

## ACCELERATOR MASS SPECTROMETRY ANALYSIS OF NON-SOLUBLE CARBON IN AEROSOL PARTICLES FROM HIGH ALPINE SNOW (MT. SONNBLICH, AUSTRIA)

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**ABSTRACT.** With an elaborate accelerator mass spectrometry (AMS) technique radiocarbon measurements have been performed with aerosol carbon filtered from high alpine snow samples gathered consecutively at the high-altitude research station Sonnblick (3106 m, Eastern Alps, Austria) during a snow storm in April 1997. The concentration of the water-insoluble carbonaceous material in the molten snow was on the average 310  $\mu\text{g C/L}$  and the total sample amounts for analysis were in the range of 35  $\mu\text{g}$  to 60  $\mu\text{g C}$ . Using a special background correction procedure tested on similar amounts of an urban particulate standard sample the accuracy of the corrected and normalized  $^{14}\text{C}/^{12}\text{C}$  isotopic ratios of the snow aerosol samples was in the order of 4% to 14% of the measured ratios. The water-insoluble carbonaceous material of five samples from Mt. Sonnblick exhibited a weighted mean of 74 pMC (percent Modern Carbon) with a range of 64 pMC to 88 pMC. Thus, it appears that about 64% of non-soluble carbon in high alpine snow from Sonnblick was of biogenic origin. The temporal variations of the  $^{14}\text{C}/^{12}\text{C}$  isotopic ratios of the snow aerosol samples were statistically significant, suggesting alterations in the contribution of specific aerosol sources.

### INTRODUCTION

Since the  $^{14}\text{C}/^{12}\text{C}$  ratio gives a unique and absolute measure of the contemporary/fossil source ratio transient atmospheric phenomena and specific source contributions can be explored (Currie et al. 1996, 1998). Especially aerosol samples of remote territories, e.g. the Arctic and the Antarctic region, and of secluded mountain ranges, e.g. the major parts of the Alps, are capable of giving new information on long-range transport phenomena. However, the very low concentration of carbonaceous aerosols makes these measurements a challenge, even for the highly sensitive method of accelerator mass spectrometry (AMS). For the extension to carbon masses less than 100  $\mu\text{g}$ , a special micro-technique for sample preparation (Verkouteren et al. 1997), combined with modifications in the operating conditions of the Vienna Environmental Research Accelerator (VERA) (Kutschera et al 1997; Priller et al. 1997; Weissenböck et al. 1998), and the data evaluation process were necessary. The reproducibility is mainly limited by counting statistics and variability of “blank” corrections. Here we use this special small sample AMS technique for investigations of the filterable carbonaceous material in snow from a high alpine site in Central Europe.

### Carbonaceous Aerosols in Air and Snow

The total carbon (TC) of atmospheric aerosols contain carbonate carbon (CC), elemental carbon (EC) and organic carbon (OC) (Saxena and Hildemann 1996). Recent data from EC/TC measurements of Mt. Sonnblick aerosol show a ratio of 0.085 to 0.19 (Hitzenberger et al. 1999). However, only 50% of the TC from aerosols in rural samples is water-insoluble (Saxena and Hildemann 1996). The filterable portion of the carbonaceous aerosol in snow includes the EC fraction and just a certain part of the OC fraction (Chylek and Srivastava 1987; Saxena and Hildemann 1996). While EC sources are known (biomass and fossil fuel combustion) (Cooke and Wilson 1996), the sources for insoluble OC are not well defined (Saxena and Hildemann 1996). Recently, biopolymers were iden-

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tified as potentially water-insoluble organic carbonaceous material. Kunit and Puxbaum reported about the occurrence of cellulose, tracer for plant debris, in atmospheric aerosols (Kunit and Puxbaum 1996). Fossil carbon is expected to have 0 pMC, whereas recent biogenic material has about 115 pMC (Levin et al. 1995).

### **SAMPLING SITE**

Mt. Sonnblick is situated in the main ridge of the Austrian Alps (Hohe Tauern, 12°57'E, 47°03'N) and is among the highest peaks in the area (3106 m). A meteorological observation station on the summit of Mt. Sonnblick (see Figure 1) was established in 1886 and is accessible year-round. It is surrounded by large glacier fields to the east and south, and has the ideal characteristics of a high alpine background station since there are in general few tourists and essentially no local sources of carbonaceous aerosols.

