

## **Supplemental Material**

### *Offset corrections between the two laboratories*

The agreement between the EDC CH<sub>4</sub> measurements performed at Bern and LGGE is generally good, but the Bern data are consistently higher by 6 ppbv because of a different contamination correction<sup>31,33</sup>. In order to construct a continuous record consistent with earlier Dome C measurements<sup>31,33</sup>, we added 6 ppbv to all concentrations measured at LGGE along the Dome C record.

### *Offset between Vostok and Dome C measured at Grenoble*

As the two ice cores were measured in the same laboratory (LGGE) but with different gas extraction system, we have checked the offset between the two ice cores by comparing the methane values on the same time scale and time resolution. The offset between uncorrected LGGE EDC measurements and published Vostok results<sup>29</sup> is  $7.74 \pm 0.52$  ppbv (corresponding to 1.5% of the average mixing ratio measured in the two cores), with EDC values higher than Vostok values. The Vostok measurements were all performed in the 90's with the previous non-automated LGGE gas extraction system. This 7.74 ppbv difference surprisingly corresponds well to the difference between contamination corrections applied with the older extraction system (around 20 ppbv) and with the new one (around 11 ppbv<sup>33</sup>). As there is no measurable drift in our standard gas, the difference may come from an overestimate (underestimate) of the contamination correction with the old (new) system. The Vostok CH<sub>4</sub> data presented in this paper are not corrected for this last offset.

### *Time resolution of the CH<sub>4</sub> measurements*

Figure S2 shows the time resolution of our record as a function of time (0-400 and 400-800 kyr). From 0 to 400 kyr, it gets lower than 1000 years only between four CH<sub>4</sub> consecutive

data pairs. From 400 to 800 kyr, the oldest part of MIS 15 as well as MIS 16 (time range 580-660 kyr) often include CH<sub>4</sub> consecutive data pairs separated by 2 to 3 kyr. The least time resolutions take place during MIS 16 (corresponding to the data published in Spahni et al. (2005)<sup>29</sup>), when it is possible that our record missed part of the millennial-scale CH<sub>4</sub> variability. The new data covering the oldest section of the EDC record (666-799 kyr BP) show a time resolution ranging between 50 and 2185 years (one occurrence), with a mean of 550 years (gaussian distribution with a sigma of 260 years) and a median of 520 years.

#### *Correlation between CH<sub>4</sub> and EDC temperature warmth maxima*

Figure S3 shows the relationship between the maximum CH<sub>4</sub> mixing ratio observed during each of the nine interglacials covered by our record and the concomitant EDC maximum temperature, deduced from  $\delta D$  measurements corrected for the changing isotopic composition of the ocean and for altitudinal changes at the drilling site<sup>34</sup>. For MIS 1, we consider the first CH<sub>4</sub> maximum taking place at the start of the Holocene. For MIS 15, two maxima are considered in the comparison. The linear regression excluding MIS 9 and MIS 19 has a R<sup>2</sup> of 0.82 (n=8).

#### *Spectral analyses*

The spectral analyses were made via the Analyseries 2.0.3 program on Macintosh<sup>35</sup> to extract the most prominent frequencies of our signal. We have used several different spectral analyses and compared them to confirm the robustness of our results. Here we will give a short description of the methods chosen to analyse our results.

