nature portfolio

Peer Review File

Turbulence-induced bubble nucleation in hydrothermal fluids beneath Yellowstone Lake

Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality.

12th Oct 21

Dear Dr Caudron,

Your manuscript titled "Turbulence-induced bubble nucleation in hydrothermal fluids beneath Yellowstone Lake" has now been seen by 3 reviewers, and I include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication.

During revision, please ensure that the revised manuscript addresses the following editorial thresholds based on the reviewer comments:

1. Provide compelling evidence to support your claim that the nucleation process described here may initiate 'catastrophic hydrothermal explosions', or alternatively rephrase these statements more conservatively.

2. Provide sufficient evidence to support your interpretation that the acoustic signals detected "represent the nucleation of bubbles in hydrothermal fluids just beneath the lake floor", and evaluate any alternative interpretations.

2. Discuss your findings in a broader context, drawing on the wider literature on underwater venting dynamics and exploring the implications of your results for other volcanic lake environments.

In addition to these editorial thresholds, please re-check any calculations of the resonant frequency, in response to the concerns raised by reviewer #3.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter) and the completed checklist:
[link redacted]
** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Emma Liu, PhD
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Communications Earth & Environment
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Joe Aslin
Associate Editor
Communications Earth & Environment

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Please refer to our data policies at http://www.nature.com/authors/policies/availability.html.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

This study uses hydroacoustic data collected from near one of many active hydrothermal vents in Yellowstone Lake to investigate the source mechanisms of recorded acoustic events. The study finds that bubble nucleation is responsible for composite events that include a high-frequency onset followed by a lower-frequency oscillatory coda. The authors interpret this signal as CO2 bubble nucleations and volume oscillation as the bubble ascended in the water column. These results are supported by ROV video imagery of bubble formation. A lower-frequency signal is also present in the hydroacoustic data that occurs more frequently, and statistically precedes, the bubble nucleation signals. The authors interpret this signal as pressure perturbation due to flow instabilities in the hydrothermal systems. The variation in pressure at or near the lake bed then sometimes triggers the nucleation of bubbles. The authors cite this as evidence that a larger pressure perturbation, possibly from an earthquake, could produce a more energetic explosion.

This is a nice use of hydroacoustic data to study source processes in hydrothermal systems. The paper is generally well-written and most of the conclusions are well-supported. Short format papers don't leave a lot of room to expand on discussion items but there were a couple places where unsupported statements were made that trended toward conjecture. The two main examples were on lines 50 and 136 when seemingly trying to tie the nucleation of small bubbles observed in this study to 'catastrophic hydrothermal explosions'. These statements are provided without real evidence or support, which raises many questions. Like, how would a larger bubble or bubbles result in a 'catastrophic explosion'? The bubbles aren't highly pressurized, correct? There is also a lot of seismic activity in the area and yet few explosions. What evidence is there that the two large explosions over the past 13 ky were related to CO2 bubbles vs. pressure build up under an impermeable hydrothermal cap? This should be toned down / removed or supported with evidence. The rest of my comments are relatively minor and listed below as well as on the attached PDF. Once these issues are addressed, this paper will make a nice contribution that will be of interest to the volcanological and geothermal communities.

Line 9: A volcanic reference that would be appropriate here is:
Lyons, J.J., Haney, M.M., Fee, D. et al. Infrasound from giant bubbles during explosive submarine eruptions. Nat. Geosci. 12, 952–958 (2019). https://doi.org/10.1038/s41561-019-0461-0

Line 24-25: While this is still somewhat true for CO2, the advancements in both ground-based and satellite sensors make this statement read as a bit out of touch with the current state of the art for other gas species, like SO2. e.g., Carn, S., Fioletov, V., McLinden, C. et al. A decade of global volcanic SO2 emissions measured from space. Sci Rep 7, 44095 (2017). https://doi.org/10.1038/srep44095

Line 43: field -> suggest replacing with a different or more descriptive term since 'field' is being used to at least 4 times in the previous few lines to describe a thermal field.

Line 43: pockmark ->
pockmarked

Line 46: When did the Mary Bay explosion occur?

Line 50: potentially explosive -> this statement needs to be backed up with references, particularly since it is stated in the same sentence that the exsolution process is poorly understood.

Figure 1. Maps are a bit blurry / low-fi in the PDF.

Line 66: am isn't necessary since you are using a 24 hr convention above. However, it should read ~02.30.

Line 67: pm -> see previous comment. Please insure all time references follow the same convention throughout the paper.

Line 68-69: What evidence is there that these signals represent bubble nucleation? Given that the rest of the interpretation hinges on this, it is surprising that no evidence is presented. If this is your interpretation, it needs to be written as such, rather than a statement of fact, which is how it currently reads.

Line 73: Please add that the gas is assumed to be solely CO_2 e.g., a 'spherical bubble of CO_2'

Line 76-77: For the non-experts, would you explain briefly (or add an appropriate reference) why a vapor bubble would be expected to condense?

Figure 2. Not quite publication ready. 1) Y-axis labels are varying font size, some are cut-off and not fully legible, and all should be aligned to the left. 2) The scales of the wavelet transform figures would show better if they spanned the height of the wavelet figure, and were all in the same location relative to the wavelet image. 3) X-axis labels are cut off at the bottom. 4) vertical dotted line between left and right panels is unnecessary. 5) Why have two different time scales (sec vs ms) for the plots in the left and right columns? 6) White space to left and right of right column waveforms should be eliminated.

Line 116: why is it that these fluxes are only 'apparently much larger'? Have they not been measured? Is this just based on observations from the ROV?

Line 136: The idea that a 'catastrophic hydrothermal explosion' could be initiated based on the nucleation of the small bubbles in this study seems like conjecture without more supporting evidence. How would a larger bubble result in a 'catastrophic explosion'? The bubbles aren't highly pressurized, correct? There is also a lot of seismic activity in the area and yet few explosions. What evidence is there that the two large explosions over the past 13 ky were related to CO_2 bubbles vs. pressure build up under an impermeable hydrothermal cap? This should be toned down / removed or supported with evidence.

Line 149: What was the height of the hydrophone off the lake floor?

Line 163: Are the hydrothermal fluids all cold / lake temp in the study area?

Reviewer #2 (Remarks to the Author):

General comments

The submitted manuscript presents an acoustic emission study focused on the recognition, and understanding, of gas fluxes (particularly CO_2) and bubble nucleation process from active underwater hydrothermal systems.

The authors performed the analyses of acoustic data recorded by a hydrophone system deployed on the floor of Yellowstone Lake to characterize the nature of gas discharge in one of the lake floor thermal fields: the 'Deep Hole' located to the southeast of Stevenson Island. A secondary vent, ca. 100 m from the main discharge vent field, was chosen for the investigation. The investigation of the acoustic data has been cross-analysed with video footages of the bubbling vent to compare the bubble sizes estimated using the acoustic signals.

Authors recognized sixty-six discrete events during a 3-hours interval, all exhibiting a characteristic waveform, with bubble rising showing a resonant frequency of 100-200 Hz. Such frequencies were used to estimate bubble diameters of ~3-6 mm, which appear to agree with visual observations.

Authors claim that the heterogeneous nucleation of CO_2 bubbles in the hydrothermal fluids is triggered by turbulent flow instabilities within the lake floor sediment. In particular they state that their acoustic data demonstrate that the CO_2 saturated fluids are unstable to the fast (~50 Pa/s), but

small (< 10 Pa), pressure perturbations associated with unsteady flow through the lake floor sediments. They finally conclude that such process could explain the formation of the pockmarks that characterize hydrothermal discharge zones on the lake floor, and it could play a role in the initiation of large, catastrophic hydrothermal explosions, as those which have occurred in the post-glacial history.
Results are significant by the fact that i) the proposed acoustic method appear to be a very interesting and novel tool able to constrain gas fluxes in underwater environments – with important implications for monitoring and measuring underwater gas fluxes from volcanic and non-volcanic sources, ii) show new insight into bubble nucleation in hydrothermal fluids, and iii) shed a quantitative light on a very important process characterizing the state of active hydrothermal systems. Therefore, the study is definitely worthy of publication, which I recommend after that the following comments (minor) have been suitably addressed.