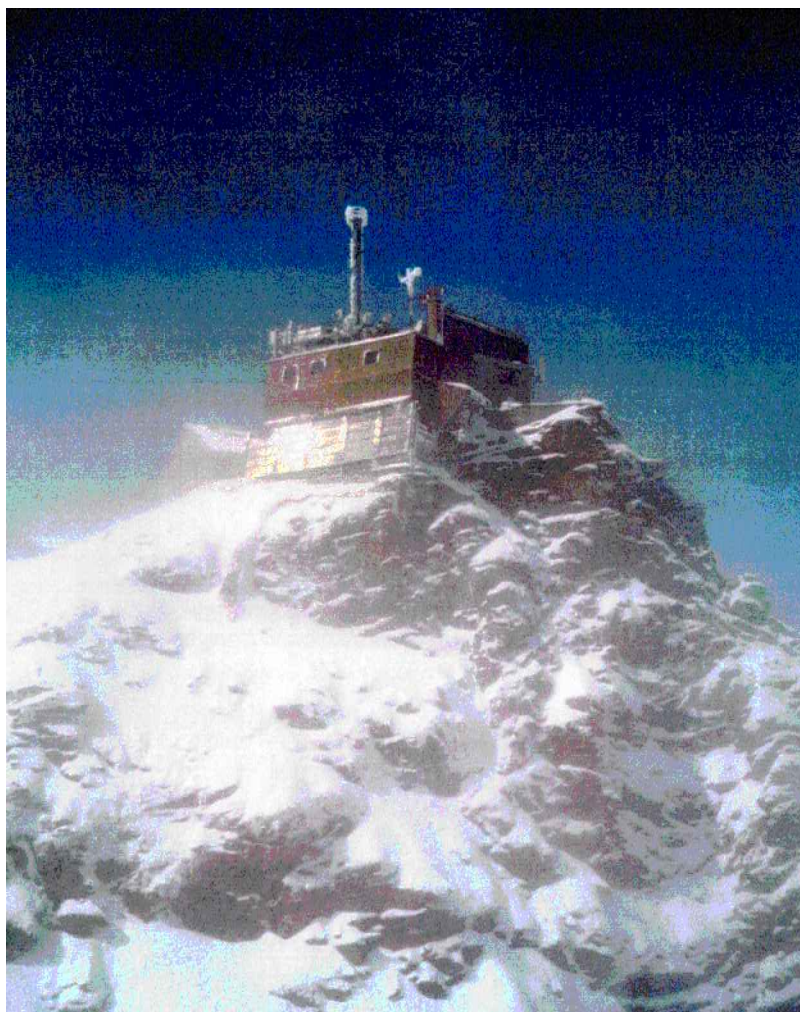


Figure 1 The high altitude research station Sonnblick (3106 m) in the Austrian Alps

## SNOW SAMPLING

During a strong snow storm in April 1997 fresh fallen snow layers of about 10 cm thickness were collected at a platform on the high altitude research station (Table 1). The snow samples were stored in pre-cooled glass bottles (1 L) and kept frozen until the time of the chemical treatment at the National Institute of Standards and Technology (NIST). For quality control purposes, two samples (S6, S9) were each divided into sub-samples at NIST.

The amount of water-insoluble carbon from the snow samples was about 310  $\mu\text{g/L}$ . Recently, Lavanchy et al. (1999) similarly gathered 380  $\mu\text{g/L}$  from snow from Colle Gnifetti (Monte Rosa, Switzerland, 4452 m).

