

We would like to thank the two anonymous reviewers and the Editor for their careful reviews of our manuscript. Below we present our point-by-point responses to all of the comments and annotations. The original comments from the reviewers are shown in black, while our responses are presented in red. Where indicated, additions to the manuscript are shown in blue and removed passages shown in red strikethrough font.

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Comments from the reviewer #1 and our responses:

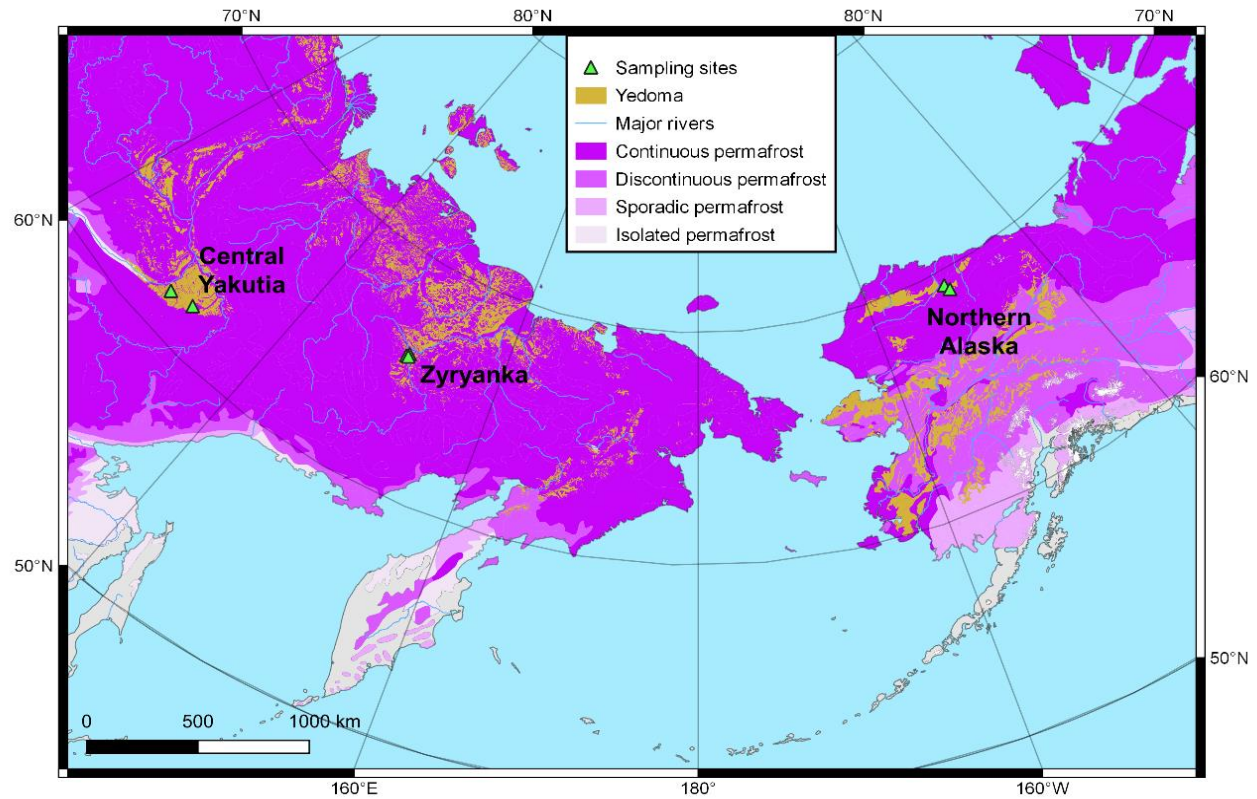
## 1) GENERAL COMMENTS

This methodological, brief communication paper reports on a comparison of different techniques (wet vs. dry extraction, with or without biocide to test microbial contamination) to extract gas (CH<sub>4</sub> and N<sub>2</sub>O) from ice wedges of Alaska and Siberia. The authors report that tested methods yield good results for the easily extractable gas fraction (bubbles), but this is not so convincing for the adsorbed phase or gas contained within soil aggregates. One of the main conclusions, therefore, is that current estimates of ground-ice gas budgets are likely underestimated, as a fraction of produced gases are not taken into account. For me, this is the main take-home message.