Cristian Montanaro

Main comments
In general, the manuscript should be “adjusted” to broader the implications of the key and important results from this study; this can be done by comparing/discussing findings from this study case in the context of other known works on degassing and venting dynamics from underwater environments.
Concerning the figures, please double check consistency between units discussed in the text and those reported in the graphs, as well make sure that no labels are missing from the x-y-axis of graphs (see also more comments below).

• In the Summary paragraph:
Line 1: expand a bit more on the effect of gases, for instance: "...including CO2, which can affect the surrounding environments, and eventually threaten health and life of humans and animals, as well as damage crops and property. Therefore, the understanding of gas fluxes remains a...”. This will strengthen more the importance of the study.

Line 11: Would be good to specify in the sentence where the instabilities occur during the rising of hydrothermal fluids. "... within sediments close to the lake floor...?

Line 19: To broaden the impact of this novel observation, it would be worthy to briefly mention how the findings from this study can improve the understanding of similar processes from other "intra-caldera lakes" (e.g. lake Rotorua in NZ) or lake sitting within actively degassing craters (e.g. Rotomahana lake in NZ, or Albano Lake in Italy). See also comment below.

• In Main – Introduction:
Line 24: Another good example could be the deadly effect of gas observed after the phreatic eruption within the Diëng plateau (Le Guern et al. 1982).

Line 26: The study from Aiuppa et al. (2019) can be also another good example of CO2 flux estimation.

Line 47: Mary Bay is more a cluster of craters or a crater field. …115 m crater field…

Line 50: I think here is worthy to mention the work of Thiéry et al. (2010), which discusses the decompression of aqueous solutions and their explosive potential.

• In Results:

Line 57: citation of Figure S1 and S2 are missing. I suggest to add the first one here: …acoustic hydrophone (Figure S1) on the floor…

Lines 63-67: I recommend to label each panel in the Figure 2 - a, c, e on the left panels, and b, d, f the right panel. Then you can cite (Figure 2 a,c,e) after oscillation, and put the reference (Figure 2 b,d, f) at the end of the next sentence. This would make it easier for the reader to follow text and figure. To be also noted that in panel c labels on the y-axis are cut and/or missing (e.g. -20 in the right panel of c).

Maybe I don't get it correctly, but by looking at the figure, it seems that the largest amplitude signals last 10 ms, while the overall duration is 50 ms? if not, please try to explain better the sentence.

Line 67: change time in 13:00 for consistency with the time format given before

Line 72: … bubble 20 (see Methods)…

Line 74: The estimated volume here should be referenced to Figure S3: in the figure the volume on the y-axis should be changed in mL for consistency with the reported median volume in the text (or vice versa text changed in L).

Line 75: The authors stated that the video observations are consistent with the estimated radius. However, from the video (.MP4 file) it is hard to have an idea of the surrounding elements sizes (e.g. rocks on the lake floor, ROV arm).

I recommend to add a figure/panel that shows one (or more) frame(s) of the video with bubbles, and including a reference scale (size of the ROV arm for example?) to which to compare the bubbles size. Such a Figure might be implemented as a panel in Figure 1, for instance.

Line 76: Somewhere before, or in the Methods it is important to specify P-depth of the investigated site, as well as the temperature (T-profile if measured with probes at the investigated site) since these are key parameters controlling the fluids behaviour and the bubble nucleation process. Moreover, the pressure is used in the Minnaert's relation used to calculate the bubble radius.

Lines 74-78: The paragraph is too long. I recommend to split it at least in two to make it easier to read and follow.

Lines 90-97: Generally, this whole paragraph is not well explained by referencing to Figure 3, where: i) units of y-axis are missing, and reported values do not match what reported in the text (for instance, the frequencies are reported and referenced as Hz in the text - not in the Figure), ii) decompression rate is in Pa/s, not reported in the figure y-axis.

Please, modify the text or provide a new Figure 3

Line 97: It might be worthy to clarify the meaning of cavitation:

…consistent with cavitation (i.e. formation of vapor-filled bubbles at low pressures) in a flowing fluid…

Line 104:...interaction with the hosting sediments could...

Lines 112-114: Too long sentence. What about:
…The bubbles we detected are associated with relatively weak venting at a site that is ~100 m from the main discharge zone containing at least 20 active vents (Figure 1). Such vents exhibited continuous bubble fluxes during periods when our visual observations could be made with the remote operating vehicle…

Lines 115-116: How did you estimated/measured the 5x10-10 kg/s flux value? Eventually specify it here, and/or add the description in the Method
…"The estimated CO2 flux associated with bubbles in out acoustic data (ca. 5x10-10 kg/s; see Methods)"…

Lines 117-118: ...through the whole 'Deep Hole' area. Indeed, with a dissolved...

Figure 4: I really like this figure, simple and effective. I also think it might benefit from adding the produced frequency by the indicated process. For e.g. in the panel a, it can be added 1-40 Hz with a line/arrow pointing to the turbulent fluid-sediment interaction. Similar thing can be done in panels b-c.
In the Figure 4 caption: “flowpath constriction” can this be produced by a localized mineral precipitation-seal? Similar obstruction can be observed at discharging spots in sub-aerial environment (see comment below). Line 125: Maybe a brief explanation of breathing and dissolution when discussing the cavitation, should be briefly mentioned in the text within the "Results" text.

Lines 127-128: I think it is also worthy to briefly discuss the effect of pressure-depth at which the process was observed, and what expected at higher depth or shallower depth (pressures). In other words, I think it can be important to consider/discuss if there is a "depth-pressure limit" for unsteady flow to be an efficient process enhancing the heterogeneous bubble nucleation.
Moreover, the unsteady flows are explained to be the result of fluid-sediment interaction: another mechanism which might amplify-cause unsteady flow, favouring CO2 bubble nucleation, can be the precipitation of minerals from hydrothermal solutions (in analogy with sub-aerial environments) that can result in the formation of constriction just below – or at the lake floor level (for example cementing clasts as those shown in the video). Mineral precipitation and cementation processes have being already discussed in Shanks et al. (2005, 2007). This process can be briefly discussed here, considering known cases from Yellowstone (above mentioned Shanks studies), or from other volcanic and geothermal areas with active sub-aerial hydrothermal manifestations (e.g. Jones & Renaut, 2003; Kennedy et al., 2020).

Lines 128-130: This is a critical point, and a very important finding of this study.

Lines 134-135: It would be more impact-full here to make comparisons with other known cases (see next comment as well), to show that results – and derived processes – observed here by using the acoustic method for the first time, might have been overlooked in underwater environments elsewhere, where instead they can represent a key mechanism explaining lake floor deformation, collapses (sinkhole-like), or the occurrence of underwater explosions.

Lines 135-137: This statement “alone” seems to be too strong. It could be expanded a bit more with a short paragraph summarizing the trigger mechanism envisaged for mid-large eruptions at Yellowstone. For instance, Morgan et al. (2009) indicate that areas affected by cataclysmic explosion

were (1) dominated by high heat flow, (2) had been repeatedly subjected to inflation and deflation of the caldera, and (3) had an active well-developed sub-lacustrine hydrothermal system with significant alteration and vein formation. Most importantly (and related to the key finding of bubbling process enhanced by small pressure perturbations) Morgan et al. suggest that a significant seismic event related to formation of the sublacustrine Lake Hotel graben may have helped trigger the explosions within and around the lake.
In this, the presence of large amount of dissolved CO2 in hydrothermal fluids within sediments, and the “fast” nucleation following larger P-perturbation, might have strongly contributed in over-pressurizing the system prior to, and/or during the explosive events – for the effect of CO2 in enhancing explosivity is also worthy to consider the works of (Thiéry and Mercury 2009; Hurwitz et al. 2016).

• In the Concluding Paragraph:
As mentioned in the comments above, and in order to broaden the impact of the interesting findings from this study, I recommend the authors to discuss the potential key role of enhanced bubbling nucleation in the formation of pockmark fields (or large sinkholes), which have also been observed at other caldera-hosted lakes, as in NZ's Rotorua (https://www.gns.cri.nz/Home/News-and-Events/Media-Releases-and-News/Scientists-and-Navy; Pearson, 2007), Italy's Albano Lake (Colli Albani District - Anzidei et al., 2008), Azores Archipelago (Andrade et al. 2019), and at other crater-hosted lakes with strongly degassing activity as at Rotomahana (de Ronde et al. 2016). At such sites, the formation of large sinkholes, and high the fluxes of CO2 from the floor are recognized at specific geological sites (e.g. fractured zones following main structural elements, vents of previous eruptions, etc.).

