Reply to the referee comments of Sapart et al., CH_4 isotopic studies from firn air at 11 sites

We thank both reviewers for the relevant and constructive comments, which we used to improve the quality of the paper. Below is our point-by-point reply, but first a general comment.

Our paper has two major goals: 1) To investigate whether a consistent $\delta^{13}CH_4$ history over the last 50 years can be constructed by combining firn air measurements from multiple sites in Greenland and Antarctica. For this purpose, we use the only existing firn air model to date that allows reconstructing atmospheric time trends from isotopic data at a large number of firn drilling sites ("multi-site" inversions). 2) To highlight the uncertainties in isotope reconstructions from firn air when the atmospheric signal is of the same order as the firn fractionation effects, as in the case of $\delta^{13}CH_4$. Moreover, recent investigations in several laboratories including IMAU show that Krypton ions interfer with $\delta^{13}CH_4$ and this may likely be an additional cause of the discrepancies observed between the different firn datasets.

As the referee comments highlighted, several processes cannot be quantified within the state of the art on firn modeling (e.g. dispersive mixing and 3D transport). It is clearly beyond the scope of our manuscript to improve the physics included in existing firn models. The model used here has performed well in many previous studies (e.g. Wang et al., 2012, Buizert et al., 2012 and Witrant et al., 2012), and was compared to other state of the art models in (Buizert et al., 2012).

Considering the comments of the referees we clarified our main goal in the revised version of the paper and we propose a new title.

Reply to referee 1:

General comment:

1) This manuscript presents a synthesis and analysis of a large body of data produced over many years, with the aim of providing the best possible estimate of the time history of atmospheric methane carbon-13 over the past century. It is an impressive

body of work, on an important topic concerning global change and future greenhouse gas forcing. The writing is generally excellent and the modeling appears sound. However, I have some concerns about the fact that the model architecture that was used only contained molecular diffusion in the lock-in zone, and did not attempt to account for dispersive or non-fractionating transport within the deep layers of the lock-in zone (discussed in detail below):

Answer 1:

First, we would like to thank the referee for the positive general evaluation. To our knowledge dispersive or non-fractionating transport within the lock-in zone is a recently developed concept first introduced by the referee (Severinghaus et al., EPSL, 2010 (supplement)). Some previous versions of this model assumed negligible diffusion in the lock-in zone (e.g. Battle et al., 1996 ; Severinghaus and Battle, 2006). On the other hand several $\delta^{13}CH_4$ firn air studies emphasize the importance of diffusional fractionation (Trudinger et al., 1997; Francey et al., 1999), which is caused by molecular diffusion but not by dispersive mixing. We would like to mention that the differences between our results and previous $\delta^{13}CH_4$ firn air studies cannot be caused by the effect of dispersive transport on the lock-in zone because this new concept was not used in the earlier studies (Francey et al., 1999, Braunlich et al., 2001, Sowers, GBC, 2005). A recent study (Trudinger et al., 2012) further indicates that dispersive mixing formulated as a Deddy term in the lock-in zone is at least very difficult to constrain (Trudinger et al., 2012 ACPD, p17804 l9-28, p17806 l12-18). Finally, while being an interesting concept, to our knowledge, there is no direct evidence that dispersive mixing really occurs in firn. As stated in the general remark above, it is beyond the scope of our paper to improve physics in our firn model. The key feature of the model that we use here is the multi-site inversion capability. Dispersive transport is further discussed below.

2) I also wonder about the effect of three-dimensional transport within the lock-in zone, in which gases could migrate horizontally by molecular diffusion but move vertically through small cracks (essentially capillaries from the standpoint of physics) via non-fractionating processes such as viscous flow.

