THE CLOUD* PROJECT: CLIMATE RESEARCH WITH ACCELERATORS

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(on behalf of the CLOUD Collaboration‡)

Abstract

The current understanding of climate change in the industrial age is that it is predominantly caused by anthropogenic greenhouse gases, with relatively small natural contributions due to solar irradiance and volcanoes. However, palaeoclimatic reconstructions show that the climate has frequently varied on 100-year time scales during the Holocene (last 10 kyr) by amounts comparable to the present warming—and yet the mechanism is not understood. Estimated changes of solar irradiance on these time scales are too small to account for the climate observations. This raises the question of whether cosmic rays, which are modulated by the solar wind, may be directly affecting the climate, providing an effective *indirect* solar forcing mechanism. Indeed recent satellite observations—although disputed—suggest that cosmic rays may affect clouds under certain conditions. However, given the many sources of variability in the atmosphere and the lack of control of the cosmic ray flux, demonstrating overall cause and effect beginning with changes in cosmic rays and ending with observations of perturbed clouds is challenging. For these reasons a novel experiment known as CLOUD has recently begun operation at the CERN Proton Synchrotron which aims to study, under controlled conditions, the effects of cosmic rays on aerosol nucleation and growth, cloud droplets and ice particles. The CLOUD project represents an unprecedented opportunity for the particle accelerator community to combine their facilities and expertise with those of the atmospheric and climate communities to address a subject of major importance for science and society.

SCIENTIFIC MOTIVATION

Among the most puzzling questions in climate change is that of solar-climate variability, which has attracted the attention of scientists for more than two centuries. Until recently, even the existence of solar-climate variability has been controversial—perhaps because the observations had largely involved correlations between climate and the sunspot cycle that had persisted for only a few decades. Over the last few years, however, diverse reconstructions of past climate change have revealed clear associations with galactic cosmic ray (GCR) variations recorded in cosmogenic isotope archives [1], providing persuasive evidence for solar or cosmic ray forcing of the climate (examples

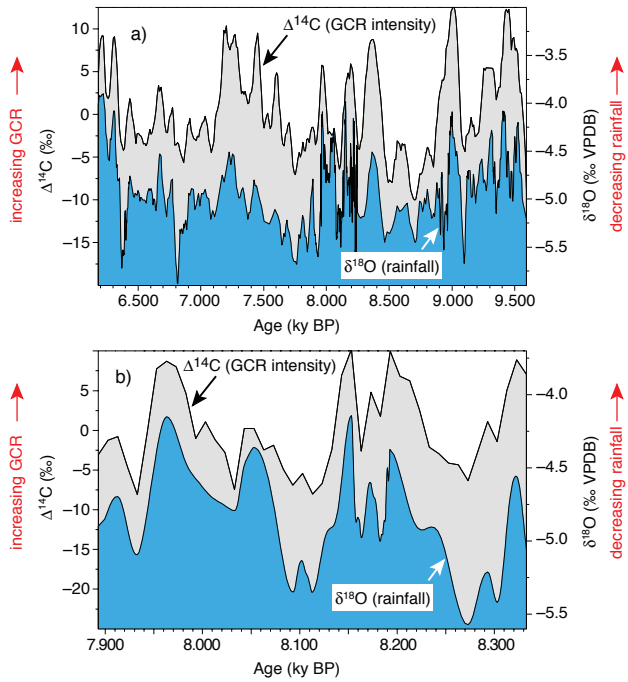

Figure 1: Profiles of δ^{18}O from a U-Th-dated stalagmite from a cave in Oman, together with Δ^{14}C from tree rings in California bristlecone pines and elsewhere, for a) the 3.4 kyr period from 9.6 to 6.2 kyr BP (before present) and b) the 430 yr period from 8.33 to 7.9 kyr BP [2].

are shown in Figs. 1 and 2). However, despite the increasing evidence of its importance, solar-climate variability is likely to remain controversial until a physical mechanism is established. Although this remains a mystery, observations suggest that cloud cover may be influenced by GCRs, which are modulated by the solar wind and, on longer time scales, by the geomagnetic field and by the galactic environment of Earth.