• In Methods:
Line 151: Please define STA/LTA (Short-Term to Long-Term moving averages)

Lines 155-156: Might be useful a reference for these methods (Morlets-Fourier).

Lines 162-163: Pressure (or depth) at the investigated site should be indicated here in the Methods, or earlier in the introduction (see also comment above). This is a key parameter used in the calculation of the bubble radius.

Cited references:
Aiuppa A, Fischer TP, Plank T, Bani P (2019) CO2 flux emissions from the Earth’s most actively degassing volcanoes, 2005–2015. Sci Rep 9:. https://doi.org/10.1038/s41598-019-41901-y
Andrade C, Cruz JV, Viveiros F, Coutinho R (2019) CO2 flux from volcanic lakes in the western group of the Azores archipelago (Portugal). Water (Switzerland) 11:1–17. https://doi.org/10.3390/w11030599
Anzidei M, Carapezza ML, Esposito A, et al (2008) The Albano Maar Lake high resolution bathymetry and dissolved CO2 budget (Colli Albani volcano, Italy): Constrains to hazard evaluation. J Volcanol Geotherm Res 171:258–268. https://doi.org/10.1016/j.jvolgeores.2007.11.024
de Ronde CEJ, Walker SL, LeBlanc C, et al (2016) Reconstruction of the geology and structure of Lake Rotomahana and its hydrothermal systems from high-resolution multibeam mapping and seismic surveys: Effects of the 1886 Tarawera Rift eruption. J Volcanol Geotherm Res 314:57–83. https://doi.org/10.1016/j.jvolgeores.2016.02.002
Hurwitz S, Clor LE, McCleskey RB, et al (2016) Dissolved gases in hydrothermal (phreatic) and geyser eruptions at Yellowstone National Park, USA. Geology 44:G37478.1.

https://doi.org/10.1130/G37478.1
Jones B, Renaut RW (2003) Hot spring and geyser sinters: the integrated product of precipitation, replacement, and deposition. Can J Earth Sci 40:1549–1569. https://doi.org/10.1139/E03-078
Kennedy BM, Farquhar A, Hilderman R, et al (2020) Pressure Controlled Permeability in a Conduit Filled with Fractured Hydrothermal Breccia Reconstructed from Ballistics from Whakaari (White Island), New Zealand. Geosciences 10:1–19. https://doi.org/10.3390/geosciences10040138
Le Guern F, Tazieff H, Pierret RF (1982) An example of health hazard: People killed by gas during a phreatic eruption: Diëng plateau (Java, Indonesia), February 20th 1979. Bull Volcanol 45:153–156. https://doi.org/10.1007/BF02600430
Morgan LA, Shanks WCP, Pierce KL (2009) Hydrothermal processes above the Yellowstone magma chamber: Large hydrothermal systems and large hydrothermal explosions
Pearson LK (2007) The nature, composition and distribution of sediment in Lake Rotorua, New Zealand. The University of Waikato, Hamilton, New Zealand.
Shanks WCP, Alt JC, Morgan LA (2007) Geochemistry of Sublacustrine Hydrothermal Deposits in Yellowstone Lake—Hydrothermal Reactions, Stable-Isotope Systematics, Sinter Deposition, and Spire Formation. US Geol Surv Prof Pap 1717:33
Shanks WCP, Morgan LA, Balistrieri L, Alt JC (2005) Hydrothermal Vent Fluids, Siliceous Hydrothermal Deposits, and Hydrothermally Altered Sediments in Yellowstone Lake. Geotherm Biol Geochemistry In: Inskee:53–72
Thiéry R, Loock S, Mercury L (2010) Explosive properties of superheated aqueous solutions in volcanic and hydrothermal systems. In: Rzoska S, Drozd-Rzoska A, Mazur V. (eds) Metastable Systems under Pressure, NATO Scien. Springer, Dordrecht, pp 293–310
Thiéry R, Mercury L (2009) Explosive properties of water in volcanic and hydrothermal systems. J Geophys Res Solid Earth 114:1–19. https://doi.org/10.1029/2008JB005742

Reviewer #3 (Remarks to the Author):

This is an interdisciplinary research on bubble acoustics and fluid flow. The authors have done an interesting field work to collect some acoustic data from the Yellowstone Lake, and were trying to have some conclusions from the observation. However, the bubble resonance frequency was wrongly calculated and the evidence of bubble nucleation phenomena was insufficient, which made the novelty of the manuscript limited.
Major issues:
1. Line 12-13: “The resonant frequency of the bubbles (100-200 Hz) is consistent with diameters of ~3-6 mm, in agreement with visual observations.”
According to Minnaert’s equation and the given values through the text (P = 882kPa, \gamma = 1.3, \rho=1000 kg/m3, bubbles with diameters of 3~6 mm, water depth 100-115 m), I obtain the resonant frequency of the bubbles as 3.5-7 kHz, which is not the described 100-200 Hz in the text. This indicates that the authors have incorrect assumption of the acoustic regime.

2. Line 17-18: “The nucleation of CO2 bubbles in hydrothermal fluids due to small pressure perturbations is a novel observation … “
There is no theory to support the claim of “the nucleation of CO2 bubbles” was existing “in hydrothermal fluids”.

3. Line 60 “The system was deployed in an area of relatively weak discharge, ~100 m north of the main vent field.”

Recent Research have shown that the acoustic signals from individual bubble film oscillation can only be detected in a few meters due to the attenuation and the low signal to noise ratio (SNR) in the field. The authors deployed only one acoustic hydrophone to monitor the acoustic signal, which is limited to improve the SNR. At 100m range, the authors won't be able to measure sounds from individual "bubble events", but a spectral frequency band if the leakage was strong. However, the authors measures some "bubble acoustic-like" acoustic waveforms (e.g., 1 ms at line 63 and top panels in Figure 2), which can only be possible that there were a number of weak gas seeps close to the hydrophone.

4. Line 79-81: "These visual observations, combined with the fact that the hydrothermal fluids discharged at the Deep Hole field are rich in magmatic gases and are saturated in CO2, lead to the conclusion that the bubbles are dominantly composed of magmatic CO2, with possibly minor components of H2S."
This is an ill-conditioned deduction. The authors won't be able to have such a strong conclusion without enough chemical information from the seep site.

5. Figure 2 "the resonance of ~200 Hz"
Again, 200 Hz could not be resonant frequency of bubbles at such depth.

6. Line 99, there is neither evidence nor theory to show there was "nucleation of non-condensable gas bubbles".

7. Line 112 "relatively weak venting at a site that is ~100 m from the main discharge zone, which contains at least 20 active vents …"
Again, the hydrophone won't be able to detect individual "bubble events" at 100 m. There have been a number of publications in the field research of underwater acoustic bubble to declare bubble acoustic detection threshold, either in the Mediterranean Sea or in the North Sea.

8. Line 138-141, Concluding paragraph: "Our data from Yellowstone Lake demonstrates the utility of acoustic methods for constraining gas fluxes in underwater environments, and provide new insight into bubble nucleation in hydrothermal fluids. "
Researchers have done a lot of theoretical and field work on gas flux quantification in underwater environments, which means that the former conclusion here is meaningless. There is neither theory nor evidence to support the nucleation of bubbles was exit in hydrothermal fluid.

Turbulence-induced nucleation in hydrothermal fluids beneath Yellowstone Lake

Corentin Caudron*, Jean Vandemeulebrouck, Robert A. Sohn

*Corresponding author

Summary paragraph

Volcanic systems generate large amounts of gas, including CO_2, and understanding gas fluxes is a fundamental aspect of volcanology and hazard mitigation. While it is challenging to measure gas fluxes in subaerial settings, underwater environments provide a unique opportunity to study gas emissions because bubbles are powerful sound sources[1]. Here we take advantage of a unique deployment of an audio-quality acoustic system on the floor of Yellowstone Lake to characterize the nature of gas discharge in a lake floor vent field. We find that turbulent flow instabilities trigger heterogeneous nucleation of CO_2 bubbles in the hydrothermal fluids. The resonant frequency of the bubbles (100-200 Hz) is consistent with diameters of ~3-6 mm, in agreement with visual observations. Our acoustic data demonstrate that the CO_2 saturated fluids are unstable to the fast (~50 Pa/s), but small (< 10 Pa), pressure perturbations associated with unsteady flow through the lake floor sediments, indicating that the system could respond catastrophically to larger-scale perturbations, such as those associated with earthquakes. The nucleation of CO_2 bubbles in hydrothermal fluids due to small pressure perturbations is a novel observation that may help explain why Yellowstone Lake has experienced numerous hydrothermal explosions in its post-glacial history.