Table 1 Data of the carbonaceous snow sample collected at the high altitude research station Sonnblick

| Sample ID <sup>a</sup> | Start date | Start time (UTC) | End date   | End time (UTC) | Volume of melted snow (mL) | Mass of insoluble carbon fraction ( $\mu\text{g}$ ) | Carbon mass per volume snow ( $\mu\text{g/L}$ ) |
|------------------------|------------|------------------|------------|----------------|----------------------------|---|---|
| S6a                    | 29-04-1997 | 09:00            | 29-04-1997 | 12:00          | 109                        | 38.1  | 350   |
| S6b                    |            |                  |            |                | 91                         | 37.3  | 410   |
| S7                     | 29-04-1997 | 12:00            | 29-04-1997 | 21:00          | 200                        | 58.6  | 293   |
| S8                     | 29-04-1997 | 21:00            | 30-04-1997 | 05:15          | 190                        | 37.7  | 198   |
| S9a                    | 30-04-1997 | 05:15            | 30-04-1997 | 18:00          | 109                        | 34.7  | 318   |
| S9b                    |            |                  |            |                | 173.5                      | 61.3  | 353   |
| S10                    | 30-04-1997 | 18:00            | 30-04-1997 | 21:10          | 178                        | 45.9  | 258   |

<sup>a</sup>Snow samples S6 and S9 were divided for quality purposes

## SAMPLE TREATMENT AND TARGET PREPARATION<sup>5</sup>

The snow samples were melted in a room-temperature bath and the resulting solutions were filtered through pre-fired Pallflex<sup>®</sup>, high purity quartz filters (Pall Gelman Laboratory<sup>TM</sup>, type: Tissuquartz 2500 QAT-UP). These had a typical aerosol retention of 99.999% for 0.5  $\mu\text{m}$  particles and 99.99% for 0.3–0.05  $\mu\text{m}$  particles respectively at standard test conditions. The retention is specified by the American Society of Testing and Materials (ASTM) designation number D-2986-71 for dioctyl phthalate smoke particles and an air flow of 32 L/min through a 100  $\text{cm}^2$  filter area. Subsequently, the filters were dried at 110  $^{\circ}\text{C}$  for 3 hr. Carbonates were then removed by exposing the filters to HCl vapors for 5–7 hr. After that, the filters were placed into a container with NaOH in order to attempt neutralizing the filter surface. Finally, the carbonaceous residue on the filter was combusted in a pre-heated quartz tube with addition of 200 mg CuO. Halides were removed with 40 mg silver wire. The individual sample  $\text{CO}_2$  was then transferred to the evacuated graphitization system (see Figure 2) and trapped with liquid nitrogen. After pumping off residual gases the  $\text{CO}_2$  mass was quantified by manometry at room-temperature. According to Verkouteren et al. (1997), the  $\text{CO}_2$  was then cryo-transferred to an evacuated quartz tube, containing 100 mg Mn-chips and 2 mg Fe-wool. The tube was then flame-sealed. The  $\text{CO}_2$  was first reduced to CO by the Mn-chips and then catalytically to

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graphite on the Fe-wool (24 hr at 600 °C). Finally, the sample Fe-C matrix was magnetically separated from the Mn-chips, the tube was filled with ultra high purity helium and the matrix was sintered at 1575 °C. The result was a solid bead of 0.5 mm to 0.9 mm diameter.

For quality purposes some “artificial” snow processing samples were generated by filtering distilled water through two carbon reference materials: SRM 1649a, the Washington, DC Urban Dust (aerosol) standard reference material (aerosol reference samples, Table 2), and SP-2, a commercial graphite material that is used as a “dead”  $^{14}\text{C}$  reference material by NIST (dead reference samples, Table 2). The filtering process, followed by closed-tube combustion of the carbon retained on the high purity quartz filter, is labeled “total chemistry”. When the same two reference materials on quartz fiber filters were submitted to closed-tube combustion, without the initial filtering step, the process was deemed “combustion”. A third preparation method used to generate “bulb” samples of HOxI (SRM 4990B) ( $^{14}\text{C}$  dating reference sample, Table 2) was designed to minimize carbon blanks associated with sample processing. It utilized direct combustion of HOxI in amounts ( $>1$  mg carbon equivalent) quite large compared to typical combustion blanks. Finally, the combustion and total chemistry blanks were prepared without the presence of SP-2 or SRM 1649a (blank samples, Table 2).

Following combustion to  $\text{CO}_2$ , samples were prepared as Fe-C bead targets which were fixed into a custom-made Al target holder and then mounted into the 40-position Al-target wheel of the negative ion sputter source of VERA. The sample labeled “HPG no chemistry” in Table 2 was an exception, in that case the carbonaceous material was used directly as an AMS target without any chemical processing. HPG is a VERA high purity graphite reference material that is  $^{14}\text{C}$ -free (“dead”). Note that all uncertainties quoted are combined standard uncertainties ( $u_c$ ) that are essentially due to counting statistics and variations of the blank carbon mass of individual samples.