It appears as an interesting short paper, although the methodology used is not in my immediate field of expertise. To my knowledge, this manuscript does not have major flaws that should ultimately prohibit its publication. It is generally well written and easy to read. I have however a few points to mention that preclude acceptance for publication as is:

- 1) I am not convinced, for now, of the general, broad-audience impact of the manuscript. Does it really « *report new developments, significant advances, and novel aspects of experimental and theoretical methods and techniques which are relevant for scientific investigations within the journal scope* »? ([https://www.the-cryosphere.net/about/manuscript\\_types.html](https://www.the-cryosphere.net/about/manuscript_types.html)). The authors have not convinced me that this work is new, innovative or represent a major advancement that is relevant to the community at large. They rather suggest that a future, novel extraction method might provide better results. I am also looking forward to that. This work might be useful for a small specialized group, however. Furthermore, the conclusion about underestimation of current gas budgets in ice-wedge terrains is itself interesting and timely.  
→ We believe that our manuscript has broad implication to the large community working on permafrost-climate interaction. Permafrost thawing is a major potential threat of future global warming, expected to input large amount of greenhouse gas (GHG) to the atmosphere. Thus, quantification of the permafrost GHG budget is important for better projection of future climate change. There are a growing number of works reporting methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) concentration either in permafrost or ground ice. Nonetheless, there has been no consensus on gas extraction methods, nor they have been tested properly. To our best knowledge, this manuscript is the first attempt to test different gas extraction methods to understand the applicability and limitations of both techniques. We believe that our study makes a significant contribution to the literature because our experiments show that existing methods allow gas extraction from the soft parts of ice, however gas adsorbed to or trapped in soil particles may not be extracted, affecting the measure GHG contents and their mixing ratios. In addition, we reveal that the microbial activities have insignificant impact on the wet extraction results. Although our manuscript does not deal with <new development>, we do believe that all these findings provide <significant advances> and <novel aspects of experimental methods>. Development of a new technique is far beyond the scope of this manuscript.

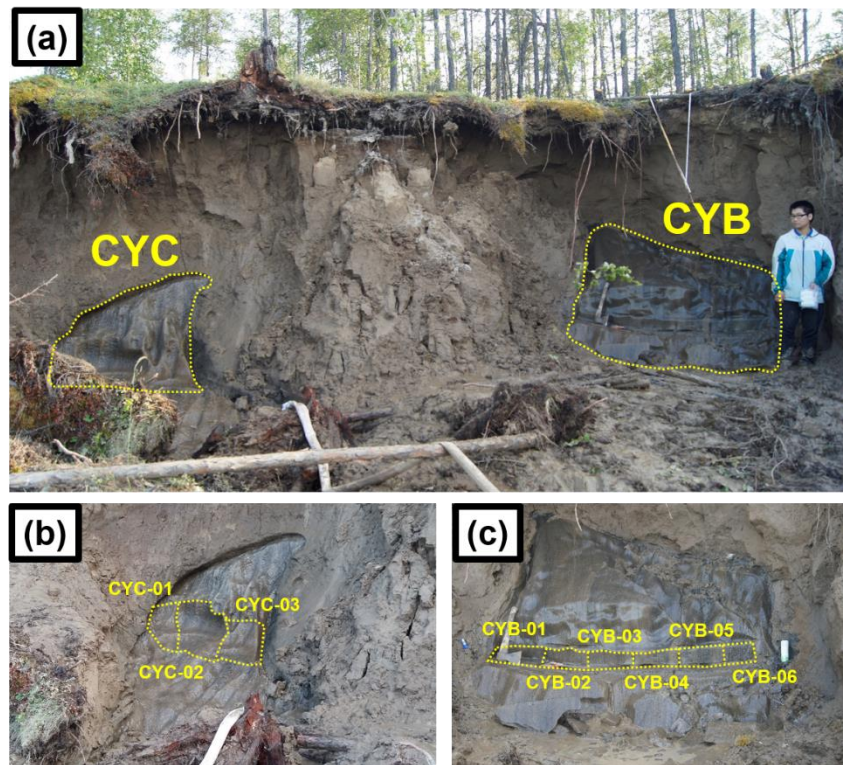
- 2) I understand that this is a brief communication and that the number of figures/tables is limited. However, it is really unfortunate that there is no map of the many study sites, and no picture or illustration of field sampling procedures, as well as lab instruments (especially the 'needle crushing system'). It would greatly help to have visual support for such investigations.
- Now we add the following maps and pictures of the sampling sites as well as those of gas extraction systems in the Supplement.



**Supplementary Figure 1.** The site locations of the ground ice samples used in this study are marked in the map of circumpolar permafrost (Brown et al., 1997), yedoma distributions (Strauss et al., 2016), and major rivers.