Main

Introduction

Gas fluxes through volcanic systems constitute an important component of the global cycle for elements such as carbon[2] and sulfur[3], and they present a significant geological hazard for communities located near volcanic systems[4]. Gas fluxes are difficult to accurately measure in subaerial environments due to the intrinsic difficulties associated with sampling, and the relatively high atmospheric concentrations of dominant species such as CO_2 [5]. Most volcanic systems, however, are located underwater [6], and many others host intra-caldera lakes, which opens the possibility of using acoustic methods to monitor gas fluxes. Gas bubbles are energetic sound sources, and recent studies have demonstrated that acoustic methods can effectively measure gas fluxes in volcanic lakes[7,8], in much the same way they are being used to monitor gas fluxes during CO_2 storage experiments[9] and in CH_4 seep environments[10].

The Yellowstone Plateau Volcanic Field (YPVF) constitutes one of the largest volcanic systems on Earth, and although it has not experienced a volcanic eruption for the past ~70 ky[11], it is presently discharging magmatic CO_2 at an estimated rate of ~32 kt d^{-1} [12]. This flux estimate, however, is uncertain by a factor of ~3 due to inaccuracies in gas accumulation chamber measurements[12], and it does not include any fluxes into Yellowstone's hydrothermally active lakes. Yellowstone Lake, which straddles the southeast margin of the 640 ka Yellowstone Caldera, is the largest lake in the YPVF and it hosts a variety of hydrothermal fields, including both liquid- and vapor-dominated systems at depths of up to ~110 m below the lake surface[13]. The gas fluxes from these sublacustrine systems are largely unknown, but a recent multidisciplinary study[14] has shown that one of the lake floor thermal fields, the 'Deep Hole' located to the southeast of Stevenson Island (Figure 1), is among the most energetic (~28 MW) thermal areas on the YPVF[15]. Fluid samples acquired from this vapor-dominated field are rich in CO_2 and H_2S dissolved gases[16], and the field itself is located within a ~~pockmark~~ field, most likely generated by sediment expulsion associated with the episodic release of gas[17]. More generally, the sublacustrine hydrothermal systems beneath Yellowstone Lake have generated more than a dozen hydrothermal explosions over the past ~13,000 years, including the Mary Bay explosion, which created a ~2.6 km wide by ~115 m deep crater on the lake floor and distributed debris over an area of ~30 km^2 [18]. The role of magmatic gases in these catastrophic explosions is unclear, but the release of trapped gas from a subsurface reservoir and the exsolution of dissolved gases from hydrothermal fluids are poorly understood, but potentially explosive, processes.

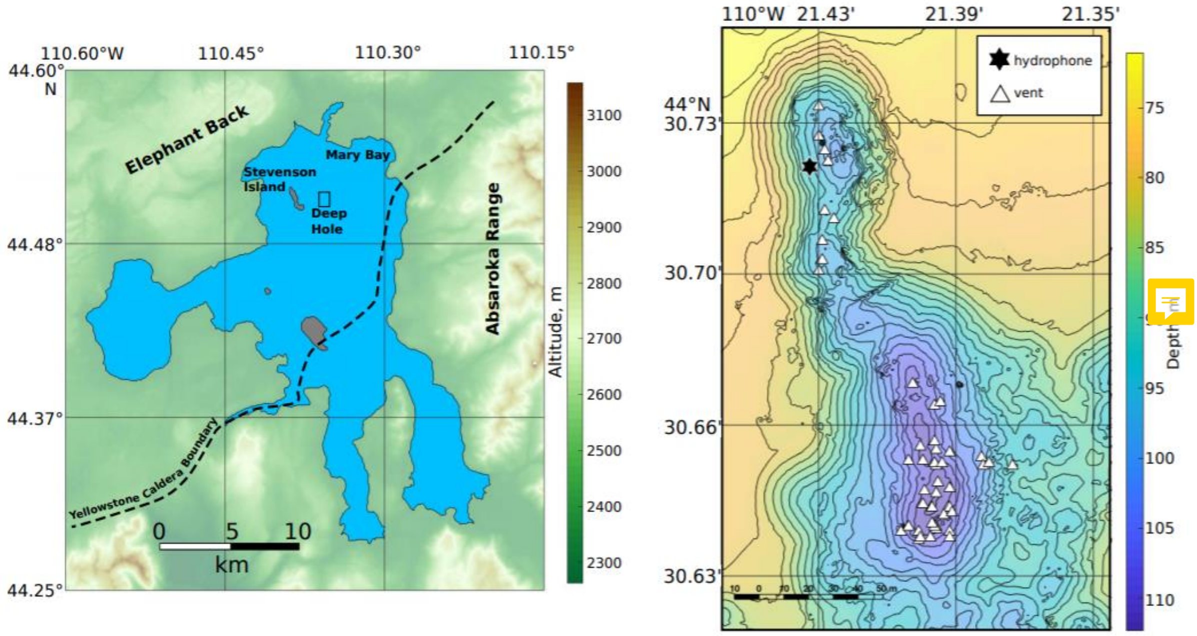

Figure 1: ***Location****: Left: Yellowstone Lake. The northern part of the lake lies inside the 640 ka Yellowstone Caldera (dashed line). Right: Bathymetry of the Deep Hole thermal area (black rectangle in left panel). White triangles indicate active venting locations observed during remotely operated vehicle dives, and the black star corresponds to the high-rate hydrophone location.*

Results

We deployed an audio-quality (44.1 kHz, 24 bit) acoustic system on the floor of the Deep Hole thermal field, located ~2 km southeast of Stevenson Island (Figure 1), to investigate the effectiveness of using acoustic measurements to monitor gas fluxes in this environment. The system was deployed in an area of relatively weak discharge, ~100 m north of the main vent field. We recorded 13 hours of continuous data, within which we observed a 3-hour interval (23:40 16 August to 02:30 17 August, UTC) of intense activity. Sixty-six discrete events were detected during this interval (see Methods), all exhibiting a characteristic waveform (Figure 2) consisting of a brief (~1 ms), high-frequency (several kHz), impulsive onset, followed by lower frequency (~100-200 Hz, Figure 2b) oscillation. The event durations scale with amplitude, with the largest amplitude signals having durations of up to 50 ms, and typical durations of ~10 ms. The intense rate of activity (~0.5 event/min) suddenly stops at ~2.30~~am~~, and no additional events were detected until the end of the recording (1~~pm~~ 17 August).

These acoustic signals, which we hereafter refer to as 'bubble events', represent the nucleation of bubbles in hydrothermal fluids just beneath the lake floor. The impulsive, high-frequency component observed at the beginning of the signals is associated with bubble nucleation, while the lower frequency component represents free oscillations of the newly formed bubble[19,20] (Figure S2). Applying Minnaert's equation for the relationship between oscillation frequency and the radius of a spherical bubble[20] yields radii estimates ranging from 1.4 - 3.1 mm, with a median volume of 2.31 x 10^{-5} mL. Videographic observations of bubble discharge from the Deep Hole field made by the Remotely Operated Vehicle *Yogi* (Supplementary Video) are consistent with these sizes, and show that the bubbles do not condense upon discharge into the ~4°C lake water, as would be expected for a vapor bubble, but rather can rise significant distances through the water column, with some rising all the way to the lake surface. These visual observations, combined with the fact that the hydrothermal fluids discharged at the Deep Hole field are rich in magmatic gases and are saturated in CO_2 [21], lead to the conclusion that the bubbles are dominantly composed of magmatic CO_2, with possibly minor components of H_2S.

