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***Interactive comment on “High-resolution  
interpolar difference of atmospheric methane  
around the Last Glacial Maximum” by  
M. Baumgartner et al.***

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GENERAL COMMENTS

The ‘interpolar difference’ in the concentration of atmospheric methane (IPD) is a key observational constraint on the geographic distribution of methane sources, the value of which can be determined at various times in the past using a combination of Greenland and Antarctic ice-core records. This is a well written paper reporting new, high quality measurements (and measurement/sampling strategies) to determine the IPD at, and around, the Last Glacial Maximum (LGM) and it offers important new insights. Subject to addressing a few concerns (some substantive, but straightforward to ad-

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dress), I fully endorse its publication.

To my mind, the main finding is that, based on new high-resolution NGRIP and EDML measurements, the IPD between 21.2 and 21.9 kyr before present (BP)) was appreciable (3.7+/-0.7%), implying boreal wetlands were still active at the LGM. This contrasts with the findings of Dällenbach et al. (2000) based on GRIP and Byrd/Vostok measurements, which implied boreal wetlands were shutdown (and hence methane sources confined to the tropics) at the LGM. What makes the measurements reported here so convincing is:

1. Their high resolution, offering improved synchronisation of Greenland and Antarctic records.
2. The measurement/sampling strategies employed: both cores were analysed in the same laboratory, subject to the same standard gases, within the same space of time (with samples from both cores - randomised to avoid systematic drift - being analysed on each day of measurements).
3. The GRIP samples reanalysed in this laboratory (subject to the same standard gases etc) show generally good agreement with the new NGRIP measurements, implying the measurements by Dällenbach et al. (2000) were around 30 ppbv too low (relative to Byrd/Vostok measurements).

The only substantive concerns I have (on which I will expand below) are as follows:

1. Much is made of the relative IPD (rIPD - normalised to polar-mean methane concentration) being lower between 21 and 28 kyr BP than between 11 and 21 kyr BP. If I am honest, I am not convinced by this caricature of the rIPD measurements and (therefore) not convinced the measurements support the possible long-term influence on the rIPD of the northern/southern summer insolation ratio. To my mind, your observation that, with notable exceptions, the rIPD shows remarkable overall stability is the first-order message.

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2. There is no differentiation between boreal and (northern) tropical sources, preventing quantitative comparison of this study's findings re the distribution of methane sources with previous studies' (e.g. Fischer et al. (2008) re the LGM and Bock et al. (2010) re Dansgaard-Oeschger events 7 and 8).

This is otherwise an excellent piece of work and I very much look forward to reading the finished article.

Dällenbach et al. (2000), Changes in the atmospheric CH<sub>4</sub> gradient between Greenland and Antarctica during the last glacial and the transition to the Holocene, *Geophys. Res. Lett.*, 27, pp 1005–1008.

Fischer, H., et al. (2008), Changing boreal methane sources and constant biomass burning during the last termination, *Nature*, 452, 864–867.

Bock, M. et al. (2010), Hydrogen Isotopes Preclude Marine Hydrate CH<sub>4</sub> Emissions at the Onset of Dansgaard-Oeschger Events, *Science*, 328, 1686, doi: 10.1126/science.1187651.

## SPECIFIC COMMENTS

1. Looking at Figure 6, assuming the error bars represent plus or minus one standard deviation, I am not convinced that the rIPD is statistically significantly lower than 1.0 between 21 and 28 kyr BP, and it appears to be only barely significantly greater than 1.0 between 11 and 21 kyr BP.

Between 21 and 28 kyr BP, there are three points at which the rIPD significantly differs from 1.0: it is greater than 1.0 at DO2 and less than 1.0 at roughly 21.5 and 25.5 kyr BP; it is almost significantly(!) greater than 1.0 again at about 30 kyr BP. Between 11 and 21 kyr BP, there is strictly only one point at which rIPD is significantly greater than 1.0 – at about 16 kyr BP.

I therefore think your own summation (page 5485, line 8) that the rIPD is, with notable exceptions (e.g. the significant rise between about 21.5 and 19 kyr BP), overall remark-

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ably stable throughout the record is the main message. This in itself is an interesting finding - perhaps the result of compensating effects of shifts in wetland distribution and shifts in the ITCZ, as you describe.

So by all means include the arguments as to why we might expect the rIPD to be generally lower before 21 kyr BP and generally higher after, but I would encourage you to keep returning to what, to my mind, your measurements indicate – remarkable overall stability (with notable exceptions).

Similarly, looking at Figure 7, I am not convinced your measurements support the long-term influence on the rIPD of the northern/southern summer insolation ratio (Ins/Iss). There are data points that appear show a correlation with Ins/Iss (e.g. between 15 and 25 kyr BP) but there are as many that do not (e.g. between 5 and 15 kyr BP – not merely the YD – and at about 27 kyr BP).

So likewise, I would suggest discussing this possible influence, and noting the correlation between Ins/Iss and the rIPD calculated based on Singarayer et al.'s (2011) model calculations, but recognising that your measurements neither support this nor rule it out; given the superposition of other influences acting on shorter timescales, more measurements spanning a longer period are needed.

2. Constrained by the IPD, delta13CH4 and deltaD(CH4) – but I expect, mostly the IPD – Fischer et al. (2008) concluded (from numerous Monte-Carlo calculations) that boreal wetland emissions were almost entirely shutdown at the LGM (3-4 Tg CH4 per year c.f. 54 in the preboreal Holocene) whilst tropical wetlands were reduced by roughly 25-45% (75-130 c.f. 130-170 Tg CH4 per year).

