We would like to thank the reviewers and the Editor for their careful review of our manuscript. Our point-by-point responses to all the comments and annotations are presented below. The comments from the reviewers are shown in black, and our responses in red. Where indicated, additions to the manuscript are shown in blue and deleted text is indicated by red strikethrough, and the pages and line numbers are based on our revised (non-track) manuscript.

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Comments from the reviewer #1 and our responses:

1) GENERAL COMMENTS

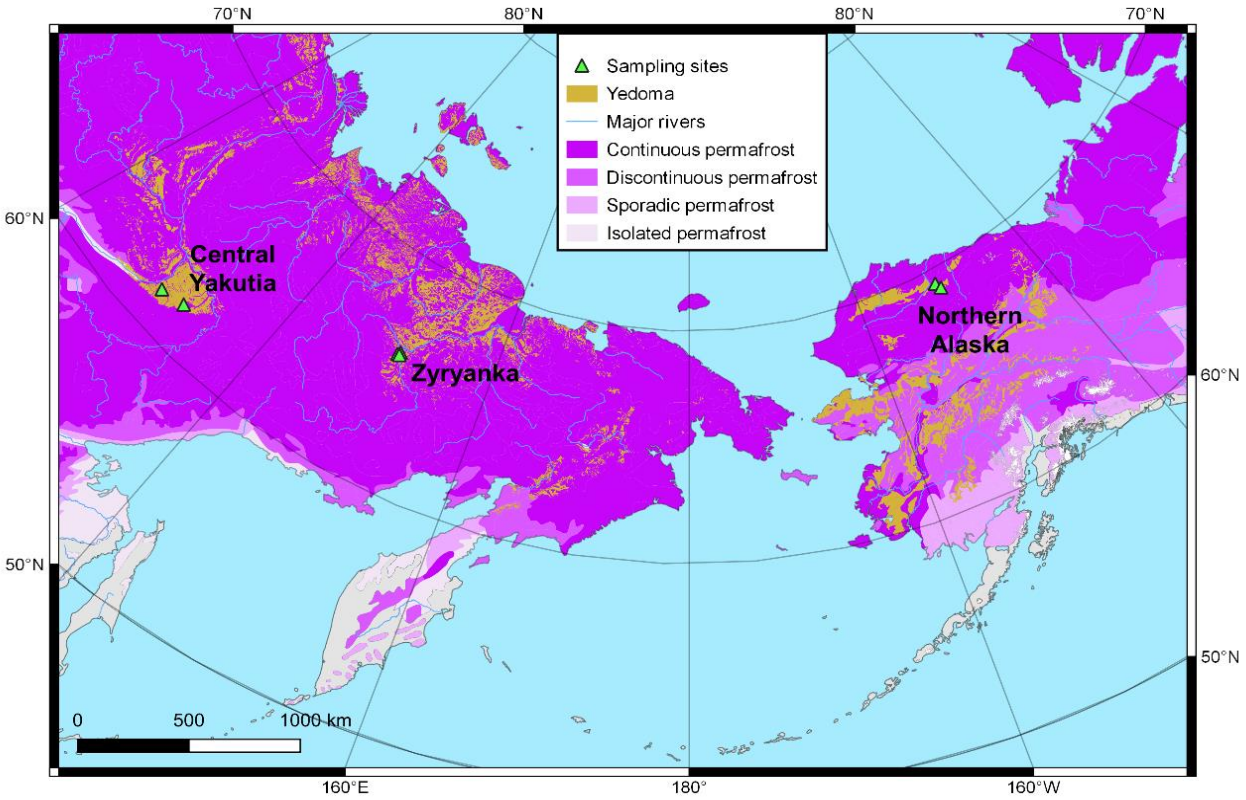
This methodological, brief communication paper reports on a comparison of different techniques (wet *vs*. dry extraction, with or without biocide to test microbial contamination) to extract gas (CH4 and N2O) from ice wedges of Alaska and Siberia. The authors report that tested methods yield good results 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). 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 implications for the large community studying on permafrost-climate interactions. Permafrost thawing is a major potential global warming threat, which is expected to input large amount of greenhouse gas (GHG) into the atmosphere. Thus, it is important to quantify the permafrost GHG budget is important for better projection of climate change. There are many works on methane (CH_4) and nitrous oxide (N_2O) concentrations either in permafrost or ground ice. However, there is currently no consensus on gas extraction methods, and no method has been tested properly. To the best of our knowledge, this study is the first 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 that while existing methods allow gas extraction from the soft parts of ice, gas adsorbed by or trapped in soil particles may not be extracted, thus affecting the measurement of GHG contents and their mixing ratios. In addition, we show that the microbial activities have a negligible effect on the wet extraction results. Although our manuscript does not deal with a <*new development*>, we believe that these findings provide <*significant advances*> and <*novel aspects of experimental methods*>. The development of a new technique is far beyond the scope of this manuscript.

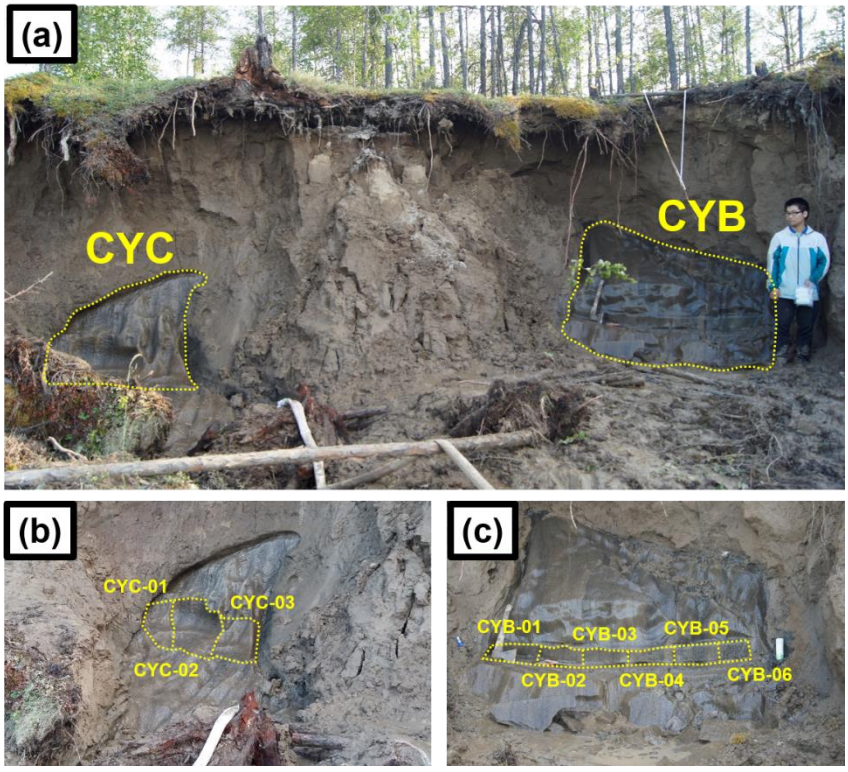
2) I understand that this is a brief communication and that the number of figures/tables is limited. However, it is really unfortunate that there is no map of the many study sites, and no picture or illustration of field sampling procedures, as well as lab instruments (especially the 'needle crushing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

➔ We add the following maps and images of the sampling sites as well as those of the gas extraction systems, in the Supplement.

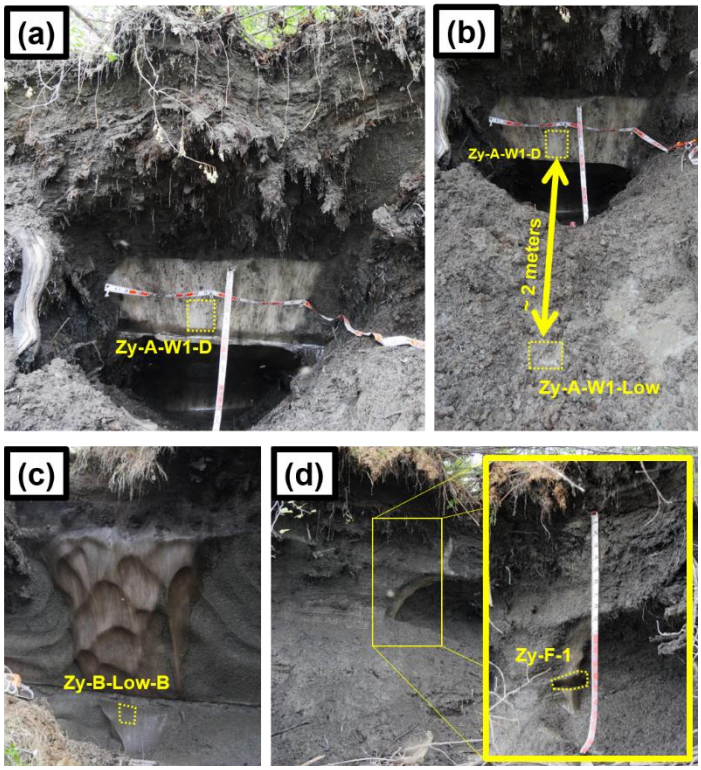


Supplementary Figure 1. The site locations of the ground ice samples used in this study are marked in the map of circum-Arctic permafrost (Brown et al., 2002), yedoma distributions (Strauss et al., 2016), and major rivers.

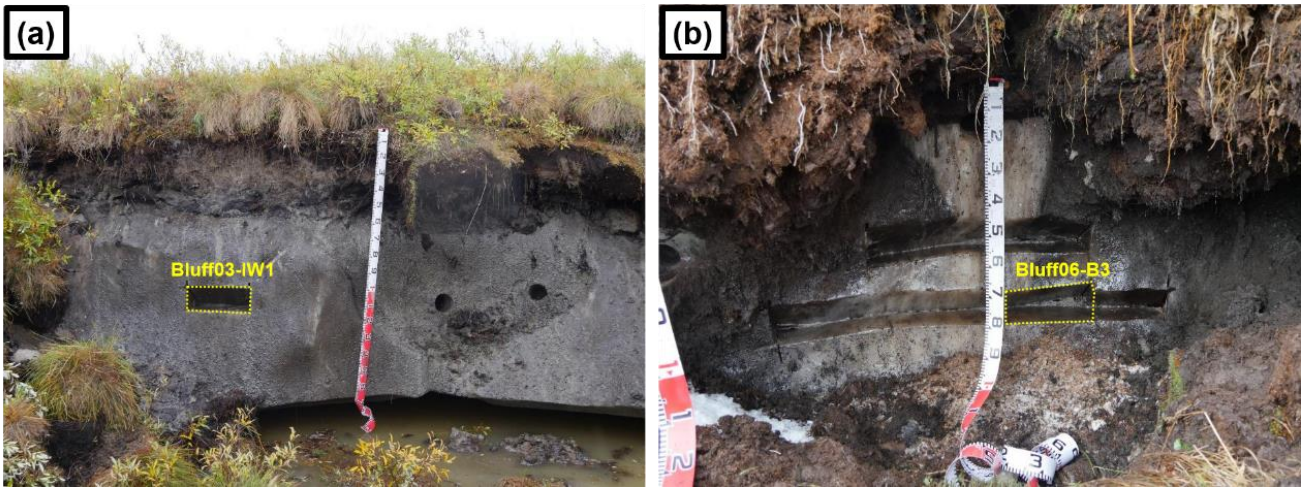
Supplementary Figure 2. Photographs of ground ice outcrops at Churapcha (central Yakutia) site. Locations of the samples used in this study are indicated by yellow dotted circles.



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.
   ➔ Our study mainly focuses on the experimental- and methodological aspects. Each set of tests was designed with logical flow. Therefore, we believe it would be easier to follow the study in the current structure than the one by the reviewer. However, as the re-arrangement does not change any fundamental content of our manuscript, we are willing to make this change if the Editor suggests 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 revise these sentences as per the specific comments below. We 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 per the reviewer's suggestions.
   ➔ (P1. L29.) Permafrost ~~soils~~ preserves 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 responses 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 above response.

P2, L35. « … which in turn can trigger positive feedbacks… ».
   ➔ The phrase is modified as per the reviewer's suggestion.
   ➔ (P2. L31-35.) 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_2), methane (CH_4), and nitrous oxide (N_2O), 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 it 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.
   ➔ This sentence is revised as follows.

➔ (P2. L37-40.) 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 CO2 production, but CH4 and N2O are generally produced under 'anaerobic' conditions, or both oxic-anoxic.

➔ This sentence is revised to include both aerobic and anaerobic respirations.

➔ (P2. L41-43.) 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_4 and N_2O to in-situ methanogenesis, nitrification, and denitrification. We note this in the sentence.

➔ (P2. L43-46.) 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.

➔ (P3. L59-62.) 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 images 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).

➔ We have shown the new maps 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.
- ➔ (P4. L88-89.) 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 believe the "ice-wedge ice" represents "the ice from ice wedges" in a more concise way. We prefer "ice-wedge ice" to just "wedge ice" for better specificity because there are wedges from different origins (e.g., sand wedges).

