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Continuous methane measurements from a late Holocene Greenland ice core: Atmospheric and in-situ signals

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ABSTRACT

Ancient air trapped inside bubbles in ice cores can now be analysed for methane concentration utilising a laser spectrometer coupled to a continuous melter system. We present a new ultra-high resolution record of atmospheric methane variability over the last 1800 yr obtained from continuous analysis of a shallow ice core from the North Greenland Eemian project (NEEM-2011-S1) during a 4-week laboratory-based measurement campaign. Our record faithfully replicates the form and amplitudes of multi-decadal oscillations previously observed in other ice cores and demonstrates the detailed depth resolution (5.3 cm), rapid acquisition time (30 m day^{-1}) and good long-term reproducibility (2.6%, 2σ) of the continuous measurement technique.

In addition, we report the detection of high frequency ice core methane signals of non-atmospheric origin. Firstly, measurements of air from the firn–ice transition region and an interval of ice core dating from 1546–1560 AD (gas age) resolve apparently quasi-annual scale methane oscillations. Traditional gas chromatography measurements on discrete ice samples confirm these signals and indicate peak-to-peak amplitudes of ca. 22 parts per billion (ppb). We hypothesise that these oscillations result from staggered bubble close-off between seasonal layers of contrasting density during time periods of sustained multi-year atmospheric methane change. Secondly, we report the detection of abrupt (20–100 cm depth interval), high amplitude (35–80 ppb excess) methane spikes in the NEEM ice that are reproduced by discrete measurements. We show for the first time that methane spikes present in thin and infrequent layers in polar, glacial ice are accompanied by elevated concentrations of carbon- and nitrogen-based chemical impurities, and suggest that biological in-situ production may be responsible.

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1. Introduction

Records of past atmospheric methane concentration derived from ancient air trapped in ice cores help to define our understanding of the global carbon cycle and its interaction with climate on many timescales (e.g., Blunier and Brook, 2001; Loulergue et al., 2008; Mitchell et al., 2011). Specifically, records of past methane variability are extremely valuable for understanding the relative strength and spatial distribution of methane sources and sinks (Brook et al., 2000) and their response to climate change (Lelieveld et al., 1998; Menviel et al., 2011; Singarayer et al., 2011). Furthermore, methane has become an important tool for global synchronization of ice core records as a result of its large and frequent variations on time scales of interest for paleoclimate reconstruction (Blunier et al., 2007).

Ice core methane mixing ratios are typically determined by gas chromatography on discrete pieces of ice using a wet (e.g., Flückiger et al., 2002; Grachev et al., 2007, 2009) or dry (e.g., Blunier et al., 1995; Etheridge et al., 1998; MacFarling Meure et al., 2006) extraction method. Discrete analysis using these methods can be both highly accurate and reproducible (Mitchell et al., 2011) but it is relatively labour- and time-intensive, and, depending on sample spacing, some true signal variability will be aliased. Recently, a powerful technique for efficient, high resolution, continuous measurement of ice core methane using laser spectroscopy has been

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developed (Stowasser et al., 2012). During two consecutive seasons at the North Greenland Eemian (NEEM) ice core project drill site, a system of gas extraction was coupled to the continuous flow analysis (CFA) setup used for chemical analysis of the NEEM ice (Bigler et al., 2011; Kaufmann et al., 2008). Gas released from bubbles in the ice, previously considered as a waste product, was extracted and analysed in real time at a precision equalling or surpassing discrete measurements (Stowasser et al., 2012). Many of the techniques and solutions devised at NEEM were employed for our analytical campaign at the Desert Research Institute (DRI), Reno, NV, USA, in which methane concentrations were measured continuously on the 410 m NEEM-2011-S1 (hereafter referred to as NEEMS1) ice core to reconstruct methane concentrations of the past 1800 yr.

Stowasser et al. (2012) reported a depth resolution (the minimal depth range for which a damped version of a periodic input signal can be detected) of 5 cm for continuous methane measurements along the main NEEM core. This is significantly better than that of the highest resolution routine discrete measurement programs, which are typically conducted at metre-scale resolution (e.g., Mitchell et al., 2011). Significantly, the excellent depth resolution of the continuous technique equates to a temporal resolution that allows detection of all "climatically relevant" variations in methane down to 1980 m depth in the NEEM core (Stowasser et al., 2012). Effectively, this means that any oscillation in atmospheric methane will be resolvable in the ice core record, providing that it survives the processes of signal smoothing, which occur in the firn pack during densification, prior to complete bubble close-off (Spahni et al., 2003). The degree of smoothing in the firn varies between ice cores, and is primarily dependent on temperature and accumulation rate (Schwander et al., 1997). For Holocene ice at NEEM the gas age distribution (width at half maximum) at the base of firn is ca. 32 yr (Buizert et al., 2012) and therefore any higher frequency variability in the atmosphere, for example, the seasonal cycle in methane concentration, cannot be preserved in the ice core gas record.