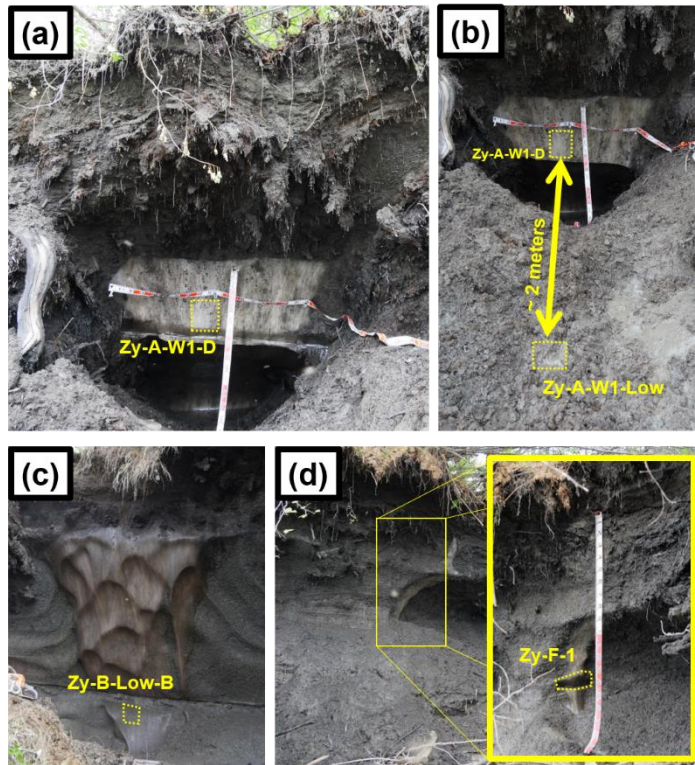


**Supplementary Figure 2.** Photographs of ground ice outcrops at Churapcha (central Yakutia) site. Locations of the samples used in this study are indicated by yellow dotted circles.

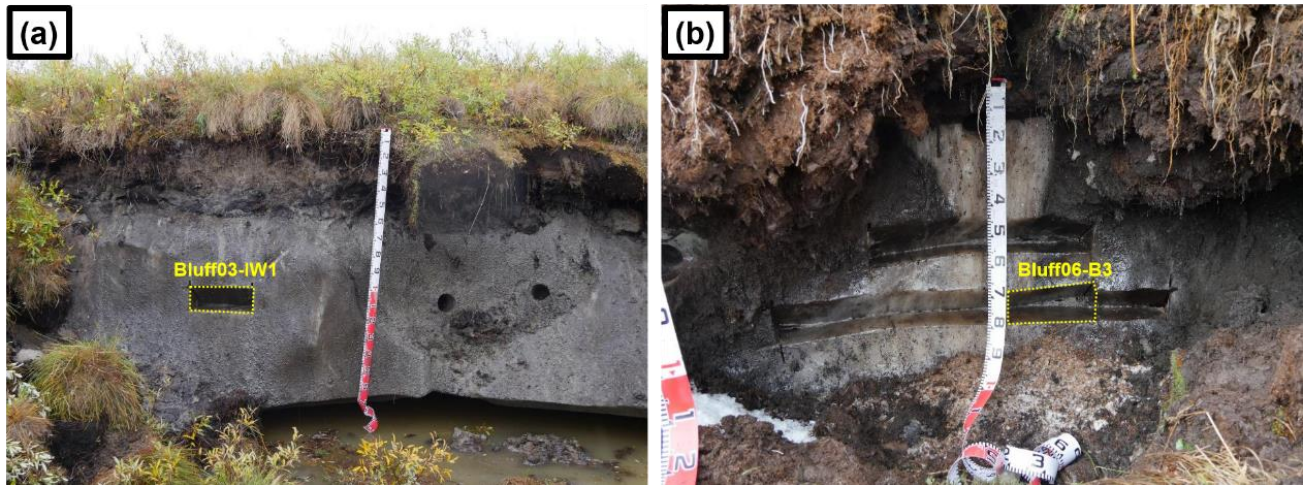


**Supplementary Figure 3.** Photographs of ground ice outcrops at Cyuie (central Yakutia) sites: (a) ice wedge outcrop, (b) CYC and (c) CYB samples. Locations of the samples used in this study are indicated by yellow dotted lines.





**Supplementary Figure 4.** Photographs of ground ice outcrops at Zyryanka sites: (a and b) Zy-A, (c) Zy-B, and (d) Zy-F. Locations of the samples used in this study are indicated by yellow dotted lines.



**Supplementary Figure 5.** Photographs of ground ice outcrops at northern Alaskan sites: (a) Bluff03 and (b) Bluff06. Locations of the samples used in this study are indicated by yellow dotted boxes.

- 3) Several sentences contained in the results/discussion section are in fact related to methods. I mention examples in the specific comments section below. The structure of the main text should therefore be re-aligned, so that methods sentences are in the methods section.
- We would like to remind that our manuscript mainly focuses on experimental- and methodological aspects. Each set of tests has been designed with logical flows. Therefore, we believe that it would be easier to read in current structure than suggested by the reviewer. However, as the re-arrangement does not change any fundamental content of our manuscript, we're willing to accept this change if the Editor suggests to do so.
- 4) Finally, some statements and conclusions in the main text are either not accompanied by a mention to the results or figure(s) they come from, or not supported by literature reference(s).
- We will revise those sentences following the specific comments below. We will also re-check the main text thoroughly.

Overall, I cannot accept this manuscript for publication as is. If the authors are willing to make major revisions (general points above and specific comments below), I would be happy to review a revised version of the manuscript.