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

- ➔ Corrected.
- ➔ (P5. L108-109.) 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 revise the following sentence accordingly.
- ➔ (P5. L122-125.) …and a modern air sample from a surface firn at Styx Glacier, Antarctica (obtained in November 2016), which was calibrated as 1758.6 ± 0.6 ppb CH_4 and 324.7 ± 0.3 ppb N_2O 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.
- ➔ (P7. L177-178.) 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 obtained from neighboring ice-wedge pieces were highly variable. Our previous work (Kim et al., 2019) showed the centimetre-scale variability of the gas mixing ratios in ice wedges. We change the relevant sentence as follows:
- ➔ (P8. L190-193.) We note~~d~~ 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.

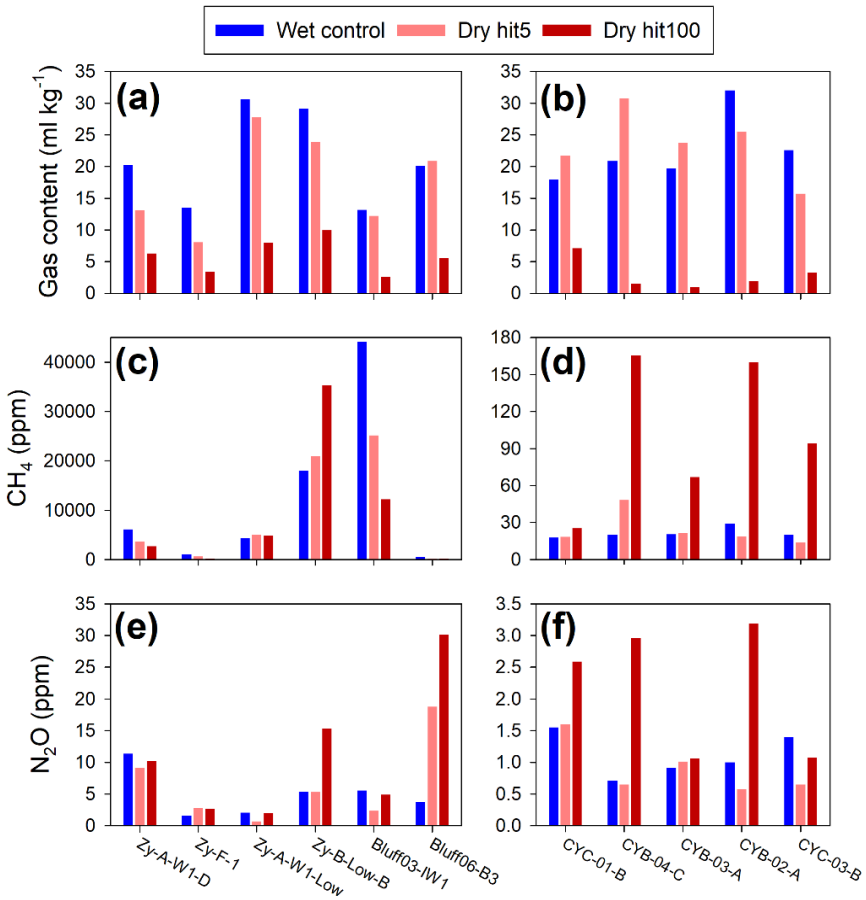
P11, L224. «… polar ice core samples… » (remove the 2nd 'ice')

➔ We delete the repetition of 'ice'.

➔ (P11. L227-228.) 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. We used a table originally because of the large ranges of CH_4 and N_2O mixing ratios, which make figures less readable. Below we have made a figure (grouped bar graphs) as an example. Because there are large variations among the samples, some data are difficult to see, particularly for samples with lower mixing ratios. Thus, we prefer use Table 1; however, we will follow the Editor's decision.

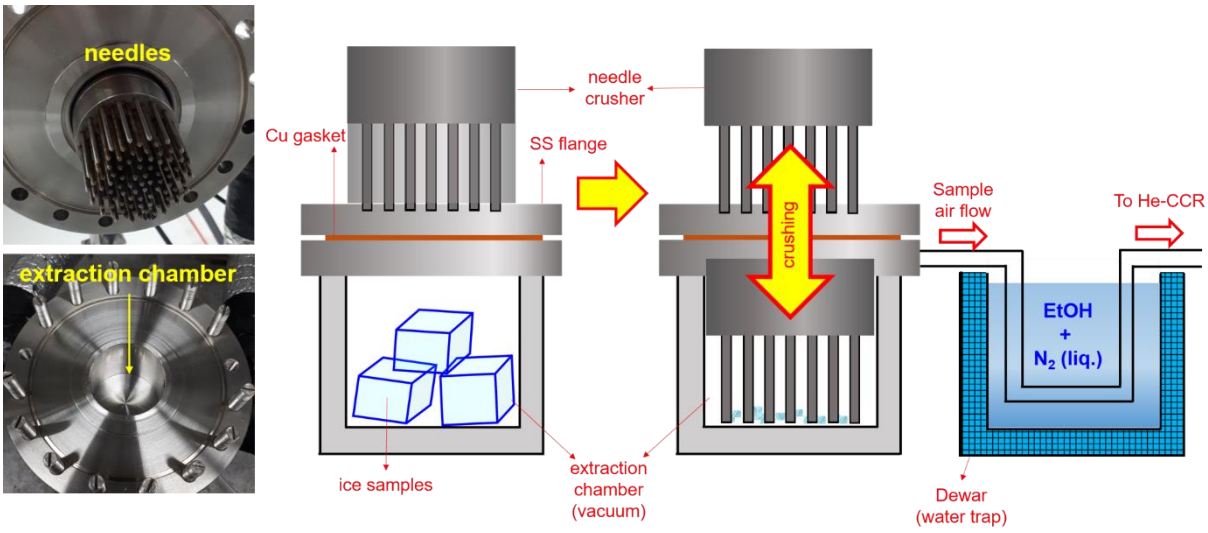
P12, L250-252. This statement is based on what result? Can we see this displayed somewhere in a figure/table? If yes, please refer to it in the text.

➔ We add that the comparisons with the dry soil content are shown both in Figures 1 and A3.

➔ (P12. L254-256.) 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.

➔ Detailed descriptions and schematic diagram of the needle crushing system can be found in Ahn et al. (2009) and Shin (2014). We add images of the needle crushing system to the Supplement.



Supplementary Figure 6. Schematic diagram of the needle-crusher method together with enlarged photographs of crushing needles (left top), and extraction chamber (left bottom).

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

➔ This sentence is our interpretation of the comparisons of hit5 with hit100 extractions, listed in Table 1. We note this at the end of the sentence.

➔ (P13. L284-287.) 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).

P13, L288-290. Please support this statement by relevant references. In fact, the simple association N2O=oxic / CH4=anoxic is not entirely and always true. For example, N2O production has been recorded under both oxic and anoxic conditions (Gil et al. 2017; *Global Biogeochemical Cycles*), as well as CH4 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 follows:

➔ (P13. L292-295.) 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 the current structure is easier to read for the same reason discussed above. Please find our response to General Comment #3.

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

➔ We wish to retain the current structure for the same reason mentioned in the response to General Comment #3.

P17, L350-352. This is indeed interesting. Do we know why N2O appears to be more extractable (or less present in the residual adsorbed phase) than CH4, 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_2O could make the adsorbed N_2O more extractable by wet extraction, compared with CH_4. To the best of our knowledge, this is reported for the first time by our manuscript. Thus, we add a sentence mentioning this as below:

➔ (P17. L355-359.) These results imply that most of the N_2O in ice wedges is extracted by three melting-refreezing cycles, such that only a small amount of N_2O is left adsorbed or entrapped in ice-wedge soils. The authors posit that this might be attributed to the high solubility of N_2O to water compared to CH_4.

P18, L375. «… easy to extract… »

➔ The typo is corrected.

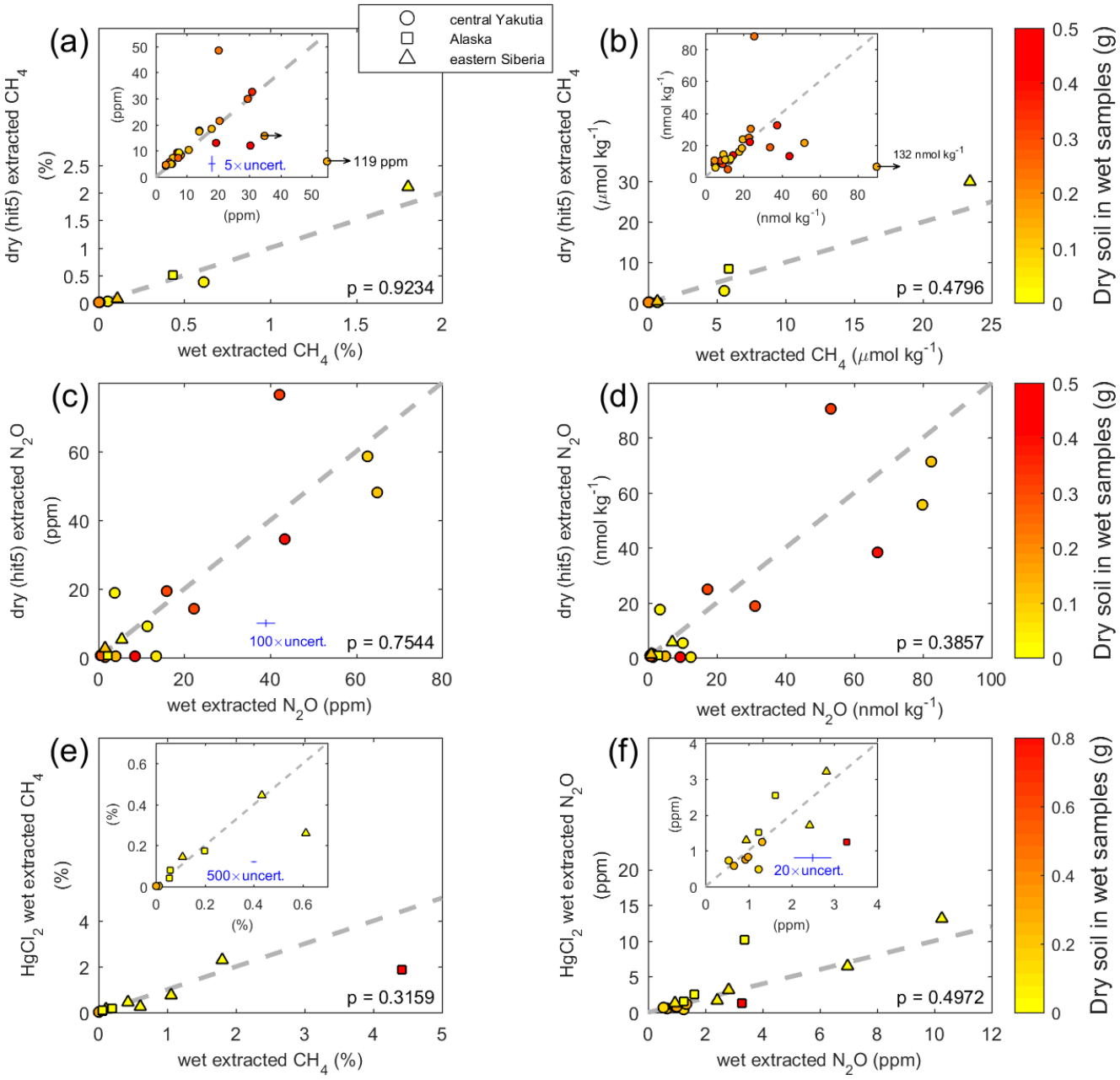
P19, L397. « Our findings indicate that ... »

➔ 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 insets better show the data points in the low ranges. The modified version of Figure 1 is shown 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 modify the figure caption for clarity. See our response below.

b) Explain what does 'hit5' mean.
   ➔ The meaning of 'hit5' and 'hit100' are already explained in the main text of Section 3.3. In addition, we add a sentence explaining 'hit5' in the figure caption for clarity.
   ➔ (P9.) **Figure 1.** Comparison of CH_4 and N_2O mixing ratios and contents obtained by different extraction methods. Shown are scatter plots between wet- and dry (hit5) extraction results of CH_4 (a and b) and N_2O (c and d), and between control- and biocide-treated wet extraction results for CH_4 (e) and N_2O (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 magnified by 5x, 20x, 100x, and 500x as 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 bottom right corner~~top~~ of each plot.

Table 1, P15.

a) It is not explained why the hit100/hit5 ratios for gas content (6th 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?
   ➔ We 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 with 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.
   ➔ (P18.) **Figure 2. Comparison of wet-extracted gas and residual gas for CH_4 and N_2O 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.

Comments from the reviewer #2 and our responses:

1) GENERAL COMMENTS

The paper reports novel aspects of experimental methods of gas extraction techniques. Currently, there are no works that conducted such a comparison, which has long been needed due to huge number of gas measurements in permafrost area conducted recently. Unfortunately, there is still no unified method for carrying out gas extraction that leads to the impossibility and impropriety of comparison the results obtained by different research groups.