Answer 2:

All currently available models of gas transport in firn are 1D models. Building multi-dimensional models is certainly a potentially productive path for future science as it has long been known that horizontal layering in firn has some impact on e.g. air content in ice (Martinerie et al., 1992), however available models of trace gas transport in firn all use monotonous (smoothed) density profiles. A major difficulty in modeling firn heterogeneities is to evaluate the horizontal extent of potentially impermeable layers and more generally to constrain the 3D properties of firn with relevant observations. Moreover, over the last decade, several firn microstructure studies using 3D tomography imaging were published. They did not show evidence for the presence of small cracks or capillaries in the deep firn (Lomonaco et al., J Glaciol., 2011 ; Spaudling et al., J. Glaciol, 2011 ; Fujita et al., JGR, 2009 ; Freitag et al., J Glaciol, 2004).

It should also be noted that the deepest $\delta^{13}CH_4$ data-points of the lock-in zone have not be used in the final multi-site reconstructions. For these reasons, 3D transport in the lock-in zone is not included in our study

3) For all these reasons, I believe the manuscript should eventually be published, but only after a very thorough rethinking of the basic assumptions employed and a major revision that may well include changes to the model architecture including perhaps incorporation of new physics.

Answer 3:

As mentioned in the general reply above, improving the physics in the firn air model is beyond the goal of our paper. It has been shown in Buizert et al., 2012, Witrant et al., 2012 and Wang et al., 2012 that the firn transport model used here performs well compared to other models. In this paper, we focus on the consistency of datasets from multiple sites and on demonstrating the importance of the firn fractionation for $\delta^{13}CH_4$.

4) In a prior work that involved many of the same authors, Buizert et al. (2012) explored the rich data set of halocarbons and other trace gases obtained from the NEEM firn air experiments, to compare different firn gas transport models. One interesting new

observation was that slow-diffusing gases such as CFC-113 were more enriched than expected based on models that only incorporated molecular diffusion in the deepest layers of the firn near the bubble close-off zone (known widely in the literature as the lock-in zone). These slow-diffusing gases have up to a factor of three lower diffusivity than methane, which immediately suggests that the sensitivity of transport to their diffusivity could in principle be quite substantial.

Answer 4:

At looking carefully at Buizert et al., 2012, we did not find a mention that the LGGE-GIPSA model had biased results for CFC-113 in the lock-in zone (LIZ) compared to the 5 other models in this inter-comparison. The influence of the LIZ representation on model results is discussed in section 4.1 of Buizert et al., 2012. It concludes that "Models that reproduce the observations equally well can have completely different parameterizations, so our analysis does not tell us which scheme is more likely to be correct." Figure 3 in Buizert et al., 2012 does not show a stronger bias of LGGE-GIPSA model than other models for CFC-113 in the LIZ. In order to further investigate the issue of the role of eddy diffusivity in correctly representing the LIZ, additional calculations were performed of discrepancies between models and data using the same performance indicator as in Buizert et al., 2012, Equation 4: "RMSD", but we applied Equation 4 only to the lock-in zone (below 63m depth) (Table 1).

Table 1: "RMSD" (Buizert et al., 2012, Equation 4) in the lock-in zone for reference gases with largely differing D/Dco_2 values (CH_4: D/Dco_2 = 1.365, CO_2: D/Dco_2 = 1, SF_6: D/Dco_2 = 0.555, CFC-113: D/Dco_2 = 0.452) for the two NEEM 2008 firn air pumping operations ("EU" and "US"). The models of gas transport in firn in column one are those compared in Buizert et al. (2012).

Model	**CH_4**		**CO_2**		**SF_6**		**CFC-113**
	EU	**US**	**EU**	**US**	**EU**	**US**	**EU**
CIC	**0.9**	**0.7**	**0.9**	**0.7**	**0.6**	**0.6**	**0.5**
CSIRO	**0.7**	**1.5**	**1.6**	**1.8**	**0.4**	**0.8**	**0.7**
INSTAAR	**0.9**	**0.6**	**1.2**	**0.6**	**0.4**	**0.5**	**0.7**
LGGE-	**0.6**	**0.8**	**0.9**	**0.9**	**0.5**	**0.3**	**0.5**

GIPSA							
OSU	**0.8**	**0.7**	**2**	**1.2**	**0.8**	**0.9**	**1.1**
SIO	**0.7**	**0.6**	**1**	**0.9**	**0.4**	**0.5**	**0.9**

Compared to other models (Table 1), the LGGE-GIPSA model shows good performances in the lock-in zone, with excellent results for CFC-113.