MECHANISMS

Aerosol particles in the atmosphere exert an important influence on climate by changing cloud albedo and lifetime; increased aerosols result in brighter clouds, with longer lifetimes. When aerosols exceed about 100 nm diameter, they constitute cloud condensation nuclei (CCN) and can activate cloud droplets when the relative humidity exceeds 100% by only a few tenths of a per cent.

Although aerosols are ubiquitous throughout the troposphere, their origins are poorly understood. Aerosol lifetimes are typically only a few days and so secondary production from trace condensable vapours is thought to be

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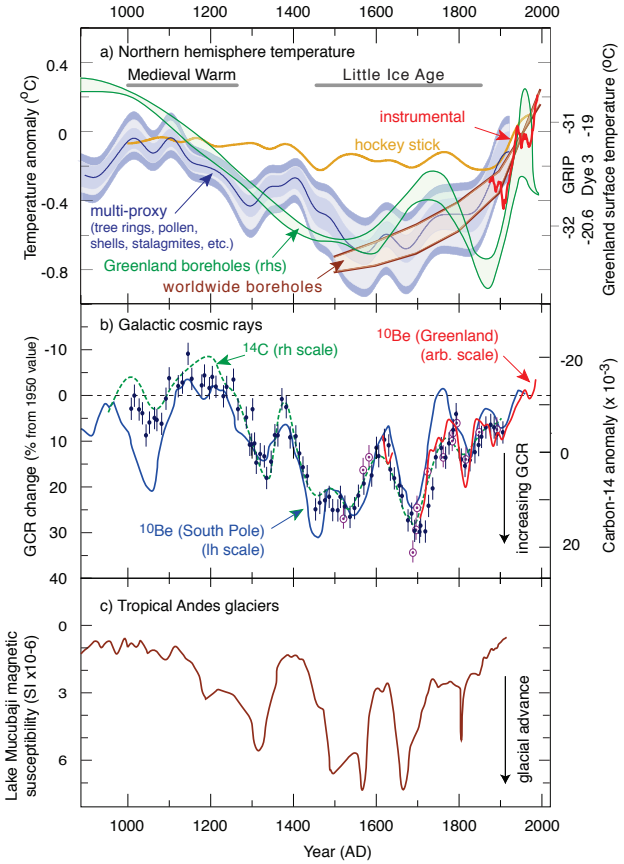

Figure 2: Comparison of variations during the last millennium of a) temperature (with respect to the 1961–1990 average), b) galactic cosmic rays (note the inverted scale; high cosmic ray fluxes are associated with cold temperatures) and c) glacial advances in the Venezuelan tropical Andes near the equator (at 3570 m altitude) [1].

the major source over large regions of the atmosphere that are remote from primary sources. Indeed, so-called *gas-to-particle conversion* (Fig. 3) may be responsible for as much as one half of global CCN. After exceeding a critical size, the embryonic molecular clusters are stable against evaporation and may grow by further condensation and coagulation to CCN sizes—although only a small fraction survive due to coagulation with existing larger aerosols. Sulphuric acid, produced by the photo-oxidation of sulphur dioxide, is thought to be the primary condensable vapour in the atmosphere. However atmospheric H_2SO_4 concentrations are generally less than 10^6–10^7 cm^{-3} which, according to classical nucleation theory, is far too low for binary homogeneous nucleation of H_2SO_4/H_2O to occur at an appreciable rate. Several mechanisms have therefore been proposed to enhance sulphuric acid nucleation at low vapour concentrations. Among these is ion-induced nucleation, where the presence of a single charge may stabilise the sub-critical cluster and accelerate the early growth rate (Fig. 3) [3].