In this study we demonstrate that our efficient and precise continuous measurements on the NEEMS1 ice core produce an ultra-high resolution atmospheric history of methane for the last 1800 yr, which replicates multi-decadal variability previously observed. Further key findings of this paper concern high frequency methane signals that cannot be of atmospheric origin. Reproducible annual scale methane oscillations that may be artifacts of heterogeneous bubble close-off in the firn are detected. We also report the presence of abrupt, high amplitude methane spikes in the ice that likely result from biological in-situ production of methane.

2. Method

2.1. NEEM-2011-S1 ice core drilling

The NEEMS1 ice core was drilled in northern Greenland to 410 m depth in 2011. The borehole location was 200 m from the main NEEM borehole (77.45°N, 51.06°W). Drill fluid was used to maintain the diameter of the borehole below 80 m depth. The ice core was cut in the field, and later also at DRI, to produce two equi-dimensional (55 × 3.4 × 3.4 cm) sticks, designated M cut and B cut, for continuous analysis.

2.2. Analytical system

The continuous melter system at DRI (McConnell and Edwards, 2008; McConnell et al., 2007, 2002) was modified to include two sealed debubblers, a gas extraction box (detailed description in Stowasser et al., 2012) and a laser spectrometer (Fig. 1, letters in parentheses in following text are displayed on figure). A mixture of melted ice and gas bubbles, extracted from the middle concentric ring of the melterhead (A) (2.73 cm^2 area), was pumped to the first sealed debubbler (B) (internal volume 1.8 cm^3). All the gas bubbles and approximately 30% of the water were pushed on towards a second debubbler (C) (internal volume

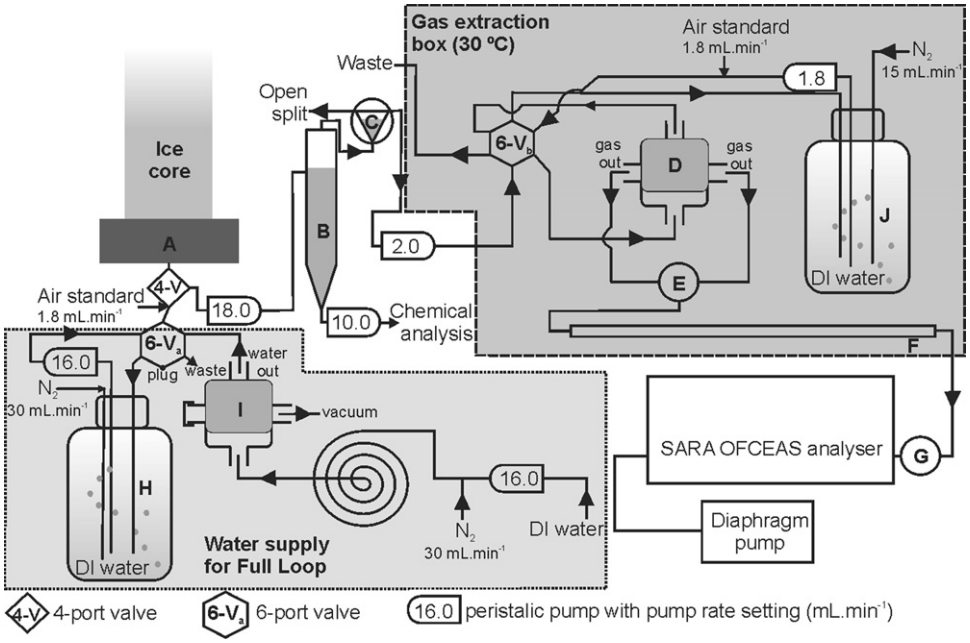

Fig. 1. Schematic of the continuous ice core gas analytical setup. Ice core melting occurs at a heated melterhead (A), sample is pumped to a debubbler (B), open split (C) and into the gas extraction box. The gas extraction system consists of a gas-permeable membrane module (Liqui-Cel, G591) (D) and a homemade Nafion dryer (2 m length, 0.3 mm inner diameter) (F). Gas flow is monitored by a pressure transducer (Omega Inc.) (E) and controlled by a back-pressure controller (IQ^+ Flow, Bronkhorst Inc.) (G). Degassed water from either a N_2-flushed 2 L bottle (H) or a gas-permeable membrane module (I) (position of 6-V_a determines this) is mixed with synthetic air standard (position of 6-V_a determines which) and introduced to the system via a 4-port valve below the melterhead. when no ice core is being melted.

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