Martinerie et al., 2009 used diffusivity profiles calculated by minimizing the model-data differences for CH_4 only for Devon Island, North GRIP, Berkner and Dronning Maud Land in order to simulate halocarbon data in firn together with "molecular only" diffusion. Results show no obvious sign of a D/Dco_2 dependent bias in the LIZ. Other studies also successfully modeled CFC-113 and low D/Dco_2 coefficient halogenated species without Deddy (e.g. Sturrock et al., JGR, 2002; Butler et al., 1999, Sturges et al., 2012, etc.).

5) Most of the models in the Buizert et al. study chose to alter their architecture to include a non-fractionating form of mixing or transport known as eddy diffusion or dispersion. This adjustment improved the fit of the model to the slow-diffusing gas data. The model used for the present manuscript, however, was not altered.

Answer 5:

No evidence showed that using Deddy in the lock-in zone was improving the fit of the slow-diffusing gas data (see Answer 4). The choice of not using eddy diffusivity in the lock-in zone in the LGGE-GIPSA model was made for three reasons:

➔ ***First, our results do not show a lower quality than other models, thus we did not see an obvious reason to modify the principle of adjusting a molecular diffusion coefficient down to the bottom of the firn used at LGGE since 1997 (Rommelaere et al., 1997).***

➔ ***Second, the eddy diffusion is defined as "a turbulent flow where the transport of momentum and energy can take place by a random motion of large turbulent eddies, or globs of fluid. This turbulent transport gives rise to effective values of viscosity and thermal conductivity defined as eddy viscosity and eddy thermal conductivity [...] much larger (typically 10 to 100 times larger) than the respective***

molecular values" (Anderson, 1991). Figure 5 in Buizert et al. (2012) indicates that there is no consensus on the values of Deddy/Dtotal to be used. Thus besides qualitative considerations, we do not find a real physical base to justify Deddy in the lock in zone.

➔ ***Third, no criterion exists to discriminate what part of the gas transport is due to molecular diffusion and what part is potentially due to dispersive mixing.***

6) Therefore it would be very relevant and helpful if the authors of the present manuscript would run a sensitivity test using a model that does include dispersion, to examine how important the neglect of dispersion is. Of course, diffusive fractionation is at the heart of the current manuscript, because of its profound influence on the isotope signals. The authors do mention at the end that their model may have produced a bias toward atmospheric histories with less change, due to this effect. But this is insufficient. Therefore I think it is fundamental and first-order to consider this issue before this manuscript can be considered for publication in ACP.

Answer 6:

We understand the point of the referee but respectfully disagree that such an exercise should be included in our study. The referee argues in terms of improving firn air modeling, which is an important issue, but our manuscript has a different focus (see general remarks above). The consideration of dispersion is a new concept that has not been clearly quantified yet, and there is no evidence that such processes actually occur in the firn. One major drawback in testing the impact of eddy diffusivity is that there is no consensus on the relative magnitude of Deddy versus Dtotal to be used (see Fig 5. and supplementary Fig.7 in Buizert et al., 2012). For example the SIO model uses a fixed Deddy/Dmolecular ratio of 27% (Buizert et al., 2012, section 4.1) whereas the CIC and OSU models progressively switch from purely molecular to purely eddy diffusion. Moreover, the CIC, INSTAAR and OSU models use different depth profiles of Deddy/Dtotal for the NEEM-EU and NEEM-US holes, thus the extrapolation to other drill sites is not obvious.

Therefore, carrying out such an exercise would require to develop a specific diffusivity optimization method and several months of computing time to re-evaluate diffusivity

for all boreholes. This could be a great scientific objective for a future firn modeling project, but it is not possible to do this within our study.

