Response to reviewer 3:

All of the reviewers provided excellent suggestions and feedback on the paper, and we think that by addressing their concerns the paper will be greatly improved. Many of their comments were complementary. Therefore we will first summarize the major revisions we plan to make to the paper before responding to each reviewer in detail:

Planned Major Revisions
1) We have revised the δ^2H-CH_4 dataset in response to comments from Reviewer 1 and Reviewer 3. (i) For peatland sites with depth stratified sampling we have decided to only include samples from the upper 50 cm, as suggested by reviewer 3, since this is the depth range that is most likely to emit CH_4 to the atmosphere. This affects a total of 8 sites. (ii) Reviewer 1 noted that an outlier sample from the Amazon River with very high δ^2H-CH_4 and δ^{13}C-CH_4 could be derived from thermogenic methane. We agree that this outlier is suspect, and therefore have decided not to include it. (iii) We also noted that one site (Mirror Lake, Florida, USA) was analyzed in two separate studies, and therefore was included twice in the dataset. We have combined the data from the two studies into one site entry.

2) As suggested by all three reviewers, we have performed much more rigorous analysis of the relationship between measured and modeled δ^2H-H_2O values. Specifically we have done the following: (i) In addition to annual precipitation δ^2H values, we now also analyze growing season precipitation δ^2H, which is defined as the amount-weighted mean δ^2H of months with mean temperature greater than 0º C. This provides an opportunity to assess whether seasonal variation in precipitation in the mid to high-latitudes is important in controlling the environmental δ^2H-H_2O value; (ii) separately analyzing inland water and wetland environments, since these are different hydrological environments and the controls on δ^2H-H_2O are potentially different.

This analysis led to the following key results (see Table R1 below for summary of results): A) growing season modeled precipitation δ^2H is a better predictor of inland water δ^2H-H_2O than annual precipitation δ^2H, in that the regression curve is indistinguishable from the 1:1 line. B) annual modeled precipitation δ^2H is a better predictor of wetland δ^2H-H_2O, in that the slope of the regression is indistinguishable from 1, and the R^2 value is higher. However, the regression line is offset from the 1:1 line by 18.6±9‰. We interpret this as an indicator of likely widespread evaporative effects on δ^2H-H_2O in wetland environments.

We use these results to then develop a 'best estimate' for comparing δ^2H-H_2O with d2H-CH4. (i) For sites with measured δ^2H-H_2O values we use the measured value. (ii) For inland water sites without measured δ^2H-H_2O we use modeled growing season precipitation, since as discussed above the regression of this against measured δ^2H-H_2O is indistinguishable from the 1:1 line. (iii) For wetland sites without measured δ^2H-H_2O we estimate the δ^2H-H_2O using the regression relationship with annual precipitation δ^2H-H_2O shown in Table R2. We feel this approach combining measured and modeled data is

most consistent with that of Waldron et al., 1999, who we note also analyzed a combination of sites with measured δ^2H-H_2O (29 out of 51 sites) and estimated δ^2H-H_2O based on precipitation isotopoic measurements (22 out of 51 sites).

Table R1: Comparison of regression relationships between modeled δ^2H_p and measured δ^2H-H_2O

	Slope	Intercept	R^2	RMSE	p	n
Inland waters						
Growing season δ^2H_p	1.05±0.09	-0.3±8	0.82	22.3	4.81E-13	33
Annual δ^2H_p	0.85±0.06	-2.1±7	0.84	20.5	3.17E-14	33
Wetlands						
Growing season δ^2H_p	1.24±0.09	14.8±10	0.87	16.5	4.46E-13	28
Annual δ^2H_p	1.057±0.08	18.6±9	0.88	15.7	1.20E-13	28