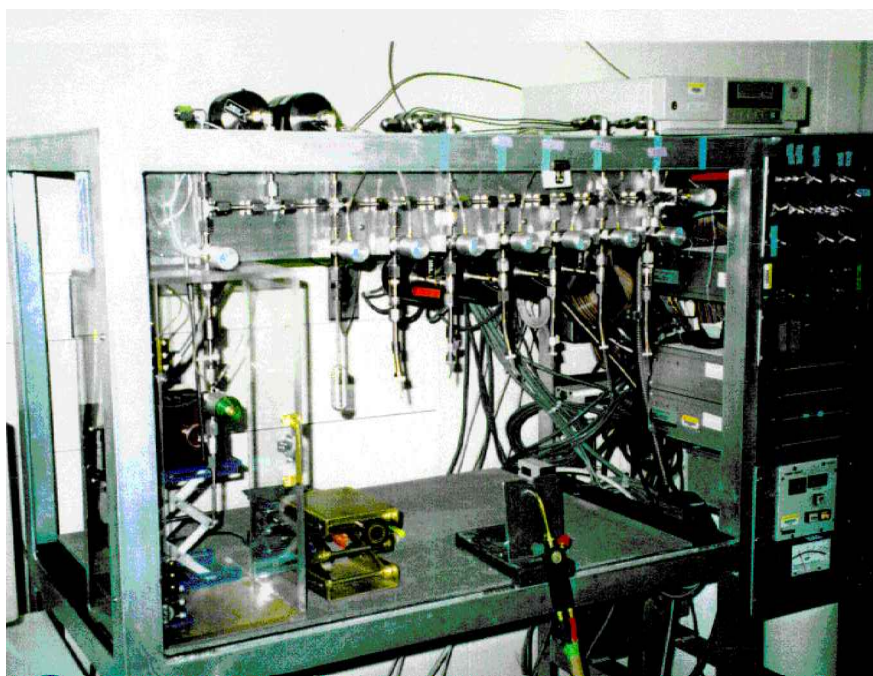


Figure 2 Photograph of the graphitization system at NIST

Table 2 Results of the carbonaceous samples measured at VERA in November 1998<sup>a</sup>

| Sample material                                | $m_s, m_b^b$<br>( $\mu\text{g}$ ) | $f_s, f_b^c$<br>( $10^{-12}$ ) | $f_{0,s}^d$<br>( $10^{-12}$ ) | $^{14}\text{C}_s^e$<br>(pMC) <sup>f</sup> |
|--|-----------------------------------|--------------------------------|-------------------------------|---|
| <i>“Dead” reference samples</i>                |                                   |                                |                               |   |
| SP2-Combusted                                  | 23.4                              | $0.253 \pm 0.13$               | —                             | —   |
| SP2-Combusted                                  | 68.9                              | $0.0458 \pm 0.0023$            | —                             | —   |
| SP2-Total chemistry                            | 18.0                              | $0.87 \pm 0.11$                | —                             | —   |
| SP2-Total chemistry                            | 51.7                              | $0.201 \pm 0.050$              | —                             | —   |
| SP2-Total chemistry                            | 91.4                              | $0.1306 \pm 0.0034$            | —                             | —   |
| HPG-No Chemistry                               | —                                 | $0.00098 \pm 0.00033$          | —                             | $0.11 \pm 0.04$                           |
| <i>“Blank” samples</i>                         |                                   |                                |                               |   |
| Combustion blank                               | 5.8                               | $1.25 \pm 0.20$                | —                             | —   |
| Total chemistry blank                          | 13.8                              | $0.983 \pm 0.023$              | —                             | —   |
| <i><sup>14</sup>C dating reference samples</i> |                                   |                                |                               |   |
| HOxI-Bulb <sup>g</sup>                         | 42.0                              | $0.9554 \pm 0.0067$            | —                             | Ref. material                             |
| <i>Aerosol reference samples</i>               |                                   |                                |                               |   |
| SRM 1649a-Combusted                            | 27.5                              | $0.603 \pm 0.010$              | $0.481 \pm 0.068$             | $52.3 \pm 7.4$                            |
| SRM 1649a-Combusted                            | 84.5                              | $0.5196 \pm 0.0092$            | $0.480 \pm 0.022$             | $52.2 \pm 2.4$                            |
| SRM 1649a-Total chemistry                      | 33.4                              | $0.6383 \pm 0.0079$            | $0.416 \pm 0.067$             | $45.3 \pm 7.3$                            |
| SRM 1649a-Total chemistry                      | 71.4                              | $0.522 \pm 0.011$              | $0.418 \pm 0.027$             | $45.5 \pm 3.0$                            |
| SRM 1649a-Total chemistry                      | 192.0                             | $0.4741 \pm 0.0062$            | $0.437 \pm 0.010$             | $47.6 \pm 1.1$                            |
| <i>Sonnblick snow samples</i>                  |                                   |                                |                               |   |
| S6-Total chemistry <sup>g</sup>                | 37.7                              | $0.798 \pm 0.018$              | $0.699 \pm 0.040$             | $76.1 \pm 4.4$                            |
| S7-Total chemistry                             | 58.6                              | $0.678 \pm 0.011$              | $0.590 \pm 0.025$             | $64.2 \pm 2.8$                            |
| S8-Total chemistry                             | 37.7                              | $0.869 \pm 0.070$              | $0.808 \pm 0.109$             | $87.9 \pm 11.9$                           |
| S9-Total chemistry <sup>g</sup>                | 48.0                              | $0.859 \pm 0.025$              | $0.812 \pm 0.037$             | $88.4 \pm 4.1$                            |
| S10-Total chemistry                            | 45.9                              | $0.773 \pm 0.014$              | $0.689 \pm 0.030$             | $75.0 \pm 3.3$                            |