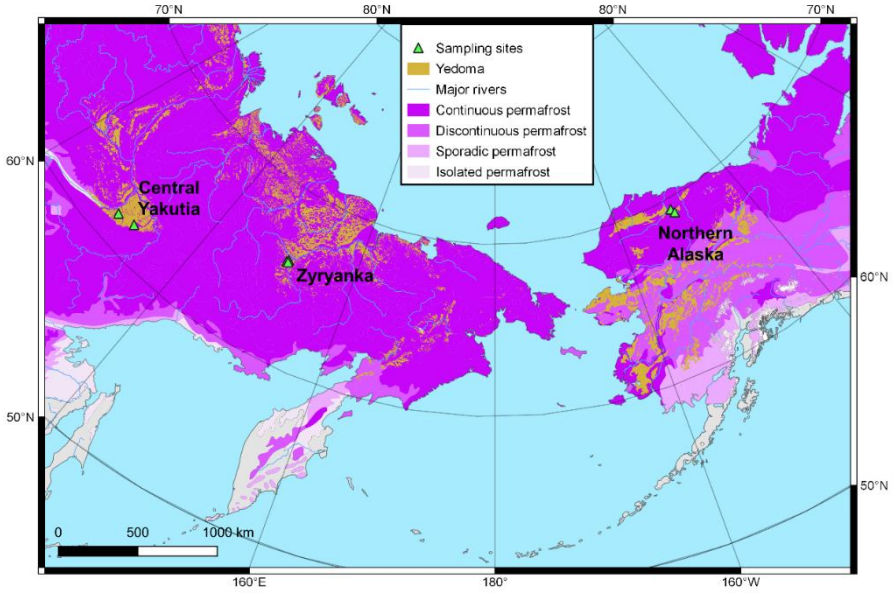
The authors tested conventional wet and dry gas extraction methods for ice wedges coming to conclusion that current estimates of ground-ice gas budgets are likely underestimated. They found insignificant effects of microbial activity during wet extraction and significant difference in extraction results from polar ice cores and ice wedges. Therefore, the manuscript is of big interest for scientific community and contributes to changing our scientific understanding of a subject as it is has to be for TC.

The results are presented in well-structured way and the paper is easy to read.

However, there are a few general suggestions that could improve the article:

1) I suggest adding a map of study sites, maybe some geological sections to get a better idea of the location and structure of ice wedges. Are they all Pleistocene?

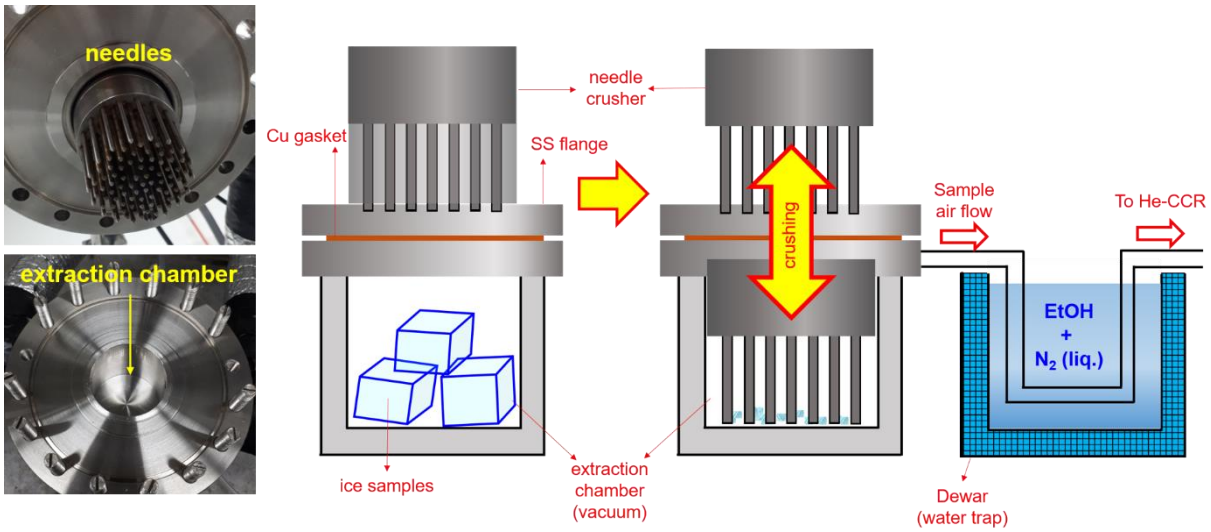
➔ We agree with the reviewer and are including a map (see below) in our Supplement. The ages of the studied ice wedges have not been analysed, as they are beyond the scope of our manuscript.



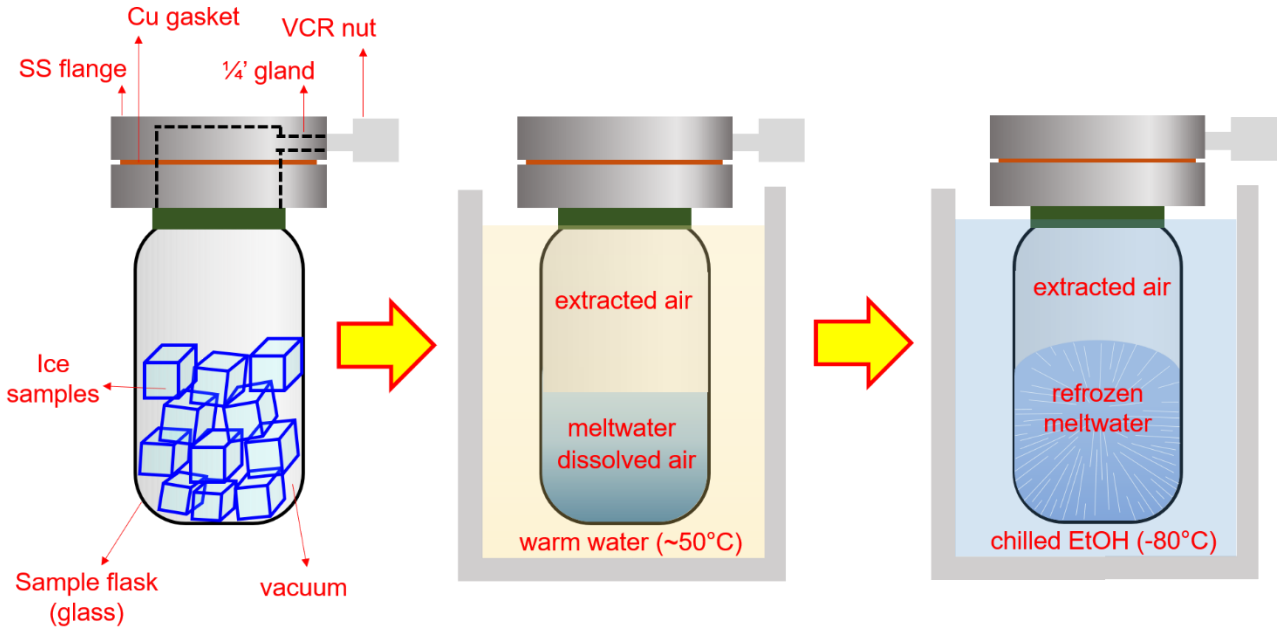
Supplementary Figure 1. The site locations of the ground ice samples used in this study are marked in the map of circum-Arctic permafrost (Brown et al., 2002), yedoma distributions (Strauss et al., 2016), and major rivers.

2) Besides it is really necessary to include the schemes of gas extraction procedures (both wet and dry techniques as well as the experiment on dry extraction efficiency and on residual gas contents after wet extraction. Due to the limitation of the number of figures both 1 and 2 can be added as supplementary material.

➔ We add figures to the Supplement for clarity. As we already cited in the text, the details of the extraction system and procedures are well described in Ahn et al. (2009) and Shin (2014) for dry extraction and Yang et al. (2017) and Ryu et al. (2018) for wet extraction.



Supplementary Figure 6. Schematic diagram of the needle-crusher method together with enlarged photographs of crushing needles (left top), and extraction chamber (left bottom).



Supplementary Figure 7. Schematic diagram of melting-refreezing (wet extraction) procedure used in this study. More details about the wet extraction line and GC systems are described in Yang et al. (2017) and Ryu et al. (2018).

3) Since the article is devoted to the comparison of methods, it would be useful to estimate the limits of applicability of the methods and measurement errors.

➔ We agree with the reviewer's comment, but the limits of applicability and measurement errors of both methods were already described in the main text (Section 3.3 and 3.4) and Appendix, respectively.

4) It is necessary to add initial data on gas content and CH_4 and N_2O mixing ratios as supplementary material to prove you main result about the same effectiveness of wet and dry extraction methods. As I see now from the Table 1 there can be 2 times difference (up to 20000 ppm) for CH_4

➔ We add our original data to our Supplement.

➔ We provide the relative amount of extracted gas in Table 1.

So the article is of big interest but needs major revision to be accepted and I would be happy to review a revision of the paper.

2) SPECIFIC COMMENTS AND EDITORIAL SUGGESTIONS

P.2 L50-51. «…ice sample was melted in a saturated sodium chloride (NaCl) solution, in order to minimize microbial activity and gas dissolution (Cherbunina et al., 2018 and references therein)». I found no mention in the article that NaCl was used in order to minimize microbial actvity, is it really there?

➔ We correct the reference as below:

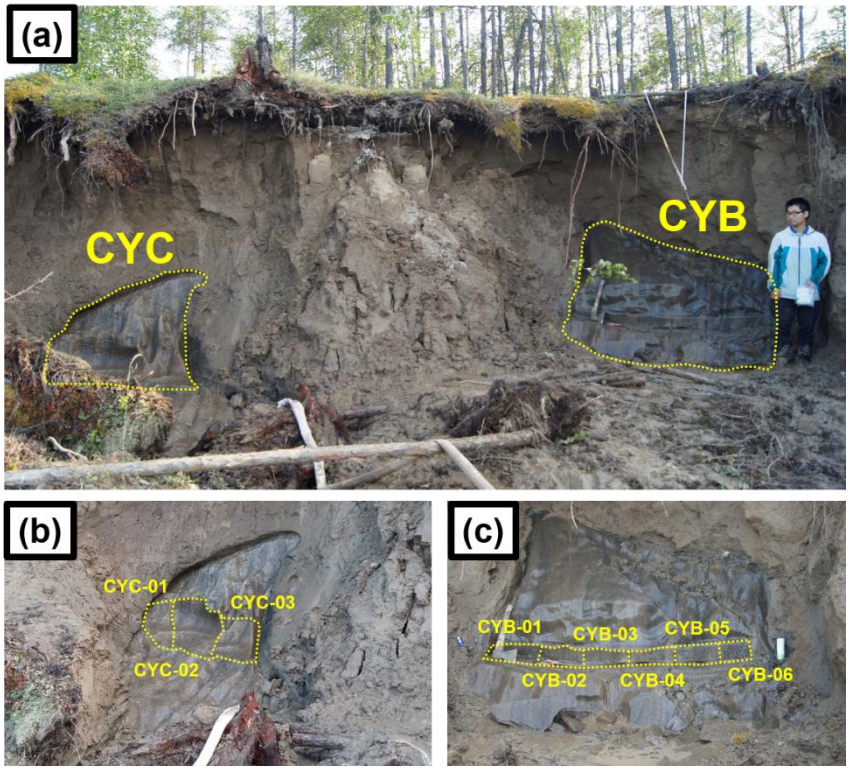
➔ (P2. L49-52.) Other studies conducted by Russian scientists used an on-site melting method in which a large (1–3 kg) block of ground ice sample was melted in a saturated sodium chloride (NaCl) solution, in order to minimize ~~microbial activity and~~ gas dissolution (Arkhangelov and Novgorodova, 1991~~Cherbunina et al., 2018 and references therein~~).

➔ (P27. L488-490.) Arkhangelov, A. A., and Novgorodova, E. V.: Genesis of massive ice at 'Ice Mountains', Yenesei River, Western Siberia, according to results of gas analyses, Permafrost Periglac. Proc., 2, 167-170, http://doi.org/10.1002/ppp.3430020210, 1991.