3). As suggested by all three reviewers, it is important to consider the effects of modeled δ^2H-H_2O on the regression between δ^2H-H_2O and δ^2H-CH_4. To do this carefully we performed the regression analysis using four different estimates of δ^2H-H_2O:
(i) the 'best-estimate' of δ^2H-H_2O as described above in Planned Major Revision 2; (ii) measured δ^2H-H_2O, only analyzing sites with this measurement; (iii) modeled annual precipitation δ^2H; and (iv) modeled growing season precipitation δ^2H. We think it is valuable to continue to include the regression relationships for modeled precipitation because these relationships could be used in future studies using Earth Systems Models to predict the distribution of δ^2H-CH_4. For each of these cases we analyzed all sites, inland waters, and wetlands. We also compare these relationships with those of Waldron et al., (1999), both for the total dataset in that study, and for the dataset that only includes sites with measurements of δ^2H-H_2O (29 out of 51 sites). A summary of the results of this analysis are shown in Table R2 below.

A key point is that we have decided to use unweighted, as opposed to weighted, regression. Comments by Reviewer 1 made us realize that weighting by standard error was causing a few sites to strongly bias the regression results. Statistical research has found that for environmental data with poorly constrained error variance unweighted regression is frequently less biased than weighted regression (Fletcher and Dixon, 2012). Using a statistical test proposed by that study we find that unweighted regression is a good choice for our dataset. Note that in Table R2 we apply unweighted regression to the dataset of Waldron et al., (1999), in part because the specific weighting methodology was not specified in that study. This produces a small difference in the regression relationship shown in Table R2 with that reported by Waldron et al., (1999), but the two regression relationships are within error.

We then used analysis of covariance (ANCOVA) to examine differences between the regression relationships shown in the table. Based on a multiple comparison test, none of

the regression relationships shown in Table R2 are significantly different from one another. Therefore we conclude that (i) using modeled δ^2H-H_2O does not have a significant effect on the estimate of the relationship between δ^2H-H_2O vs δ^2H-CH_4; (ii) Differences in the slope of this relationship between inland waters and wetland sites are not conclusive; and (iii) that since all of the regression relationships using the larger dataset produce a flatter slope than that of Waldron et al., (1999), the true global slope is likely to be flatter than inferred in that study, but confirmation of this flatter global slope will require more data and further analysis.

Table R2: Comparison of regression relationships between δ^2H-H_2O and δ^2H-CH_4 using different estimates of δ^2H-H_2O

	Slope	Intercept	R^2	RMSE	p	n
Best Estimate δ^2H-H_2O						
All	0.44±0.05	-298±5	0.42	27.4	1.33E-16	129
Wetlands	0.51±0.06	-300±5	0.59	23.7	7.85E-12	55
Inland Waters	0.42±0.07	-295±7	0.34	29.1	6.72E-08	74
Measured δ^2H-H_2O						
All	0.5±0.08	-292±8	0.43	28.6	1.22E-08	61
Wetlands	0.53±0.11	-298±13	0.44	26.2	6.58E-05	28
Inland Waters	0.42±0.1	-291±10	0.37	28.9	0.000156	33
Modeled Annual δ^2H_p						
All	0.42±0.04	-293±5	0.44	26.9	1.17E-17	129
Wetlands	0.57±0.06	-287±6	0.65	21.9	1.01E-13	55
Inland Waters	0.37±0.06	-293±7	0.36	28.4	1.25E-08	74
Modeled Growing Season δ^2H_p						
All	0.51±0.05	-292±5	0.41	27.6	2.55E-16	129
Wetlands	0.71±0.07	-285±6	0.63	22.4	4.05E-13	55
Inland Waters	0.44±0.07	-294±8	0.33	29.3	1.03E-07	74
Waldron et al. (1999)						
All data	0.74±0.1	-284±6	0.5	26.3	6.56E-09	51
Measured δ^2H-H_2O						
only	0.79±0.2	-279±10	0.44	29.6	9.21E-05	29

4) We then used the 'best-estimate' δ^2H-H_2O values and the regression based on those values, shown in Table R2, to calculate a revised δ^2H-$CH_{4,w0}$ value for each site. These

analyses were then applied in the subsequent analyses in the paper shown in Figures 5,8 and 9. We also calculated an alternate value for sites with measured δ^2H-H_2O, using the values and regression curve for those sites.