## 2) SPECIFIC COMMENTS AND EDITORIAL SUGGESTIONS

P= page number, L = line number.

P1, L29. To avoid repetition (soil): choose either « Permafrost preserves large amounts of soil carbon and nitrogen... », or « Permafrost soils preserve large amounts of carbon and nitrogen... ».

- The sentence is revised as suggested by the reviewer.
- Permafrost ~~soils~~ preserve large amounts of soil carbon (C) and nitrogen (N) in a frozen state (e.g., Hugelius et al., 2014; Salmon et al., 2018), temporarily removing this frozen carbon (C) and nitrogen (N) from active global cycles.

P2, L30. I suggest adding 'temporarily': «... temporarily removing this frozen carbon... ».

- We add 'temporarily' to the sentence. Please find our response above.

P2, L30-31. (C) and (N) should be put at the beginning of the section (P1, L29), i.e. the first time that the words 'carbon' and 'nitrogen' are mentioned.

- We revise the sentence following the reviewer's comment. Please refer to our response above.

P2, L35. « ... which in turn can trigger positive feedbacks... ».

- The phrase is modified as the reviewer suggested.
- Therefore, future projections of permafrost stability are of great interest, particularly because thawing permafrost may lead to decomposition and/or remineralization of the buried soil C and N and their abrupt emission into the atmosphere in the form of greenhouse gases (GHGs) – carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and nitrous oxide (N<sub>2</sub>O), which in turn can trigger positive feedbacks (e.g., Salmon et al., 2018).

P2, L38-40. This might be true for Yedoma regions (eastern Siberia, Alaska, Yukon), but not all permafrost is necessarily ice-rich. It should be specified in the paragraph, otherwise we have the impression that permafrost all over the Arctic contains 40-90% of ground ice.

- The concerning sentence is revised as below.

- However, the processes responsible for in-situ C and N remineralization and GHG production in ground ice are poorly understood, despite the fact that ground ice accounts for a substantial portion ~~of the upper permafrost:~~ (up to approximately 40–90% by volume) of Pleistocene ice-rich permafrost, or Yedoma (e.g., Kanevskiy et al., 2013; Jorgenson et al., 2015).

P2, L40. I suggest adding 'Pleistocene': « ... volume of Pleistocene ice-rich permafrost, or Yedoma ».

- We add 'Pleistocene' as suggested. Please find our response above.

P2, L42. « ... evidence for in-situ microbial aerobic respiration... ». Why just 'aerobic' conditions? This might be relevant for CO<sub>2</sub> production, but CH<sub>4</sub> and N<sub>2</sub>O are generally produced under 'anaerobic' conditions, or both oxic-anoxic.

- The sentence is revised to include both aerobic and anaerobic respirations.
- The gases trapped in ground ice allow unique insights into the origin of ground ice and evidence for in-situ microbial aerobic and anaerobic respirations (e.g., Boereboom et al., 2013; Kim et al., 2019; Lacelle et al., 2011).

P2, L43-44. « ... detailed information on in-situ biogeochemical processes responsible for GHG production... ». Which biogeochemical processes? Methanogenesis? Respiration? Other processes?

- The cited literatures – Boereboom et al. (2013) and Kim et al. (2019), attributed the elevated mixing ratios of CH<sub>4</sub> and N<sub>2</sub>O to in-situ methanogenesis, nitrification, and denitrification. We note this in the sentence.
- Among others, the GHGs in ground ice may provide detailed information on in-situ biogeochemical processes responsible for GHG production (i.e., methanogenesis, nitrification, and denitrification) (e.g., Boereboom et al., 2013; Kim et al., 2019).

P3, L58-59. «... because ice wedges are one of the most abundant morphological features... ».

- Corrected.
- Ice-wedge samples from Alaskan and Siberian permafrost were used because ice wedges are ~~the ice wedge is~~ one of the most abundant morphological features of massive ground ice, consisting of approximately 5 to 50% by volume of the upper permafrost (Kanevskiy et al., 2013; Jorgenson et al., 2015).

P3, L73-79. For the reader not familiar with the study sites and ground-ice sampling protocols in permafrost landscapes, I strongly suggest adding 1) a map of the study sites (Siberia and Alaska); 2) pictures of an outcrop and sample collection (drilling). This way, the reader would have a much better idea of what the samples and sites look like.

- We add the maps showing the sampling sites along with permafrost and yedoma distributions as well as the photographs of the outcrops where our samples were taken in the Supplement. Please find our response to the general comment above and the Supplement material of the revised manuscript below.

P4, L80-97 and L88-95. Again, all these descriptions and distances would make much more sense if they were accompanied by a map (with sampling sites labeled on the map).

- Please find the new maps we added above.

P4, L87. «... on the first terrace of the river... ». Do the authors mean the younger (i.e. lower) terrace?