7) A less serious issue concerns the treatment of the two hemispheres in the global inversion, in which it was assumed that the Inter-Polar Gradient (IPG) in isotopes was unchanged with time. As detailed below, a more physically realistic approach would be to employ a simple two-box atmospheric model with mixing parameters derived from halocarbon and krypton-85 observations, so that northern and southern hemisphere sites could be inverted simultaneously. This seems like a way potentially to reduce the size of the error bars on the ultimate best-guess reconstruction of atmospheric δ13C(CH4).

Answer 7:

This is another interesting suggestion, but such a feature (coupling to a two-box model) is not included in any firn inversion model to date. We have considered including a free parameter for the IPG of $\delta^{13}C$ in the inversion. However, we decided against it because the IPG for $\delta^{13}CH_4$ is very small and temporal changes in the IPG are unfortunately not possible to quantify on this timescale because the lack of constraints (see recent paper by Kai et al. (Nature, 2011) and comment by Levin et al. (Nature, 2012)). Following comments of the two referees, the global (NH+SH) inversion has been removed.

8) One important point that is glossed over by the authors is that firn air studies do not sample the MEAN composition of air in the firn at any one particular depth. They sample only what is extracted from the open porosity, neglecting what resides in the bubbles and in macro-pores that are not necessarily bubbles but that have lost large-scale interconnectivity and so cannot be pumped in a firn air experiment. In this connection, it is relevant to reflect upon the fact that closed porosity is measured on hand samples of decimeter scale, which is small enough that many of these macro-pores may have been cut, such that they are recorded as "open porosity". In the deepest samples, the extractable air might only comprise 10% or less of the total air present at any one level. Thus it is important to consider the fact that the bubble records may be a better measure of the atmosphere, as bubbles sampled in aggregate by a typical large sample (such as

used in the Law Dome studies, 500 g) indeed do closely represent the MEAN composition of firn air at any one particular depth. Also, Severinghaus and Battle (2006) have shown that close-off fractionation can severely alter the composition of this remaining residual of air that is extracted in a firn air experiment, to a much larger degree than the mean bubble composition. The composition of this residual air can evolve quite cumulatively and substantially in a sort of Rayleigh-distillation process.

Answer 8:

To our knowledge, all models of gas transport in firn consider close and open porosity, no model uses a third type of porosity such as macro pores. Two specific processes producing isotopic fractionation in the LIZ are considered in some studies (Battle et al., 2011 and references therein): molecular size-dependent fractionation and fractionation due to pressure gradients. Severinghaus and Battle (2006) estimated that size-dependent fractionation is negligible for methane; pressure gradient fractionation is evaluated as ~0.015‰, one order of magnitude smaller than $\delta^{13}CH_4$ variations in the lock-in zone.

As explained in section 5.4 of the paper, the data points from the deepest firn were barely used to constrain the multi-site reconstructions, thus what occurred in the deepest part of the close-off zone cannot be the cause of the discrepancies between the different datasets. A sensitivity test excluding all data points in the LIZ (Supplementary Section 4 and Figure S4) further shows consistent results with our main multi-site scenarios.

9) It is also not inconceiveable that there is some isotopic fractionation of 13C(CH4) during this close-off process. Battle et al. (2011) have in fact identified isotope fractionation of epsilon = 5 per mil in 18O of O2 in WAIS Divide firn air, during the close-off fractionation process. Can the authors safely disregard such a fractionation process? To do so seems hazardous, absent better information. One consolation is that methane is a larger molecule than O2 and as such might be expected to be immune to the isotopic effects of close-off fractionation, somewhat like argon appears to be (Battle et al., 2011, Controls on the movement and composition of firn air at the West Antarctic Ice Sheet Divide. Atmos. Chem. Phys. 11, 18633-18675. doi:10.5194/acp-11-11007-2011).

Answer 9:

In their Section 9.1, Battle et al. (2011) attribute the anomalous isotopic fractionation of O_2 to a molecular-size related permeation through ice. They demonstrate that other atoms/molecules such as N_2, Kr and Xe are not affected. The molecular size of methane being higher than the one of Krypton (Severinghaus and Battle, 2006), methane should also be unaffected. We did not find a discussion of close-off fractionation for methane in Battle et al. (2011).