Notably, for the comparison between δ^2H-$CH_{4,w0}$ and α_C we have found that there continues to be evidence for a segmented linear relationship. However, the breakpoint of this relationship is not consistent when analyzing all sites or only sites with measured δ^2H-H_2O. Furthermore, the regression relationships for the two components of the segmented linear relationship were weaker than in our original analysis, and were not consistently statistically significant. Therefore in our revised analysis we will place less emphasis on this result, and less emphasis on the relationship between methanogenic pathway and δ^2H-CH_4 generally, as suggested by reviewer 1. Instead we will discuss four processes or variables that have the potential to influence δ^2H-CH_4 in freshwater environments: (i) differences in methanogenic pathway, including possible use of methanol as a substrate; (ii) methane oxidation; (iii) isotopic fractionation due to diffusion; and (iv) differential thermodynamic favorability of methanogenesis, or differential enzymatic reversibility. Ultimately, our conclusion is that δ^{13}C-CH_4 or α_C cannot fully resolve the effects of these processes on δ^2H-CH_4 on a global basis, and other approaches will be necessary to determine their relative importance, or the possible importance of other processes.

We will continue to present these results in Figure 5, given that we feel it is important to show co-variation, or lack thereof, between these isotopic measurements. Given the findings mentioned above, we will substantially revise Figure 6. Instead of distinguishing samples by inferred methanogenic pathway in this figure, we will distinguish samples by environment (wetland vs inland water), and also show available data for cow rumen and landfills. We may reverse the order of Figures 5 and 6.

5) Reviewer 2 made numerous comments about the representativeness of our δ^{13}C-CH_4 dataset. We want to make clear that to our knowledge this is the largest database of freshwater methane δ^{13}C-CH_4 currently compiled. For comparison, the second largest dataset, that of Sherwood et al., (2017), includes 48 freshwater sites (including rice paddies), of which 16 are also included in our database. However our δ^{13}C-CH_4 database is not comprehensive (unlike the δ^2H-CH_4 database), in that it does not include many measurements that are not paired with δ^2H-CH_4 measurements and that have not yet been compiled into a database. It is also probably not representative, because some important environments, namely C_4 plant dominated ecosystems, are not well represented.

Since the primary focus of this paper is δ^2H-CH_4, it is not within its scope to provide a comprehensive database of freshwater δ^{13}C-CH_4, although that would be a worthwhile goal for future research. In order to make our analysis as complete as possible, in our revised manuscript we will include the 32 freshwater sites from Sherwood et al., (2017) that were not included in our original analysis in our calculations for the upscaling exercise, as well as Figures 7, 8, and 9. We will also carefully discuss the likely biases in

this dataset, especially in terms of C_4 plant environments, and their implications for our interpretations.

6) Both reviewers 2 and 3 expressed some concerns with the upscaling analysis. We acknowledge that the upscaling analysis is relatively simplistic, and that some of the interpretations were speculative. However, we still think it is valuable to use the estimates of freshwater CH_4 isotopic composition, differentiated by latitude, produced in this study to estimate global source δ^2H-CH_4 and $\delta^{13}C$-CH_4, and to compare that with other estimates. We wish to make clear that given uncertainties and complexity in estimating sink fractionations, particularly for δ^2H-CH_4, we are not attempting to estimate atmospheric values, but instead the integrated source δ^2H-CH_4 and $\delta^{13}C$-CH_4 prior to sink fractionations. We think there is value in comparing this with (i) previous bottom-up estimates of these values; and (ii) with the top-down estimates reported by Rice et al., (2016). We concur with Reviewer 2 that the discussion of alternate emissions scenarios is too speculative and simplistic, and therefore we will remove this discussion. Instead, we will focus on likely sources of error in the isotopic source signatures, and the best ways to address these errors in future studies.

We disagree with Reviewer 2 that the error estimates for isotopic source signatures are generally too optimistic, which we will discuss in more detail in our response to that reviewer.