Except for contributions from radioactive isotopes near the land surface, GCRs are responsible for generating all the fair-weather ionisation in the troposphere and lower stratosphere, where clouds form. Ion-induced nucleation is therefore a candidate mechanism for solar-cosmic ray-climate variability. A second class of mechanisms concerns the influence of GCRs on the global electrical circuit in the atmosphere. This affects the space charge above and below cloud boundaries, leading to more highly charged aerosols which may affect ice nucleation and other cloud microphysical processes [3].

THE CLOUD EXPERIMENT

Definitive laboratory measurements of the fundamental physical and chemical processes underlying these mechanisms have not yet been made, and their climatic significance is unknown. The CLOUD experiment [4] has been designed to answer these important open questions.

The concept is to expose a large chamber, filled with ultra-pure humidified air and selected trace gases, to an adjustable "cosmic ray" beam from the CERN Proton Synchrotron. By means of sensitive analysing instruments attached to sampling probes, the nucleation and growth of aerosol particles inside the chamber can be monitored and analysed. In separate experiments, small (up to 200 mbar) adiabatic expansions can be made to operate the chamber as a classical Wilson cloud chamber for studies of cloud droplets and ice particles. CLOUD will investigate the effects of cosmic rays on a wide range of microphysical cloud processes: the creation of aerosols by trace condensable vapours; aerosol growth up to CCN sizes; the activation of CCN into cloud droplets; aerosol-droplet interactions; and finally the creation and dynamics of ice particles in clouds.

The CLOUD experiment (Fig. 4) is optimised to study ion-aerosol-cloud interactions. Technical input for the design was obtained in a pilot experiment in 2006 [5]. The chamber is a 3m-diameter electro-polished 316L stainless-steel cylinder (26.1 m^3). A field cage comprising two transparent electrodes at voltages of up to +30 kV and -30 kV, respectively, is installed inside the chamber to allow the rapid removal of ions, when required. The contents of the chamber are irradiated by ultra violet (UV) light in the range 250-400 nm. The UV is introduced via 240 optical fibre vacuum feedthroughs installed on top of the chamber. The chamber is surrounded by an insulated thermal housing. The temperature is controlled by precisely regulating the temperature of the air circulating in the space between the chamber and the thermal housing. Experimental runs can be performed at highly-stable temperatures (near 0.01°C) between 40°C and -30°C (to be extended to -90°C in 2011). In addition, the chamber can be raised to 100°C for bakeout.

The chamber is exposed to a 3.5 GeV/c secondary pion beam from the CERN PS, corresponding to the characteristic energies and ionisation densities of cosmic ray muons in the lower troposphere. The beam intensity can be adjusted over the natural range from ground level to the stratosphere.

