



Temporal changes of inorganic ion deposition in the seasonal snow cover for the Austrian Alps (1983–2014)



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HIGHLIGHTS

- SO_4^{2-} , NO_3^- and H^+ in high alpine snow decrease over 30 years of observations.
- Decrease in concentrations and depositions are lower than decrease in emissions.
- Seasonal patterns show maximum concentrations during spring for NH_4^+ , SO_4^{2-} and H^+ .
- Anthropogenic and crustal influence are the main sources for measured ions.

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ABSTRACT

A long-term record of inorganic ion concentrations in wet and dry deposition sampled from snow packs at two high altitude glaciers was used to assess impacts of air pollution on remote sites in central Europe. Sampling points were located at Wurtenkees and Goldbergkees near the Sonnblick Observatory (3106 m above sea level), a background site for measuring the status of the atmosphere in Austria's Eastern Alps. Sampling was carried out every spring at the end of the winter accumulation period in the years 1983–2014. Concentrations of major ions (NH_4^+ , SO_4^{2-} , NO_3^- , Ca^{2+} , Mg^{2+} , K^+ , Na^+ and Cl^-) were determined using ion chromatography (IC) as well as atomic absorption spectroscopy (AAS) in the earlier years. Concentration of H^+ was calculated via the measured pH of the samples.

Trends in deposition and concentration were analysed for all major ions within the period from 1983 to 2014 using Kendall's tau rank correlation coefficient. From 1983 to 2014, total ion concentration declined ~25%, i.e. solutions became ~25% more dilute, indicating reduced acidic atmospheric deposition, even at high altitude in winter snow. SO_4^{2-} and NO_3^- concentrations decreased significantly by 70% and 30%, respectively, accompanied by a 54% decrease of H^+ concentrations. Ionic concentrations in snow-pack were dominated by H^+ and SO_4^{2-} in the earliest decade measured, whereas they were dominated by Ca^{2+} by the most recent decade. SO_4^{2-} and H^+ depositions, i.e. concentrations multiplied by volume, also showed a significant decrease of more than 50% at both sites. This reflects the successful emission reductions of the precursor gases SO_2 and NO_x . Seasonal values with significantly elevated spring concentrations of NH_4^+ , SO_4^{2-} and H^+ compared to fall snow reflects the beginning of vertical mixing during spring. All other ions do not show any seasonality. Source identification of the ions was performed using a principal component analysis (PCA). One anthropogenic cluster (SO_4^{2-} , NO_3^- and NH_4^+) coming from road traffic or fossil fuel combustion and animal husbandry, one crustal cluster (Ca^{2+} , Mg^{2+}) originating from local geological input or Saharan dust events as well as one cluster of unknown origin with episodic character (Na^+ , K^+ and Cl^-) was found.

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1. Introduction

Determination of the ionic composition of high alpine snow packs plays an important role in estimating the input of nutrients as

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well as eutrophic and acidifying components in terrestrial and aquatic ecosystems. Atmospheric trace gases as well as aerosol particles are removed from the atmosphere and accumulated in the snow cover via different deposition processes like the removal by precipitation (wet deposition) or by direct (dry) deposition. Besides atmospheric precipitation, wind-caused snowdrifts and avalanches play a relevant role for snow accumulation during winter. Schöner et al. (2009) studied the annual course of snow depth for the same glacier fields investigated in this paper. Based on their findings the winter accumulation period can be clearly defined as the period from September to the end of April. Due to spatial and temporal variation of snowfall, the chemical composition of the snow cover is heterogeneous.

During the winter season the deposited compounds accumulate in the snow cover and will be preserved as long as melting does not occur. If melting occurs, the corresponding percolation of meltwater through the snow cover causes a change in the concentration and distribution of solutes, leading to a washout of ions. This is controlled by various processes such as leaching of solutes from snow crystals and grains, meltwater-particulate interactions and microbial activity (Jones, 2001). Additionally, physical and chemical processes further modify the chemical concentration of the snow cover after deposition. Jones (2001) lists potential interactions between the dry snow cover (without melting) and the atmosphere, such as dry deposition, volatilization and snow metamorphism, as main processes, which could alter the concentration or could redistribute ions within the snow cover. Volatilization seems to be of minor importance for non-volatile compounds, but dry deposition is a dominant source for elevated concentrations in the snow cover, especially in rural and remote areas as the Sonnblick region (Davies et al., 1991). Snow metamorphism seems to be unimportant for absolute ion concentrations within the snow pack, but might lead to losses or gains of ions at specific grain interfaces since solutes can be redistributed and concentrated on the snow grain surfaces or boundaries.