Given comments from all three reviewers we will revise Figure 10 to only include panel C, and make the comparison with other estimates of global source δ^2H-CH_4 and $\delta^{13}C$-CH_4 clearer in this figure.

Specific Responses to Reviewer 3: Reviewer comments are in plain text. **Responses are in bold text.**

During the past two decades, there has been limited progress in advancing understanding of controls on **d2H(CH4)** values in freshwater environments and improving estimates of **d2H** values of CH_4 emissions. This study: (i) updates and attempts to refine the relationship between **d2H(H2O)** and **d2H(CH4)** first reported by Waldron et al. (1999b), (ii) evaluates the extent to which factors other than **d2H(H2O)** may influence **d2H(CH4)** values in freshwater environments, (iii) uses the refined relationships to estimate new **d2H** values for CH_4 emissions from freshwatersources, and (iv) weights CH_4 fluxes reported by Saunois et al. (2020) with a mixture of old and new **d2H** and **d13C** values to estimate global **d2H** and **d13C** values for atmospheric CH_4. In my opinion, the study offers new insights that are worthy of publication pending revision.

We thank Dr. Hornibrook for his detailed review, and we are heartened to hear his opinion that the study is worthy of publication pending revision.

Site level mean values - The study has produced a thorough compilation of stable isotope data related to CH_4 from freshwater environments. The availability of **d2H(CH4)** values presumably was the key criterion for inclusion in the data base. The supplemental file

contains a summary of the data, showing the number of samples from each site and site-level mean isotopic values as described in section 2.3.1. While I appreciate the motivation to avoid introducing bias towards sites that have larger datasets, this approach does limit the extent to which the study can comment meaningfully on differences between environments. **d₂H(CH₄)**, **d₁₃C(CH₄)** and **d₁₃C(CO₂)** values all exhibit significant ranges and trends with depth in the subsurface of wetlands. That information is lost when profiles of **d**-values are averaged. In peatlands where CH₄ production pathways change with depth or CH₄ oxidation occurs, **d**-values determined from an average of shallow and deep layers has little meaning in the context of production pathways or evidence for CH₄ alteration. The pooled **d**-values also do not take into account differences in the amount of CH₄ or CO₂ at different depths. Moreover, **d**-values from deep peat typically will have little bearing on the stable isotope composition of CH₄ emitted from a wetland. Venting of accumulated gas bubbles from deep peat can occur (e.g., Glaser et al, 2004) but there is little evidence that such events are common. The bulk of CH₄ production occurs at shallow depths (from water table level to ~50 cm depth) where the supply of labile substrates from plant roots is greatest and temperature is highest during summer. The residence time of CH₄ at those depths is shortest (e.g., Lombardi et al., 1997; Bowes and Hornibrook, 2006) and most of the CH₄ produced seasonally is either consumed or evaded to the atmosphere. If subsurface data must be averaged to avoid bias, then I suggest using a consistent depth range (e.g., 0 to 50 cm) to (i) generate mean **d**-values that are more likely to represent **d**-values of CH₄ emissions, and (ii) enable analysis of **a**C and **a**H values that are more likely to be related to one methanogenic pathway or exhibit the influence of methane oxidation rather than a blend of pathways and processes across a range of depths. An important advance in this study was the attempt to discern the relative impact of factors other than **d₂H(H₂O)** on **d₂H(CH₄)** values. Use of site level means for **d**-values raises concern about the validity of the **a**C and **a**H values calculated to assess breakpoints in CH₄ production pathways and oxidation.

The reviewer raises an important point about δ^2H-CH₄ variability with depth in peatlands, and potential biases that are introduced by averaging values across depth profiles. The primary goal of our study is to investigate spatial variability between sites, and therefore we think it is important to provide a single value for each site. In addition, one of the key goals is to characterize the δ^2H values of CH₄ emitted to the atmosphere. Therefore, we agree with the reviewer's suggestion to use a consistent depth range (0-50 cm) when averaging data from peatlands with depth-resolved sampling. See Planned Major Revision 1 above. This change affects 8 sites, from 5 publications (Hornibrook et al., 1997; Waldron et al., 1999; Chasar et al., 2000; Chanton et al., 2006; Alstad and Whiticar, 2011). Other studies included in our dataset sampled peatlands at shallow depths. To our knowledge all studies in other wetland environments also sampled shallow (< 50 cm) soils.