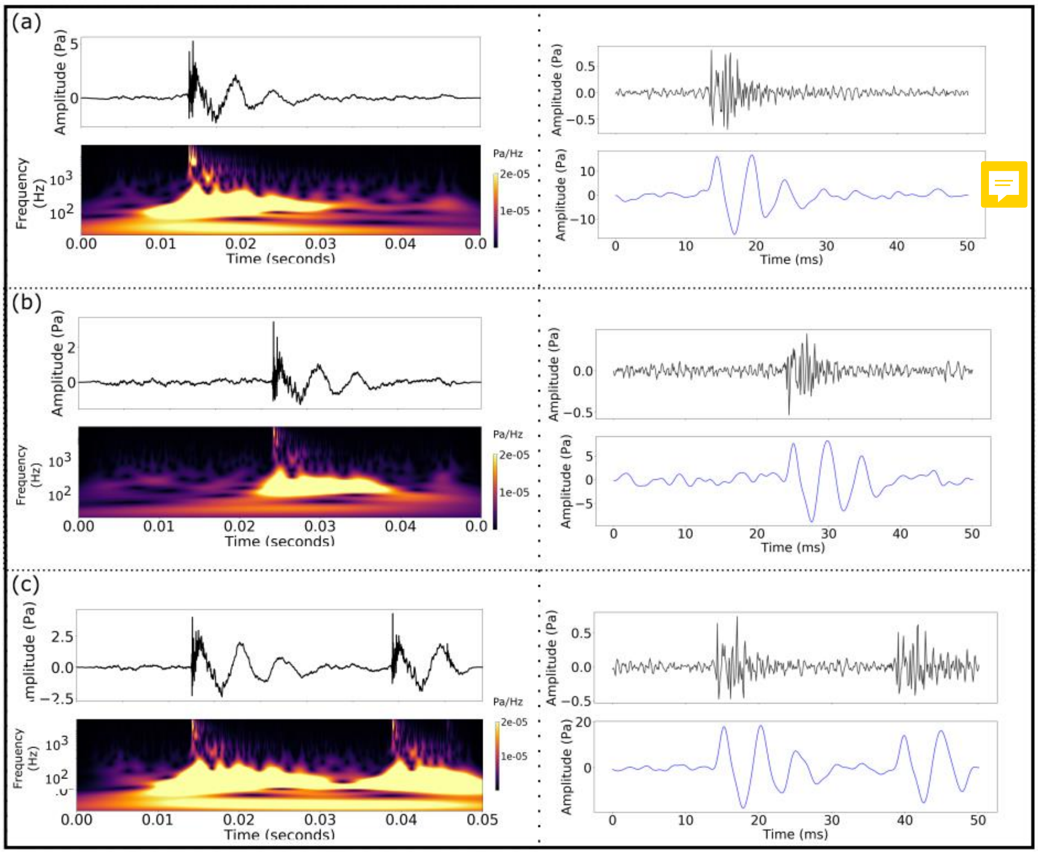

*Figure 2: **Time-frequency characteristics of bubble events**: Left: typical waveform and corresponding time-frequency evolution (wavelet transform) of four bubbles (panel (c) contains two events). Right: filtering to isolate the impulsive and resonant components of the waveforms. Top panel (black) shows the impulsive onset (high-pass filtered above 600 Hz), and bottom panel (blue) shows the resonance of ~200 Hz (low-pass filtered below 600 Hz). All the waveform amplitudes are in Pascal.*

Visual inspection of the records reveals that most (> 70%) of the bubble events are immediately preceded (< 1 s) by short (~400 ms) pressure pulses at frequencies below ~40 Hz (Figure 3). These signals occur more frequently (5/min) than the bubble events (0.4/min), but a statistical analysis confirms that there is a non-random association between the event times, with bubble events preferentially occurring after a low-frequency event (see Methods). A typical pulse begins with a rise to a pressure maximum (compression), followed by a drop to a pressure minimum (decompression), and the bubble events preferentially occur when the decompression rate is maximal (~50 Pa/s) during the initial pressure drop (Figure 3), consistent with cavitation in a flowing fluid [22].

We thus find a systematic relationship between the two types of acoustic signals, with low-frequency pressure pulses triggering the heterogeneous nucleation of non-condensable gas bubbles in the hydrothermal fluids. Of all the potential mechanisms by which hydrothermal fluid flow may generate noise[23], unsteady flow provides the most likely explanation for the low-frequency pulses we observed. The Reynolds number of the hot hydrothermal fluids being discharged at the Deep Hole field is on the order of $\sim 1 \times 10^5$, which is in the turbulent flow regime[24] . The turbulent interaction of the fluids with the sediments hosting flow could produce pulses of unstable flow that radiate acoustic energy in the 1-40 Hz band, and these turbulent instabilities can in turn trigger heterogeneous nucleation of bubbles in the flowing liquids[25] (Figure 4). The triggering process is stochastic, since during the 3-hour interval of interest about one out of every ten low-frequency events triggers a bubble nucleation event.

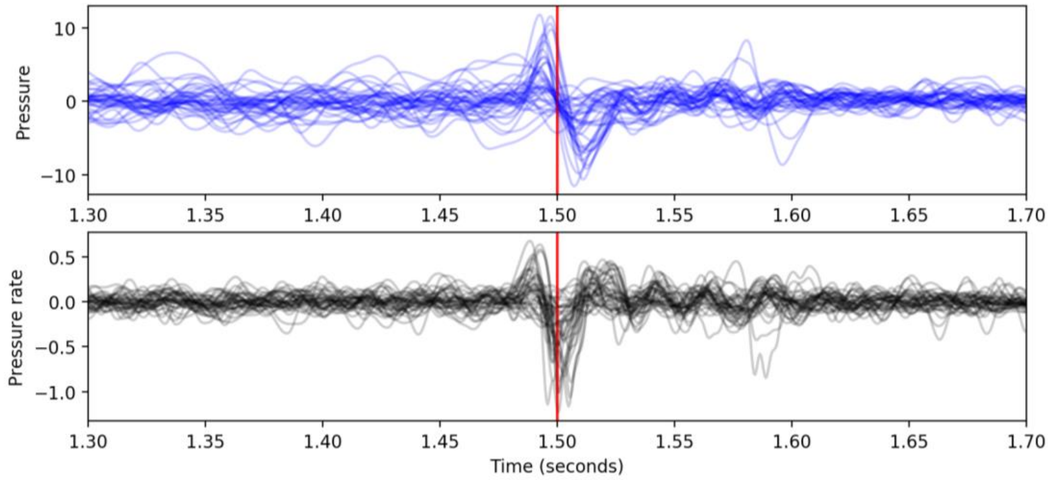

*Figure 3: **Nucleation of bubbles**: A subset of the low-frequency pressure signals (blue), and rate (black), aligned by time of bubble initiation (red lines). The amplitudes have been normalized by removing the mean and dividing by the standard deviation.*

The bubbles we detected are associated with relatively weak venting at a site that is ~100 m from the main discharge zone, which contains at least 20 active vents (Figure 1), many of which exhibited essentially continuous bubble fluxes during periods when visual observations could be made during remotely operated vehicle dives. The CO_2 flux associated with bubbles in our acoustic data (~5 x 10^{-10} kg/s) is thus likely insignificant compared to the apparently much larger fluxes through the main part of the vent field, and it is certainly insignificant compared to dissolved CO_2 fluxes through the main vent field. With a dissolved CO_2 concentration of ~7-18 mmol/kg[13], and a total discharge rate of ~56 kg/s [24], the Deep Hole field discharges ~46 - 119 kg/s of dissolved CO_2 into the lake.