- Yes, this is the lowest terrace. The sentence is revised accordingly.
- Most of the outcrops that were sampled for ground ice were on the first (lowest) terrace of the river.

P4, L96. « The ice-wedge ice... ». This phrase is weird. Suggestions: « The ice from ice wedges is different from polar ice cores, in that... », or « Wedge ice is different...».

→ Here we disagree. We think the “ice-wedge ice” represents “the ice from ice wedges” in a more concise way. We prefer “ice-wedge ice” to just “wedge ice” in order to better specify because there are wedges from different origins (e.g., sand wedges).

P5, L107. «... 8~13 g of ice sample were crushed... ».

→ Corrected.

→ In brief, 8~13 g of ice sample ~~was~~were crushed in a cold vacuum chamber (extraction chamber).

P5, L121-123. Which year for this modern air sample? Please specify.

→ The modern air samples used as standard in this study were collected in November of 2016. We specify it in the sentence.

→ ...and a modern air sample from a surface firn at Styx Glacier, Antarctica (in November of 2016), which was calibrated as  $1758.6 \pm 0.6$  ppb CH<sub>4</sub> and  $324.7 \pm 0.3$  ppb N<sub>2</sub>O by the National Oceanic and Atmospheric Administration (NOAA).

P7, L175. I suggest ‘thoroughly’ or ‘vigorously’ instead of ‘well’. («... shaken flasks were shaken thoroughly/vigorously... »

→ Revised as suggested.

→ After the control-wet extractions were complete, the sample flasks were shaken thoroughly ~~well~~ and the meltwater samples were each poured into a 50 mL conical tube.

P8, L187-189. What is meant by this statement about the heterogeneous distribution of samples? How is it shown on Fig. 1?

→ The gas mixing ratios in the neighboring ice pieces from an ice wedge are highly variable. Our previous work (Kim et al., 2019) showed the centimeter-scale variability of the gas mixing ratios in ice wedges. We change the relevant sentence as follows:

→ We noted that the heterogenous distribution of gas mixing ratios of in centimeter scales (Kim et al., 2019) may not have been completely smoothed out by our sub-sample selection, although we randomly chose 8-12 ice cubes for each measurement.

P9, L195-196. This is a busy figure, see comments below (section 3) FIGURES). Some elements could be removed to enhance clarity.

→ We revise Figure 1. Please refer to our response to 3) FIGURES AND TABLES below.

P10, L208-211. This reads more like methods, not a results and discussion section.

→ Please find our response to the General Comment #3.

P10, L211-212. How about the Eastern Siberia samples? (triangles in Fig1) Did they also « not show significant differences » between the two sets of tests?

→ The complementary tests using BES were carried out only with Central Yakutian- and Alaskan samples. The results are plotted in Figure A3 in Appendix, rather than Figure 1.

P11, L224. «... polar ice core samples... » (remove the 2<sup>nd</sup> ‘ice’)

→ We delete the repetition of ‘ice’.

→ The gas extraction efficiency of the SNU needle crusher system has been reported as ~80–90% for polar ice core ~~ice~~-samples (Shin, 2014).



P11, L230-237. This reads more like a paragraph about methods.

→ Again, we hesitate to do so for the reason we mentioned before. Please find our response to the General Comment #3.

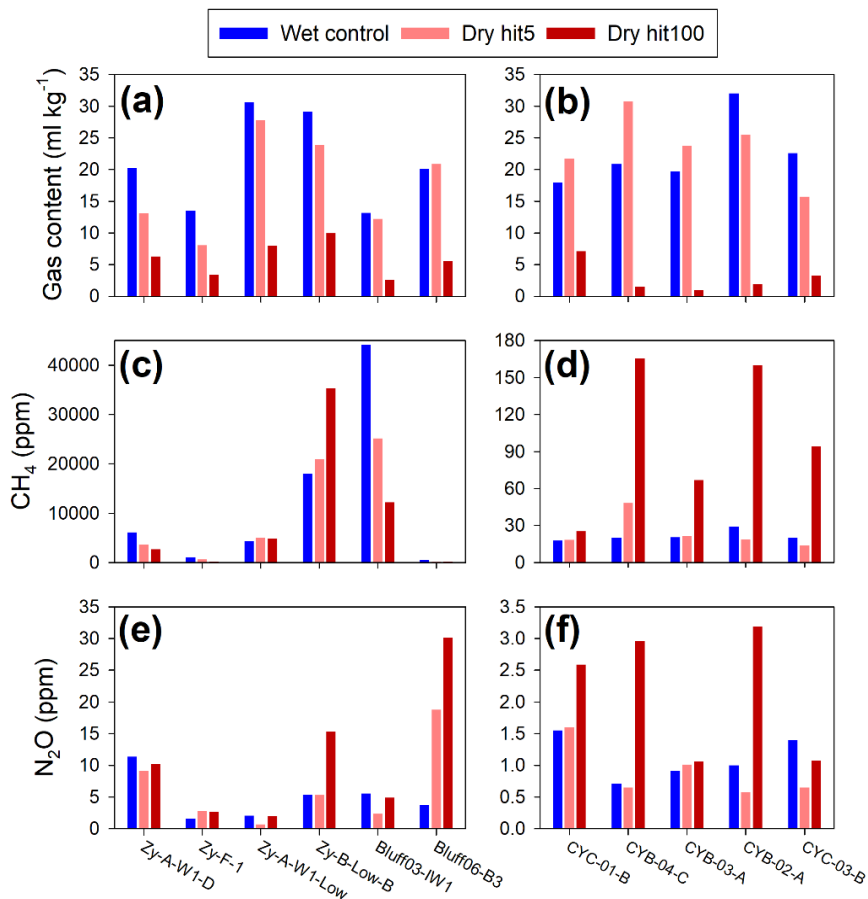
P11, L236-237. This information would be better displayed and more appealing in a figure.