'Bottom-up' mixing model - I appreciate that considerable effort was invested in attempting to upscale d₂H(CH₄) and d₁₃C(CH₄) values; however, it is questionable whether that portion of the manuscript has potential to advance discourse on global isotope-weighted CH₄ budgets. A more valuable outcome of this work would have been

the one identified by that authors in lines 441- 443: "A logical next step in predicting global freshwater δ_2H-CH$_4$ source signatures would be to combine high-resolution mapping of wetlands and inland waters, maps of the global distribution of δ_2H$_p$, and regression relationships between δ_2H-CH$_4$ vs. δ_2H$_p$." In my view, production of a global gridded map of **d**$_2$H(CH$_4$) values for freshwater environments would have a more suitable application of the outcomes from the data analysis. It would provide a useful counterpart to the **d**$_{13}$C(CH$_4$) global map for wetlands published by Ganesan et al. (2018). I realize at this stage in the process that would take the second half of the manuscript in a very different direction. As things stand, the weighted atmospheric **d**$_2$H(CH$_4$) and **d**$_{13}$C(CH$_4$) values that were calculated are difficult to reconcile with atmospheric data and KIEs associated with sinks for atmospheric CH$_4$. It's possible that the values may be offering new insights but it seems more likely that there are issues with attribution of **d**$_2$H and **d**$_{13}$C values to CH$_4$ sources.

We recognize the reviewer's concerns that the upscaling results presented in this paper may not advance discourse on isotope weighted CH$_4$ budgets. Reviewer 2 made similar comments. We note that we specifically did not try to resolve these results with atmospheric data, given the uncertainties related to sink KIEs, but instead compared them with past estimates of global source isotopic values that are based on atmospheric data and previously published models of sink fractionations (Rice et al., 2016, Figure 10C in the original manuscript). We will make this clearer in the revised manuscript, and highlight the associated uncertainties to a greater degree.

We have decided to substantially revise this part of the manuscript. See Planned Major Revision 6. We think it is still worthwhile to present estimates of global methane source δ^2H and δ^{13}C that include the results of our data analysis, and to compare this with previous bottom-up estimates of global isotopic source signatures, as well as the top-down estimates from Rice et al., (2016) mentioned above. We will then focus on an assessment of the largest areas of uncertainty in the isotopic source signatures, and not dwell on uncertainties in sink fractionations, since these are not the focus of this paper. We will mention possible errors in flux inventories, but will devote less focus to this than possible biases in isotopic signatures. In particular we will direct more focus on the problem of a lack of data from C$_4$ plant dominated ecosystems in synthetic datasets, which may compromise data-based estimates of freshwater δ^{13}C-CH$_4$ signatures.

Creating a gridded map of freshwater δ^2H-CH$_4$ values entails a substantial amount of work and additional expertise in GIS methods, and this is beyond the scope of the revisions for this paper, which as reviewer 1 noted is already quite extensive and ambitious. However, this is the goal of collaborative research that is currently in development. This research in development will also look more closely at comparisons with atmospheric data.

Citations within the text do not appear to be listed consistently either alphabetically or chronologically.

We thank the reviewer for noting this. It is probably a problem with the EndNote citation style, and we will check this carefully in the revised version.

Line 38: 'clearly' = 'unequivocally' ?

We agree this would make this sentence clearer and will make the change.

Lines 51-52: 'recent technological developments'. An additional sentence or two about laser based methods would be helpful for a broader readership.

That is a good idea and we will add a sentence or two about new laser based methodologies.

Lines 53-57: Rigby et al. (2012) also demonstrated the utility of a multi-isotope approach for global methane cycle characterization.

We thank the reviewer for bringing this paper to our attention. We will revise this paragraph to include this conclusions of that study.