The consistency of $\delta^{13}CH_4$ measurements performed on the Law Dome ice cores, firn air and the Cape Grim air archive also argues against a significant methane isotopic fractionation due to permeation through ice (Ferretti et al., 2005, Supplement, Ice core integrity Section).

10) Finally, this manuscript and the problem it treats has much in common with the study of 13C in atmospheric CO2. Similar problems have plagued this isotopologue in firn air studies. It seems important that the present authors read and incorporate any lessons learned from firn air studies of 13CO2. Specifically I recall that South Pole firn air 13CO2 was never successfully reconciled with the bubble record from Law Dome and the Cape Grim archive record. It seems likely that there are fundamental problems with the treatment of diffusive fractionation in firn air, especially in the deep layers of the firn where only a small fraction of the total porosity is open porosity and the firn air that is extracted represents a small residual that is dwarfed by the amount of air contained in bubbles around it. The work of Francey, Trudinger, Bender, Etheridge and others comes to mind. It seems to me that a paper that only focuses on 13C(CH4) is missing part of the story. It is highly likely that the same model problems apply to both species.

Answer 10:

We note again that the goal of our manuscript is not a model evaluation, but a reconstruction of $\delta^{13}CH_4$ using data from multiple sites.

We are not aware of a published study about $\delta^{13}CO_2$ only in South Pole firn air. To our knowledge, only Trudinger et al., (1997) considered both $\delta^{13}CH_4$ and $\delta^{13}CO_2$ and more recent work led by Mauro Rubino (paper in preparation) suggests that there is no significant difference that might be attributed to dispersion in the lock-in zone.

It is important to note that the magnitude of diffusional fractionation is much larger for $\delta^{13}CH_4$ than for $\delta^{13}CO_2$. Francey et al. (1999) pointed out that the correction for $\delta^{13}CH_4$ is about 10 times larger than the correction for $\delta^{13}CO_2$ at DE08-2. Because of the relatively larger mass difference between the $^{13}CH_4$ and $^{12}CH_4$ molecules, the diffusion coefficients of $^{13}CH_4$ and $^{12}CH_4$ are more different than those of $^{13}CO_2$ and $^{12}CO_2$ (see e.g. Supplementary Table 5 in Buizert et al., 2012).

We performed simulations of $\delta^{13}CO_2$ at DE08-2 with the LGGE-GIPSA model (Figure 1). They show a good consistency with the CSIRO model results and the atmospheric scenario from Francey et al. (1999b) based on the Cape Grim air archive for its recent part.