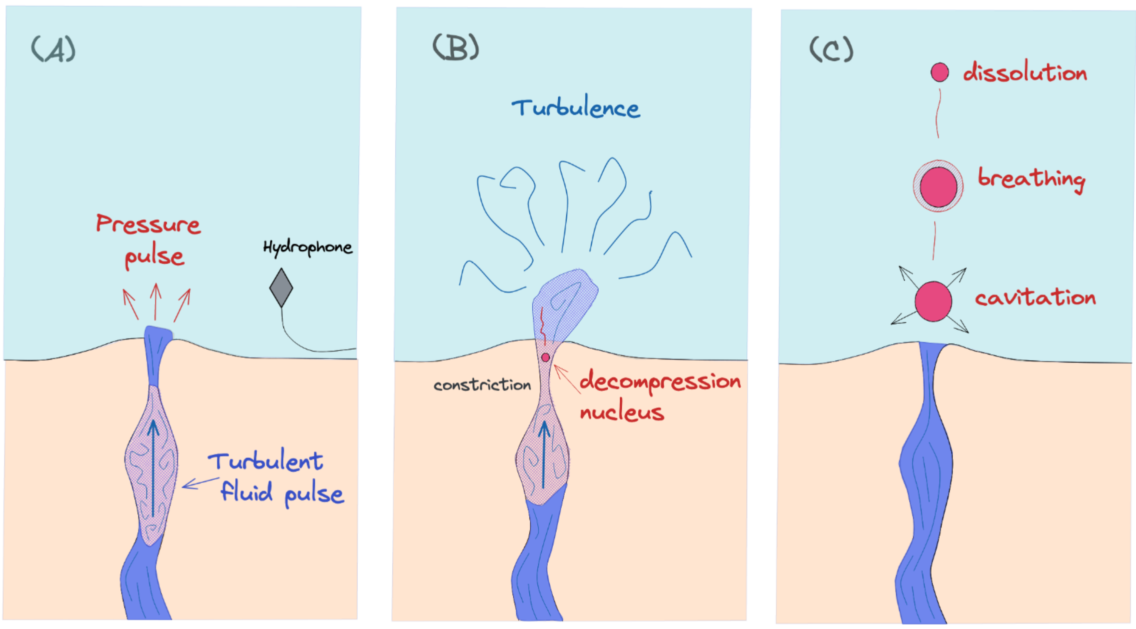

*Figure 4: **Conceptual model**: (A) A flow instability generated by the interaction of the hydrothermal fluid with the sediment matrix (e.g., a flowpath constriction), momentarily depressurizes the fluid, triggering the nucleation of a CO_2 bubble in the saturated fluid (B). The bubble oscillates at its*

natural frequency before dissolving into the under-saturated lake water or rising to the lake surface (C).

The heterogeneous nucleation of gas bubbles due to unsteady flow has important implications for the stability of the lake floor hydrothermal systems, and their potential to generate explosions. The bubble events we observed were triggered by instabilities with typical amplitudes of 10^{-1}-10 Pa (or 100-140 dB ref. 1 μPa) and maximum decompression rates of ~50 Pa/s. The fact that these relatively small pressure perturbations can trigger bubble nucleation in the hydrothermal fluids implies that larger, and more sustained, pressure perturbations, such as could be generated by earthquakes or rapid changes in hydrostatic load, for example, could trigger wholesale nucleation of CO_2 bubbles in the permeable sediments hosting fluid flow. Such an event could readily explain the formation of the pockmarks that characterize hydrothermal discharge zones on the lake floor [17], and it could play a role in the initiation of large, catastrophic hydrothermal explosions, which have occurred at least twice in the post-glacial (~13 ky) history of Yellowstone Lake [18].

Concluding paragraph

Our data from Yellowstone Lake demonstrates the utility of acoustic methods for constraining gas fluxes in underwater environments, and provide new insight into bubble nucleation in hydrothermal fluids. The observation of CO_2 bubble nucleation in hydrothermal fluids due to small-scale flow instabilities has important implications for the stability of the sublacustrine hydrothermal system and the processes associated with hydrothermal explosions. Considering that more than 75% of Earth's volcanism occurs underwater[6], our results support the idea that acoustic methods have the potential to play an important role in monitoring and measuring volcanic gas fluxes[7] at the global scale, which is one of the centennial grand challenges highlighted by the scientific community[26].

Methods

The hydrophone sensor was a High Tech HTI-90-U hydrophone (Figure S1), with a nominal frequency response of 2 Hz to 20 kHz. The acoustic records were first visually inspected before choosing the STA/LTA (STA of 0.3 second, LTA of 5 seconds) parameters for event detection. Each detection event was verified manually to remove any spurious detections from the final catalogs. To characterize the signals, we first resampled them to 2000 Hz because the characteristic signals have dominant frequencies around 200 Hz (so-called breathing). For the low-frequency signals, we low-passed the signals below 50 Hz before computing the STA/LTA. We then automatically calculated the dominant frequencies for the 66 bubble signals using both wavelet (Morlet) and Fourier transforms. We removed the mean and the linear ~~tend~~, and tapered the signal with a Hanning window (10%) before conducting spectral analyses. The data processing was done using Obspy[27].

Based on the dominant frequency, we compute the radius (r) using Minnaert's[20] relation, assuming an adiabatic equation of state for the gas in the bubble:

$$r = \frac{(3P\gamma/\rho)^{1/2}}{2\pi f}$$

Where P is the hydrostatic pressure (882 kPa), γ is the ratio of specific heats of the gas in the bubble (1.3 for CO_2), ρ is the density of the liquid (1000 kg/m^3) and f is the dominant frequency (in Hz).

Uniformity plots (Figure S4) demonstrate that the bubble events can be modeled as a Poisson process, where the event times are uniformly distributed over the observation interval. Assuming a Poisson process, we used a parametric bootstrap test to assess the association between the bubble events (66) and the low-frequency events (665), with the test statistic equal to the mean delay time between each bubble event and the immediately preceding low-frequency event. To implement the test, we generated 1000 bootstrap replicates of the bubble event catalog sampled using a uniform distribution, and calculated the mean delay time for each replicate catalog, keeping the low-frequency event times fixed. Only two of these replicate catalogs had mean delays times less than the observed value, yielding a *p*-value of 0.002, with an approximate error of ±0.001. This test demonstrates that the bubble events are associated with a preceding low-frequency event to a very high degree of confidence.

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Acknowledgements

The authors thank Alan Gardner for development of the hydrophone and audio recording package, Andy Solow for helpful statistical advice, and The Global Foundation for Ocean Exploration for logistical support during field work. We thank the Yellowstone National Park Resources office for the support required to conduct research within Yellowstone National Park. This research was supported by the National Science Foundation grant EAR-1516361 to Robert A. Sohn. All work in Yellowstone National Park was completed under an authorized research permit (YELL-2018-SCI-7018).

Author Contributions

R.S. acquired the data. J.V. and C.C. analyzed the recordings. All the authors contributed to the interpretation and writing.

Competing interests

We do not have any competing interest.

Materials & Correspondence

The data that support the findings of this study are available from R. Sohn. The computer codes generated are available from the corresponding author and will be freely made available to the scientific community.

Extended data

Supplementary figures and videos

A video showing bubbles and fluids discharged into the lake is provided as supplementary material.

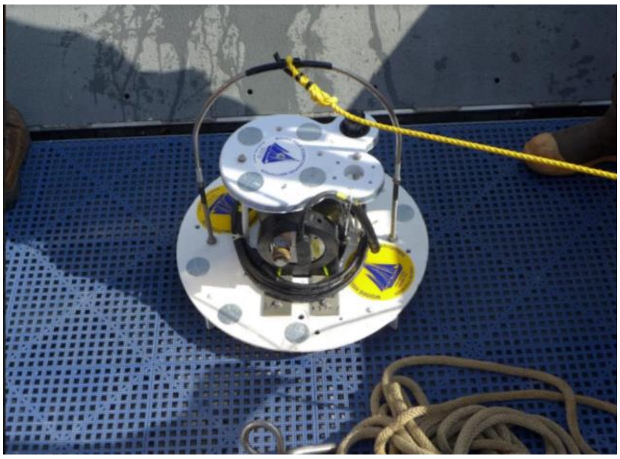

Figure S1: Hydrophone used in this study

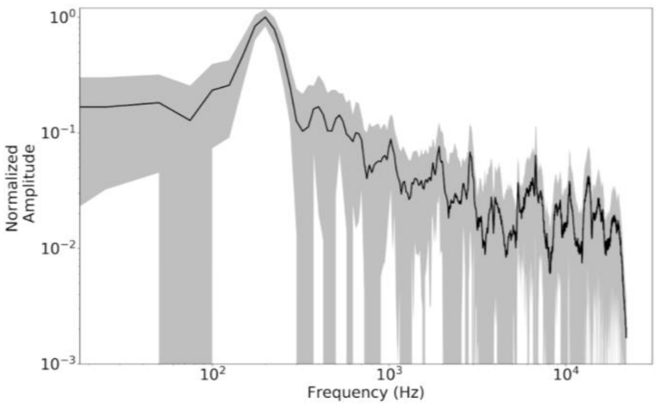

Figure S2: Stacked spectra of bubbles showing the breathing mode of the bubbles (dominant frequency of ~200 Hz) and the higher frequencies (from 500 Hz to Nyquist (20500 Hz)) corresponding to nucleation.