Lines 87-88 (and elsewhere): 'data is' should be 'data are'

We will adjust this here and throughout the manuscript.

Line 105: A citation for Coplen (2011) could be added for the definition of delta that (correctly)
does not include a 'x 1000' factor.

We will add the suggested citation

L129: The citation for John Lansdown's thesis should be:
Lansdown J. M. (1992) The carbon and hydrogen stable isotope composition of methane released from natural wetlands and ruminants. Ph.D. dissertation, Univ. of Washington.
(The citation can be confirmed at: https://dggs.alaska.gov/pubs/id/28259)

We thank the reviewer for this correction, and will edit the references and citations

L156 – Is the annual estimate of δ_2H_p weighted by the relative amounts of precipitation during different seasons?

Yes, the annual estimates from the model are amount-weighted values (See Bowen and Wilkinson 2002 for specifics on the methodology).

L200: d_2H (superscript missing)

We will fix this error

L258-L259 "55 sites are classified as wetlands, including 16 bogs, 14 swamps and marshes, 12fens, and 8 rice paddies."
>> Are the classifications for bogs and fens based upon pore water chemistry and vegetation surveys? The word 'bog' sometimes is used in site names that are other wetland types, in particular, fens.

This is a good point. We have done our best to be careful about the wetland classifications, but we have primarily relied on the classification of the original study. Of the 16 bog sites, 14 came from studies that specifically differentiate between bogs and fens (Chanton et al., 2006; Lansdown, 1992 (thesis), Alstad and Whiticar, 2011, Waldron et al., 1999; Chasar et al., 2000), or provide detailed information on vegetation and/or soil pH (Lansdown et al., 1992; Hornibrook et al., 1996). One other paper (Whiticar et al., 1986) provides data from Volo Bog, Illinois, which based on other references is an ombrotrophic, sphagnum-dominated bog. The only remaining bog site is a West Virginia Bog, from Wahlen, (1994), which did not provide enough information to verify this classification. Given that this original classification is all we have to go on we continue to use it for this sample.

Table 1: Origins of some data are unclear. When indicated as 'no specific measurement indatabase', what does it mean to say 'we used the isotopic values and uncertainties for X'? Which literature source? Also, only C3 $\mathbf{d}_{13}$C values appear to be used for biomass burning. Grassland and savanna wildfires presumably generate CH_4 that has more positive $\mathbf{d}_{13}$C values from burning of C4 grasses.

Thank you for raising these ambiguities in Table 1. Reviewer 2 has brought up similar concerns and we will make this table and the underlying data clearer in the revised manuscript. The database being referred to is the Gas Geochemistry Isotope Database (Sherwood et al., 2017), as referenced in section 2.4. This was the source for all isotopic estimates, with the exception of biogenic marine methane, which we derived from Whiticar et al., (1999).

The Global Gas Geochemistry Database was our basis for the biomass burning $\delta^{13}C$-CH_4 values. Out of 24 biomass burning $\delta^{13}C$-CH_4 values, only 2 are ostensibly from C_4 plants and have a higher $\delta^{13}C$-CH_4 value. These were included in our analysis. In keeping with our data centered approach, and the lack of definitive estimates of the relative proportion of biomass burning CH_4 emissions from C_4 plants, we did not attempt to weight these values in our analysis. However, in the revised manuscript we will mention this as a possible source of error in our discussion, and highlight the importance of more data on methane from C_4 plant ecosystems, both for biomass burning and microbial emissions.

L266-L271 The comparison of modelled δ_2H_p values and measured $\mathbf{d}_2H(H_2O)$ values for 62 sites is important for validating the approach on which estimating $\mathbf{d}_2H(CH_4)$ relies. The text is not clear though with respect to causes in deviation from a 1:1 relationship. Presumably "$\mathbf{d}_2H$-H_2O is generally higher" means $_2H$-enrichment is evident in the measured data. Is the statement about 'overall smaller water volumes' meant to infer

evaporative enrichment of $_2$H?