#### *The multi-tapered method*

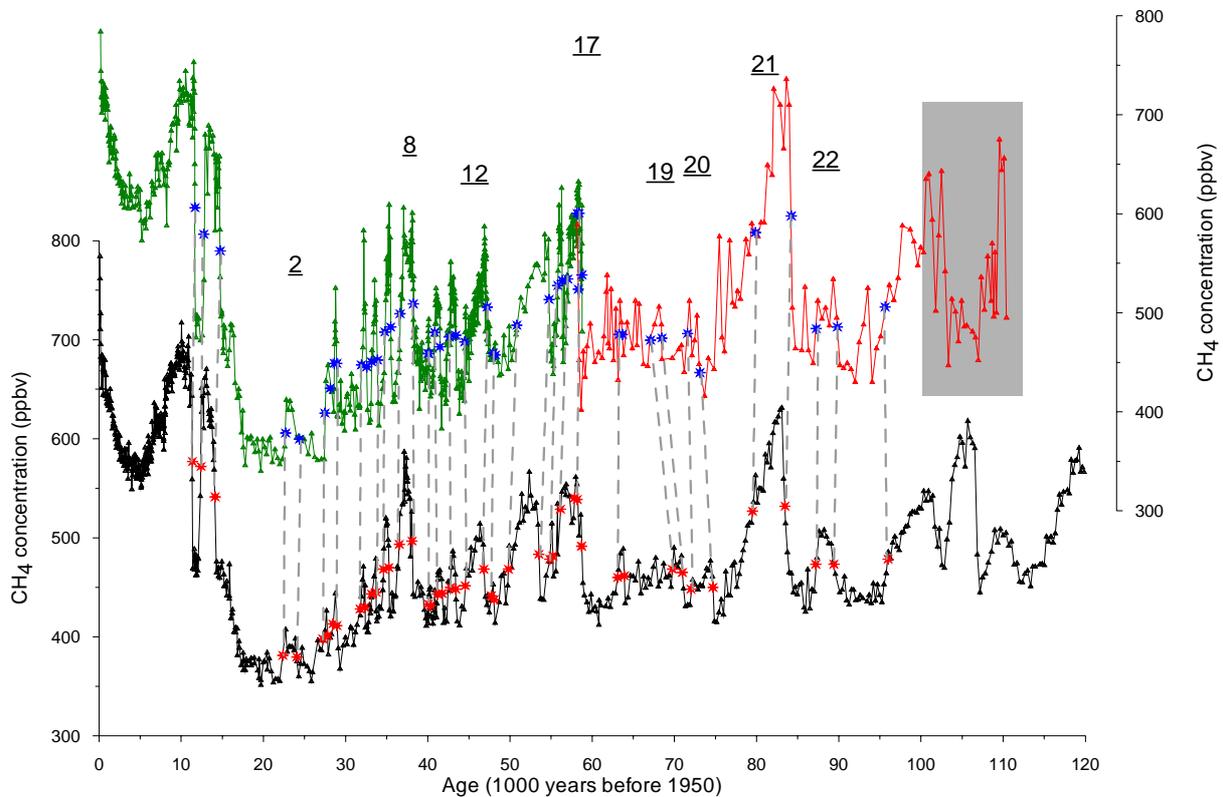
In Figure 2a, the EDC methane record is analyzed with this method. We have chosen it to define the amplitude of the different orbital frequencies and compare it on different time scales. This method is independent of the spectral power, even small amplitude oscillations may be considered as significant. This is not the case with a Blackman-Tukey method. The significance of each spectral peak is determined through a F-test ( $>0.95$ ).