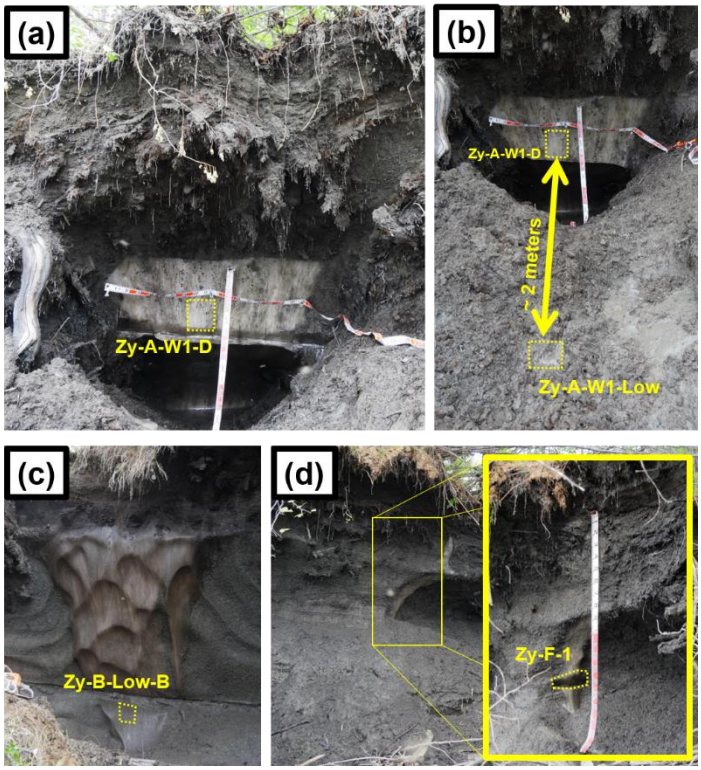
P.2 L72. Please specify the size of the samples, add the site map and geological sections with sampling location

➔ A map of sampling sites is added as a Supplement Figure 1 (please refer to our response to General Comment #1 above). We also add images of sampling site outcrops.

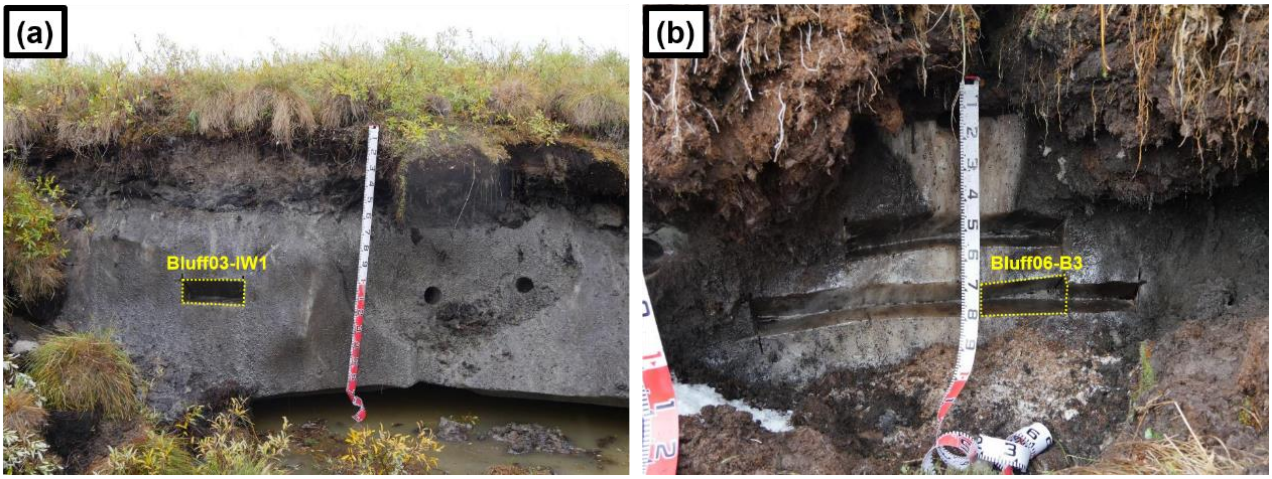
Supplementary Figure 2. Images of ground ice outcrops at Churapcha (central Yakutia) site. Locations of the samples used in this study are indicated by yellow dotted circles.



Supplementary Figure 3. Images 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. Images 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. Images 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.

P.2 L104. Please include the schemes of gas extraction procedures

➔ Please refer to our response to General Comment #2.

P.2 L108. Why it is used precisely 5 times, not 100, can you reason it somehow?

➔ We have empirical knowledge that the polar ice core samples are well crushed within five times hitting during our dry extraction (needle-crusher) at SNU. For consistency of the analytical setup, we used an identical procedure for the dry extraction of ice-wedge samples to test whether the dry extraction method is applicable. In the meanwhile, tests with 100-times hitting were designed to understand the gas extraction efficiency and differences in gas mixing ratios for easily- and hardly crushed portions of ice wedges.

P.2 L144. It is not clear where did you get the dry soil mass before the extraction. «Taking the dry soil mass of the analysed samples (0.33 g) into account, we added 24 µL of saturated HgCl2 solution (at 20°C) to the sample flasks» Were there used the data on dry soil mass from the other samples? Because later you say « Dry soil content was measured using the leftover meltwater from the control-wet extraction tests. »

➔ We obtained the dry soil mass (0.33 g) from the leftover meltwater samples of the **previous** wet extractions, which was done to compare dry- and wet extractions. We revised the sentence for clarity.

➔ (P2. L145-150.) For biocide-treated tests, 1.84 mmol of mercuric chloride ($HgCl_2$) was applied per unit kilogram of soil, following established procedures for soil sterilization (Fletcher and Kaufman, 1980). We obtained the average dry soil mass (0.33 g) from the leftover meltwater samples of the previous wet extractions, which were carried out for comparison between dry- and wet extractions. Taking the average dry soil mass ~~of the analysed samples~~ (0.33 g) into account, we added 24 µL of saturated $HgCl_2$ solution (at 20°C) to the sample flasks.

P.12 L249. Please specify what do you mean by «ice hardness» here. As in L 249 «the extraction efficiency of the needle crusher not only depends on site characteristics, but also on the individual ice sample hardness», and later L. 251 «no relationship was observed between the dry soil content and the extraction efficiency», but L.274 « soil-rich ice has greater hardness than the soil-poor ice». I guess this is the matter of «soil aggregates» as you mention later, so the hardness in this case is defined by this parameter? Is it possible to quantify this?

➔ We observed that the samples with large-sized soil aggregates were difficult to crush with our needle-crusher system. Because no significant relationship was found between dry soil content and the extraction efficiency, the important parameter controlling the hardness may be the presence of large-sized aggregates, rather than just the soil content. Unfortunately,

we do not have quantitative measurements of size (or volume) of each soil aggregate. Further study with three-dimensional image analysis will be useful to address this.

➔ To clarify this issue, we reword the sentence L.274 “soil-rich ice has greater hardness than the soil-poor ice” as follows:

➔ (P13. L276-280.) Thus, the hit5 CH_4 mixing ratios of the Cyuie samples may more reflect the gas mixing ratios in bubbles, while the hit100 results reflect more of the contribution from gas adsorbed on soil and trapped within soil aggregates than the hit5 results because ~~soil-rich ice~~the ice sample containing larger-sized aggregates has greater hardness than ~~the soil-poor ice~~those with smaller aggregates or fine particles.

P.12 L255. Please specify the size range for «This is because the large-sized uncrushed soil aggregates or particles may have prohibited the needle crusher from crushing the small-sized ice flakes or grains». As the presence of the aggregates is one of the main limits to use the technique, is it possible to make at least a rough estimate of the amount of gas that can remain there?

➔ Although we have no quantitative measure of the size of soil aggregates, the aggregates in the studied ice wedges were observable by the naked eye as there was a clear contrast of darkness when back-lighted. Empirically, the size of observable aggregates ranged from millimetres to centimetres.

➔ However, it was not possible to estimate the amount of gas remaining in the uncrushed portion (mostly Zyryanka and Bluff samples), because the amount of entrapped gas is unknown. In contrast, for easily crushed samples (i.e. Cyuie samples), the hit100/hit5 ratio of gas content could be used to estimate the gas amount in soil aggregates. However, we cannot recommend this estimation because a certain portion of gas in soil aggregates could have been extracted by the hit5 procedures, and the amount of soil aggregates that are crushed or uncrushed after the hit5 procedures remains unknown.

P.12 L259. «Therefore, we do not recommend using a needle crusher system to measure gas contents in ice-wedge samples». Can you estimate the efficiency of the method in % in the same way it has been done for polar ice core ice samples (80–90%) (Shin, 2014)? As I see from the Table 1 the procedure «Hit5+Hit100» in most cases allows to extract more gas then the wet method even if uncrushed aggregates still occur. Can you recommend using dry extraction method in this modification?

➔ We cannot recommend this method. To measure the gas content precisely, near-perfect gas extraction from ice wedge samples is required. As the reviewer noted, the amount of gas extracted from both hit5 and hit100 procedures is generally higher than wet extraction. However, we hesitate to make a general statement because uncrushed soil aggregates may still exist even after hit100 extraction, depending on sampling locations and individual samples.

P.12 L272. Since you talk about gas in bubbles here : «the hit5 CH_4 mixing ratios of the Cyuie samples may more reflect the gas mixing ratios in bubbles, while the hit100 results reflect more of the contribution from gas adsorbed on soil and trapped within soil aggregates than the hit5 results» and further, may be it would be useful to get the data on ice porosity to compare with the results of extracted volume of gas since the volume of gas normalized to layer pressure approximately corresponds to porosity.

➔ This is a great idea. However, the relationship between the porosity and the normalized volume is applicable only when the gas extraction efficiency is close to 100%, or near constant. The gas extraction efficiency of the ice wedge is highly variable and difficult to measure precisely, limiting the estimation of the porosity.

P.16 L320. Please explain if I understand correctly the next paragraph:

«To examine how well the gas is extracted by wet extraction, we applied the dry extraction method to refrozen ice-wedge samples after wet extraction. We first prepared degassed ice-wedge samples that had undergone repetitive wet extractions (wet-degassed ice hereafter). Once the wet extraction experiments were completed, we repeated two cycles of melting-refreezing and evacuation procedures to degas the ice melt. After degassing by a total of three cycles of wet extraction and evacuation, the outermost surfaces (~2 mm) of the wet degassed ice were trimmed away in the walk-in freezer at SNU on the morning of experiments. The wet-degassed ice was then inserted into the needle crusher and the crusher chamber was evacuated. A specific amount of standard air was injected. Then, the wet-degassed ice samples were hit 20 or 60 times by the needle crusher.»

After the first freezing-melting cycle the sample gas in the headspace of the flask was collected and gas content, CH_4 and N_2O ratio was measured. Then the two cycles were conducted. (and my question here is what happened to the gas in the flask-was it collected and measured or just evacuated) and next step was measuring the gas content in degassed ice through needle-crusher procedure.

- ➔ The reviewer has understood the text correctly. Regarding the question, we did not collect the gas extracted by second and third cycles of melting-refreezing procedures.

P.16 L328. Explain please why were such parameters chosen if in the previous dry extraction procedure you used 5 and 100 hits: « Then, the wet-degassed ice samples were hit 20 or 60 times by the needle crusher»

- ➔ The main goal of the tests with wet-degassed ice samples was to know if any gas remained after three cycles of wet extraction. We chose hitting 20 times instead of 5 times because significant amount of gas was already extracted during the three cycles of wet extractions. We also chose hitting 60 times to check if there is a significant difference in the amount of the extracted gas between hitting 20- and 60 times.

P.16 L331. The tests using the wet-degassed ice show an additional gas extraction of 43 to 88% of the amount of gas extracted during the initial wet extraction. I suggest to add this information to the conclusions as it is of big significance as well as if I get it right the best way of degassing the sample according to your manuscript is to combine three cycles of wet extraction with dry extraction for the residual gas. I think this has to be one of the main conclusion.

- ➔ The more number of wet and dry extractions, the better the gas extraction efficiency. We would prefer to leave the conclusions as is, since we cannot specify the best combination in numbers of the two extraction methods.

P.19 L391. Please specify what do you mean by «relatively soft ice wedges».

- ➔ Here we refer to the ice wedges that are more easily crushed than others by the hit5 procedure. To specify this, we revise the sentence as follows:
- ➔ (P19. L400-403.) In the meantime, we propose that both existing techniques may be suitable for gas mixing ratio measurements for bubbles in relatively soft ice wedges (i.e., easily crushed ice wedges by a hit5 extraction, e.g., Cyuie ice wedges in this study).

P.19 L 392 It seems to me that you have very good results of applying the method of three times wet extractions + residual gas extracted by a needle crusher for N_2O and I don't get why there is in conclusions «Exceptionally, the N2O content in ice wedges may be measured by using repeated wet extractions, but this is not the case for determining the N2O mixing ratio»

- ➔ As the reviewer pointed out, our results indicate that repeated melting-refreezing procedures extract most of the N_2O from ice wedges. However, we cannot guarantee the N_2O mixing

ratio because the relative extraction efficiency for each gas species may be variable. To clarify, we add below sentences in the main text.

➔ (P18. L374-376.) It should be noted that combination of repetitive wet extractions with dry extraction does not guarantee reliable estimation of N_2O mixing ratio, because extraction efficiency of the other gas components may be different from that of N_2O.

3) FIGURES AND TABLES

Table 1. Add a column of dry soil content as in table 2

➔ We revise Table 1 as suggested (see next page).

The table with the data used for Fig.1 need to be added to get the difference between wet and dry method results.

➔ We add a table for the data used for Fig. 1 in the Supplement.