This comment, as well as those of reviewers 1 and 2, make it clear that we need to more thoroughly evaluate the relationship between emipirical δ^2H-H$_2$O and modeled δ^2H$_p$ values in this paper. We have done so, including considering wetlands and inland waters separately, and examining whether modeled annual precipitation or growing season precipitation is a better predictor of the empirical δ^2H-H$_2$O values. See our Planned Major Revision 2.

The comment about higher δ^2H-H$_2$O in mid-latitude sites was based on our expectation that in wetlands the residence time of water is lower, and therefore there is more seasonal variability in δ^2H-H$_2$O. Since almost all samples were collected in summer, when δ^2H$_p$ is higher than average in higher-latitude settings, this would lead these values to be higher than annual precipitation. However, our more detailed analysis does not support this contention, and instead implies that evaporation is likely leading to water δ^2H-H$_2$O values that are higher than precipitation in wetlands specifically. See Planned Major Revision 2 and Table R1.

L282-L283 "Both relationships result in a large amount of unexplained residual variability, implying the importance of other variables in controlling δ_2H-CH$_4$."

I'll expand here on the point raised in my general comments. The extent to which residual variability exists is likely underestimated because of the use of site-level means. There are relatively few data sets globally that contain subsurface profiles of both **d$_2$H(H$_2$O)** and **d$_2$H(CH$_4$)** values. Four of those data sets are shown in the enclosed figure which was published in Hornibrook and Aravena (2010): Turnagain Bog (open triangles; Chanton et al. 2006), Sifton Bog (open diamonds; Hornibrook et al. 1997), Point Pelee Marsh (open circles; Hornibrook et al. 1997) and Ellergower Moss (open squares; Waldron et al. 1999a). The arrows indicate the direction of increasing depth in peat for Turnagain Bog, Sifton Bog, Point Pelee Marsh and Ellergower Marsh. The figure also includes **d$_2$H** values of coexisting CH$_4$ and H$_2$O values from Alaskan peatlands along a N-S transect (filled triangles; Chanton et al. 2006) and regression equations (Table 6.2 from Hornibrook and Aravena, 2010 also enclosed) from a number of studies including Waldron et al. (1999b; line 5) and Whiticar et al. (1986; lines 1 and 2).
The approach of using site-level means reduces each of those depth trends to a single point in **d$_2$H(H$_2$O)** vs. **d$_2$H(CH$_4$)** space. The **d$_2$H** values of CH$_4$ emitted to the atmosphere are likely to be similar to the most $_2$H-depleted values in each trend which corresponds to CH$_4$ in shallow peat near the water-air interface and within the root zone where CH$_4$ may be transported to the atmosphere via plant aerenchyma. Averaging **d$_2$H(CH$_4$)** values from all depths (2 m for Sifton Bog and Pelee Marsh; 6 m for Ellergower moss) yields a mean that is substantially more $_2$H-rich. Again, I appreciate the goal of not biasing the analysis to these larger data sets but a single mean for each site does not reflect the considerable residual variability that exists with depth as **d$_2$H(CH$_4$)** values shift away from the global **d$_2$H(H$_2$O)** vs. **d$_2$H(CH$_4$)** regression line. Moreover, the **d$_{13}$C(CH$_4$)** and **d$_{13}$C(CO$_2$)** depth trends from these sites yield systematic shifts in **a$_C$** values that are lost when the **d$_{13}$C** values similarly are reduced to unitary site-level means.

We thank the reviewer for the detailed explanation of their argument on this issue. As we discussed above, the primary goals of this paper are to explore inter-site geographic variability in the δ^2H-CH$_4$ emitted to the atmosphere. Therefore, while intra-site variability is of great interest, we do not want to add an additional layer of complexity to this paper by considering this. We feel the reviewer's earlier suggestion of limiting samples from the upper 50 cm of peat is a good solution to this issue, and we have followed this suggestion in our revised analysis. See Planned Major Revision 1.