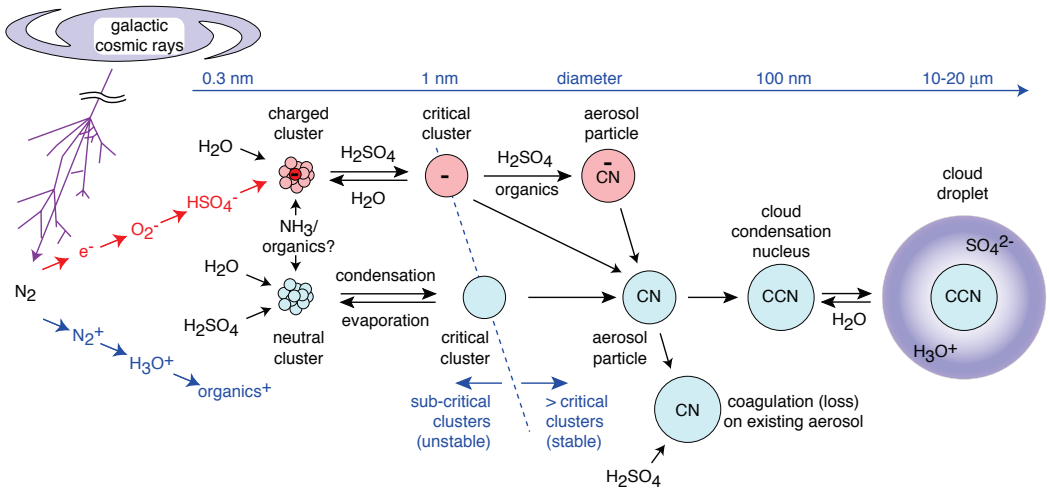

Figure 3: Nucleation and growth of new particles from trace condensable vapours and water in the atmosphere. The ion-induced nucleation pathway (red) is energetically favoured over the neutral pathway (blue) but limited by the ion production rate from galactic cosmic rays, and by the ion lifetime. Cosmic rays may also accelerate the early growth rate, which controls the survival fraction of embryonic clusters to reach the critical size.