#### *The Blackman-Tukey method*

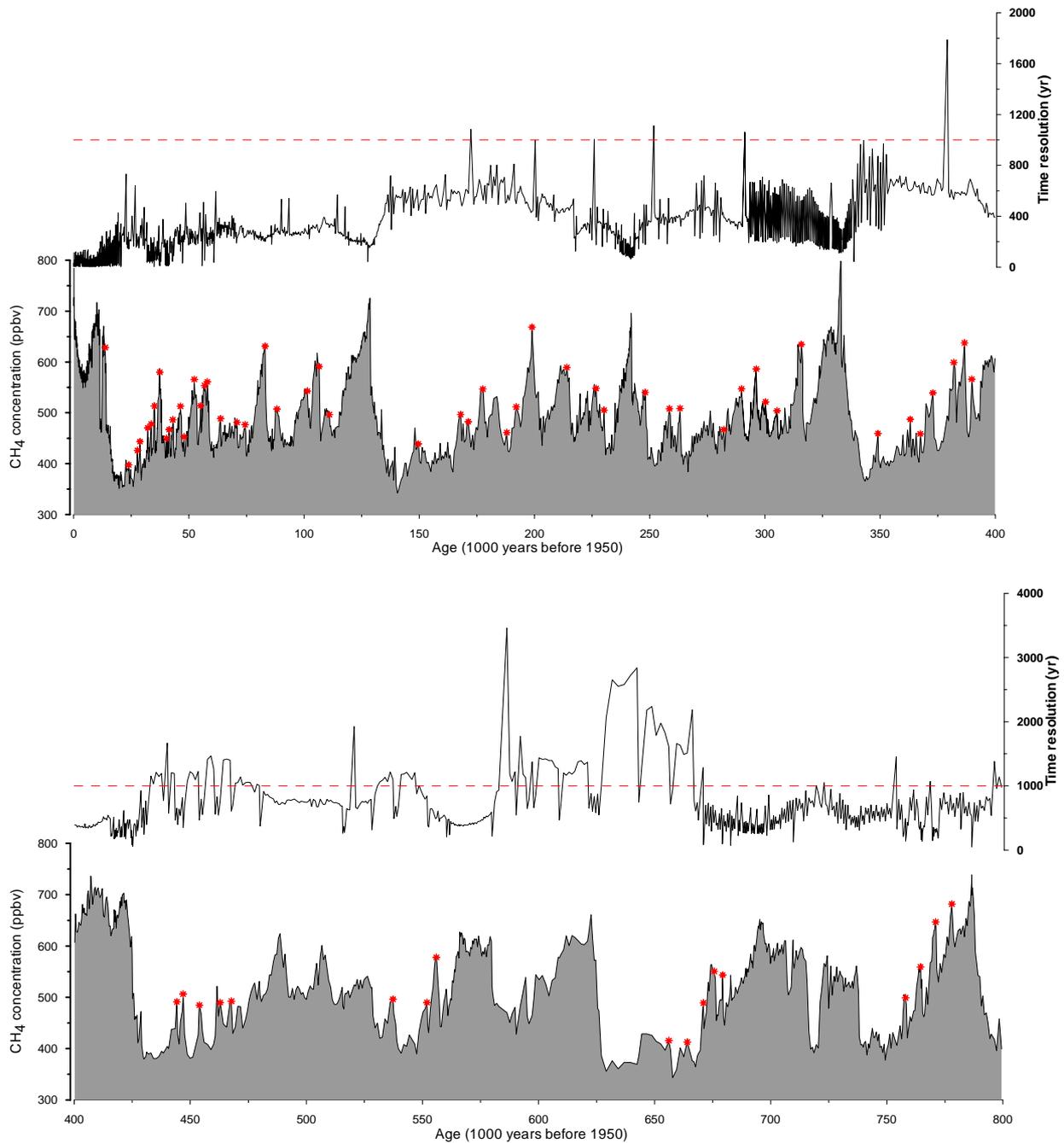
This is the classical method for spectral analysis. We have used it to determine the phase relationship between two sets of data, using two different filters to get the best coherence for any given frequency. For this method, we have resampled the different datasets, using a time resolution of 500 yr.

