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.

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. We add a map in our Supplement (see below). The ages of the studied ice wedges have yet been analyzed, and 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., 1997), 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 in the Supplement for readers. 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 needle-crusher method together with enlarged photographs of crushing needles (left top), and extraction chamber (left bottom). The detailed descriptions about the SNU dry extraction system can be found elsewhere in Ahn et al. (2009) and Shin (2014).



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

- ➔ As we mentioned above, we will upload our original data in a public data repository (PANGAEA) once our manuscript is accepted.
- \rightarrow 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 (<u>Cherbunina et al., 2018</u> and references therein)». I found no mention in the article that NaCl was used in order to minimize microbial activity, is it really there?

- ➔ We correct the reference as below:
- → 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 (<u>Arkhangelov and Novgorodova, 1991Cherbunina et al., 2018 and references therein</u>).
- 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

➔ We add the map of sampling site as Supplement Figure 1 (please refer to our response to General Comment #1 above). We also add photos of outcrops where our samples were taken.



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.

- P.2 L104. Please include the schemes of gas extraction procedures
 → Please refer to our response to the General Comment #2 above.
- 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 by our dry extraction (needle-crusher) at SNU. Therefore, for consistency of analytical setup, we applied the identical procedure of dry extraction to ice-wedge samples to test whether the dry extraction method is applicable. In the meanwhile, the tests with 100-times hitting were designed to understand gas extraction efficiency and difference in gas mixing ratios in between 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 μ L 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 wrere carried out for comparison between dry- and wet extractions. To clarify we will revise the words in the revision.

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 containing large-sized soil aggregates are hardly crushed by our needle-crusher system. Since no significant relationship was found between dry soil content and the extraction efficiency, it may imply that the important parameter controlling hardness is the presence of large-sized aggregate, rather than just soil content. Unfortunately, we have no quantitative measure 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 will reword the sentence L.274 "soil-rich ice has greater hardness than the soil-poor ice" like as follows:
- → Thus, the hit5 CH₄ 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 icethe ice sample containing larger-sized aggregates has greater hardness than the soil-poor icethose with smaller aggregates or fine particles.

P.12 L255. Please specify the size range for «This is because <u>the large-sized uncrushed soil aggregates</u> 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 size of soil aggregate, the soil aggregates in the studied ice wedges are observable by naked eyes as they have clear contrast of darkness when back-lighted. Empirically, the size of observable aggregates is in millimeter-scale, while some of them reached a centimeter.
- ➔ However, it is not possible to estimate the amount of gas remained in the uncrushed portion (mostly Zyryanka and Bluff samples), because it is unknown how much gas is entrapped there. On the other hand, in case for the 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 also been extracted by hit5 procedures, and we have no information on how much of soil aggregates are crushed or uncrushed after the hit5 procedures.

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 can't. To measure gas content precisely, it requires a near-perfect gas extraction from ice wedge samples. As the reviewer pointed out, it is true that 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 the 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 CH₄ 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 indeed. However, the relationship between the porosity and the normalized volume would work only when gas extraction efficiency is near 100%, or

near constant. The gas extraction efficiency of the ice wedge is highly variable and difficult to be precisely measured, limiting estmation 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 understands correctly. Regarding the question, we didn't collect the gas extracted by 2nd and 3rd 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 the wet-degassed ice samples is to know if there is gas remained after three-cycles of wet extraction. We chose 20-times hitting instead of 5 times because significant amount of gas was already extracted from the 3-cycled wet extractions. We also chose 60-times hitting to see if there is a significant difference in amount of the extracted gas between the 20- and 60-times hittings.

P.16 L331. <u>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.</u> 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 gas extraction efficiency. We think we better leave the conclusions as they are because 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 will revise the sentence like below:
- → 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 N₂O 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 the repeated melting-refreezing procedures extract most of the N₂O from ice wedges. However, we cannot guarantee the N₂O mixing ratio because the relative extraction efficiency for each gas species may be variable. To clarify, we will add words in the main text.

3) FIGURES AND TABLES

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

→ We revise the 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 will add a table for the data used for Fig. 1 in Supplement.

Site Location	Sample	<u>soil</u> content	gas content				CH4 mixing ratio				N ₂ O 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
		<u>wt. %</u>	ml/kg	ml/kg	ml/kg		ppm	ppm	ppm		ppm	ppm	ppm	
Zyryanka, Northeastern Siberia	Zy-A-W1-D	<u>0.155</u>	20.2	13.1	6.3	0.48	6138	3713	2721	0.7329	11.37	9.10	10.15	1.12
	Zy-F-1	<u>0.618</u>	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	<u>0.049</u>	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	<u>0.107</u>	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	<u>2.07</u>	13.2	12.2	2.6	0.21	44160	25230	12240	0.4851	5.58	2.36	4.93	2.09
	Bluff06-B3	<u>0.078</u>	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	<u>0.252</u>	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	<u>0.498</u>	20.9	30.7	1.5	0.049	20.2	48.4	165.6	3.42	0.71	0.65	2.96	4.5
	СҮВ-03-А	<u>0.420</u>	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	<u>0.403</u>	32.0	25.5	1.9	0.073	29.1	18.7	159.8	8.55	1.00	0.58	3.19	5.5
	СҮС-03-В	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