from the road by 2-m-high wall; the effect of this wall on fine and coarse aerosol particles, however, was not assessed. During sampling, typical daily average temperature ranged from –2 to +4°C, daily average relative humidity from 55 to 75%. The daily average wind speed was <1 m s^{–1}.

2.2. Source Apportionment of Carbonaceous Aerosols With ¹⁴C

[7] Fossil and non-fossil sources of OC and EC were distinguished using ¹⁴C determinations as described in detail elsewhere [Szidat *et al.*, 2006]. In brief, OC was separated from the filters by oxidation in pure oxygen at 340°C, whereas EC was isolated by removal of OC with water extraction and oxidation in air at 390°C for a period of 4 h, followed by carbon dioxide formation from the remaining filter material at 650°C in O₂ [Szidat *et al.*, 2004c]. In contrast to earlier studies, we increased the temperature of OC elimination for EC isolation from 375°C to 390°C, as we observed OC to remain after the thermal treatment on selected filters at the lower temperature. This interference was overcome at the higher temperature. CO₂ from OC and EC was cryogenically trapped and transformed into graphite targets for ¹⁴C determination with accelerator mass spectrometry [Szidat *et al.*, 2004a; Jenk *et al.*, 2007].

[8] Radiocarbon measurement results are expressed as fractions of modern carbon (f_M), which is the ¹⁴C/¹²C isotopic ratio of the sample related to that of the reference year 1950. Fossil material is characterized by $f_M = 0$ due to its age and the extinction of ¹⁴C. Contemporary sources show slightly elevated f_M levels compared to the theoretical value of 1 as a consequence of the nuclear bomb excess: $f_M = 1.24 \pm 0.05$ was set for carbonaceous aerosols from residential wood burning using 30–50-year-old plants and $f_M = 1.072 \pm 0.015$ for present biogenic OC [Szidat *et al.*, 2006]. In contrast to the procedure of Szidat *et al.* [2006], it was not possible to separate biogenic OC from wood-burning OC due to the overwhelming contribution of aerosols from the latter. Consequently, we report on fossil and non-fossil OC in this work with $f_M = 1.16 \pm 0.05$ for non-fossil OC, assuming a rectangular-distributed probability of both non-fossil sources. For EC, on the other hand, we distinguish between fossil and wood-burning EC (with $f_M =$

Table 2. Concentration and Isotopic Measurements^a

Campaign	Daytime	Date of 2005	PM10, $\mu\text{g m}^{-3}$	TC, $\mu\text{g m}^{-3}$	OC, $\mu\text{g m}^{-3}$	EC, $\mu\text{g m}^{-3}$	EC/TC	$f_M(\text{OC})$	$f_M(\text{EC})$
I	Morning	13/14 Jan	20.4	14.3	11.3	3.0	0.21	1.005 ± 0.017	0.474 ± 0.037
I	Evening	13/14 Jan	32.3	18.4	15.1	3.3	0.18	1.068 ± 0.012	0.511 ± 0.023
I	Morning	19/20 Jan	20.7	9.9	7.4	2.5	0.25	0.940 ± 0.027	0.530 ± 0.024
I	Evening	19/20 Jan	41.9	23.1	18.8	4.4	0.19	1.098 ± 0.016	0.887 ± 0.022
II	Morning	04/05 Feb	46.9	17.1	10.6	6.4	0.38	0.707 ± 0.016	0.060 ± 0.020
II	Evening	04/05 Feb	66.6	20.7	14.6	6.1	0.29	0.861 ± 0.012	0.223 ± 0.009
II	Morning	08/09 Feb	76.9	22.6	16.2	6.3	0.28	0.759 ± 0.009	0.211 ± 0.010
II	Evening	08/09 Feb	101.9	33.7	24.2	9.5	0.28	0.847 ± 0.009	0.171 ± 0.010
III	Evening	02/03 Mar	48.5	18.2	15.0	3.3	0.18	1.002 ± 0.014	0.654 ± 0.017
III	Evening	08/09 Mar	16.8	9.9	8.5	1.4	0.14	1.051 ± 0.024	0.909 ± 0.045
III	Morning	09/10 Mar	6.8 ^b	5.8 ^b	4.9 ^b	1.0 ^b	0.17 ^b	0.358 ± 0.019^b	0.239 ± 0.034^b
III	Morning	11/12 Mar	24.4	7.0	5.6	1.4	0.20	0.697 ± 0.026	0.324 ± 0.025