→ Here we hesitate to replace Table 1 with a figure. The reason why we use a table here instead of a figure is the large ranges of CH<sub>4</sub> and N<sub>2</sub>O mixing ratios that make figures less readable. Below we made a figure (grouped bar graphs) as an example. Since there are large variations among the samples, some of the data can hardly be seen, particularly for the samples having lower mixing ratios than others. Thus, we prefer to keep the Table 1, but we'll follow the Editor's decision.

P12, L250-252. This statement is based on what result? Can we see this displayed somewhere in a figure/table? If yes, please refer to it in the text.

→ The comparisons with the dry soil content are shown both in Figures 1 and A3. We note it in the sentence.

→ When compared with the dry soil content measured from the sub-samples used for wet extraction, no relationship was observed between the dry soil content and the extraction efficiency (Figures 1 and A3).



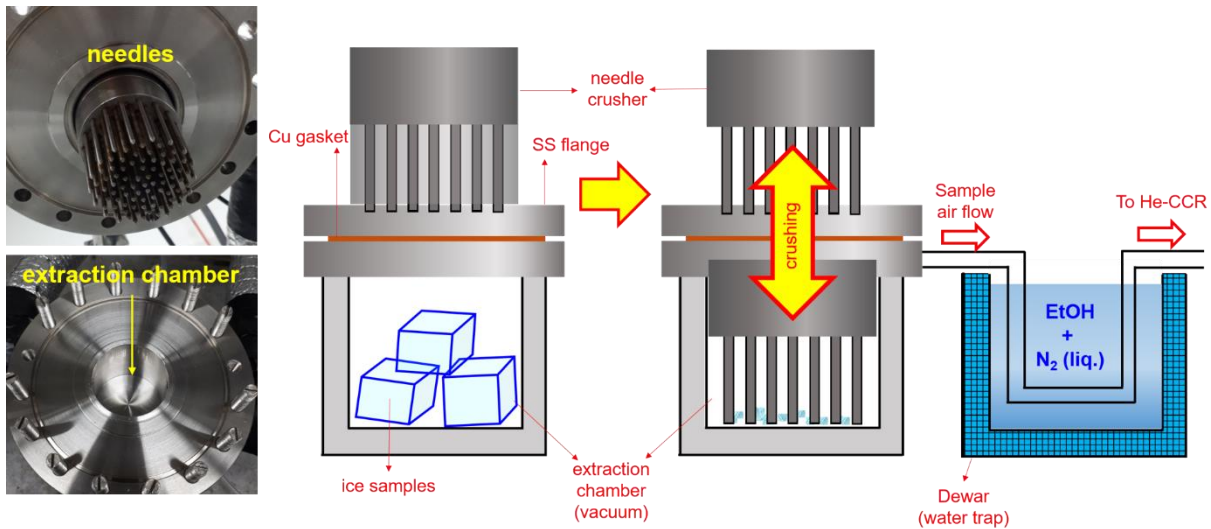


P12, L255-260. For the reader not familiar with the needle crushing system, a picture or a sketch of what the apparatus looks like might help. The sentences in this paragraph would be more easily understood.

- The detailed descriptions and schematic diagram of the needle crushing system used in this study can be found elsewhere in Ahn et al. (2009) or Shin (2014). However, for convenience of the reader, we add the photographs of the needle crushing system in the Supplement.

P13, L280-282. Please refer to results (figure or table) to support this statement.

- This sentence is our interpretation of the results from comparisons between hit5 and hit100 extractions, listed in Table 1. We note this at the end of the sentence.
- In the meanwhile, in the Bluff and Zyryanka samples, the hit5 results reflect the mixing ratios of the gases from the crushed portions, regardless of their origin: bubbles, particle adsorption, or microsites in aggregates (Table 1).



Supplementary Figure 7?. Schematic diagram of the needle-crusher method together with enlarged photographs of crushing needles (left top), and extraction chamber (left bottom).