#### *Filtering*

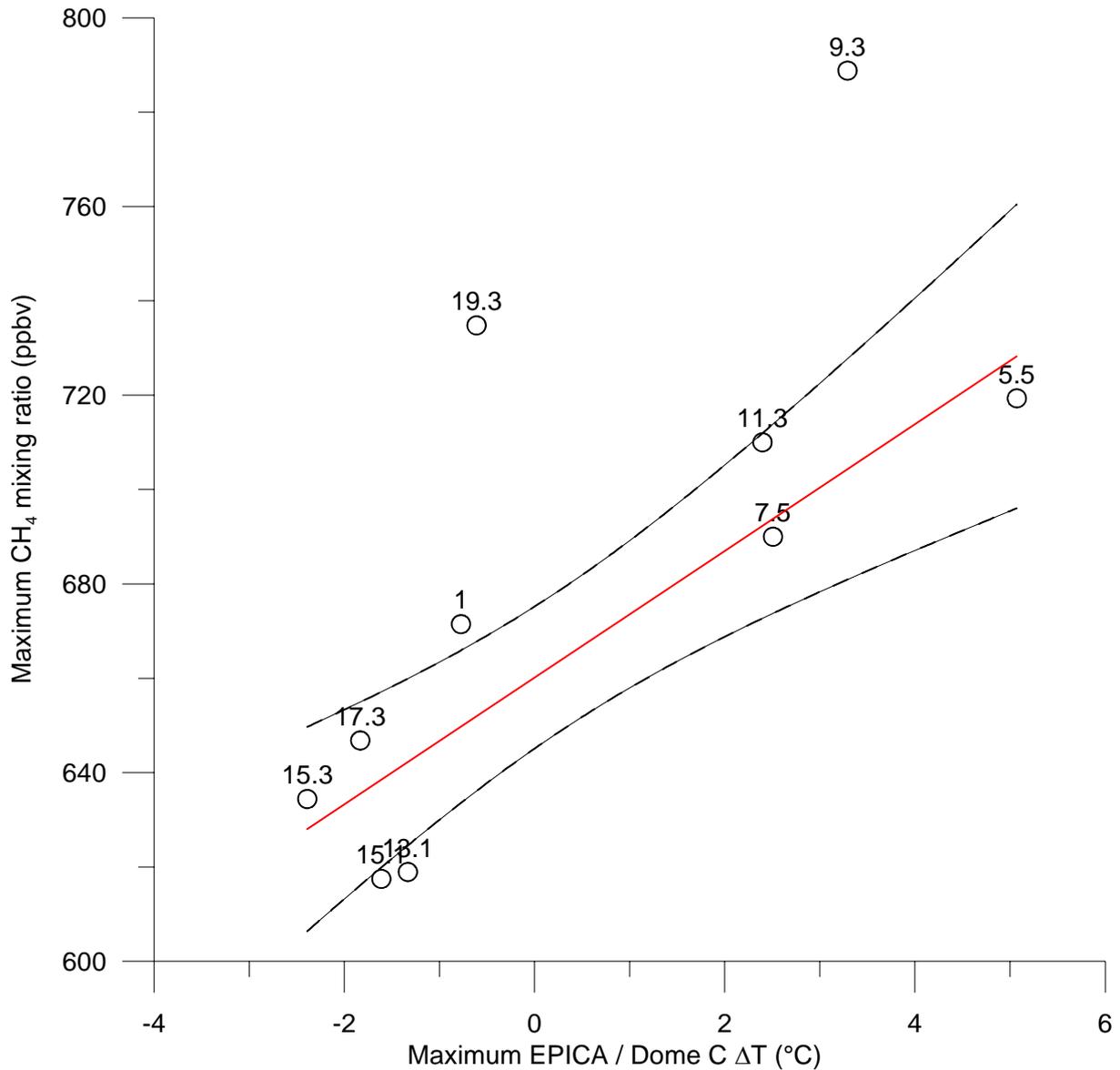
Filtering is performed using a band-pass Gaussian filter with frequency centre determined by the orbital parameter (Figure 2 with the description of frequency and width). The methane record is centered to perform such filtering.