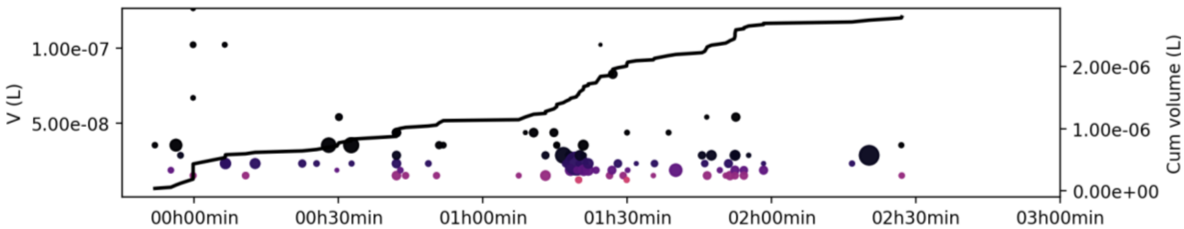

Figure S3:estimated volume for individual bubbles (circles, in L) based on Minnaert's relation[20] *and degassing rate (cumulated volumes, black line)*

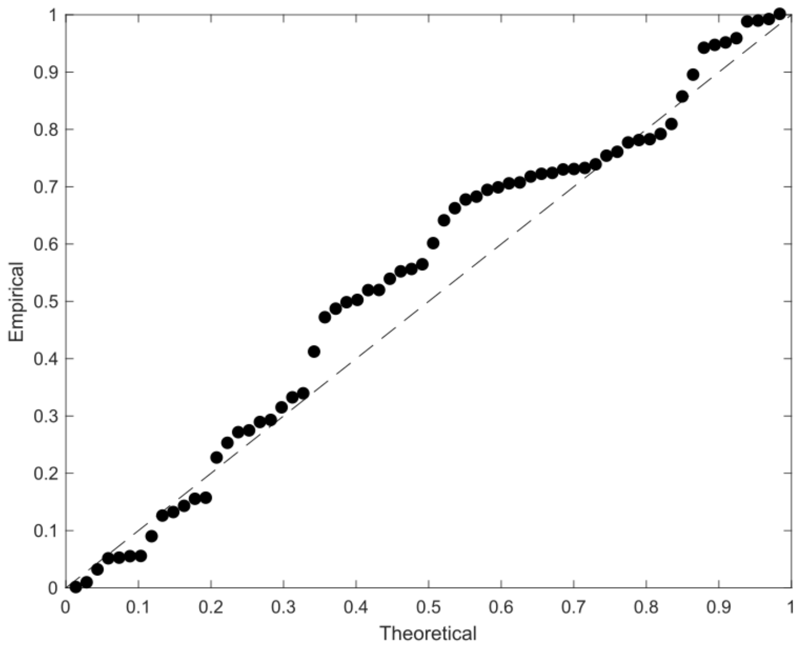

Figure S4: Uniformity plot for bubble event catalog. The empirical distribution of the bubble events during the 3-hour interval of activity (black dots) is compared to the theoretical uniform distribution (dashed line), demonstrating that the assumption of a uniform distribution (Poisson process) is valid for this event (point process) catalog.

Data availability

The datasets generated during and/or analysed during the current study are available from the corresponding author. The data that support the findings of this study are available from R. Sohn.

General comments

The submitted manuscript presents an acoustic emission study focused on the recognition, and understanding, of gas fluxes (particularly CO_2) and bubble nucleation process from active underwater hydrothermal systems.

The authors performed the analyses of acoustic data recorded by a hydrophone system deployed on the floor of Yellowstone Lake to characterize the nature of gas discharge in one of the lake floor thermal fields: the 'Deep Hole' located to the southeast of Stevenson Island. A secondary vent, ca. 100 m from the main discharge vent field, was chosen for the investigation. The investigation of the acoustic data has been cross-analysed with video footages of the bubbling vent to compare the bubble sizes estimated using the acoustic signals.

Authors recognized sixty-six discrete events during a 3-hours interval, all exhibiting a characteristic waveform, with bubble rising showing a resonant frequency of 100-200 Hz. Such frequencies were used to estimate bubble diameters of ~3-6 mm, which appear to agree with visual observations. Authors claim that the heterogeneous nucleation of CO_2 bubbles in the hydrothermal fluids is triggered by turbulent flow instabilities within the lake floor sediment. In particular they state that their acoustic data demonstrate that the CO_2 saturated fluids are unstable to the fast (~50 Pa/s), but small (< 10 Pa), pressure perturbations associated with unsteady flow through the lake floor sediments. They finally conclude that such process could explain the formation of the pockmarks that characterize hydrothermal discharge zones on the lake floor, and it could play a role in the initiation of large, catastrophic hydrothermal explosions, as those which have occurred in the post-glacial history.

Results are significant by the fact that i) the proposed acoustic method appear to be a very interesting and novel tool able to constrain gas fluxes in underwater environments – with important implications for monitoring and measuring underwater gas fluxes from volcanic and non-volcanic sources, ii) show new insight into bubble nucleation in hydrothermal fluids, and iii) shed a quantitative light on a very important process characterizing the state of active hydrothermal systems. Therefore, the study is definitely worthy of publication, which I recommend after that the following comments (**minor**) have been suitably addressed.

Cristian Montanaro

Main comments

In general, the manuscript should be “adjusted” to broader the implications of the key and important results from this study; this can be done by comparing/discussing findings from this study case in the context of other known works on degassing and venting dynamics from underwater environments.

Concerning the figures, please double check consistency between units discussed in the text and those reported in the graphs, as well make sure that no labels are missing from the x-y-axis of graphs (see also more comments below).

- In the ***Summary paragraph:***

Line 1: expand a bit more on the effect of gases, for instance: "...*including CO_2, which can affect the surrounding environments, and eventually threaten health and life of humans and animals, as well as damage crops and property. Therefore, the understanding of gas fluxes remains a...*". This will strengthen more the importance of the study.

Line 11: Would be good to specify in the sentence where the instabilities occur during the rising of hydrothermal fluids. "... *within sediments close to the lake floor*...?

Line 19: To broaden the impact of this novel observation, it would be worthy to briefly mention how the findings from this study can improve the understanding of similar processes from other "intra-caldera lakes" (e.g. lake Rotorua in NZ) or lake sitting within actively degassing craters (e.g. Rotomahana lake in NZ, or Albano Lake in Italy). See also comment below.

- In ***Main – Introduction***:

Line 24: Another good example could be the deadly effect of gas observed after the phreatic eruption within the Diëng plateau (Le Guern et al. 1982).

Line 26: The study from Aiuppa et al. (2019) can be also another good example of CO_2 flux estimation.

Line 47: Mary Bay is more a cluster of craters or a crater field. *…115 m crater field…*

Line 50: I think here is worthy to mention the work of Thiéry et al. (2010), which discusses the decompression of aqueous solutions and their explosive potential.

- In ***Results***:

Line 57: citation of Figure S1 and S2 are missing. I suggest to add the first one here: *...acoustic hydrophone (Figure S1) on the floor…*

Lines 63-67: I recommend to label each panel in the Figure 2 - a, c, e on the left panels, and b, d, f the right panel. Then you can cite (Figure 2 a,c,e) after oscillation, and put the reference (Figure 2 b,d, f) at the end of the next sentence. This would make it easier for the reader to follow text and figure. To be also noted that in panel **c** labels on the y-axis are cut and/or missing (e.g. -20 in the right panel of c).
Maybe I don't get it correctly, but by looking at the figure, it seems that the largest amplitude signals last 10 ms, while the overall duration is 50 ms? if not, please try to explain better the sentence.

Line 67: change time in 13:00 for consistency with the time format given before

Line 72: … *bubble* [20] *(see Methods)*…

Line 74: The estimated volume here should be referenced to *Figure S3*: in the figure the volume on the y-axis should be changed in *mL* for consistency with the reported median volume in the text (or vice versa text changed in *L*).

Line 75: The authors stated that the video observations are consistent with the estimated radius. However, from the video (.MP4 file) it is hard to have an idea of the surrounding elements sizes (e.g. rocks on the lake floor, ROV arm).
I recommend to add a figure/panel that shows one (or more) frame(s) of the video with bubbles, and including a reference scale (size of the ROV arm for example?) to which to compare the bubbles size. Such a Figure might be implemented as a panel in Figure 1, for instance.