Site Location	Sample	soil content	gas content				CH_4 mixing ratio				N_2O mixing ratio			
			Wet control	Dry hit5	Dry hit100	hit100/hit5	Wet control	Dry hit5	Dry hit100	hit100/hit5	Wet control	Dry hit5	Dry hit100	hit100/hit5
		wt. %	ml/kg	ml/kg	ml/kg		ppm	ppm	ppm		ppm	ppm	ppm	
Zyryanka, Northeastern Siberia	Zy-A-W1-D	0.155	20.2	13.1	6.3	0.48	6138	3713	2721	0.7329	11.37	9.10	10.15	1.12
	Zy-F-1	0.618	13.5	8.1	3.4	0.42	1080	655.6	173.5	0.2646	1.57	2.81	2.65	0.942
	Zy-A-W1-Low	0.049	30.6	27.8	8.0	0.29	4309	5073	4818	0.9497	2.07	0.69	2.02	2.9
	Zy-B-Low-B	0.107	29.1	23.9	10.0	0.418	18030	21010	35290	1.680	5.37	5.32	15.36	2.89
Northern Alaska	Bluff03-IW1	2.07	13.2	12.2	2.6	0.21	44160	25230	12240	0.4851	5.58	2.36	4.93	2.09
	Bluff06-B3	0.078	20.1	20.9	5.6	0.27	558.7	164.2	219.5	1.337	3.74	18.78	30.14	1.605
Cyuie, Central Yakutia	CYC-01-B	0.252	18.0	21.7	7.1	0.33	18.0	18.3	25.4	1.39	1.55	1.60	2.59	1.62
	CYB-04-C	0.498	20.9	30.7	1.5	0.049	20.2	48.4	165.6	3.42	0.71	0.65	2.96	4.5
	CYB-03-A	0.420	19.7	23.7	1.0	0.041	20.5	21.5	67.1	3.12	0.91	1.01	1.06	1.05
	CYB-02-A	0.403	32.0	25.5	1.9	0.073	29.1	18.7	159.8	8.55	1.00	0.58	3.19	5.5
	CYC-03-B	0.830	22.6	15.7	3.3	0.21	20.3	13.9	94.5	6.80	1.40	0.65	1.08	1.7

Brief Communication: The reliability of gas extraction techniques for analysing CH_4 and N_2O compositions in gas trapped in permafrost ice-wedges

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Abstract. Methane (CH_4) and nitrous oxide (N_2O) compositions in ground ice may provide information on their production mechanisms in permafrost. However, existing gas extraction methods has not been well tested. We test conventional wet and dry gas extraction methods using ice-wedges from Alaska and Siberia. We find that both methods extract gas from the easily extractable parts of the ice (e.g., gas bubbles), and yield similar results for CH_4 and N_2O mixing ratios. We also find insignificant effects of microbial activity during wet extraction. However, both techniques are unable to fully extract gas from the ice, presumably because gas molecules adsorbed onto or enclosed in soil aggregates are not easily extractable. Estimation of gas production in subfreezing environment of permafrost should consider the incomplete gas extraction.

1. Introduction

Permafrost ~~soils~~ preserves 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. 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_2), methane (CH_4), and nitrous oxide (N_2O), which in turn can trigger positive feedbacks (e.g., Salmon et al., 2018). In addition, the projected polar amplification (e.g., Masson-Delmotte et al., 2013) may strengthen these positive feedbacks. 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).

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). 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). However, analytical methods remain poorly scrutinized. Boereboom et al. (2013) utilized the conventional melting-refreezing method (wet extraction) used in polar ice core analyses. In this technique, the ice samples were melted under a vacuum to liberate the enclosed gases, then refrozen to expel the dissolved gases present in the meltwater. Other studies conducted by Russian scientists used an on-site melting method in which a large (1–3 kg) block of ground ice sample was melted in a saturated sodium chloride (NaCl) solution, in order to minimize ~~microbial activity and~~ gas dissolution (Arkhangelov and Novgorodova, 1991~~Cherbunina et al., 2018 and references therein~~). A recent study instead used a dry

extraction technique to prevent the microbial activity during wet extraction (Kim et al., 2019), which employed a needle-crusher in a vacuum to crush approximately 10 g of ice sample without melting (Shin, 2014).

In this study, for the first time we test the reliability of both wet and dry extraction methods for CH_4 and N_2O mixing ratios and contents (volume or moles of gas in a unit mass at standard temperature and pressure conditions (STP)) using permafrost ground ice samples. Ice-wedge samples from Alaskan and Siberian permafrost were used because ~~the ice-wedge is~~ice wedges are 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). More specifically, this study aims to address the following scientific questions: 1) Do wet and dry extraction methods yield different results? 2) Are the melting-refreezing results affected by microbial activity during gas extraction? 3) How effectively does the wet/dry extraction extract gases from ice wedges? To address the first question, CH_4 and N_2O results from dry and wet extractions were compared. For the second question, we applied the wet extraction method to both biocide-treated and control samples. Finally, for the third question we carried out tests with and without extended number of hitting ice with a needle system in a crushing chamber, as well as additional dry extraction from ice samples that had been degassed by our wet extraction method.

2. Materials and Methods

2.1.Ice samples and sample preparation

The ice-wedge samples used in this study were collected from Churapcha, Cyuie (central Yakutia), and Zyryanka (north-eastern Yakutia) in Siberia, as well as from northern Alaska (Supplementary Figure 1). The Churapcha site (61.97°N, 132.61°E) is located approximately 180 km east of Yakutsk. The Cyuie site (61.73°N, 130.42°E) is located

approximately 30 km southeast of Yakutsk. The Cyuie samples were collected from two outcrops (CYB and CYC) (Kim et al., 2019). At each site, 30 cm long ice-wedge cores were drilled perpendicular to the outcrop surface (Supplementary Figures 2 and 3).

Zyryanka is located in the southern boreal region of the Kolyma River, at the junction of the Chersky and Yukaghir Ranges, in a region affected by thermokarst development (Fedorov et al., 1991). Site A (Zy-A) is located on a tributary of the Kolyma River, approximately 22 km north of Zyryanka. Site B (Zy-B) is approximately 14 km west of the start of the Kolyma tributary, which begins ~11 km north of Zyryanka. Site F (Zy-F) is located approximately 4 km west of the tributary that leads to site B. The ground ice samples were collected from riverbank walls exposed by lateral erosion using a chainsaw (Supplementary Figure 4). Most of the outcrops that were sampled for ground ice were on the first (lowest) terrace of the river.

For the Alaskan sampling locations, Bluff03 (69.40°N, 150.95°W) and Bluff06 (69.14°N, 150.61°W) are located in the Alaska North Slope region, approximately 120 and 150 km from the Arctic Ocean, or 100 and 70 km northwest of the Toolik Field Station (68.63°N, 149.59°W), respectively. Samples from Bluff03 were collected from the bluff walls that had developed by gully formations on a gentle slope of the Yedoma using a chainsaw. Samples of Bluff06 were collected from outcrops within eroded frozen peatland in a thaw lake basin (Supplementary Figure 5). All the ice-wedge samples used in this study were stored in a chest freezer at < -18°C before analysis.

The ice-wedge ice is most different from polar ice cores, in that their gas mixing ratios are not homogeneous (e.g., Kim et al., 2019), which may hinder exact comparison with results from adjacent ice samples. We therefore randomly mixed sub-samples to reduce the effect of the heterogeneous gas composition distribution (random cube method hereafter). Approximately 100–200 g of an ice-wedge sample was cut into 25 to 50 cubes of 3–4 g each,

and for each experiment, ~10 to 12 cubes were randomly chosen so that the total weight of the sub-sample was ~40 g.

2.2.Gas extraction procedures

Dry extraction (needle crusher)

For dry extraction, we used a needle-crusher system at the Seoul National University (SNU, Seoul, South Korea) (Shin, 2014). In brief, 8~13 g of ice sample ~~was~~ were crushed in a cold vacuum chamber (extraction chamber). The ice samples were usually hit five times by the needle set. The temperature within the extraction chamber was maintained at -37°C by using a cold ethanol-circulating chiller. The extracted gas was dried by passing it through a water vapor trap at -85°C and cryogenically trapping it in a stainless-steel tube (sample tube) at approximately -257 °C using a helium closed-cycle refrigerator (He-CCR). Since the extraction chamber cannot accommodate ~40 g of ice at once, the ~40 g of random cube sub-samples were extracted using three sequential extractions and the gas liberated from each extraction was trapped in a sample tube.

Following extraction, the sample tubes were detached from the He-CCR, warmed to room temperature (~20°C), and attached to a gas chromatograph (GC) equipped with an electron capture detector (ECD) and a flame ionization detector (FID) to determine the mixing ratios of CH_4 and N_2O. Details of the GC system are given in Ryu et al. (2018). The daily calibration curves were established using working standards of 15.6 ± 0.2 ppm CH_4, 10000 ± 30 ppm CH_4, 2960 ± 89 ppb N_2O, 29600 ± 888 ppb N_2O, and a modern air sample from a surface firn at Styx Glacier, Antarctica (obtained in November 2016), which was calibrated as 1758.6 ± 0.6 ppb CH_4 and 324.7 ± 0.3 ppb N_2O by the National Oceanic and Atmospheric Administration (NOAA).

130 ***Wet extraction (melt-refreeze)***

131 For the control and $HgCl_2$-treated wet extraction experiments, a melting-refreezing wet
132 extraction system at SNU was employed (Yang et al., 2017; Ryu et al., 2018). The gas
133 extraction procedure is identical to the procedure described in Yang et al. (2017) and Ryu et al.
134 (2018), except for the sample gas trapping procedure (see below). Ice-wedge sub-samples of
135 ~40 g (composed of 10–12 ice cubes for each) were placed in a glass container welded to a
136 stainless-steel flange (sample flask), and the laboratory air inside the sample flasks was
137 evacuated for 40 min. The sample flasks were then submerged in a warm (~50°C) tap water
138 bath to melt the ice samples. After melting was complete, the meltwater was refrozen by
139 chilling the sample flasks with cold ethanol (below -70°C). The sample gas in the headspace
140 of each sample flask was then expanded to the volume-calibrated vacuum line to estimate the
141 volume of extracted gas, and trapped in a stainless-steel sample tube by the He-CCR device.
142 In this study, we attached the He-CCR device to our wet extraction line and the gas samples in
143 the flasks were cryogenically trapped. The reasons for using He-CCR instead of direct
144 expansion to a GC are twofold: 1) to better compare the dry and wet extraction methods by
145 applying the same trapping procedure, and 2) to maximize the amount of sample gas for GC
146 analysis, because the gas expansion from a large flask allows only a small fraction of gas to be
147 measured by the GC.

148 For biocide-treated tests, 1.84 mmol of mercuric chloride ($HgCl_2$) was applied per unit
149 kilogram of soil, following established procedures for soil sterilization (Fletcher and Kaufman,
150 1980). We obtained the average dry soil mass (0.33 g) from the leftover meltwater samples of
151 the previous wet extractions, which were carried out for comparison between dry- and wet
152 extractions. Taking the average dry soil mass ~~of the analysed samples~~ (0.33 g) into account, we
153 added 24 µL of saturated $HgCl_2$ solution (at 20°C) to the sample flasks. The flasks with $HgCl_2$
154 solution were then frozen in a deep freezer at < -45°C to prevent the dissolution of ambient air

into the solution during ice sample loading. After the wet extraction procedure was complete, the extracted gas was trapped in a sample tube and the CH_4 and N_2O mixing ratios were determined using the same GC-ECD-FID system as the dry-extracted gas. The resulting CH_4 and N_2O mixing ratios have not been corrected for partial dissolution in ice melt in the flasks, because CH_4 and N_2O trapped in refrozen ice are negligible compared to the ranges of the systematic blanks (see Appendix).

2.3.Gas content

The analytical methods described previously are for determining the mixing ratios of CH_4 and N_2O in the extracted gas. To convert these mixing ratios into moles of CH_4 and N_2O per unit mass of ice-wedge sample (CH_4 and N_2O content, respectively, hereafter) requires data regarding the amount of gas extracted. The gas content is a measure of gas volume enclosed in a unit mass of ice sample at STP (in mL kg_{ice}^{-1}). Thus, the CH_4 and N_2O contents can be calculated using the gas content, the total mass of the random cube ice, and the gas mixing ratio. The gas content in the control and $HgCl_2$-treated wet extraction experiments was calculated from the temperature and pressure of the extracted gas and the internal volume of the vacuum line. The details of the extraction system and correction methods used for estimating gas content are described in Yang (2019). Similarly, the gas content of the dry extraction samples was also inferred from the volume and pressure of gas inside the vacuum line once the sample tube was attached to the line for GC analysis. The uncertainties of the calculated CH_4 and N_2O contents were calculated by using error propagation of the blanks and gas content uncertainties (see Appendix for uncertainty estimation of the blank corrections and gas contents).

2.4.Dry soil content

Dry soil content was measured using the leftover meltwater from the control-wet extraction tests. After the control-wet extractions were complete, the sample flasks were shaken ~~well~~ thoroughly and the meltwater samples were each poured into a 50 mL conical tube. The meltwater and soils were separated by a centrifugal separator at 3000 rpm for 10 min. The separated wet soils were wind-dried in evaporating dishes at approximately 100°C for 24 hours. The weight of each individual evaporating dish was pre-measured before use. The dry soil content was calculated by subtracting the weight of the evaporating dish from the total weight of the dried soil sample plus the evaporating dish.