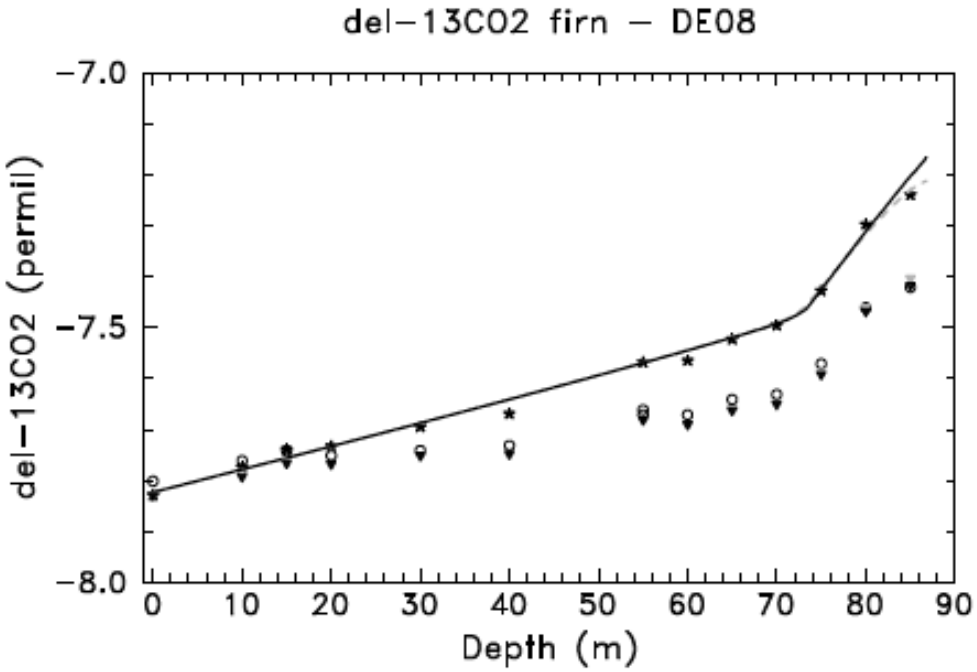

Figure 1: Simulations of $\delta^{13}CO_2$ in the DE08-2 firn. Stars: $\delta^{13}CO_2$ measurements in the DE08-2 firn (D. Etheridge, private communication, 2012). Open circles: DE08-2 firn data corrected for the effect of diffusional fractionation with the CSIRO model (Francey et al., 1999b), triangles: DE08-2 firn data corrected for the effect of diffusional fractionation with the LGGE-GIPSA model. Lines: LGGE-GIPSA forward model results using as input a spline fit to Cape Grim air archive and Law Dome ice core data (Francey et al., 1999b and corresponding file at Australian Antarctic Data Centre), shifted by -0.037‰ to account for the Cape Grim to DE08 gradient (Trudinger et al., 1997). Black lines/symbols show the results

using the diffusivity constrained with all DE08-2 reference gas data, the grey symbols show the results using the diffusivity constrained without the deepest CO_2 data point.

11) I would like the authors to give serious thought to all of these issues, and I do need to see the manuscript again before it can be considered publishable. The authors should prepare a detailed response to this review, addressing each point in turn and justifying in a convincing way the choices made, including the editorial points raised below.

Answer 11:

We would like the referee and the editor to acknowledge that our study is an experimentally motivated work, where the aim was to combine measurements at a large number of sites in a multisite inversion using the only model worldwide that has such a capacity at the moment.

We have thought and discussed about the general points raised by the referee in great depth, but come to the conclusion that many of the points that the referee raises can only be answered by a future dedicated project that aims at studying the physics incorporated in firn air models in detail. An important challenge for a future modeling study aiming at providing new constraints on deep firn physics will be to find an objective criterion allowing quantifying the ratio of molecular diffusion versus eddy dispersion in the lock-in zone. It would therefore have a very different focus than the project that we have carried out and wish to publish in the present manuscript.

Other points to be addressed individually:

We thank the referee for his help in improving the writing style of our manuscript.

Pg 9589, Line 4 "…helps IN reconstructing…"

This has been corrected in the revised manuscript.

Pg 9590, line 15 "…REFERENCES therein)"

This has been corrected in the revised manuscript.

Pg 9590. Line 20 "…stable isotope RATIO measurements…"

This has been corrected in the revised manuscript.

Pg 9590, line 24 "…anthropogenic 13C-enriched…"

This has been corrected in the revised manuscript.

Pg 9591, line 3 At some point in this discussion, it should be pointed out that the expectation is that all sites should agree, because the atmosphere is well-mixed on relevant timescales. This is an implicit assumption in this discussion, which should be made explicit. [Actually, the validity of this assumption is perhaps not immediately as clear as one would like. Is it really true that local CH4 sources upwind of Greenland do not affect the firn air record? I certainly would doubt it, but it perhaps needs some justification to disregard this possibility. You could cite the recent airborne trace gas sampling campaign HIPPO as justification, for example, that mid-troposphere air at 70 N is well-mixed and not sensitive to local sources.]

One sentence has been added to clarify this point at the end of the introduction.

Pg 9591, line 4 "math/mismatch" seems a bit unclear. Perhaps you could find a better word? Perhaps "..mathematical aspects of the mismatch of these firn air results…"? I'm not sure exactly what you are trying to say here.

The word "consistency" in place of match/mismatch is used in the revised manuscript.

Pg 9592, line 3 "…trace GAS samples…"

This has been corrected in the revised manuscript.

Pg 9592, line 18 "…which STRONGLY reduce…"

This has been corrected in the revised manuscript.