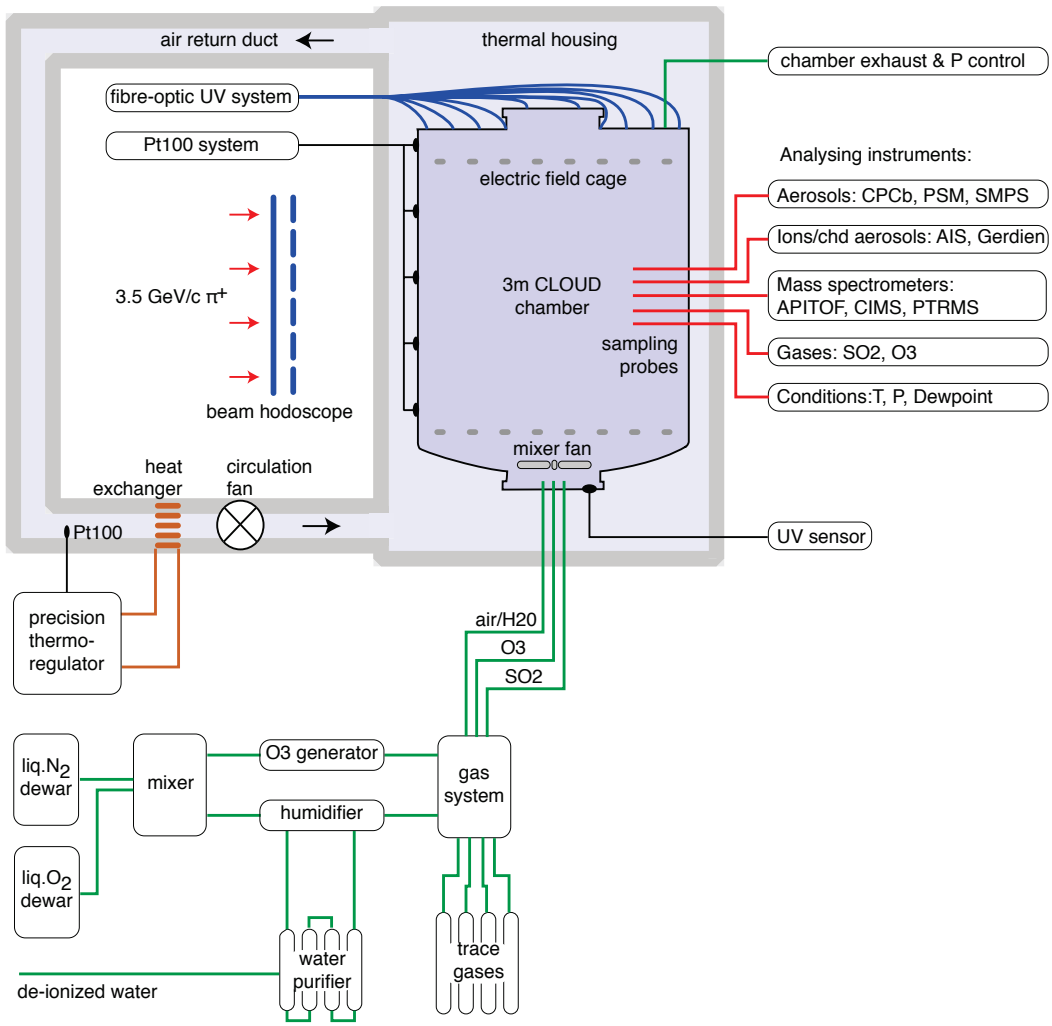

Figure 4: Schematic diagram of the CLOUD experiment in 2009.

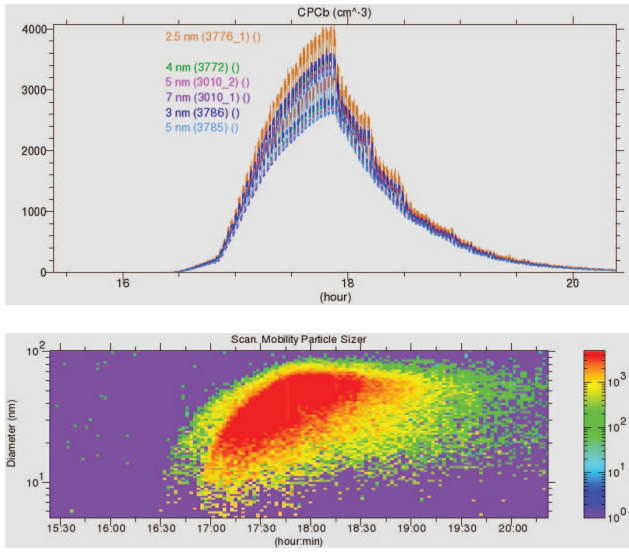

Figure 5: An example of a one of the first nucleation bursts recorded by CLOUD. The upper panel shows the time development of the aerosol particle concentration, measured by the CPC battery. The lower panel shows the time-development of the aerosol size spectrum measured in the SMPS, with a detection threshold at ∼8 nm. The pion beam was turned on at 16h45 and produced a sharp increase in the aerosol nucleation rate, detected shortly afterwards (seen as a break near the start of the curves in the upper panel). This is a clear observation of ion-enhanced nucleation.

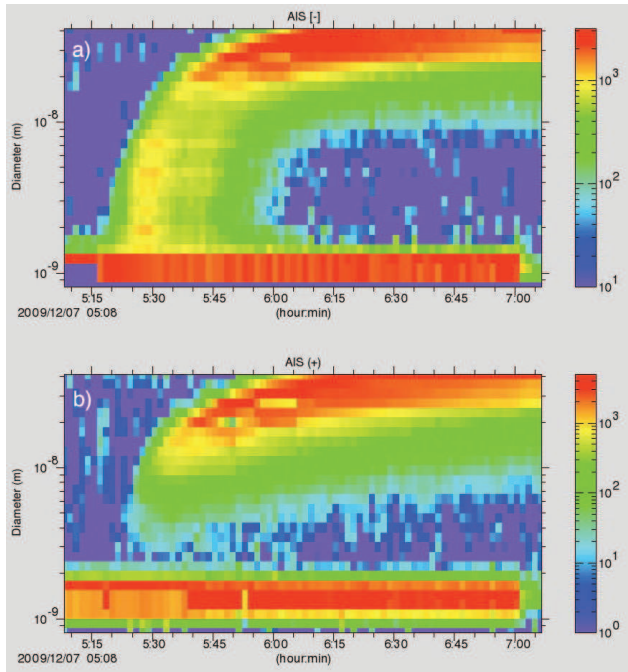

Figure 6: A nucleation ("banana") event seen in the AIS: the time evolution of the size spectrum for a) negatively charged particles, and b) positively charged particles. Ion-induced nucleation by negatively-charged particles is clearly seen.