3. Results and Discussion

3.1.Comparison between wet and dry extraction methods

The results from the wet and dry extractions were compared using 23 ice-wedge samples (21 for N_2O) from Alaska and Siberia. In both the CH_4 and N_2O mixing ratio analyses, we found that the wet and dry extraction results did not differ significantly ($p > 0.1$), regardless of sampling site or soil content (Figure 1, a to d). We note~~d~~ that the heterogeneous distribution of gas mixing ratios of in centimetre 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. Some previous studies have avoided using the wet extraction method because of potential reactivation of microbial CH_4 and/or N_2O production in ice melt (e.g., Cherbunina et al., 2018; Kim et al., 2019). Assuming that activation of microbial metabolism is unlikely during dry extraction at a temperature of -37°C in the extraction chamber for < 1 h, our findings may imply that wet extraction does not stimulate microbial reactivation to a measurable extent.

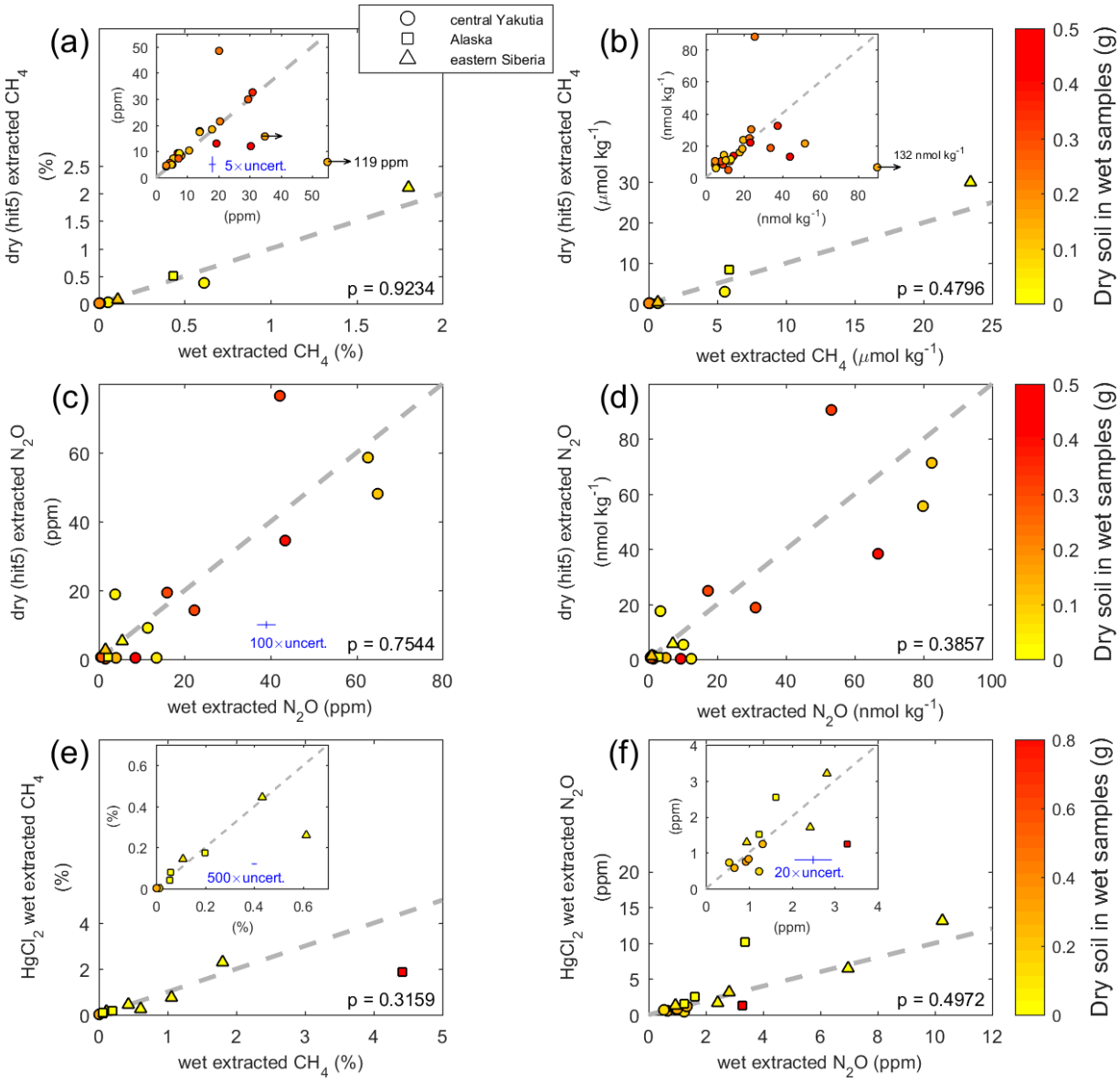

Figure 1. Comparison of CH_4 and N_2O mixing ratios and contents obtained by different extraction methods. Shown are scatter plots between wet- and dry (hit5) extraction results of CH_4 (a and b) and N_2O (c and d), and between control- and biocide-treated wet extraction results for CH_4 (e) and N_2O (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 magnified by 5x, 20x, 100x, and 500x as 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 bottom right corner~~top~~ of each plot.

205 **3.2.Testing microbial alteration during wet extraction**

206 To test the microbial production of CH_4 and N_2O during wet extraction more accurately,
207 we conducted wet extraction experiments on samples treated with $HgCl_2$, a commonly used
208 effective biocide (e.g., Torres et al., 2005), and compared the results with those of untreated
209 (control) wet extractions. We prepared 12 additional ice-wedge samples using the random cube
210 method for these tests (see Materials and Methods section). We found no significant differences
211 between the control and $HgCl_2$-treated wet extraction results for both CH_4 and N_2O mixing
212 ratios (Figures 1e and 1f), indicating that the bias due to microbial activity during
213 approximately an hour of the melting-refreezing procedure is not significant. This is further
214 supported by tests on an additional 12 ice-wedge samples (using the random cube protocol)
215 treated with 2-bromo-ethane-sulfonate (BES), a specific methanogenesis inhibitor (e.g., Nollet
216 et al., 1997) (Figure A3). Similar to the $HgCl_2$-treated experiments, 25 μL of a saturated BES
217 solution was added to each sample flask. These additional tests were carried out only for CH_4.
218 The two-sided t-test for the CH_4 data indicates an insignificant difference between the two
219 results ($p > 0.9$). Data from individual sampling sites also do not show significant differences
220 ($p > 0.9$ for the Alaskan samples and $p > 0.5$ for the central Yakutian samples).

221 According to microbial sequencing studies that have shown the presence of viable
222 microbes in permafrost and ground ice (e.g., Katayama et al., 2007), it is likely that culturable
223 microbes exist in the ice-wedge samples used in this study. However, considering that at least
224 14 days and up to 3 months of culturing was required to identify microbe colonies extracted
225 from ground ice (Katayama et al., 2007; Lacelle et al., 2011), our melt-refreeze time of an hour
226 was insufficient for microbial activity to resume and produce CH_4 and N_2O.

227

228 **3.3.Dry extraction efficiency and gas mixing ratios**

229 One limitation of our needle crushing dry-extraction technique is the inability to

completely extract gas from ice samples, because small ice particles and/or flakes placed in the space between the needles are not fully crushed. The gas extraction efficiency of the SNU needle crusher system has been reported as ~80–90% for polar ice core ~~ice~~ samples (Shin, 2014). However, the gas extraction efficiency has not been tested for ice-wedge samples. Depending on the extraction efficiency, the needle crushing method could underestimate the gas contents if the gas is not completely extracted. Another possible bias in the gas mixing ratios arises if the CH_4 and N_2O compositions are different between the crushed and uncrushed portions of the ice-wedge samples.

To estimate the biases arising from incomplete gas extraction, we designed a series of tests to identify the differences of the CH_4 and N_2O mixing ratios and contents between the crushed and uncrushed sample portions. Each ice-wedge sample that was randomly collected was first crushed by the regular dry extraction procedure (by hitting it five times with the needle system, 'hit5'), and the gas liberated from the sample was trapped in a sample tube. Then we performed an additional 100 hits on the leftover ice ('hit100'), monitored the amount of additional gas liberated, and trapped the additional gas in a separate sample tube. Comparisons between the hit5 and hit100 results are summarized in Table 1.

Here we regard the ratio of gas content of hit100 to that of hit5 (hit100/hit5 ratio hereafter) as a measure of the gas extraction efficiency of the needle crusher system. The results demonstrate an average hit100/hit5 ratio of gas content of 0.40 ± 0.07 for the Zyryanka samples, 0.24 ± 0.07 for the Bluff samples, and 0.14 ± 0.11 for the Cyuie samples (Table 1). Despite the fact that the number of samples was limited, the ice-wedge samples from the different sites show distinct hit100/hit5 ratios of the amount of extracted gas. However, we observed that the leftover ice from the Bluff and Zyryanka samples were not well-crushed, even after 100 hits with the needle crusher. This was especially true if the ice sub-samples contained soil aggregates: the frozen soil aggregates were barely crushed. In contrast, the Cyuie samples were

255 relatively well-crushed, and the leftover samples were apparently finer-sized ice flakes. We
256 also observed that the hit100/hit5 ratios of gas content are highly variable within samples from
257 a particular site, implying that the extraction efficiency of the needle crusher not only depends
258 on site characteristics, but also on the individual ice sample hardness. When compared with the
259 dry soil content measured from the sub-samples used for wet extraction, no relationship was
260 observed between the dry soil content and the extraction efficiency (Figures 1 and A3). In
261 addition, in the case of samples uncrushed by the hit100 test, it is difficult to estimate the
262 extraction efficiency using the hit100/hit5 ratio of gas content, as the hit100 tests liberated only
263 a marginal portion of gas from these samples. This is because the large-sized uncrushed soil
264 aggregates or particles may have prohibited the needle crusher from crushing the small-sized
265 ice flakes or grains. The needles move up and down together, as they are fixed to a pneumatic
266 linear motion feedthrough device, thus if there is a sizable soil clod that cannot be crushed, it
267 blocks the needle crusher from moving further down. Therefore, we do not recommend using
268 a needle crusher system to measure gas contents in ice-wedge samples.

269 The hardness of the ice samples may also affect the gas mixing ratio analysis in the hit5
270 and hit100 procedures. The hit100/hit5 ratio of CH_4 mixing ratio of Bluff and Zyryanka
271 samples are less than 1 in four out of six samples, yielding an average of 0.9 ± 0.5. However,
272 all five samples from the Cyuie ice-wedges have ratios greater than 1, with an average of 4.7
273 ± 2.6 (Table 1). The higher hit100/hit5 ratio of CH_4 mixing ratios of Cyuie samples indicates
274 that the gases extracted via the hit100 procedure have higher CH_4 mixing ratios than the gases
275 extracted via the hit5 procedure. Considering these results with those discussed previously, we
276 speculate that there are three ways gas can be trapped in ice-wedge ice: enclosed in bubbles,
277 adsorbed on soil particles, and entrapped in soil aggregates. The better-crushed leftover ice
278 flakes in the Cyuie samples may have allowed most of the gas in bubbles and part of the CH_4
279 molecules adsorbed on soil particles and/or trapped in microsites within soil aggregates to be

liberated. Thus, the hit5 CH_4 mixing ratios of the Cyuie samples may more reflect the gas mixing ratios in bubbles, while the hit100 results reflect more of the contribution from gas adsorbed on soil and trapped within soil aggregates than the hit5 results because ~~soil-rich ice~~the ice sample containing larger-sized aggregates has greater hardness than ~~the soil-poor ice~~those with smaller aggregates or fine particles. If this is the case for the Cyuie samples, we can infer that CH_4 is more concentrated in soil particles and in microsites within soil aggregates, compared to in bubbles in the ice. This is partly supported by evidence that ice-wedge layers exhibit relatively trace amounts of CH_4 compared to the surrounding permafrost soil layers (Rivkina et al., 2007); however, this needs to be further evaluated by detailed microbial and chemical analyses. 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). Given that some of the Bluff and Zyryanka ice-wedge samples were not fully crushed by the hit100 tests, it may require additional hits or another extraction technique. Unlike CH_4, the N_2O mixing ratios from the hit100 extractions are higher than the hit5 in ten out of eleven samples, regardless of the sampling site. The hit100/hit5 ratios of N_2O mixing ratios of the Bluff and Zyryanka samples (1.9 ± 0.8 on average) are not significantly different ($p = 0.32$) from those of the Cyuie samples (2.9 ± 1.8 on average). 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~~in relatively oxic conditions~~, while CH_4 is produced as the final product of methanogenesis~~strictly in anoxic conditions~~.