<sup>a</sup>All isotopic uncertainties (last 3 columns) are combined standard uncertainties ( $u_c$ )

<sup>b</sup>Carbon masses measured by manometry

<sup>c</sup>Isotopic ratio  $^{14}\text{C}/^{12}\text{C}$  of the individual sample  $s$  or blank  $b$  (normalized to  $-25\text{‰}$ )

<sup>d</sup>Isotopic ratio  $^{14}\text{C}/^{12}\text{C}$  of the individual sample  $s$  (background corrected)

<sup>e</sup>Relative  $^{14}\text{C}$  content of an individual sample  $s$  (background corrected)

<sup>f</sup>pMC = percent Modern Carbon (defined as 100 for 1950 AD)

<sup>g</sup>Weighted mean of two sub-samples

## <sup>14</sup>C MEASUREMENT

VERA (see Figure 3) is designed for fast sequential injection of the carbon isotopes  $^{12}\text{C}^-$ ,  $^{13}\text{C}^-$  and  $^{14}\text{C}^-$ . This is achieved by applying different voltages to the insulated vacuum chamber (“bouncer”) of the injection magnet (Priller et al. 1997). The 3-MV Pelletron tandem accelerator was operated with 2.7 MV terminal voltage and Ar-gas stripping. At the high-energy analyzing magnet,  $^{12}\text{C}^{3+}$  and  $^{13}\text{C}^{3+}$  currents were measured in the respective offset Faraday cups. Typical  $^{12}\text{C}^{3+}$  currents were in the range of 6 nA to 1800 nA compared to 9  $\mu\text{A}$  to 12  $\mu\text{A}$  for mg-sized archaeological samples (see also Rom et al. 1998; Wild et al. 1998). These low currents required an adjustment of the timing of the sequential injection system, since the gain and integration times of the Faraday cup amplifiers had to be enhanced for a reliable current measurement. For a complete isotope cycle  $^{12}\text{C}^-$  was therefore injected for 3 ms,  $^{13}\text{C}^-$  for 126 ms and  $^{14}\text{C}^-$  for 1000 ms. The  $^{14}\text{C}^{3+}$  ions were counted with a solid-state surface barrier detector. Each run consisted of 180 cycles.