They did so based on GRIP/EDML measurements suggesting a near-zero IPD at the LGM, similar to Dällenbach et al. (2000). It strikes me as crucial that, based on your NGRIP/EDML measurements showing an appreciable IPD at the LGM, you 'follow through' on what the implications are in terms of boreal and tropical emissions - if and how they differ from Fischer et al.'s (2008).

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I recognise the problem is underconstrained; you have only two poles of data so, in principal, can only apportion methane sources to two regions. However, both Fischer et al. (2008) and Bock et al. (2010), using a model with just two tropospheric boxes (northern and southern hemispheres) differentiated between boreal and tropical sources - presumably making certain assumptions.

Unless you are prevented from doing so by a lack of isotopic data, I strongly encourage you to state the necessary assumptions and, subject to those, present your best estimates of the relative strengths of boreal and tropical sources at the LGM. I also encourage you to do similarly for DO events 2, 3 and 4 for comparison with Bock et al.'s (2010) conclusions regarding DO events 7 and 8.

## SPECIFIC COMMENTS CONT.

Page 5472, lines 4-5 Strictly speaking, I don't think the IPD is an 'additional' constraint over and above Greenland and Antarctic methane concentrations, but it is a valuable product derived from these. I suggest replacing 'valuable additional parameter which allows to constrain' with simply 'valuable constraint on'; you can also remove 'the responsible'.

Page 5473, line 29 This is not correct. I would urge you to replace 'that the effect of BVOC is negligible' with 'the effect of changes in BVOC emissions to be all but negated by the effects of changes in air temperatures on humidities and gas-phase chemical kinetics'. You might also like to reference a very recent extension of our earlier work that reinforces this message:

Levine et al. (2012), Controls on the tropospheric oxidizing capacity during an idealized Dansgaard-Oeschger event, and their implications for the rapid rises in atmospheric methane during the last glacial period, *Geophys. Res. Lett.*, 39, L12805, doi:10.1029/2012GL051866.

Page 5476, lines 23-24 Same as for page 5472, lines 4-5 above.

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Figure 2 Does the size of the light blue circles (corresponding to GRIP reanalyses) reflect uncertainties in methane concentration and/or age? If not, I suggest these be made smaller to allow closer comparison with the NGRIP data they overlap.

Page 5483, lines 19-21 Unless I am missing something, rIPD does not strictly decrease with increasing atmospheric lifetime  $\tau$  as a result of increasing northern and southern polar methane concentrations  $c_n$  and  $c_s$ . rIPD decreases with increasing  $\tau$  because, for a given interhemispheric exchange time  $t_{ex}$ , the extent to which methane is mixed between hemispheres, and hence the extent to which its concentration is homogenised globally, increases with increasing  $\tau$ . You could replace these two sentences with:

‘For short exchanges times,  $t_{ex}$ , the IPD is not especially sensitive to the atmospheric lifetime,  $\tau$ . However, for a given value of  $t_{ex}$ , rIPD decreases with increasing  $\tau$  as the extent to which methane is mixed between hemispheres (and hence its concentration homogenised globally) increases.’

Page 5485, line 1-4 It is not just methane sources that influence  $\delta^{13}C_{CH_4}$ . Just as changes in the relative strengths of difference methane sources (with different isotopic signatures) affect  $\delta^{13}C_{CH_4}$ , so do changes in the relative strengths of different methane sinks (e.g. oxidation by atomic chlorine c.f. the hydroxyl radical) that show greater/lesser preference for reaction with, and hence removal of,  $^{12}CH_4$  over  $^{13}CH_4$ . I suggest you add the following right at the end of this sentence:

‘; besides changes in the relative strengths of different methane sources, changes in the relative strengths of different methane sinks (e.g. oxidation by atomic chlorine c.f. the hydroxyl radical) showing greater/lesser preference for removing  $^{12}CH_4$  over  $^{13}CH_4$  can also influence  $\delta^{13}C_{CH_4}$  (e.g. Allan et al., 2001; Levine et al., 2011).’

Allan et al. (2001), Active chlorine in the remote marine boundary layer: Modeling anomalous measurements of  $\delta^{13}C$  in methane, *Geophys. Res. Lett.*, 28 (17), pp 3239-3242.

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Levine et al. (2011), The role of atomic chlorine in glacial-interglacial changes in the carbon-13 content of atmospheric methane, *Geophys. Res. Lett.*, 38, L04801, doi:10.1029/2010GL046122.

Page 5488, lines 14-16 Although you subsequently note that the interpretation of  $\delta^{13}\text{CH}_4$  is 'not yet unambiguous', the phrasing 'which points to' implies a causal link. I would replace this with 'could point to' or similar; see comments above re Page 5485, line 1-4.

Page 5488, lines 18-19 Following on from the comments above, some further acknowledgement here should be made of the possible influences of biomass burning and methane sinks on  $\delta^{13}\text{CH}_4$ . You could for instance add the following right at the end of this sentence:

' , as could an increase in biomass burning – a particularly rich source of  $^{13}\text{CH}_4$  – or in principal an increase in the fraction of methane oxidised by atomic chlorine, which shows a particularly strong preference for removing  $^{12}\text{CH}_4$  over  $^{13}\text{CH}_4$  (see, e.g., Levine et al., 2011)'

See above for citation.

#### TECHNICAL CORRECTIONS

Page 5480, line 26 Move comma from just after 'both', to just before 'both'.

Page 5484, line 3 Replace 'on' at the beginning of this line with 'to'.

Page 5484, line 15 For clarity, I would add commas immediately after 'interpolar' and 'interhemispheric'.

Page 5485, line 12 Insert 'statistically significantly' between 'not' and 'different'.

Page 5486, line 8 Replace 'on' towards the end of this line with 'at'.

Page 5488, line 9 Remove comma after 'both'.

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Page 5489, line 7 Would it be accurate to add 'increase in' between 'the' and 'boreal'?

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