For our study region at elevations above 3000 m a.s.l. precipitation is almost exclusively solid during the accumulation period. Snow stratigraphy from multi-year snow pits at altitudes higher than 3000 m a.s.l. show that during this period melt events are rare and restricted to the built up of surface crusts only. Hence, the ionic loads are preserved in the snow cover and the chemical components found in high alpine snow are stored in layers and are released or percolated during the melt period in spring within only a few weeks (Williams and Melack, 1991). As high alpine snow is known to be slightly acidic (Maupetit and Delmas, 1994a, b) snow melt in spring causes an acidifying ionic pulse in melt water and thus for downstream ecosystems. Melt water enriched in inorganic nutrients will also serve as a nutrient supply for microbial communities. The acidification of natural ecosystems observed in the 1980s spawned the idea to investigate the impact and sources of the acidifying compounds incorporated in the snow as well as their neutralizing compounds. Now long-term trends can be deduced.

The investigation of alpine snow chemistry is of major interest since the Alps are a densely populated area, located between highly industrialized countries (Italy, France, Germany and Poland) and the Mediterranean region, thus with intense human impact. Nickus et al. (1997) summarized the results for the SNOSP (snow sampling campaign of the ALPTRAC project) study. It covered 17 high-alpine sampling sites of snow chemistry, ranging from the Southwestern French Alps to the Eastern Austrian Alps, investigated twice a year in March and May from 1990 to 1994. All sites showed a slightly acidic snow cover and NH_4^+ , NO_3^- as well as SO_4^{2-} concentrations were two to three times higher in spring snow than in winter snow. This finding can be well explained by the low vertical mixing height during winter and the increasing number of events with vertical

atmospheric mixing up to 3000 m a.s.l. level in spring (Kuhn et al., 1998). Furthermore, concentrations of most of the ions feature a west to east increase whereas no regional patterns were found for ionic loads. Snow pack studies on glaciers were performed also by other studies, for example in France (Maupetit and Delmas, 1994a), Italy (Filippa et al., 2010) and the USA (Turk et al., 2001) trying to improve the understanding of the physical and chemical processes responsible for the accumulation of major ions in high alpine snow covers. All of these studies covered a maximum time span of four years at the most whereas there are no long-term studies on chemical analysis of high alpine snow packs. One exception is the study from Ingersoll et al. (2008), examining seasonal snow pack chemistry data for the Rocky Mountain region covering the period 1993 to 2004. It shows a significant decrease of SO_4^{2-} concentration and deposition, whereas NH_4^+ and NO_3^- increased significantly in the Central and Southern Rockies. No trend was observed for the Northern Rockies. Regarding not only high alpine sites, Avila (1996) reported a significant decrease of SO_4^{2-} concentration and deposition from 1983 to 1994 in precipitation samples collected in the Montsey mountains in NE Spain, as well as a pH increase. A similar study for Austria combining snow and rain samples by Puxbaum et al. (1998) shows also a significant decrease of SO_4^{2-} and H^+ in ion concentration and deposition. Both authors explained this decrease as an indication that the reduction of SO_2 emissions has an effect on the SO_4^{2-} concentration and deposition over NE Spain as well as over Austria. The first 10 years (1983–1993) of the time series presented in this paper have already been published by Winiwarter et al. (1998). These authors clearly identified SO_4^{2-} and NO_3^- ions as predominantly responsible for acidification, but they did not provide a statistical analysis to isolate robust temporal trends.

Within this paper, we assess the status and temporal changes of the ionic composition in a high alpine snowpack for a multi-decade period using two sampling sites in the Austrian Alps. Due to the elevation of more than 3000 m a.s.l. ionic loads observed in the snow cover of the Sonnblick region are regarded as being driven by air of the mid or free troposphere. The unique time series from 1983 to 2014 is used to investigate trends and seasonal patterns for the accumulation period (autumn, winter and spring) and to determine sources of the ions found in the snow cover. Then the ion concentrations and depositions are compared with emission inventories for Austria and neighboring countries. Finally, we conclude our work with respect to the relevance of findings for high-mountain environment and changes of the status of the atmosphere in the European Alps.

2. Experimental

2.1. Study area

Samples were collected at two different sites, Goldbergkees (GOK) and Wurtenees (WUK), both located in the Austrian National Park Hohe Tauern in the Eastern Alps (see Fig. 1) at an elevation of more than 3000 m a.s.l. These sites are distant to any anthropogenic activity and probably among the most remote areas to be found in Central Europe. At GOK samples were taken annually from 1987 to 2014, whereas at WUK sampling started slightly earlier in 1983 and lasted until 2012. The GOK sampling site is close to the meteorological observatory located at the summit Hoher Sonnblick (SBO), which is part of the Global Atmosphere Watch Program (GAW) of the World Meteorological Organization (WMO). Near the WUK sampling site a ski resort opened in 1985. As the sampling site was situated clearly outside the skiing region we assume that there is no significant influence on ion concentration and snow depth from the production of artificial snow. Sampling at

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