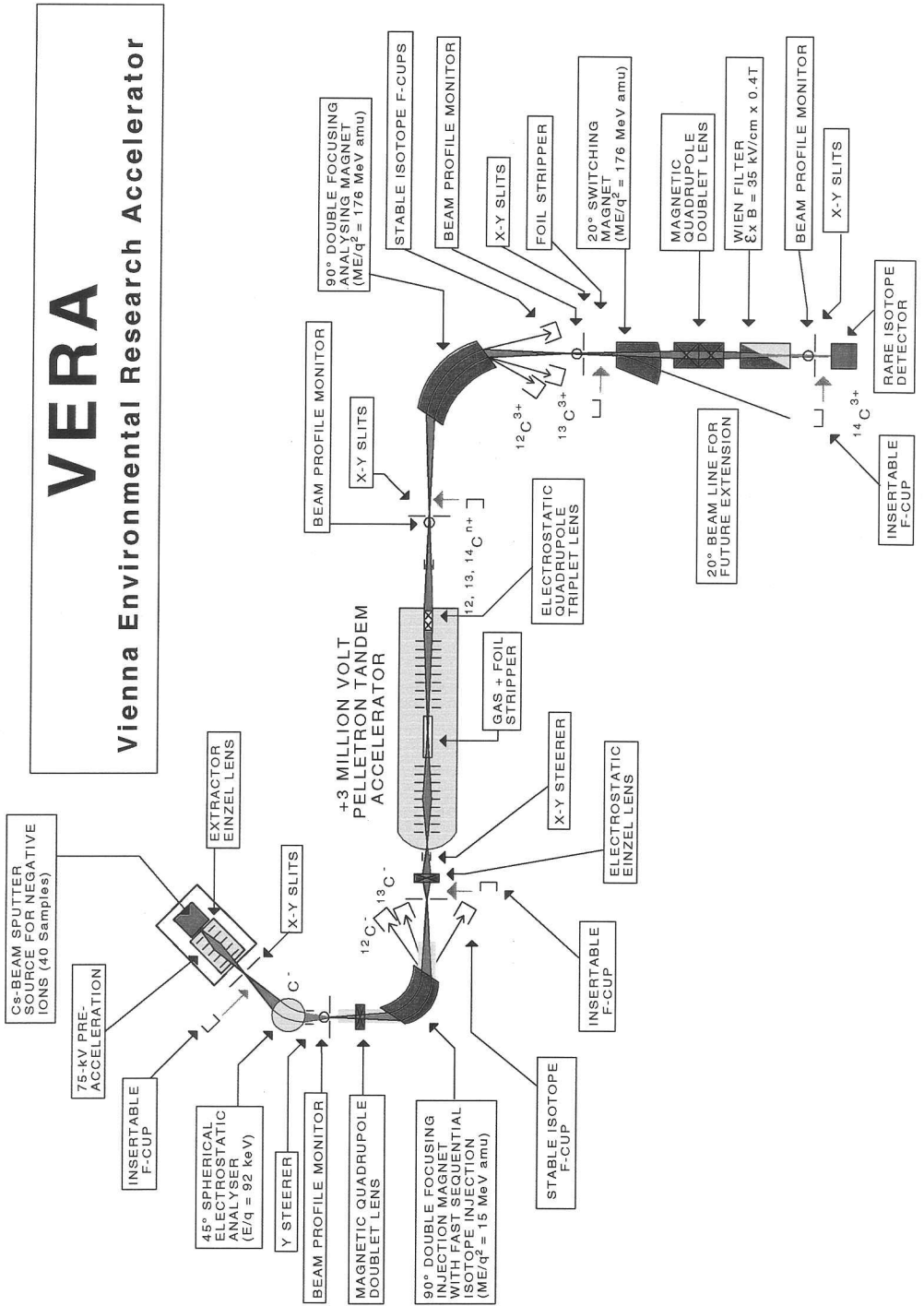


Figure 3 Schematic layout of the AMS facility in Vienna

## DATA ANALYSIS, RESULTS AND DISCUSSION

HOxI was used as  $^{14}\text{C}$  reference material for both  $\delta^{13}\text{C}$  and  $^{14}\text{C}$  content determinations. Whenever more than one target per sample was available, the weighted mean calculated from the individual targets were used. The  $^{14}\text{C}/^{12}\text{C}$  isotopic ratio  $f$  of all samples  $S$  (except HOxI) were normalized to  $\delta^{13}\text{C} = -25\text{‰}$  according to (Stuiver and Polach et al. 1977)

$$f_s = \left( \frac{^{14}\text{C}}{^{12}\text{C}} \right)_{S,-25} = \left( \frac{^{14}\text{C}}{^{12}\text{C}} \right)_{S,\text{meas}} \left[ 1 - \frac{2(25 + \delta^{13}\text{C}_S)}{1000} \right] \quad (1)$$

