Ice core based reconstruction of past climate conditions and air pollution in the Alps using radiocarbon

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Summary

Climate change, the variation of global and regional climate over time, coupled with changes in atmospheric conditions, has become an important issue. Climate models are a fundamental tool in the discussion of this topic. For their construction and calibration, information of past climate conditions is essential, allowing identification and understanding of climate relevant processes. Therefore, natural archives of past climate such as ice cores are of major importance. So far, numerous factors (so-called climate forcings), which influence our climate system were identified. While they were of purely natural origin in the past, many of them were now realized to be enhanced due to human related (anthropogenic) activities. Model runs clearly indicated that the observed warming during the 20th century can only be explained with the additional forcing introduced by human activity (anthropogenic forcing). However, not all of the different forcing factors, such as the so-called aerosol forcing, are quantified and understood at present time. Thus, forcing by aerosols, and carbonaceous particles in particular, is still not included in climate models widely used today. High altitude glaciers allow reconstruction of past climate parameters such as accumulation and temperature, but can also contribute to the question of human induced climate forcing as they are close to anthropogenic emission sources. However, a profound understanding of the individual archive is required as each of them preserves climate information in its characteristic way. To obtain a meaningful interpretation, proxies must be carefully calibrated against instrumental records for a certain time period to establish a reliable relationship between a proxy indicator and the climatic variable or variables it is assumed to represent. For this study, an ice core from the Fiescherhorn-glacier in the Bernese Oberland was extracted in December 2002 (Swiss Alps, 46°33'3.2''N, 08°04'0.4''E; 3900 m asl.). Using this glacier-archive, the aim was on the one hand the reconstruction of past Alpine temperature and precipitation and on the other hand the investigation of aerosol species which are related to climate forcing, paying special attention to quantify the additional anthropogenic contribution over time. Thereby, the main focus lied on the investigation of carbonaceous particles - a constituent of the aerosol - for which the scientific interest increased over the last years. Although they were recognised to have influence on the radiation balance of the earth, a lack of data exists about their concentrations and sources (i.e. natural, anthropogenic) in the past. Aiming to overcome a major limitation of high altitude glacier-archives arising from imprecise dating in the oldest part, samples from another ice core which was extracted in September 2003 from Colle Gnifetti (Swiss - Italian Alps, 45°55'50"N, 7°52'33"E, 4455 m asl) were analysed. So far, strong layer thinning occurring in the deepest parts of high altitude glaciers did not allow receiving

exact chronologies with conventional methods. The same technique that was developed for the source apportionment analysis of carbonaceous particles could be applied to find a remedy.

The chronology of the Fiescherhorn core was established using i) annual layer counting of seasonal varying signals (NH_4^+, NO_3^-, SO_4^{2-}, δD, melt layers), ii) detected reference horizons attributed to documented events of Saharan dust falls (2000, 1977, 1947) or volcanic eruptions (Katmai 1912), and iii) the years 1956 and 1963 could be fixed by 'wiggle matching' of the δD record with the δD record of the Fiescherhorn 1989 ice core in which maxima in the analysed ^{36}Cl (1956) and ^{3}H (1963) activity, originating from nuclear weapon tests were observed. For the new core presented here, a bottom age of 340 ± 30 years was derived and the resulting time scale covers the time period 1660-2002, extending the one derived by the earlier Fiescherhorn-glacier ice core.

In the records of most major ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, F^-, CH_3COO^-, $HCOO^-$, $CH_3SO_3^-$, Cl^-, NO_3^-, SO_4^{2-}, $C_2O_4^{2-}$) influence from melt water could be recognized. The observed smearing of the signal resulted from the occurrence of snow melt during summer when temperatures are rising above 0°C and liquid water subsequently percolates to deeper layers before it refreezes again. During this process, also the water soluble species are at least partly eluted and shifted to greater depths which results in the described signal in the records. As expected but contrary to this, water matrix components (e.g. δD, $\delta^{18}O$) showed not to be influenced by such events. Despite the described disturbance of the records, main emission sources for the different species could be identified and the anthropogenic input was quantified. For NH_4^+, an industrial maximum (1970-2000) to pre-industrial (1680-1850) ratio of 3.0 was observed and attributed to increased agricultural activities. The ratio of 3.4 for NO_3^- seems to reflect the increased traffic whereas the ratio of 4.2 found for SO_4^{2-} is a result of enhanced consumption of fossil fuels. The highest ratio was found for F^- (14.0), most probably related to HF emissions from the aluminium industry located in the Wallis, nearby to the study site. Anthropogenic emissions of HCl (ratio of 2.1) were related to the combustion of hard coal and in more recent years to the chemical industry and waste combustion.