Pg 9593, line 4 Witrant et al., (2011) is cited here. My understanding is that this is a Discussion paper, and did not pass peer review. Under present editorial policy of ACP, Discussion papers are fully citable. This is perhaps a philosophical question, but I wonder if these kind of citations, to literature that has not passed the peer review process, can be potentially misleading to readers? Isn't there a danger that the casual reader (which means most of us these days, in practice, since we are all so short of time) will misunderstand and come to view these papers as equivalent to published works? One possible remedy might be to include some sort of additional information, such as "Witrant et al. (Discussion, 2011)". This is stylistically somewhat analogous to the present use of "(E. Witrant, personal communication, 2011)", but of course with the vast advantage that readers can easily download the information in question. This situation has arisen because of the computer age, and perhaps our time-honored traditions of citation and scholarship have yet to fully catch up to the new technological situation and

its wonderful advantages.

Witrant et al., (2012) has in the meantime been accepted for publication in ACP. The model physics and model results used here (diffusivity profiles) are the same in the ACPD and ACP articles.

Pg 9593, line 11 "…is not ONLY affected by the micro-structure…but is ALSO related to…" Surely you do not believe that micro-structure has no impact on tortuosity? It is well known that clay particles (i.e. plate-like particles) create a porous media with very much higher tortuosity than do spheres. In the particular case of firn, wind-packed snowflakes near the surface can have a much higher tortuosity than the porous medium containing rounded, quasi-spherical firn grains found at 5 meters depth. Furthermore, tortuosity is affected by the prevalence of "dead-end" or "cul-de-sac" pores, because these cause the average path length of molecule transport to increase due to "detours".

This sentence refers to the second paragraph of Section 4 in Fabre et al. (2000). As firn micro-structure is not the main subject of our manuscript, this sentence was removed.

Pg 9593, line 24 "…Green's function…" for background see http://en.wikipedia.org/wiki/Green%27s_function

This typo has been corrected in the revised manuscript.

Pg 9593, line 26 "…to calculate the probability OF HAVING air of a certain age.."

This has been corrected in the revised manuscript.

Pg 9595, line 4 "THE DC and SPO-95 based SCENARIOS are flatter…"

This has been corrected in the revised manuscript.

Pg 9595, line 20 "…even in THE absence of A …trend…"

This has been corrected in the revised manuscript.

Pg 9595, line 23 "… using a firn model RUN IN A forward mode…"

This has been corrected in the revised manuscript.

Pg 9595, line 24 "…based on THE NOAA-ESRL…"

This has been corrected in the revised manuscript.

Pg 9595, lines 24-27 This sentence is too long and too confusing. Chop it in half perhaps? It seems that what you are trying to say is that the NOAA-ESRL methane concentration histories were used to isolate and model the effects of diffusion

fractionation, with constant isotopic ratios in the atmosphere. As pointed out earlier, this calculation must be done separately for 12CH4 and 13CH4, and this was done in this case. It is not made clear in this sentence why Buizert et al 2011 is needed for the northern hemisphere, and Witrant et al., 2011 for the southern hemisphere. Are the concentration histories different in these works, from the NOAA-ESRL histories? Or did you just adopt the run results from these works, rather than running new experiments? Overall clarification is needed.

Witrant et al. (2012) is used for the SH scenario, because Buizert et al.,(2012) only provides a NH scenario. This sentence has been divided in 2 to clarify the main message.

Pg 9597, line 6 "..only PROVIDED AN a posteriori CONSTRAINT ON THE DIFFUSIVITIES DERIVED FROM CO2."

This has been corrected in the revised manuscript.

Pg 9598, line 1 "…leads to a SMALLER fractionation…"

This has been corrected in the revised manuscript.

Pg 9598, line 2 Eliminate redundant parenthetical expression

This has been corrected in the revised manuscript.

Pg 9598, line 4 add a comma after "NM-09"

This has been corrected in the revised manuscript.