^a $^{14}\text{C}/^{12}\text{C}$ determinations of OC and EC are given as fractions of modern (f_M). All values are corrected for filter blanks, which typically amounted to 5%. Typical uncertainties are 5% for TC, 10% for OC, and 20% for EC as well as EC/TC. For details of sampling conditions see Table 1. PM was collected during 8 hours each on two consecutive days (see Table 1). Note that the given dates mark beginning of sampling and that evening sampling ended at 2 am on the following day.

^bValues are biased by a short event (8 min) dominated by local fossil emissions from road work (see chapter 3).

1.24 ± 0.05 for the latter fraction according to Szidat *et al.* [2006]), as biogenic sources can be neglected for EC.

[9] Concentrations of TC, OC, and EC as well as EC/TC ratios were determined offline using a stepwise thermal desorption/combustion OC/EC analyzer (Sunset Laboratory Inc.) with thermal optical transmission (TOT), which enables the correction of charring artifacts. Different OC fractions are released at multiple temperature stages less than 840°C under helium atmosphere. EC is combusted and converted into CO_2 at two temperature stages between $550\text{--}850^\circ\text{C}$ in helium with 10% oxygen. According to Turpin and Lim [2001], the following factors were used for conversion of OC and EC concentrations to the total mass of these fractions, i.e. organic mass (OM) and elemental carbon mass (ECM): 1.4 for fossil OC, 2.25 for non-fossil OC (average of non-urban and wood-smoke aerosols), and 1.1 for all EC sources. These conversion factors bear large uncertainties depending on conditions of emission and atmospheric transformation. Total carbonaceous matter (TCM) is the sum of OM and ECM.

3. Results

[10] Due to the strong, winter-time inversion conditions with boundary layer heights of several tens to several hundreds of meters at Roveredo, local aerosol emissions accumulate within a small air volume leading to elevated concentrations of PM10 and carbonaceous particles for Campaigns I and III (Table 2). Average PM10 during measurement periods amounted to $29 \mu\text{g m}^{-3}$ and $24 \mu\text{g m}^{-3}$ for Campaigns I and III, respectively, and average TC concentrations were $16 \mu\text{g m}^{-3}$ and $10 \mu\text{g m}^{-3}$, respectively. For the second site, Moleno, inversion layers are less stable. PM10 and TC levels, however, were higher for Campaign II with $73 \mu\text{g m}^{-3}$ and $23 \mu\text{g m}^{-3}$, respectively. In winter, two major sources are responsible for these high concentrations: traffic emissions from the traversing motorways and wood burning from traditional residential heating. Both processes follow diurnal trends. Whereas transit traffic occurs mainly during daytime, heating of private houses is intensified during the evening hours. We observed generally higher PM loads in the evening: for Campaigns I and II, where morning and evening sampling occurred on the same day,

PM10, TC, OC, and EC concentrations were higher by 47%, 50%, 59%, and 28%, respectively, during evening time. The average EC/TC ratio for Campaign II was 0.31, substantially exceeding the 0.21 ratio for Campaign I with 20% and 12% lower values during the evening for Campaigns I and II, respectively.