Subsequently, the individually normalized  $^{14}\text{C}/^{12}\text{C}$  ratios were background corrected. The blank corrected (“true”) isotopic ratio  $f_{0,s}$  of the individual sample was determined by

$$f_{0,s} = \frac{m_s f_s - m_b f_b}{m_s - m_b} \quad (2)$$

assuming that the measured carbon mass  $m_s$  was the sum of the “true” carbon mass  $m_0$  and the blank carbon mass  $m_b$  (see also Verkouteren et al. 1997). The latter is essentially independent of  $m_s$  (Weissenböck et al. 1998) and was derived *directly* (i.e. without adding any sample carbon) by measuring the pressure of the residual  $\text{CO}_2$  from both combustion only and total chemistry (Table 2), but also *indirectly* by using  $f_s$  and  $m_s$  from the nominally  $^{14}\text{C}$ -free SP-2 samples ( $f_{0,s} = 0$ ) and  $f_b$  from the corresponding “blank” samples. The mean total chemistry blank was  $13.1 \pm 2.3 \mu\text{g}$  and the mean combustion blank was  $4.35 \pm 1.67 \mu\text{g}$ . These values were used for background correction of the aerosol “standard” samples and the Sonnblick snow samples (Table 2). The above uncertainties are arithmetic standard deviations from the mean. The carbon contamination of the combustion blank mainly originated from the CuO used for the combustion to  $\text{CO}_2$  (Weissenböck et al. 1998).

The relative  $^{14}\text{C}$  content  $^{14}\text{C}_s$  of the individual samples given in pMC was determined by

$$^{14}\text{C}_s = 100 \frac{f_{0,s}}{0.95 f_{0,\text{HOxI}}} \quad (3)$$

The individual relative  $^{14}\text{C}$  contents of the “combusted” and “total chemistry” SRM 1649a reference samples were in the range of 45.3 pMC to 52.3 pMC (Table 2). Although the weighted means of the three total chemistry samples ( $47.3 \pm 1.0$  pMC) were lower than those of the two combustion samples ( $52.2 \pm 2.3$  pMC), their 95% confidence intervals (CI) overlapped.<sup>6</sup> (In fact, the entire set of 5 was self-consistent within the stated uncertainties, based on a Student  $t$  test:  $t/\text{df}=0.234$ ,  $p=0.53$ .) On the other hand, only the weighted mean of the two combustion samples was consistent with the NIST reference value ( $50.9 \pm 1.3$  pMC). Carbonate removal process appears to be the main reason for this discrepancy.

$^{14}\text{C}_s$  of the filterable aerosol from snow samples collected at Mt. Sonnblick in April 1997 varied between 64 and 88 pMC (see Figure 4) with a weighted mean of  $73.7 \pm 1.7$  pMC. This result indicates that about 64% of filterable carbon in high alpine snow from Sonnblick was of biogenic origin. The

<sup>6</sup>All  $^{14}\text{C}$  uncertainties represent combined standard uncertainties, representing counting statistics and variations of the carbon blank mass. Uncertainties in volumes in Table 1 and masses in Tables 1 and 2 were less than one unit in the last digit.

overall variations were statistically significant ( $\chi^2/df = 6.7$ ,  $p < 0.0001$ ), suggesting variations in the contribution of specific aerosol sources. In order to assess the variability of the data an attempt was made to locate the possible origin of the aerosols. We calculated 96 hr backward trajectories of air masses that reached Mt. Sonnblick at 12:00 and 0:00 UTC (Universal Time Coordinate), respectively, during the time of interest (see also Rom et al., 1999). In all cases, the station backward trajectories showed contact with the earth's surface 24–36 hr earlier, so that the air masses transported to Mt. Sonnblick had the opportunity to pick up pollution from sources near the ground. In terms of fact, the earlier trajectories for 28 April 1997, 0:00 UTC to 30 April 1997, 0:00 UTC show surface contact at the Atlantic ocean; the later trajectories for 30 April 1997, 12:00 UTC, and 1 May 1997, 0:00 UTC show surface contact at continental areas in northern Europe where industrial emissions are likely.