Pg 9598, line 10 high accumulation sites have thicker, not thinner diffusive column heights, all else being equal. Please revise this sentence. Perhaps the key point is the faster bubble trapping, which precludes an extended period of molecular diffusion in the highly tortuous lock-in zone, during which time the fractionation occurs.

This has been corrected in the revised manuscript. "Due to a thinner diffusive column height and" was removed.

Pg 9598, line 17 "…at DML, DESPITE THE FACT THAT it belongs to the…"

This has been corrected in the revised manuscript.

Pg 9599, line 10. You need to cite Buizert et al. 2011 in connection with this statement. Something like this would be appropriate: "…are strongly dependent on the diffusivity profile used, SIMILAR TO THE FINDING OF BUIZERT ET AL. (2011)."

This has been corrected in the revised manuscript.

Pg 9599, line 12 insert commas before and after the phrase “at least in the deep firn”

This has been corrected in the revised manuscript.

Pg 9599, line 16 “…leads SOMETIMES TO inconsistent…”

This has been corrected in the revised manuscript.

Pg 9601, line 3 “…no discrete age EXISTS for a given firn air sample.”

This has been corrected in the revised manuscript.

Pg 9601, line 15 It is not very physically realistic for the two hemispheres to have independent atmospheric histories. Would it not be more physically realistic to use a simple two-box atmospheric model to perform your global multiple-site inversion? Because the methane residence time is roughly 8 times longer than the interhemispheric mixing time, it is very difficult for the two hemispheres to have radically different histories. Rather, the well-known atmospheric interhemispheric mixing parameters (based on halocarbon and krypton-85 observations) can be used with high confidence to force the two sets of results to be consistent with each other within the known constraints, taking advantage of the accurate methane concentration records that exist for the separate hemispheres. It seems likely that your ultimate error bars would be reduced by this approach because it brings additional constraints to bear.

As mentioned earlier, combining a two-box atmospheric model with the multiple-site inverse firn model would be a very complex model development. As stated in the next sentence in our manuscript, the NH and SH scenarios agree within the uncertainty envelopes. See also our answer to comment 7 above: uncertainties are a major limitation in estimating the inter-polar gradient and its time variations.

Pg 9601, line 20 “…envelopes, the shape of the BEST ESTIMATE SCENARIO IS SLIGHTLY DIFFERENT for the two hemispheres…”

This has been corrected in the revised manuscript.

Pg 9602 line 1 It is not really clear from this discussion why it is a reasonable assumption that the IPG is fixed in time. Certainly, the fact that methane concentration trends flattened in the 1990s, suggests that one should not expect the IPG in isotopes to be constant in time. Perhaps the use of a two box model would be more appropriate?

The IPG for $\delta^{13}CH_4$ is very small and temporal changes in the IPG are unfortunately not possible to quantify because of a lack of constraints. So making the IPG change

over time would not reduce error bars on the final multisite scenario. Following comments of the two referees, the global (NH+SH) inversion has been removed.

Pg 9604 line 18 "…constrained by FEWER species."

This sentence has been removed in relation with a comment from Referee 2.

Pg 9605 line 8 "…and in ice BUBBLES…"

This has been corrected in the revised manuscript.

Pg 9605 line 11 "…conclusions OBTAINABLE from the existing.."

This sentence has been removed in relation with a comment from Referee 2.

Pg 9606 line 8 "…how WELL the diffusivity profiles…"

This has been corrected in the revised manuscript.

Pg 9606 line 17 "…models ARE important as well."

This sentence has been removed in relation with a comment from Referee 2.

Pg 9607 line 1 "We THANK the team…"

This has been corrected in the revised manuscript.

Pg 9607 line 14 "…participants IN the field work.."

This has been corrected in the revised manuscript.

Pg 9612 line 7 "…depth at which the open/total porosity RATIO becomes…"

This has been corrected in the revised manuscript.

Pg 9615 line 1 (figure caption) "…separation ON 13CH4 fractionation."

This has been corrected in the revised manuscript.

Supplementary online information: 3. Sensitivity to the regularization term (page 3) The last word should be CONSTRAINED not constrained.

This has been corrected in the revised SI.

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