[11] Measurements of ^{14}C in OC and EC are given as f_M values in Table 2. $f_M(\text{EC})$ (i.e., f_M of the EC fraction) ranged from 0.06 (nearly fossil) to 0.91 (almost contemporary). The latter level exceeds all our previous measurements of ambient $f_M(\text{EC})$ by far, e.g. by a factor of ~ 3 compared to a winter episode influenced by wood burning in Zurich [Szidat *et al.*, 2006]. Moreover, $f_M(\text{EC})$ values are generally lower than corresponding $f_M(\text{OC})$ determinations. The comparison of Campaigns I and II shows >3 times higher $f_M(\text{EC})$ and 30% higher $f_M(\text{OC})$ for Roveredo, indicating larger relative influence from wood burning at this site. The $f_M(\text{OC})$ values for Campaign II reveal that even at the motorway station of Moleno substantial wood burning contributions were observed. For Campaigns I and II, $f_M(\text{EC})$ and $f_M(\text{OC})$ values were $\sim 40\%$ and $\sim 14\%$ higher during the evening compared to morning hours, respectively, independent of the different conditions at both sites.

[12] Concentrations of fossil and non-fossil OM as well as fossil and wood-burning ECM are shown in Table 3 and average contributions to TCM are summarized in Figure 1. By conversion of OC and EC to OM and ECM, the total mass of the organic aerosol including all other atoms than carbon, e.g. oxygen, hydrogen, and nitrogen, are determined. This allows a direct calculation of the burden of anthropogenic organic pollutants to the PM mass. In general, $\text{OM}_{\text{nonfossil}}$ was the largest fraction of TCM with a maximum contribution of 87%. As for $f_M(\text{OC})$, $\text{OM}_{\text{nonfossil}}$ contributions were higher during evening hours. On the other hand, $\text{OM}_{\text{fossil}}$ showed higher percentages during the morning, whereas during evening hours, values decreased and even fell below the detection limit for two samples at Roveredo. The non-fossil fraction of TCM (comprising $\text{OM}_{\text{nonfossil}}$ and ECM_{wood}) was 91–94% and 81–85% for Campaign I during evening and morning times, respectively. For Campaign II, these values amounted to $\sim 70\%$ and 55–65%, respectively. It is remarkable that ECM_{wood} was

Table 3. Source Apportionment^a

Campaign	Daytime	Date of 2005	OM _{nf} , $\mu\text{g m}^{-3}$	OM _f , $\mu\text{g m}^{-3}$	ECM _{wood} , $\mu\text{g m}^{-3}$	ECM _f , $\mu\text{g m}^{-3}$	TCM, $\mu\text{g m}^{-3}$	TCM _{nf} /TCM	TCM/PM10
I	Morning	13/14 Jan	22.2	2.1	1.2	2.0	27.5	0.85	1.35
I	Evening	13/14 Jan	31.5	<3.2 ^b	1.5	2.1	36.7	0.90	1.14
I	Morning	19/20 Jan	13.5	1.9	1.2	1.6	18.2	0.81	0.88
I	Evening	19/20 Jan	40.1	<4.2 ^b	3.4	1.4	46.2	0.94	1.10
II	Morning	04/05 Feb	14.7	5.8	0.3	6.7	27.5	0.55	0.59
II	Evening	04/05 Feb	24.5	5.2	1.2	5.5	36.4	0.71	0.55
II	Morning	08/09 Feb	24.0	7.8	1.2	5.8	38.7	0.65	0.50
II	Evening	08/09 Feb	39.9	9.0	1.4	9.0	59.4	0.70	0.58
III	Evening	02/03 Mar	29.2	2.8	1.9	1.7	35.6	0.87	0.73
III	Evening	08/09 Mar	17.4	1.1	1.1	0.4	19.9	0.93	1.19
III	Morning	09/10 Mar	3.4	4.7 (2.0)	0.2	0.9 (0.4)	9.2 (6.0)	0.39 (0.60)	1.34
III	Morning	11/12 Mar	7.7	3.1	0.4	1.1	12.3	0.65	0.51

^aOM is separated into non-fossil (nf) and fossil (f), ECM into wood-burning (wood) and fossil sources. TCM represents the sum of all sub-fractions with $\text{TCM}_{\text{nf}} = \text{OM}_{\text{nf}} + \text{ECM}_{\text{wood}}$. The numbers in the brackets are corrected for an 8 min short event dominated by local fossil emissions from road work (see chapter 3). Final uncertainties are typically 20-30% due to large uncertainties of measurements and model parameters. For details of sampling conditions see Tables 1 and 2.