P13, L288-290. Please support this statement by relevant references. In fact, the simple association  $N_2O$ =oxic /  $CH_4$ =anoxic is not entirely and always true. For example,  $N_2O$  production has been recorded under both oxic and anoxic conditions (Gil et al. 2017; *Global Biogeochemical Cycles*), as well as  $CH_4$  production from oxic waters (Grossart et al. 2011; *PNAS*). It depends on several parameters, including local hydrology (e.g., water-logged soils). This should be acknowledged in the text.

- We revise the sentence as below:
- This can probably be explained by the fact that the  $N_2O$  mixing ratio is not necessarily higher in soil-rich ice because  $N_2O$  is an intermediate product of denitrification and nitrification ~~in relatively oxic conditions~~, while  $CH_4$  is produced as the final product of methanogenesis ~~strictly in anoxic conditions~~.

P13, L292 to P14, L298. This statement is highly speculative. Unless I missed something, this was not tested for real in this study. This paragraph should be supported by real data or removed.

- Deleted.

P14, L302-303. Again, this is methodology, not results/discussion.

- We believe that current structure is easier to read for the same reason above. Please find our response to the General Comment #3.

P16, L320-330. Again: methodology, not results/discussion.

→ Again, we retain the current structure for the same reason we answered to the General Comment #3.

P17, L350-352. This is indeed interesting. Do we know why N<sub>2</sub>O appears to be more extractable (or less present in the residual adsorbed phase) than CH<sub>4</sub>, at least based on the wet extraction technique? Was this already observed elsewhere and reported in the literature?

→ Currently we speculate that higher solubility (to water) of N<sub>2</sub>O could make the adsorbed N<sub>2</sub>O more extractable by wet extraction, compared to CH<sub>4</sub>. To our best knowledge, this is reported for the first time by our manuscript. We added a sentence mentioning this as below:

→ These results imply that most of the N<sub>2</sub>O in ice wedges is extracted by three melting-refreezing cycles, such that only a small amount of N<sub>2</sub>O is left adsorbed or entrapped in ice-wedge soils. [It might be attributed to the high solubility of N<sub>2</sub>O to water compared to CH<sub>4</sub>.](#)

P18, L375. «... easy to extract... »

→ The typo is corrected.

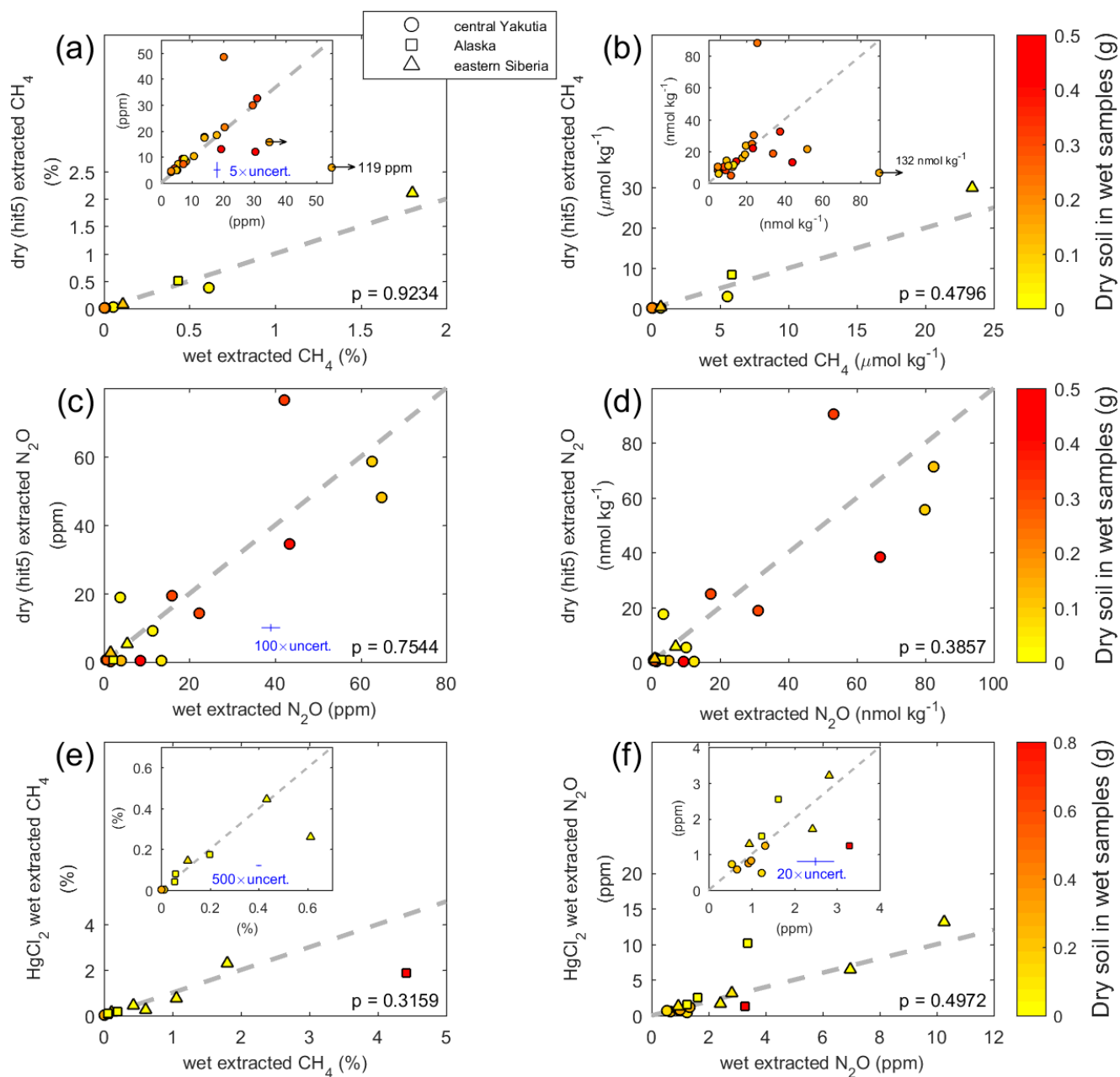
P19, L397. « Our findings indicate that ... »