L308-L309 "We do not find evidence for a piece-wise linear relationship between δ_{13}C-CH$_4$ and δ_2H-CH$_{4,w0}$ (Fig. 5a), nor did we find a significant simple linear correlation between these variables."
>> It may be worth exploring whether any relationships exist in the full data sets rather than site level means.

This is an interesting suggestion, though we have concerns that such an analysis might be biased by over-representing sites that have a large number of measurements. It will also require a large amount of additional data analysis, since the δ^2H-CH$_{4,w0}$ data are not currently disaggregated on a per sample basis. Given that the focus of this work is on variability between sites, we will leave this analysis for future work focused on intra-site isotopic variation.

L441-L443: "A logical next step in predicting global freshwater δ_2H-CH$_4$ source signatures would be to combine high-resolution mapping of wetlands and inland waters, maps of the global distribution of δ_2H$_p$, and regression relationships between δ_2H-CH$_4$ vs. δ_2H$_p$.">> I agree with the authors and suggest this would be a worthwhile output to include in this manuscript instead of the global upscaling estimate.

We appreciate this suggestion. As mentioned above, adding this output to this manuscript would entail substantial additional work, as well as additional expertise beyond that of the authors. We have however begun a collaboration with another research group to perform this analysis, and this will be the focus of a future publication.

L445-L464 Section 4.2. This section would benefit from acknowledging and discussing the study by Rigby et al. (2012).

We thank the reviewer again for this suggestion. We will acknowledge and discuss this work in the revised manuscript.

L500-L504 In addition to the caveat noted that CH$_4$ data exhibiting $_2$H-enrichment due to methane oxidation are uncommon, the amount of CH$_4$ emitted to the atmosphere bearing the effects of methanotrophy is likely to be small. Bacteria oxidation is highly efficient in the subsurface of wetlands and little CH$_4$ tends to escape to the atmosphere via diffusion through porewater. This comment applies to peatlands. The situation is different in inland

water environments.

This is an important point, and we will revise this section to make this clear. As noted in Planned Major Revision 4, we are now less confident that the observed variation in δ^2H-CH$_{4,w0}$ can be primarily ascribed to differences in methanogenic pathway. Therefore our discussion of relative importance of these mechanisms, as well as other possibly influential processes, will be quite different in the revised manuscript.

L510–L518 I was pleased to see incorporation of these alternate explanations for relationships between **d2H** and **d13C** values of CH4. Methanogenic pathways are not the only potential explanation.

We are glad to see that there is a positive reception to this. Based on this comment and those of reviewer 1 we are planning to focus on alternate explanations to a greater degree in the revised manuscript. See Planned Major Revision 4.

L592-L593 – Bellisario et al. (1999) provides a good example of how **d13C(CH4)** values vary along a trophic gradient in a wetland complex. Differences in **d13C** values of CH4 emissions and porewater CH4 values in minerotrophic vs. ombrotrophic wetland are demonstrated in Hornibrook and Bowes (2007) and Hornibrook (2009). Landscape scale measurements (atmospheric inversions and aircraft measurements; Fisher et al., 2017) also show that northern wetlands contain sources of 13C-poor CH4 that differ from values of ~-62 to -58 permil typically attributed to northern peatlands in isotope-weight CH4 budgets. Characterization of sites as ombrotrophic or minerotrophic on the basis of water chemistry and vegetation surveys is essential for making these distinctions.

We thank the reviewer for these insights. We will expand this paragraph to include the ideas and references mentioned by the reviewer. While it is difficult for us to make these distinctions in this dataset, we will note these points. In addition to the absence of C$_4$ plant ecosystems, this is an additional potential bias in the d13C database assembled in this study, and we will acknowledge this and discuss how it could be addressed with future research.

L617 to L622 It is unclear how a more negative than expected value for estimated **d13C(CH4)** can be explained by (2) source signatures being biased toward more positive **d13C** values.

This was a mistake. We meant to say '^{13}C *depleted* values' and '^{13}C *depleted* sources'. Regardless, this section of the discussion will be heavily revised based on the suggestions of reviewers 2 and 3, with less emphasis on discrepancies with atmospheric measurements. See planned major revision 6.

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