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Measurement of atmospheric ¹⁴CH₄ in Antarctic ice over the agro-industrial period: a status report.

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During the 2005 Austral summer, an ANSTO-lead expedition using a thermal ice drill collected a large (20 cm) diameter ice core from near the summit of Law Dome, East Antarctica. The drill reached a depth of 260 m where the ice dated from \sim 1620 AD and the air, contained as bubbles in the ice, from \sim 1680 AD.

This marked completion of the first stage of an ambitious collaborative project to learn more about the sources of the important greenhouse gas methane [CH₄] over the agro-industrial period (from ~1700 AD on). Whilst it is well known that methane has increased from 700 ppb to 1,700 ppb over this time as a result of human activities, the development of the different methane sources is poorly understood. Knowledge of how ¹²CH₄, ¹³CH₄ and ¹⁴CH₄, as well as CH₃D, have changed in time permits validation of postulated methane source scenarios. We are attempting to make well time-resolved measurements of ¹⁴C of CH₄ over this period. With just 100 mL of air obtainable from each 1 kg of ice, combined with the low concentration of methane in the air, special techniques are required to undertake such demanding ¹⁴C measurements, even when employing the sensitive technique of accelerator mass spectrometry [AMS].

Core chronology is being established from a combination of techniques. Seasonal ice layers are identified by measurements of Δ^{18} O of ice, glacio-chemicals and major ions. CO₂ and CH₄ mixing ratios and δ^{13} CH₄ will also be measured along the core. Diffusion of atmospheric gases through the firn and bubble-trapping at the firn-ice transition will be numerically modelled, with the tritium and ¹⁴CO₂ "bomb-pulses" used to constrain model parameters.

Each air sample will be derived from ~ 100 kg of ice, melted in 25 kg blocks in a

specially-constructed stainless steel melting tank. The air will be stored in glass flasks until the CH₄ is separated and converted to CO₂, stored in glass break-seals. The CO₂ will then be converted to graphite for ionisation in the AMS ion source. Approximately 5 μ g of carbon will be derived from each 100 kg of ice, too little for the capability of most radiocarbon AMS labs. As part of this project, a laser-heated reaction vessel, optimised for extremely small samples, has been developed to produce the graphite. Additionally, improvements have been made to the ANTARES accelerator to further improve its small-sample capability. A summary of our progress towards these goals will be presented.