One may expect that a different crushing technique might be more suitable for ice-wedge samples. However, none of the existing dry extraction techniques - centrifugal ice microtome (Bereiter et al., 2013), mechanical grater (Etheridge et al., 1988), or ball-mill crusher (Schaefer et al., 2011) is more advantageous for ice-wedge analysis compared to the

needle crusher system used in this study. The hard portion of ice wedges (e.g., frozen soil aggregates, large soil particles) could easily damage the metal blades of the centrifugal ice microtome and mechanical grater devices, or block the space within the ball-mill chamber, limiting the movement of the milling balls.

It is worth noting that friction between stainless steels could produce CH_4 with carbon from the damaged stainless-steel surface and hydrogen gas (Higaki et al., 2006). If needle crushing causes contamination in this way, the dry extraction results should be affected by the number of hits. To check the impact of the needle crushing procedure on ice-wedge CH_4 and N_2O measurements, we carried out blank tests by changing the numbers of hits from 5 to 100. The results of these tests show no systematic offset among the experiments with different numbers of hits (Figure A2), which implies that the crushing procedure does not affect the dry extraction results for CH_4 and N_2O. Even though a small of contamination does exist, its effects have already been subtracted via blank correction and taken into account in the overall error estimation (see Appendix). Therefore, we consider that our findings are not artefacts of metal friction during crushing.

To summarize, from the hit5 and hit100 comparison tests, we found that 1) the needle crusher method is not able to fully crush the ice-wedge ice samples and thus is unsuitable for measuring gas contents in a unit mass of ice, and that 2) weak crushing (e.g., a small number of hits by the needle crusher system) may better reflect gas mixing ratios of the soft parts of the samples (such as air bubbles) than strong crushing (e.g., a greater number of hits).

Table 1. Results of dry extraction tests with 5- and additional 100 times hitting ice-wedge samples, denoted as 'hit5' and 'hit100', respectively. 'hit100/hit5' is the ratio in extracted gas content or gas mixing ratio of 'hit100' to 'hit5' cases. Also shown are gas content results from both experiments, where the hit100 values are given both in the unit of ml kg^{-1} at STP conditions and µmol/kg (in parenthesis). It should be noted that the 'hit100' gas content results indicate the additional amount of gas extracted after 'hit5' crushing and evacuation.

Site Location	Sample	soil content	gas content				CH_4 mixing ratio				N_2O mixing ratio			
			Wet control	Dry hit5	Dry hit100	hit100/hit5	Wet control	Dry hit5	Dry hit100	hit100/hit5	Wet control	Dry hit5	Dry hit100	hit100/hit5
		wt. %	ml/kg	ml/kg	ml/kg		ppm	ppm	ppm		ppm	ppm	ppm	
Zyryanka, Northeastern Siberia	Zy-A-W1-D	0.155	20.2	13.1	6.3	0.48	6138	3713	2721	0.7329	11.37	9.10	10.15	1.12
	Zy-F-1	0.618	13.5	8.1	3.4	0.42	1080	655.6	173.5	0.2646	1.57	2.81	2.65	0.942
	Zy-A-W1-Low	0.049	30.6	27.8	8.0	0.29	4309	5073	4818	0.9497	2.07	0.69	2.02	2.9
	Zy-B-Low-B	0.107	29.1	23.9	10.0	0.418	18030	21010	35290	1.680	5.37	5.32	15.36	2.89
Northern Alaska	Bluff03-IW1	2.07	13.2	12.2	2.6	0.21	44160	25230	12240	0.4851	5.58	2.36	4.93	2.09
	Bluff06-B3	0.078	20.1	20.9	5.6	0.27	558.7	164.2	219.5	1.337	3.74	18.78	30.14	1.605
Cyuie, Central Yakutia	CYC-01-B	0.252	18.0	21.7	7.1	0.33	18.0	18.3	25.4	1.39	1.55	1.60	2.59	1.62
	CYB-04-C	0.498	20.9	30.7	1.5	0.049	20.2	48.4	165.6	3.42	0.71	0.65	2.96	4.5
	CYB-03-A	0.420	19.7	23.7	1.0	0.041	20.5	21.5	67.1	3.12	0.91	1.01	1.06	1.05
	CYB-02-A	0.403	32.0	25.5	1.9	0.073	29.1	18.7	159.8	8.55	1.00	0.58	3.19	5.5
	CYC-03-B	0.830	22.6	15.7	3.3	0.21	20.3	13.9	94.5	6.80	1.40	0.65	1.08	1.7

3.4.Residual gas mixing ratios and contents after wet extraction

To examine how well the gas is extracted by wet extraction, we applied the dry extraction method to refrozen ice-wedge samples after wet extraction. We first prepared degassed ice-wedge samples that had undergone repetitive wet extractions (wet-degassed ice hereafter). Once the wet extraction experiments were completed, we repeated two cycles of melting-refreezing and evacuation procedures to degas the ice melt. After degassing by a total of three cycles of wet extraction and evacuation, the outermost surfaces (~2 mm) of the wet-degassed ice were trimmed away in the walk-in freezer at SNU on the morning of experiments. The wet-degassed ice was then inserted into the needle crusher and the crusher chamber was evacuated. A specific amount of standard air was injected. Then, the wet-degassed ice samples were hit 20 or 60 times by the needle crusher. The amount of gas and gas mixing ratio of the additionally extracted gas from the wet-degassed ice are shown in Figure 2 and Table A1.

The tests using the wet-degassed ice show an additional gas extraction of ~12 to 20 ml kg_{ice}^{-1}, which is ~43 to 88% of the amount of gas extracted during the initial wet extraction. The additionally extracted gas from the dry extraction is referred to as residual gas hereafter. This is remarkably in contrast to the less than 1% residual fraction of the SNU wet extraction system for ice from polar ice sheets. If such a considerable amount of gas is left intact by repeated wet extractions, the composition of the additional gas is important to understand how much the conventional wet extraction results are biased.

Figure 2 and Table A1 show the mixing ratios and contents of CH_4 and N_2O in the residual gas. The mixing ratios of the residual gas were estimated using mass balance calculations with observed mixing ratios and the amounts of the injected standard and extracted residual gas. The CH_4 mixing ratios of the residual gas range from 10.37 to 23.78 ppm, which is similar to the range of the wet extracted gas. This evidence indicates that CH_4 in ice-wedges cannot be fully extracted by a melting-refreezing procedure. We suspect two possible reasons

for this: (1) During wet extraction, the ice-wedge samples melted and the soil particles settled at the bottom of the sample flask without any physical impact to the soil particles, causing the adsorbed CH_4 molecules on the soil particles to remain adsorbed. (2) During refreezing, the soils accumulated at the bottom of the flasks are crumpled around the centre of the refrozen ice, because the sample flasks are chilled from outside, which facilitated gas entrapment within the frozen soil aggregate. In contrast, the N_2O mixing ratios of the residual gas exhibit very low values compared to those from the initial wet extraction (Figure 2 and Table A1). These results imply that most of the N_2O in ice wedges is extracted by three melting-refreezing cycles, such that only a small amount of N_2O is left adsorbed or entrapped in ice-wedge soils. The authors posit that this might be attributed to the high solubility of N_2O to water compared to CH_4. However, it needs further investigation to better understand this.

In this section, we found that a certain amount of gas remained in ice wedges, even after three cycles of wet extraction, which is extractable instead by needle crushing. This implies that, unlike polar ice cores, wet extraction of ice-wedges does not guarantee near-complete gas extraction, and therefore, precise measurements of the gas content of ice wedges are difficult to obtain. The difficulty in measuring gas content imposes a large uncertainty in estimating CH_4 and N_2O contents. Furthermore, we found that the residual gas has a similar order CH_4 mixing ratio as the gas extracted by initial melting-refreezing, indicating that a comparable amount of CH_4 still remains unextracted in ice-wedges. Hence, a novel extraction method is required to produce reliable gas content and gas mixing ratios in ice wedges. In contrast, our results show that the N_2O content of the residual gas is at trace levels, which may suggest that most of the N_2O in ice-wedges is extractable during initial melting-refreezing. Therefore, wet extraction could be applicable for estimating the N_2O content of ice wedges. However, given that the above evidence resulted from three consecutive cycles of melting-refreezing and evacuation, it is unclear how many melting-refreezing cycles are required to extract most of

the N_2O from ice wedges. It should be noted that combination of repetitive wet extractions with dry extraction does not guarantee reliable estimation of N_2O mixing ratio, because extraction efficiency of the other gas components may be different from that of N_2O. Our findings imply that previous estimates of CH_4 budget in ground ice based on wet extraction principle (e.g., Boereboom et al., 2013; Cherbunina et al., 2018) might have been underestimated, and that the CH_4 production within subfreezing permafrost environment could be larger than previously estimated. Future study should be devoted to a novel extraction method which is ~~able~~ easy to extract gas molecules from ice effectively.

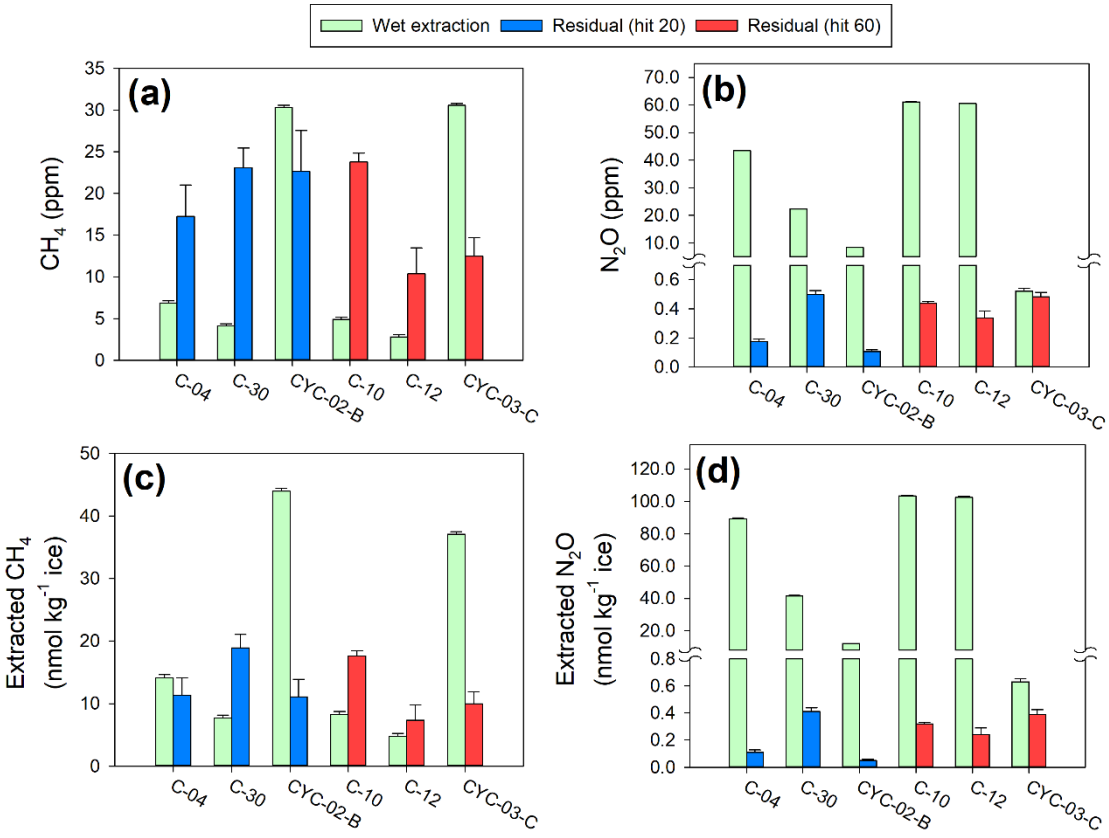

Figure 2. Comparison of wet-extracted gas and residual gas for CH_4 and N_2O mixing ratios (a and b) and contents (c and d). The residual gas was extracted from the dry extraction method using the wet-degassed ice samples. The light green bars show the results of initial wet extraction, and the blue 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.

4. Conclusions

In this study we carried out comparisons between wet and dry extractions, between untreated and biocide-treated wet extractions, and gas extraction from the easily to extract and difficult to extract parts of ice-wedge ice to better understand the characteristics of each extraction method, in order to adequately analyse CH_4 and N_2O mixing ratios and gas contents from permafrost ice wedges. Based on these comparisons, our major findings are summarized as follows:

1) Existing wet and dry extraction methods allow gas extraction from the soft parts of ice (e.g., ice bubbles) and show insignificant differences in CH_4 and N_2O mixing ratios.
2) Wet extraction results are unlikely to be affected by microbial production of CH_4 and N_2O during the melting-refreeze procedure.
3) Both dry and wet extraction methods are not able to fully extract gas from ice wedge samples, presumably due to gas adsorbed on soil particles or enclosed within soil aggregates, which may have different gas mixing ratios compared to the gas in bubbles. Further research is required to develop a proper method to quantify and extract adsorbed and enclosed gases. In the meantime, we propose that both existing techniques may be suitable for gas mixing ratio measurements for bubbles in relatively soft ice wedges (i.e., easily crushed ice wedges by hit5 extraction, e.g., Cyuie ice wedges in this study). Exceptionally, the N_2O content in ice wedges may be measured by using repeated wet extractions, but this is not the case for determining the N_2O mixing ratio.
4) Our ~~results~~ findings indicate that previous estimates of ground ice CH_4 and N_2O budget might be underestimated, implying that the greenhouse gas production in subfreezing environment of permafrost is larger than our current understanding.