The lack of a simple correlation of the snow-aerosol  $^{14}\text{C}$  with the backward trajectories points of (land, ocean) contact may seem surprising. A simplistic interpretation may be misleading, however, because of 1) lacking information on the surface aerosol  $^{14}\text{C}$  at the particular points and times of contact, 2) imperfections in backward trajectories models and input meteorological data, particularly for extended paths and time intervals, 3) the disjoint relation between air mass history and snow mass formation altitude and transport, and 4) the observed differences in organic aerosol carbon composition of simultaneously sampled air and snow (Gröllert et al. 1996). Although significant  $^{14}\text{C}$  variations were observed, it is clear from Figure 4 that the aerosol carbon contained in all five snow episodes was predominantly biogenic.

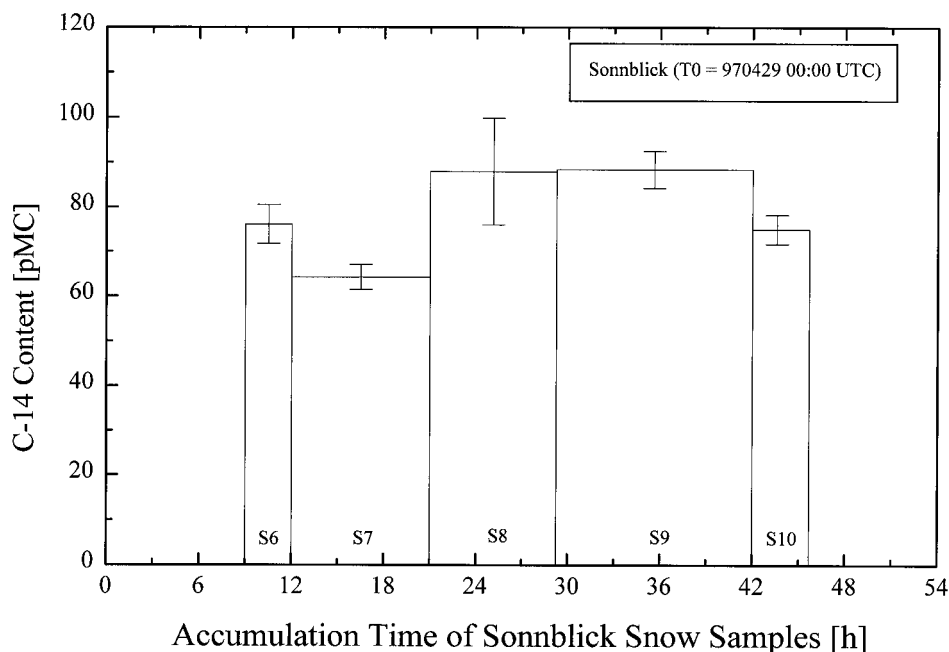


Figure 4  $^{14}\text{C}$  results of the carbonaceous snow samples collected at the high altitude research station Mt. Sonnblick in April 1997



## CONCLUSION

In this paper, we demonstrated that it is possible to measure the  $^{14}\text{C}$  content of below 100  $\mu\text{g}$  carbon samples with AMS at VERA. As result carbonaceous aerosols extracted from snow at the high-altitude research station Mt. Sonnblick in the Eastern Alps, where little carbonaceous material accumulates, can be studied. The  $^{14}\text{C}$  content of the aerosols is not simply correlated with the surface contact points of the corresponding air mass trajectories. Such a behavior is, however, expected for a background site at the mid tropospheric level (see e.g. Tschewenka et al. 1998). The result of 64% biogenic carbon in the filterable (water insoluble) part of the background aerosol at the 3-km level over Europe is indicative of the presence and dominance of biogenic polymeric material. Since the number of samples was quite limited in this study it would be very desirable to perform  $^{14}\text{C}$  investigations in high Alpine aerosols 1) during different seasons, and 2) including the water soluble part.

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