**Figures:**

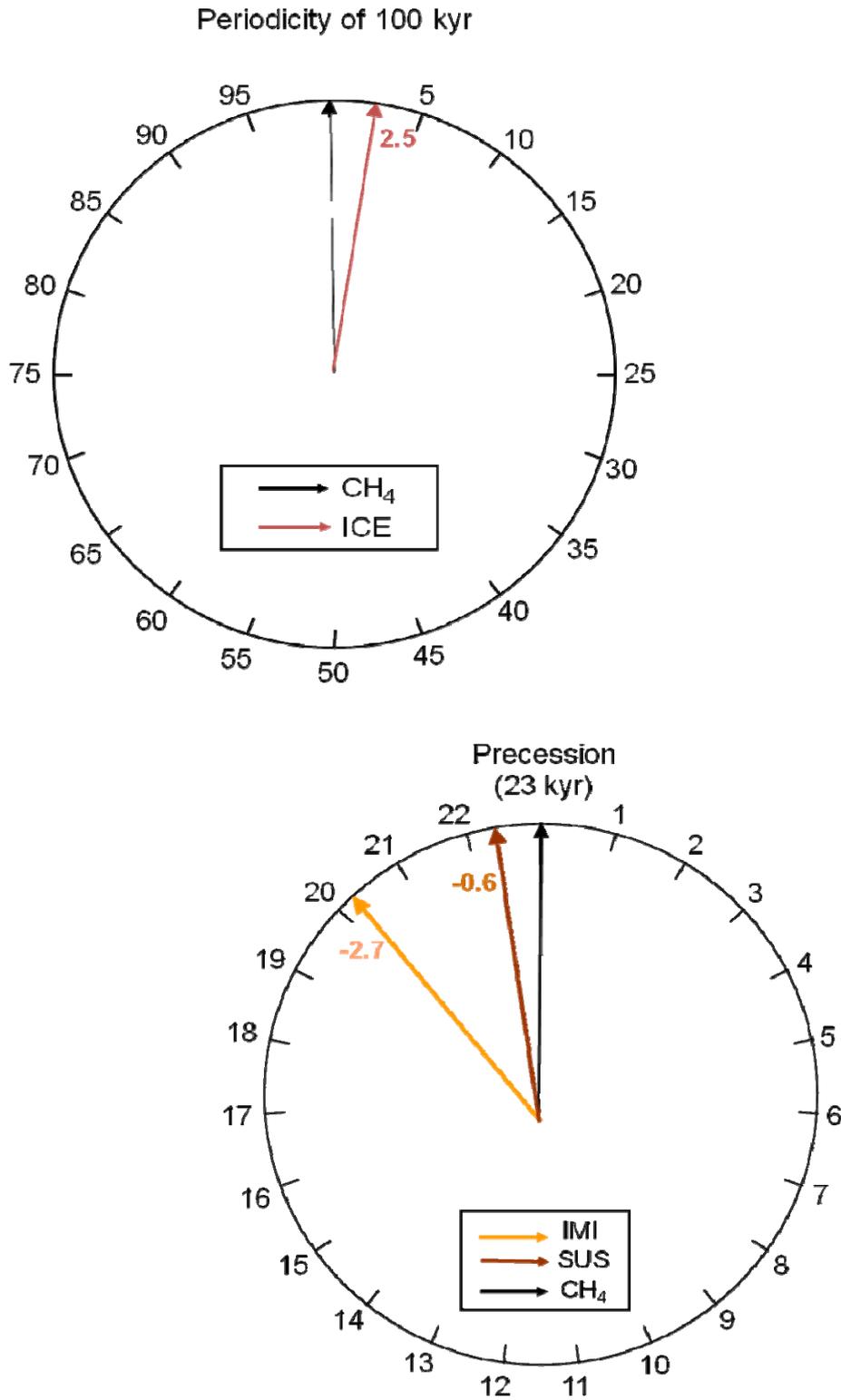
**Figure S1:** Methane values over the last 120 kyr from Greenland composite data<sup>36,37</sup> (in green, upper part), GISP 2<sup>38</sup> (in red, upper part) and EDC<sup>1</sup> (in black, bottom part). The grey shaded area in the upper right part highlights the section of the GISP2 record where stratigraphic disturbances complicate the interpretation (below 2750 m of depth). The blue and red stars locate the mid-transition of 22 Dansgaard-Oeschger events defined in the Greenland CH<sub>4</sub> record<sup>39,40,41</sup>. The records are on their own timescale. Dashed lines show the correspondence between Greenland and Antarctic CH<sub>4</sub> records. The mean difference of age of any given CH<sub>4</sub> event between EDC and the Greenland composite is 270 yr, and 432 yr for GISP2. The overlap between the Greenland composite and GISP 2, at the onset of DO 16 shows a shift of 300 yr.



**Figure S2:** Time resolution of the CH<sub>4</sub> record as a function of time, compared with the millennial-scale CH<sub>4</sub> variability defined from the amplitude of the CH<sub>4</sub> changes and their association with an Antarctic Isotope Maximum (with peak-to-peak synchronicity). The upper panel covers the time range 0-400 kyr and the lower panel 400-800 kyr. The dashed line corresponds to a 1000 year time resolution between two consecutive CH<sub>4</sub> samples.



**Figure S3** : Relationship between the maximum CH<sub>4</sub> mixing ratio during each of the last nine interglacials (two maxima are considered for MIS 15) and the concomitant maximum EDC temperature warmth<sup>34</sup>. The Marine Isotope Stage numbering is given for each interglacial. The linear regression (red line) does not include MIS 9 and MIS 19. The two black curves correspond to the 95% confidence interval of the regression.



**Figure S4:** Phase wheels showing the phase relationship between insolation, CH<sub>4</sub> and other proxies. The full circle covers one full period (time given in kyr). Calculations are performed using a Blackman-Tukey method with the Analyseries 2.0.3 program<sup>35</sup>

On the 100 kyr periodicity, the inverse of ice volume (ICE, directly deduced from the stack isotopic record of benthic foraminifera, LR04) show a lag of 2.5 kyr with respect to CH<sub>4</sub>. Note that modelling studies<sup>42</sup> show that areal changes of the Fennoscandian ice sheet lead by 2 to 4 kyr its volume changes.