5) Our finding indicates that the saturated NaCl solution is unnecessary to prevent microbial activity during melting, as employed by, e.g., Cherbunina et al. (2018). However, it remains as an open question how effectively the adsorbed gas molecules can be extracted by the method.

Appendix. Systematic blank correction and uncertainty estimation

Since the SNU dry extraction systems, including the sample tubes, were originally designed for CO_2 measurements from polar ice cores, these systems have not been tested for CH_4 and N_2O analysis. We therefore carried out a series of tests to estimate the systematic blank, which is defined here as blanks.

The systematic blanks were tested with bubble-free ice (BFI) and standard air in a cylinder calibrated by NOAA. The BFIs were prepared as described in Yang et al. (2017). A major difference is that the BFI block was cut into small BFI pieces of 3–4 g, to mimic the random cube sampling protocol (see Materials and Methods section in the main text). The systematic blanks for the dry extraction method were tested as follows. A total of ~45 g of BFI cubes was placed into the crushing chamber, sealed with a copper gasket, and evacuated until the gas pressure inside the chamber dropped lower than ~60 mTorr, because of the vapor pressure formed by sublimation of the BFI. After evacuation was completed, standard gas was injected into the crushing chamber. The amount of standard injected was controlled by a volume calibrated vacuum line in the dry extraction system. Then the BFI samples were hit with the needle system 5 to 100 times, and the gases in the chamber were passed through a water trap and cryogenically pumped into the sample tubes, using the He-CCR. The number of hits did not significantly affect the systematic blank (Figure A2) and the regression curve for blank correction was fitted to the entire set of data points (red dashed curve in Figure A1).

For the wet extraction, a total of ~45 g of BFI cubes was placed into each sample flask. The flasks were connected to the wet extraction line and sealed with a copper gasket, then evacuated. Once a vacuum was established, a known amount of standard gas was injected into each flask and the flasks were submerged into a warm water bath for ~40 min to melt completely. The flasks were then submerged into the cold ethanol bath, which was chilled to -80°C, to refreeze. For the $HgCl_2$ and Sodium 2-bromo-ethane-sulfonate (BES) treated

445 experiments, we first prepared the saturated solutions of $HgCl_2$ and BES at room temperature
446 (20°C) and added 24 μL of $HgCl_2$ or 20 μL of BES solution into the empty flasks in a fume
447 hood. Then we placed the flasks in a deep freezer, maintained at -45°C for 20 min, to freeze
448 the solutions before the BFI pieces were placed.

449 The results of the blank experiments are shown in Figure A1. The systematic blanks
450 appear to be inversely correlated with the gas pressure in the sample tube. The systematic blank
451 test results were fitted using exponential regression curves (dashed lines in Figure A1), and
452 these regression curves were then used for systematic blank correction in our ice-wedge sample
453 analyses.

454 To calculate uncertainties of the blank corrections, the blank test data were fitted with
455 exponential regression curves (Figure A1). The root-mean-square-deviations (RMSD) of the
456 data from the regression curves are taken as the uncertainties of blank corrections (Figure 1).
457 Since the ice-wedge data used in this study showed the pressure in GC sample loop of about 8
458 ~ 50 torr, the RMSD were estimated from the blank test data within this pressure range. The
459 uncertainty of the gas content measurement is calculated by error propagation from those of
460 pressure, line volume, and mass of ice samples.
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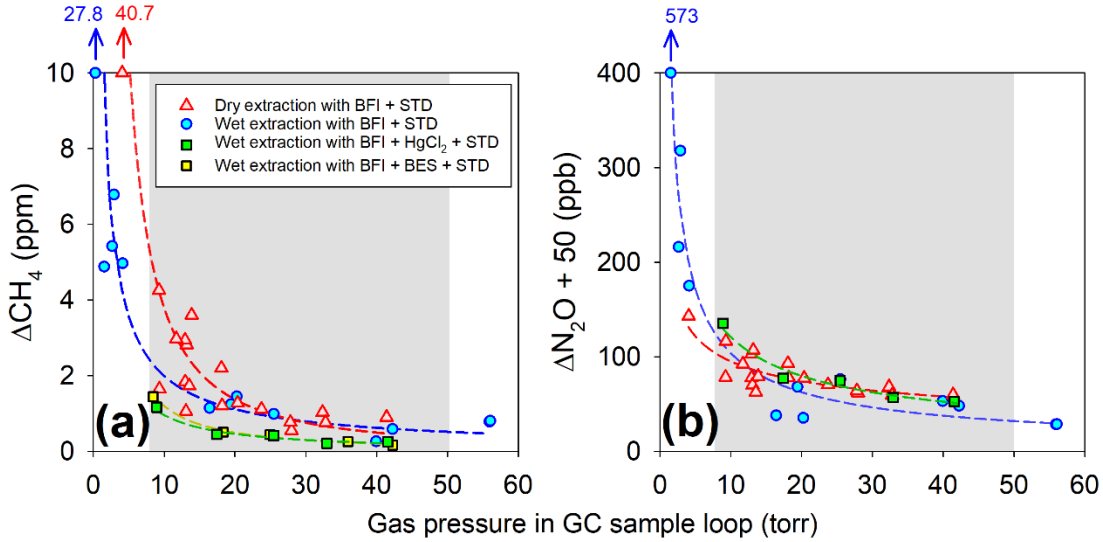

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Figure A1. Systematic blank of the needle crushing (dry extraction) and melting-refreezing (wet extraction) methods for (a) CH_4 and (b) N_2O measurements in control and biocide ($HgCl_2$) treated experiments. Also plotted are the CH_4 blanks of BES-treated wet extractions. The dashed lines represent exponential regression curve fittings. Note that all data are plotted against the amount of gas trapped in the sample tube, presented here as the pressure in the GC sample loop when the sample gas is expanded. The grey shaded areas indicate the range of ice-wedge samples used in this study (see main text). The big-delta (Δ) notion in the y-axes indicate the offset from the values of the standard used.

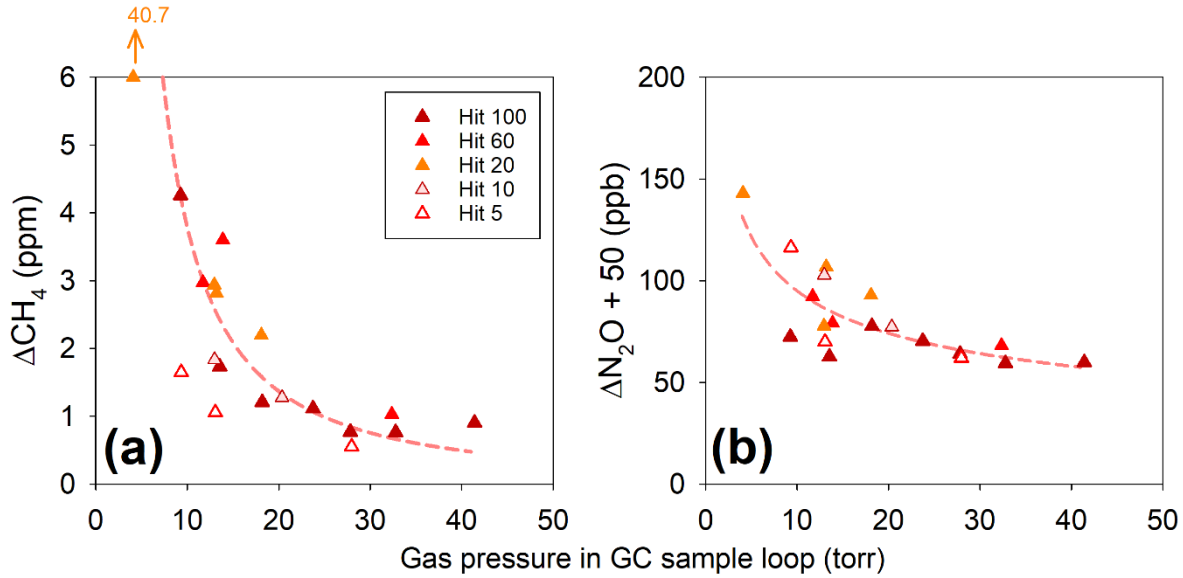

Figure A2. Influence of different number of hitting on the systematic blank of the needle crushing (dry extraction) system for (a) CH_4 and (b) N_2O measurements. Note that all data are plotted against the amount of gas trapped in the sample tube, presented here as the pressure in the GC sample loop when the sample gas is expanded (see main text). The big-delta (Δ) notion in the y-axes indicate the offset from the values of the standard used.

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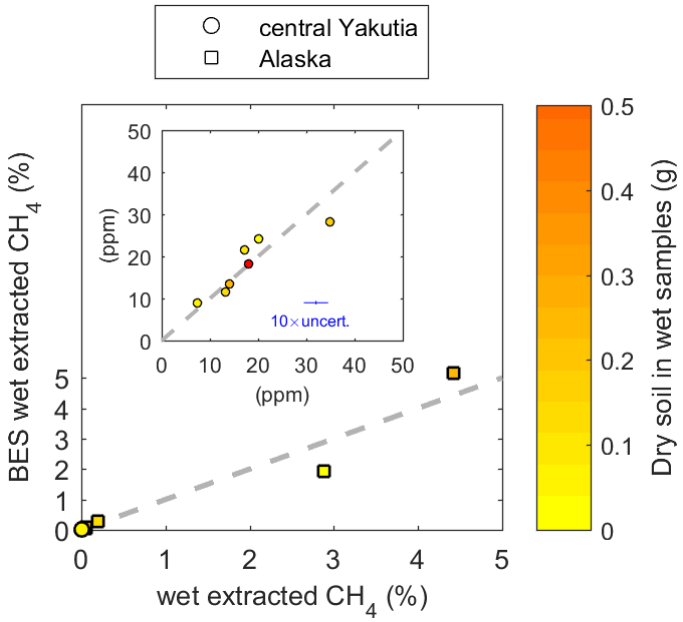

Figure A3. Comparison between control- and BES-treated wet extraction results for CH_4. The sampling area is indicated by different symbols. The color of each data point indicates the dry soil weight in the subsamples used in control wet extraction. The grey dashed lines are 1:1 reference line. The blue error bar indicates the 1-sigma uncertainty of mixing ratios magnified by 10x.

Table A1. Comparison of results from extracted gas from the conventional wet extraction method and the residual gas in ice after 3-times wet extraction. The residual gas was extracted by a needle crusher (see section 3.4 for details of the mehtods)

Site location	Sample	soil content	Wet extraction					Residual gas				
			gas content	CH_4 mixing ratio	N_2O mixing ratio	CH_4 content	N_2O content	gas content	CH_4 mixing ratio	N_2O mixing ratio	CH_4 content	N_2O content
		wt. %	ml/kg	ppm	ppm	nmol/kg	nmol/kg	ml/kg	ppm	ppm	nmol/kg	nmol/kg
Churapcha, central Yakutia	C-10	0.524	37.9	4.9	61.13	8.3	103	16.6	23.8	0.437	17.6	0.324
Churapcha, central Yakutia	C-30	1.03	41.7	4.1	22.28	7.7	41.5	18.4	23	0.50	19	0.41
Cyuie, central Yakutia	CYC-03-C	1.09	27.2	30.5	0.52	37.1	0.63	17.9	12.5	0.48	10.0	0.39
Churapcha, central Yakutia	C-04	1.38	46.0	6.9	43.46	14	89.2	14.7	17	0.17	11	0.11
Cyuie, central Yakutia	CYC-02-B	1.12	32.5	30.3	8.34	44.0	12.1	11.0	23	0.11	11	0.053
Churapcha, central Yakutia	C-12	0.370	38.0	2.8	60.47	4.8	103	15.9	10	0.34	7.3	0.24

Data availability

The data will be uploaded on the public data repository of Pangaea after publication.

Author contributions

JWY and JA conceived the research and designed the experiments. GI, JA, KK, and AF drilled the ice-wedge ice samples from Alaska and Siberia. JWY, JA, SH, and KK conducted the laboratory experiments. JWY and JA led the manuscript preparation with inputs from all other co-authors.

Competing interests

The authors declare no conflict interest.

Acknowledgements

The authors greatly acknowledge those who contributed to collect ice-wedge ice samples. We thank Gwangjin Lim and Jaeyoung Park for their help in sample preparations and gas extraction experiments, and Min Sub Sim for kind advice on inhibition experiment for methanogen.

Financial support

This project was supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF) (NRF-2018R1A2B3003256 and NRF-2018R1A5A1024958) and the NASA ABoVE (Arctic Boreal and Vulnerability Experiment; grant no. NNX17AC57A).

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