Ultra-pure air is obtained from the evaporation of cryogenic liquid N_2 and liquid O_2, mixed in the ratio 79:21, respectively. The air is humidified with a Nafion humidifier through which ultra-pure water re-circulates in a Millipore Super-Q system. Ozone is added to the air by UV irradiation. Trace gases such as SO_2 are added from trace gas cylinders using N_2 pressurised gas. In order to compensate for sampling losses, there is a continuous flow of fresh gases into the chamber of 78 l/min, resulting in a dilution lifetime of 5.6 h. An internal magnetically-coupled fan rapidly mixes the fresh gases and beam ions, ensuring good uniformity inside the chamber.

The contents of the CLOUD chamber are continuously analysed by instruments connected to sampling probes that project 0.5 m into the chamber. The chamber instrumentation for the first run comprised mass spectrometers (Chemical Ionisation Mass Spectrometer [CIMS] for H_2SO_4 concentration, Proton Transfer Reaction Mass Spectrometer [PTRMS] for organic materials concentrations and Atmospheric Pressure Interface Time Of Flight [APITOF] mass spectrometer for chemical analysis of the positive and negative ions up to 2000 amu); particle counters and size analysers (Condensation Particle Counter [CPC] battery, Particle Size Magnifier [PSM] at a size threshold of 1.5–2 nm, and Scanning Mobility Particle Sizer [SMPS]); ion size spectrometers (Atmospheric Ion Spectrometer [AIS] and Gerdien counter); gas analysers (O_3 and SO_2); and chamber operating conditions (dew-point sensor, UV intensity, temperature and pressure).

RESULTS FROM THE FIRST RUN

CLOUD was installed at the CERN PS in 2009 and a very successful first run was carried out in November-December to study ion-induced and neutral binary nucleation of H_2SO_4/H_2O, at 292 K. Shortly after the start of the run, a clear ion-enhancement of the nucleation rate was established by the reproducible observation of a sharp increase of the nucleation rate when the beam was turned on (examples are shown in Figs. 5 and 6). The detector showed excellent technical performance, and a large amount of high quality data were recorded during the two-week run (some examples are provided in Figs. 7-10). The data from the first run are currently being analysed and a journal paper on the key new results is in preparation, for expected publication later this year.

Acknowledgements

On behalf of the CLOUD collaboration I would like to thank CERN PH-DT, EN-MME, EN-MEF and TE-VSC for their excellent support in the design, construction and installation of CLOUD and, in addition, to thank the CERN machine team for their efficient operation of the PS.

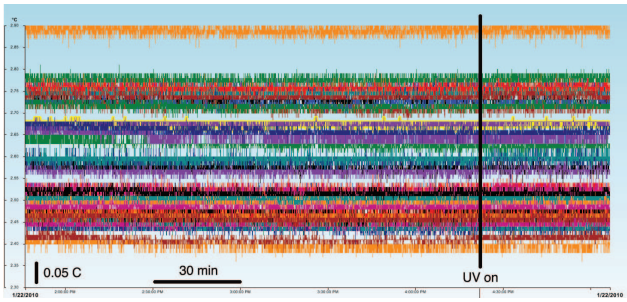

Figure 7: An example trace of the individual Pt100 thermometers recording the temperatures of the CLOUD chamber walls over a 3.3 hour period (2.6°C mean temperature). After the UV light was turned on at full intensity, no increase of temperature was observed on any Pt100, including an internal sensor that measures the gas temperature (purple trace, highlighted in yellow). The individual Pt100 resolution is 0.01°C.