^bValue below detection limit caused by the small difference between $f_{\text{M}}(\text{OC})$ and the level of pure non-fossil OC (i.e. 1.16 ± 0.05).

elevated by 92% for the evening compared to the morning for Campaigns I and II, whereas $\text{ECM}_{\text{fossil}}$ concentrations rose only marginally (12%) during the day for both sites. Furthermore, spatial variabilities between both sites of the two ECM sources differ from each other considerably: average ECM_{wood} concentrations, on the one hand, are higher for Campaign I by a factor of 1.8 compared to Campaign II; average $\text{ECM}_{\text{fossil}}$ concentrations for Campaign II, on the other hand, exceeded those of Campaign I by a factor of 3.8.

[13] Results of Campaign III have to be evaluated carefully, as PM collection was conducted with a different size cut-off than Campaigns I and II (see Table 1), sampling of morning and evening filters was not possible on the same days, and one sample (09/10 March, morning) was biased by local emissions. For this sample, excess fossil OM was

produced during construction work at the sampling site, which lasted for 8 minutes. Aerosol mass spectrometry measurements, which were performed simultaneously to PM sampling (M. R. Alfarra, A. S. H. Prévôt, S. Szidat, J. Sandradewi, S. Weimar, D. Schreiber, M. Mohr, and U. Baltensperger, Identification of the mass spectral signature of organic aerosols from wood burning emissions, submitted to *Environmental Science and Technology*, 2006, hereinafter referred to as Alfarra et al., submitted manuscript, 2006), showed that this episode comprised 33% of the total loading of this filter, for which it was corrected (see Table 3). Taking these special conditions of Campaign III into account, we observed a similar source pattern for evening samples compared to Campaign I with $\text{TCM}_{\text{nf}}/\text{TCM}$ ratios of ~ 0.9 . For morning samples, this ratio was smaller for March (~ 0.63) than for January (~ 0.83) as a

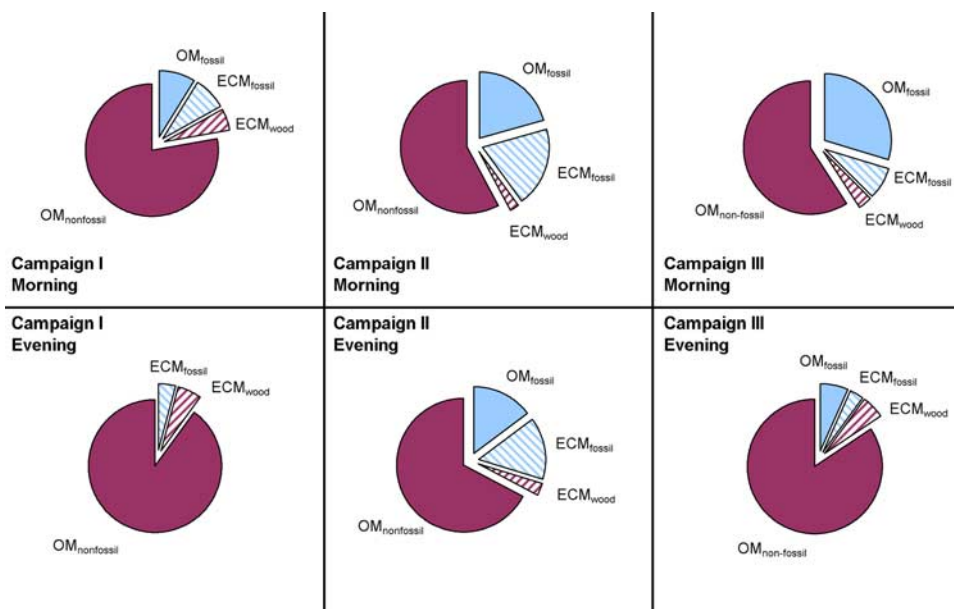


Figure 1. Average contributions of ¹⁴C-deduced sources to total carbonaceous matter (TCM) differentiated into fossil and non-fossil organic matter (OM) and fossil and wood-burning elemental carbon matter (ECM). The fraction of OM_{fossil} was insignificant for Campaign I, evening. For Campaign III, morning, corrected values are considered for one sample as given in Table 3. For details of sampling conditions see Table 1.

consequence of the influence of higher midday temperatures at Roveredo at the end of winter.