On the 23 kyr periodicity, the magnetic susceptibility of Chinese Loess (SUS<sup>43</sup>) and CH<sub>4</sub> show a similar lag of 2.7 kyr (within 600 yr) on the monsoon index (IMI) of Leuschner and Sirocko (2003)<sup>44</sup>.

Phase wheel uncertainties between CH<sub>4</sub>, ICE, SUS and IMI depends on the dating uncertainties for each proxy. Parrenin et al.<sup>45</sup> indicate for instance a maximum error of 6 kyr when comparing the EDC3 timescale with LR04. Between 0 and 400 kyr, it varies between -1.5 and +3 kyr. The absolute dating uncertainties of the EDC3 timescale is 6 kyr<sup>45</sup>.

## References

29. Spahni, R. et al. Atmospheric Methane and Nitrous Oxide of the late Pleistocene from Antarctic ice cores. *Science* **310**, 1317-1321 (2005).
30. Chappellaz, J. et al. Changes in the atmospheric CH<sub>4</sub> gradient between Greenland and Antarctica during the Holocene. *Journal of Geophysical Research* **102**, 15987-15997 (1997).
31. Flückiger, J. et al. High resolution Holocene N<sub>2</sub>O ice core record and its relationship with CH<sub>4</sub> and N<sub>2</sub>O. *Global Biogeochemical Cycles* **16**, article no. 1010 (2002).

32. Flückiger, J. et al. N<sub>2</sub>O and CH<sub>4</sub> variations during the last glacial epoch: insight into global processes. *Global Biogeochemical Cycles* **16**, article no. GB1020 (2004).
33. Bellier, B. PhD manuscript ; Etude des variations du cycle du carbone au cours de l'holocène à partir de l'analyse couplée CO<sub>2</sub>-CH<sub>4</sub> piégés dans les glaces polaires. *Laboratoire de glaciologie et de géophysique de l'environnement* (Université Joseph Fourier-Grenoble 1, Spécialité : Sciences de la Terre et de l'Univers, Grenoble, 2004).
34. Jouzel, J. et al. Orbital and millennial antarctic climate variability over the past 800000 years. *Science* **317**, 793-796 (2007).
35. Paillard, D., Labeyrie, L. & Yiou, P. Macintosh program performs time-series analysis. *EOS Trans. AGU* **77**, 379 (1996).
36. Blunier, T. et al. Synchronization of ice core records via atmospheric gases. *Climate of the past* **3**, 325-330 (2007).
37. EPICA, C. M. One-to-one coupling of glacial climate variability in Greenland and Antarctica. *Nature* **444**, 195-198 (2006).
38. Brook, E. J., Sowers, T. & Orchardo, J. Rapid variations in atmospheric methane concentration during the past 110000 years. *Science* **273**, 1087-1091 (1996).
39. North Greenland, C. m. High resolution record of Northern Hemisphere climate extending into the last interglacial period. *Nature* **2805**, 1-5 (2004).
40. Dansgaard, W. et al. Evidence for general instability of past climate from a 250-kyr ice-core record. *Nature* **364**, 218-220 (1993).
41. Grootes, P., Stuiver, M., White, J. W. C., Johnsen, S. J. & Jouzel, J. Comparison of oxygen isotope records from the GISP2 and GRIP Greenland ice cores. *Nature* **366**, 552-555 (1993).

42. Peyaud, V., Ritz, C. & Krinner, G. Modelling the Early Weichselian Eurasian Ice Sheets: role of ice shelves and influence of ice-dammed lakes. *Climate of the past* **3**, 375-386 (2007).
43. Sun, Y. et al. East Asian monsoon variability over the last seven glacial cycles recorded by a loess sequence from the northwestern Chinese Loess Plateau. *Geochem. Geophys. Geosyst.* **7**, article no. Q12Q02 (2006).
44. Leuschner, D. C. & Sirocko, F. Orbital insolation forcing of the Indian Monsoon, a motor for global climate change. *Palaeogeography, Palaeoclimatology, Palaeoecology* **197**, 83-95 (2003).
45. Parrenin, F. et al. The EDC3 age scale for the EPICA Dome C ice core. *Climate of the past* **3**, 485-497 (2007).