

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

Answers to J. G. Levine (Referee)

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 address), 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.
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 CH₄ 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 CH₄ Emissions at the Onset of Dansgaard-Oeschger Events, *Science*, 328, 1686, doi: 10.1126/science. 1187651.

Thank you very much for your detailed and constructive review comments, which have substantially improved the presentation of the material.

The two most important changes we applied to produce the revised version of the manuscript are the following:

- *Additionally to the already discussed two-box model run, we also performed a three-box model run (model from Chappellaz et al.,(1997)) to estimate the methane source distribution of the past. This allows to differentiate between boreal and tropical methane sources and to compare our two-box model run with an accepted model used in studies about the inter-polar difference of methane.*
- *We changed the structure of the discussion part to point out more clearly the stability of the relative inter-polar difference of methane (overall stability, with notable exceptions).*

The specific comments are answered in detail below.

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.

The assumption that the error bars represent plus or minus one standard deviation is correct. We agree that the variations in the rIPD are very small, especially, smaller than previously reported. Nonetheless we think it would be wrong to just ignore the small variations observed. We changed the titles of sections 5.1.1 (Lower rIPD state in the LGM) and 5.1.3 (Higher rIPD state during Termination 1) to just “rIPD around the LGM” and “rIPD during Termination 1”. This removes the weight on the high/low statements and allows to discuss the rIPD stability with notable exception, as you suggested below. Further, section 5.1.2 (‘rIPD increase around 21 kyrBP’) was integrated to section 5.1.1.

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.

Concerning the interval between 21 and 28 kyr BP we mentioned that DO2 is an exception of the low rIPD state and referred to section 5.1.4 (DO events). Further the point at 30 kyr BP, which is significantly greater than one was not in the considered interval between 21 and 28 kyr BP. We agree that between 11 and 21 kyr BP there is strictly only one point significantly greater than 1.0. However, we should not forget that this point stands for a rather long time interval and results from 94 CH4 measurements. Thus, we should be careful with the formulation ‘only one point’. Nonetheless, we agree that the concept with the low and high rIPD state was a bad choice and changed the discussion part of the manuscript according to your suggestion below.

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 remarkably 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.

Thank you for this very good suggestion. We changed the structure of the discussion accordingly (overall stability, with notable exceptions).

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).

In addition to the changes of the titles mentioned above, we have rewritten the first part of section 5 (Discussion). We replaced the lines 18-21 (page 5485) with: ‘To second order, we identify notable exceptions in the overall stability of the rIPD. The most outstanding feature is the low rIPD in the interval 21.9-21.2 kyr BP after DO event 2. Both the decrease in the rIPD from DO event 2 to this interval and the subsequent increase in the rIPD from this interval to the interval 20.4-17.8 kyr BP are statistically significant. The same is true for the increase in the rIPD from the interval 27.0-24.4 kyr BP to the DO event 2. In the following we discuss in detail the time span around the LGM, the Termination 1 (T1), and the DO events.’

Similarly, looking at Figure 7, I am not convinced your measurements support the longterm influence on the rIPD of the northern/southern summer insolation ratio (Ins/lss). There are data points that appear show a correlation with Ins/lss (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).

We are neither convinced about the existence of the long-term trend and we are not too conclusive in this (We do not mention anything about the long-term trend in the conclusion). But we still think it is worth to discuss it (motivated by the study of Singarayer et al., (2011)) and if a longer dataset comes up it should be analysed. We stated: ‘Nonetheless, if such a long-term trend exists in our data it is very weak and our new data set goes not far enough back in time to enable a comparison with the time scale of the precessional cycle.’

So likewise, I would suggest discussing this possible influence, and noting the correlation between Ins/lss 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.

We followed your suggestion and concluded the section on the long-term trend as the following: ‘In summary, our data neither support nor fully rule out a possible long-term influence of northern summer insolation on the rIPD. The limited range of our data set combined with the weak variation and the superimposed processes on millennial time scales do not allow for any conclusive remarks on this topic. High-resolution records produced in the way presented here from both poles, and over the whole last glacial cycle, are needed to address this question. The importance of the rIPD as a constraint for models is a strong motivation for future high-resolution measurements.’

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).

We added a detailed comparison with Fischer et al., (2008) to chapter 5.1.3.

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.

This issue is solved by the application of the three-box model from Chappellaz et al., (1997) (requested by other reviewer, L. Mitchell). The alternative run with this three-box model is compared to the two-box model run in the new chapter 4.2 and illustrated in the additional new Figure. With the three-box model, it is possible to quantitatively separate boreal and tropical sources. Note that in the three-box model, the southern source (30°S-90°S) is fixed at a constant level. This is necessary because the problem is underconstrained, as you mentioned correctly.

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.

The comparison of relative strengths of the boreal and tropical sources is now included (three-box model run). Further, concerning the conclusions about DO8, Bock et al, (2010) is cited. We did not run the four-box model from Fischer et al., (2008) to estimate the sources based on isotopic signatures. As you assumed correctly, there is currently a lack of isotopic data (DO event 2 and older).

For the data from Fischer et al., (2008), we added a qualitative discussion to section 5.1.3.

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'.

Changed.

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.

We apologise for this incorrect statement. We replaced it with the suggested sentence. We further added: 'Levine et al., (2012) confirm this statement also for DO events, and suggest that the concentration changes in CH4 are mainly source driven.'

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

Changed.

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.

The size of the light blue circles (GRIP remeasurements) neither reflects the uncertainty in methane concentration nor the uncertainty in the age. We made these symbols smaller, however, they are still bigger than the other symbols, since they represent the main point of Figure 2. We applied the same procedure to the TALDICE remeasurements (big yellow diamonds).

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.'

For a fixed s_n , s_s and t_{ex} , the IPD depends on tau like $\tau/(1+\tau)$, i.e. the IPD increases with increasing tau (equation 8). From equation 9 we know that the rIPD decreases with increasing tau. Taking both informations and looking at equation 2, it is the concentration that must increase to produce the decreasing rIPD, since it can not be the IPD, because the latter one increases with tau.

Nevertheless, we prefer your formulation, which is more directly related to equation 9 and easier to follow. We thus replaced the original sentences with your suggestion.

Page 5485, line 1-4 It is not just methane sources that influence $\delta^{13}CH_4$. Just as changes in the relative strengths of different methane sources (with different isotopic

signatures) affect $\delta^{13}\text{CH}_4$, 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, $^{12}\text{CH}_4$ over $^{13}\text{CH}_4$. 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 $^{12}\text{CH}_4$ over $^{13}\text{CH}_4$ can also influence $\delta^{13}\text{CH}_4$ (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 $\delta^{13}\text{C}$ in methane, *Geophys. Res. Lett.*, 28 (17), pp 3239-3242.

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.

We inserted the suggested sentence into the text.

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.

Due to changes in the discussion part, this sentence has been deleted.

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.

We inserted the suggested sentence into the text, but we replaced ‘increase in biomass burning’ with ‘decrease in biomass burning’.

TECHNICAL CORRECTIONS

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

Done.

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

Done.

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

Done.

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

Done.

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

Done.

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

Done.

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

Changed.