[14] TCM/PM10 ratios at Roveredo were about twice as high as at Moleno. The average value for Roveredo is 1.12, which indicates a discrepancy between both methods of mass determination. As PM10 was determined on filters other than those used for the measurements of the carbonaceous particles, different sampling conditions may have caused this discrepancy. Furthermore, the usage of the conversion factors for scaling up OC to OM are highly uncertain and may have led to an overestimation.

4. Discussion

[15] We observed an overwhelming impact of carbonaceous aerosols from residential wood burning on particulate matter in Alpine valleys during winter-time, which substantially surpassed traffic-related emissions even close to busy motorways. Especially at Roveredo, wood-burning emissions were dominant in spite of considerable transit traffic and aerosol sampling very close to the motorway. During evenings, when new wood logs are fed into the stoves, TCM_{nr}/TCM ratios rose up to 0.94, indicating a strong diurnal cycle of wood-burning PM. On the other hand, the spatial variability between both sites of ECM from this source is less pronounced (1.8 times elevated for Campaign I) compared to that of ECM from traffic-related sources (3.8 times elevated for Campaign II). This demonstrates that air pollution due to wood burning is of similar importance at both sites and, thus, probably in many Alpine valleys, whereas traffic becomes substantial only at locations with very high traffic fluxes. The impact from traffic at Moleno is indicated by lower TCM_{nr}/TCM ratios (0.55–0.71) and corroborated by increased EC/TC ratios. The latter ratios are generally higher for fossil than for wood-burning sources, e.g. by a factor of ~ 3 as determined from Szidat et al. [2006]. This gives rise to the fact that OM is less sensitive to the influence from traffic compared to ECM and, thus, was dominated by non-fossil emissions even at Moleno. It must be noted that these results are mass-related and do not represent ultra-fine particles, which often exhibit larger numbers for traffic. Although the ultra-fine PM fraction has a small mass, it may have a considerable impact on human health.

[16] These conclusions are valid with the assumption that anthropogenic wood burning is the exclusive non-fossil source in this study. Two processes are potentially concurring but, in fact, can be neglected. a) Natural wildfires may be interpreted as an anthropogenic combustion. These fires are not important for industrialized mid-latitude areas [Penner, 1995], especially during winter [van der Werf et al., 2006], and were not noticed in the surroundings during PM sampling. b) Certain amounts of biogenic secondary organic aerosols (SOA) may be reported as OC contributions from anthropogenic wood burning, as both emissions stem from modern plant material and, therefore, are on the contemporary ^{14}C level. This interference can be neglected as well, because aerosol mass spectra at Roveredo compared very well to the spectra measured during laboratory experiments of inefficient burning of wood in a small stove (Alfarra et al., submitted manuscript, 2006) and differed to those of organic aerosols dominated by biogenic SOA [Alfarra et al.,

2006]. Nevertheless, biogenic SOA cannot be excluded completely so that we defined OC on the contemporary ^{14}C level as “non-fossil”, whereas the respective fraction of EC unambiguously originates from wood-burning activities.

[17] Both stations represent typical Alpine valley sites for central and southern Switzerland. It is a main outcome of this investigation that regulatory air quality measures are necessary for both PM sources, traffic and wood burning. However, only little attention was paid to the latter process for a long time, because the contribution of wood burning to the total energy consumption in industrialized European countries is only marginal. The methodological concept of the AEROWOOD project combining ^{14}C determinations separately for OC and EC with aerosol mass spectrometry has made it possible to quantify the contribution of particles from wood burning emissions to ambient PM in this study.

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