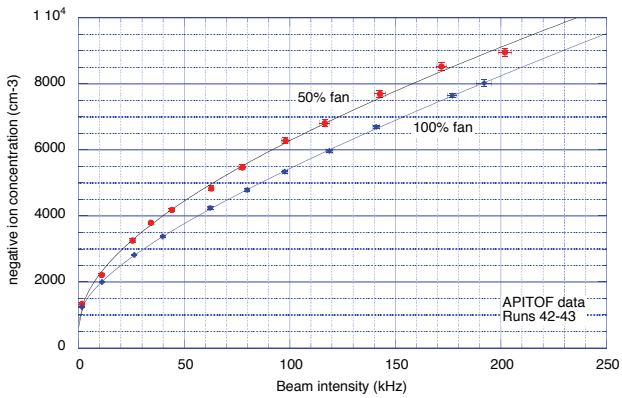

Figure 8: Negative ion concentration in the chamber, measured with the APITOF, as a function of beam intensity and at two different fan speeds. The curves have the expected shapes based on the known ion-ion recombination coefficient and on the measured wall loss rates.

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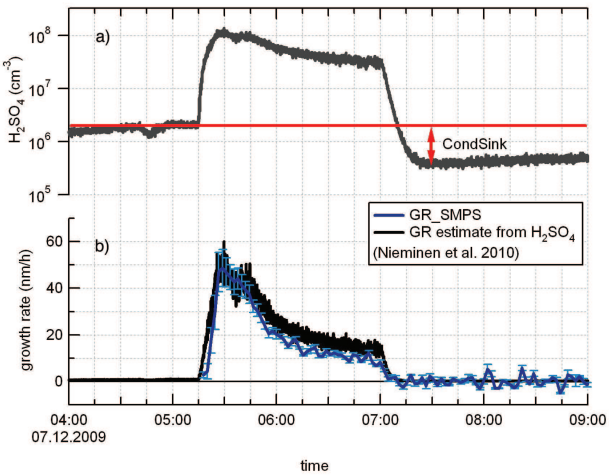

Figure 9: Time evolution of the measured and calculated aerosol particle growth rates during the same nucleation run as seen in Fig. 6: a) sulphuric acid concentration measured by the CIMS and b) comparison of the growth rate measured by the SMPS (blue curve) with that calculated from the measured sulphuric acid concentration (black curve). The good agreement indicates that the CIMS is well calibrated. The UV light was turned on at 05h15, maintained at a constant 50% intensity and then turned off at 07h00. Depletion of the H_2SO_4 concentration due to the aerosol condensation sink can be seen.

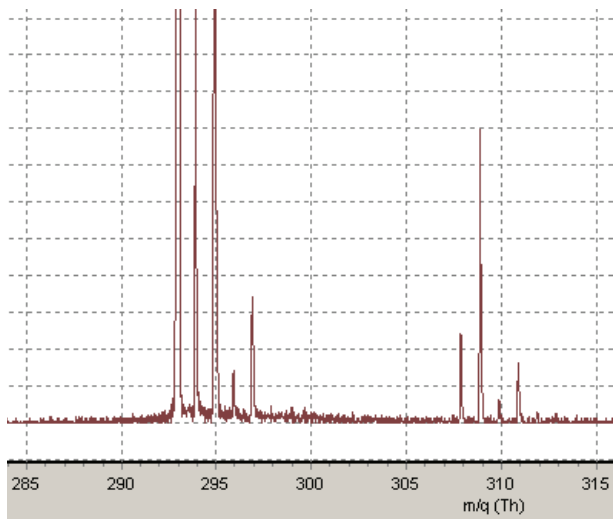

Figure 10: An example of the high resolution chemical analysis of ions from the APITOF. The first group of peaks starting at 293 amu corresponds to the charged sulphuric acid trimer, $(H_2SO_4)_2.HSO_4^-$; five isotopes of S and O are visible, in the expected relative abundance. The second group starting at 308 amu corresponds, respectively, to $(H_2SO_4)_2.SO_5^-$, $(H_2SO_4)_2.HSO_5^-$, and two isotopes.