During the calibration period (1864-2002), the reconstruction of past accumulation resulted in a record which is in general agreement with instrumental data of precipitation measured north of the Alps. However, it revealed a period (before 1910) when the transport path-ways of moisture, finally accumulated at Fiescherhorn, most probably were different from the ones observed today. This was supported by a record of reconstructed annual, summer (JJA) and winter (DJF) temperatures. For this reconstruction two different approaches were applied. In a first approach, a δD-temperature relation of 5.8 ± 0.3‰/°C could be defined, based on the δD ice core record and instrumental measurement of the air temperature performed in the nearby high-alpine research station Jungfraujoch. The second approach was based on the derived melt percent record, allowing the reconstruction of summer temperature. Results derived from the second approach were in good agreement with the observation data for the entire calibration period, whereas the reconstruction based on the precipitation dependent δD record showed severe deviation from the observation data before 1910, probably related to what was observed in the reconstructed accumulation. Nevertheless, both proxies reveal a similar trend of increasing temperatures from 1900-2002, i.e. +2.6°C in summer (JJA) temperatures and +2.0 ± 0.2°C in annual temperatures, respectively.

Carbonaceous particles were extracted from ice core samples with subsequent separation of the two main constituents into organic carbon (OC) and elemental carbon (EC) for the determination of their concentrations in the past. Thereby, a distinction and quantification of natural, biogenic and anthropogenic sources could be achieved, using the fraction of modern carbon (f_M) determined by additional microgram analysis of $^{14}C/^{12}C$ ratios in OC and EC by accelerator mass spectrometry (AMS). An existing method used for the $^{14}C/^{12}C$ analysis in aerosol samples was therefore adapted for ice samples. Procedural blanks of the adapted method were reproducible and resulted in carbon masses of 1.3 ± 0.6 µg OC and 0.3 ± 0.1 µg EC per filter. The determined f_M for the OC blank was 0.61 ± 0.13. In agreement with consensus values, analysis of processed IAEA-C6 and IAEA-C7 reference material resulted in f_M = 1.521 ± 0.011 and $\delta^{13}C$ = -10.85 ± 0.19‰, and f_M = 0.505 ± 0.011 and $\delta^{13}C$ = -14.21 ± 0.19‰, respectively. A first long-term OC and EC concentration record along with the corresponding f_M could finally be derived from an ice core. For this purpose, Fiescherhorn samples were taken from below the firn/ice transition down to bedrock, covering the time period 1650-1940 and thus the transition from the pre-industrial to the industrial era. Before ~1850, OC was approaching a purely biogenic origin with a mean concentration of 24 µg kg^{-1} and a standard deviation of 7 µg kg^{-1}. In 1940, OC concentration was about a factor of 3 higher than this biogenic background, almost half of it originating from anthropogenic sources, i.e. from the combustion of fossil fuels. In 1940, the additional anthropogenic input of atmospheric EC was about 50 µg kg^{-1}. In contrary, the biogenic EC concentration was nearly constant over the examined time period with 6 µg kg^{-1} and a standard deviation of 1 µg kg^{-1}.

As the organic carbon fraction was shown to be of purely biogenic origin (vegetation emissions and biomass burning) before ~1850, it can be assumed to reflect the $^{14}C/^{12}C$ ratio of the living biosphere at the time of photosynthesis and was thus used for ^{14}C dating. Several samples from Colle Gnifetti were analysed and the derived ^{14}C results indicated that this ice archive covers more than 10’000 years. For the first time the age of the oldest ice in the Alps (which is useful for climate research) could be revealed. This new technique may allow for improvement as well as extension of ice core chronologies in the future and might have wide implications for ice core based palaeo-climate research.

Ice with known age, recovered in August 2004 from the Pakitsoq ice sheet margin in West Greenland (69°25.83’N, 50°15.20’W, 370-380 m asl.) was analysed for a calibration of the new ^{14}C dating method. Analysis of a few samples only could be achieved due to very high dust content encountered in this ice. The derived calibrated ^{14}C age (1σ range) for TC was 10’000-7’000 yrs BP and 16’600-12’800 yrs BP for EC, respectively. These results were different from the expected age of 12’600 ± 400 yrs BP. The fact that OC and EC showed different ages is not understood and needs further investigation.