→ Corrected.

### 3) FIGURES AND TABLES

Figure 1, P9.

- ➔ This is a pretty busy figure. We don't necessarily need the 3 legends (identical) in the middle. By removing them, more space could be created to enlarge the graphs a bit, because for now they are quite small. Also: what is the purpose of the insets (a-b, e-f)?
- ➔ The purpose of the insets is to better show the data points in the low ranges. The Figure 1 is now modified as below.



a) I don't get the thing about the error bars (in blue). Are these 5x, 100x or 500x larger or smaller in 'real life' than displayed on the graphs? Not clear.

→ The data uncertainties are too small to be plotted. Thus, the blue error bars in Figure 1 are magnified by 5x, 100, and 500x. We will add words in the figure caption so that we better clarify the numbers.

b) Explain what does 'hit5' mean.

→ The meaning of 'hit5' and 'hit100' are already explained in the main text of Section 3.3. However, we add a sentence explaining 'hit5' in the figure caption for better readability.

→ **Figure 1.** Comparison of CH<sub>4</sub> and N<sub>2</sub>O mixing ratios and contents obtained by different extraction methods. Shown are scatter plots between wet- and dry (hit5) extraction results of CH<sub>4</sub> (a and b) and N<sub>2</sub>O (c and d), and between control- and biocide-treated wet extraction results for CH<sub>4</sub> (e) and N<sub>2</sub>O (f). The 'hit5' denotes the dry extraction with five times hitting (see Section 3.3). Left panels (a, c, and e) and (f) present in mixing ratios of gas in bubbles, while right (b) and (d) panels in moles of gas in a unit mass of ice (gas content). The sampling locations are indicated by different symbols. The color of each data point indicates the dry soil weight in the subsamples used in control wet extraction. The 1-sigma uncertainties of the mixing ratios (a, c, e, and f) are denoted as blue error bars (see Appendix). The error bars are not visible where the error bars are smaller than markers. The grey dashed lines are 1:1 reference line. Note that the units of the axes of the insets in (e) and (f) are identical to the original plots. The p-value of two-sided Students' t-test of each comparison is denoted at the top of each plot.

Table 1, P15.

a) It is not explained why the hit100/hit5 ratios for gas content (6<sup>th</sup> column) are much lower for most of the central Yakutia samples (Cyuie), compared to the other sites? This is indeed interesting, but why? Less soil aggregates in ice-wedge samples from this site, so relatively more bubbles and thus more extracted gas?

→ Indeed, we already addressed this in the main text of Section 3.3. The low hit100/hit5 ratios of gas content in Cyuie samples are attributed to the easier-crushing characteristics of the Cyuie samples compared to the others, so that much of the enclosed gases are extracted by hit5 extraction.

Figure 2, P18.

a) Where do the samples come from? CYC-02-B and CYC-03-C likely refer to Central Yakutia (Cyuie), but what about the other samples (C-04, C-30, C-10, C-12)? Please specify somewhere.

→ The samples of 'C-##' come from Churapcha site. We modified the figure caption to specify the sample origin.

→ **Figure 2.** Comparison of wet-extracted gas and residual gas for CH<sub>4</sub> and N<sub>2</sub>O mixing ratios (a and b) and contents (c and d). The residual gas was extracted from the dry extraction method using the wet-degassed ice samples. The light green bars show the results of initial wet extraction, and the blue and red bars indicate the dry extraction of wet-degassed ice with 20- and 60-times hitting, respectively. The Cyuie samples are denoted as 'CYC', while 'C' indicates the Churapcha samples.