Line 76: Somewhere before, or in the Methods it is important to specify P-depth of the investigated site, as well as the temperature (T-profile if measured with probes at the investigated site) since these are key parameters controlling the fluids behaviour and the bubble nucleation process. Moreover, the pressure is used in the Minnaert's relation used to calculate the bubble radius.

Lines 74-78: The paragraph is too long. I recommend to split it at least in two to make it easier to read and follow.

Lines 90-97: Generally, this whole paragraph is not well explained by referencing to Figure 3, where: i) units of y-axis are missing, and reported values do not match what reported in the text (for instance, the frequencies are reported and referenced as Hz in the text - not in the Figure), ii) decompression rate is in Pa/s, not reported in the figure y-axis.
Please, modify the text or provide a new Figure 3

Line 97: It might be worthy to clarify the meaning of cavitation:
…consistent with cavitation (i.e. formation of vapor-filled bubbles at low pressures) in a flowing fluid…

Line 104:...*interaction with the hosting sediments could...*

Lines 112-114: Too long sentence. What about:
…The bubbles we detected are associated with relatively weak venting at a site that is ~100 m from the main discharge zone containing at least 20 active vents (Figure 1). Such vents exhibited continuous bubble fluxes during periods when our visual observations could be made with the remote operating vehicle…

Lines 115-116: How did you estimated/measured the *$5x10^{-10}$ kg/s* flux value? Eventually specify it here, and/or add the description in the Method

…*"The estimated CO_2 flux associated with bubbles in out acoustic data (ca. $5x10^{-10}$ kg/s; see Methods)"*…

Lines 117-118: *...through the whole 'Deep Hole' area. Indeed, with a dissolved...*

Figure 4: I really like this figure, simple and effective. I also think it might benefit from adding the produced frequency by the indicated process. For e.g. in the panel a, it can be added 1-40 Hz with a line/arrow pointing to the turbulent fluid-sediment interaction. Similar thing can be done in panels b-c.

In the **Figure 4 caption**: *"flowpath constriction"* can this be produced by a localized mineral precipitation-seal? Similar obstruction can be observed at discharging spots in sub-aerial environment (see comment below). **Line 125:** Maybe a brief explanation of *breathing* and *dissolution* when discussing the cavitation, should be briefly mentioned in the text within the "Results" text.

Lines 127-128: I think it is also worthy to briefly discuss the effect of pressure-depth at which the process was observed, and what expected at higher depth or shallower depth (pressures). In other words, I think it can be important to consider/discuss if there is a "depth-pressure limit" for unsteady flow to be an efficient process enhancing the heterogeneous bubble nucleation.

Moreover, the unsteady flows are explained to be the result of fluid-sediment interaction: another mechanism which might amplify-cause unsteady flow, favouring CO_2 bubble nucleation, can be the precipitation of minerals from hydrothermal solutions (in analogy with sub-aerial environments) that can result in the formation of constriction just below – or at the lake floor level (for example cementing clasts as those shown in the video). Mineral precipitation and cementation processes have being already discussed in Shanks et al. (2005, 2007). This process can be briefly discussed here, considering known cases from Yellowstone (above mentioned Shanks studies), or from other volcanic and geothermal areas with active sub-aerial hydrothermal manifestations (e.g. Jones & Renaut, 2003; Kennedy et al., 2020).

Lines 128-130: This is a critical point, and a very important finding of this study.

Lines 134-135: It would be more impact-full here to make comparisons with other known cases (see next comment as well), to show that results – and derived processes – observed here by using the acoustic method for the first time, might have been overlooked in underwater environments elsewhere, where instead they can represent a key mechanism explaining lake floor deformation, collapses (sinkhole-like), or the occurrence of underwater explosions.

Lines 135-137: This statement "alone" seems to be too strong. It could be expanded a bit more with a short paragraph summarizing the trigger mechanism envisaged for mid-large eruptions at Yellowstone. For instance, Morgan et al. (2009) indicate that areas affected by cataclysmic explosion were (1) dominated by high heat flow, (2) had been repeatedly subjected to inflation and deflation of the caldera, and (3) had an active well-developed sub-lacustrine hydrothermal system with significant alteration and vein formation. Most importantly (and related to the key finding of bubbling process enhanced by small pressure perturbations) Morgan et al. suggest that a significant seismic event related to formation of the sublacustrine Lake Hotel graben may have helped trigger the explosions within and around the lake.
In this, the presence of large amount of dissolved CO_2 in hydrothermal fluids within sediments, and the "fast" nucleation following larger P-perturbation, might have strongly contributed in over-pressurizing the system prior to, and/or during the explosive events – for the effect of CO_2 in enhancing explosivity is also worthy to consider the works of (Thiéry and Mercury 2009; Hurwitz et al. 2016).

- In the ***Concluding Paragraph***:

As mentioned in the comments above, and in order to broaden the impact of the interesting findings from this study, I recommend the authors to discuss the potential key role of enhanced bubbling nucleation in the formation of pockmark fields (or large sinkholes), which have also been observed at other caldera-hosted lakes, as in NZ's Rotorua (https://www.gns.cri.nz/Home/News-and-Events/Media-Releases-and-News/Scientists-and-Navy; Pearson, 2007), Italy's Albano Lake (Colli Albani District - Anzidei et al., 2008), Azores Archipelago (Andrade et al. 2019), and at other crater-hosted lakes with strongly degassing activity as at Rotomahana (de Ronde et al. 2016). At such sites, the formation of large sinkholes, and high the fluxes of CO_2 from the floor are recognized at specific geological sites (e.g. fractured zones following main structural elements, vents of previous eruptions, etc.).

- In ***Methods***:

Line 151: Please define STA/LTA (Short-Term to Long-Term moving averages)

Lines 155-156: Might be useful a reference for these methods (Morlets-Fourier).

Lines 162-163: Pressure (or depth) at the investigated site should be indicated here in the Methods, or earlier in the introduction (see also comment above). This is a key parameter used in the calculation of the bubble radius.

Cited references:

Aiuppa A, Fischer TP, Plank T, Bani P (2019) CO2 flux emissions from the Earth's most actively degassing volcanoes, 2005–2015. Sci Rep 9:. https://doi.org/10.1038/s41598-019-41901-y

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Hurwitz S, Clor LE, McCleskey RB, et al (2016) Dissolved gases in hydrothermal (phreatic) and geyser eruptions at Yellowstone National Park, USA. Geology 44:G37478.1. https://doi.org/10.1130/G37478.1

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Pearson LK (2007) The nature, composition and distribution of sediment in Lake Rotorua, New Zealand. The University of Waikato, Hamilton, New Zealand.

Shanks WCP, Alt JC, Morgan LA (2007) Geochemistry of Sublacustrine Hydrothermal Deposits in Yellowstone Lake—Hydrothermal Reactions, Stable-Isotope Systematics, Sinter Deposition, and Spire Formation. US Geol Surv Prof Pap 1717:33

Shanks WCP, Morgan LA, Balistrieri L, Alt JC (2005) Hydrothermal Vent Fluids, Siliceous Hydrothermal Deposits, and Hydrothermally Altered Sediments in Yellowstone Lake. Geotherm Biol Geochemistry In: Inskee:53–72

Thiéry R, Loock S, Mercury L (2010) Explosive properties of superheated aqueous solutions in volcanic and hydrothermal systems. In: Rzoska S, Drozd-Rzoska A, Mazur V. (eds) Metastable Systems under Pressure, NATO Scien. Springer, Dordrecht, pp 293–310

Thiéry R, Mercury L (2009) Explosive properties of water in volcanic and hydrothermal systems. J Geophys Res Solid Earth 114:1–19. https://doi.org/10.1029/2008JB005742

Reviewer #1

"This is a nice use of hydroacoustic data to study source processes in hydrothermal systems. The paper is generally well-written and most of the conclusions are well-supported. Short format papers don't leave a lot of room to expand on discussion items but there were a couple places where unsupported statements were made that trended toward conjecture. The two main examples were on lines 50 and 136 when seemingly trying to tie the nucleation of small bubbles observed in this study to 'catastrophic hydrothermal explosions'. These statements are provided without real evidence or support, which raises many questions. Like, How would a larger bubble or bubbles result in a 'catastrophic explosion'? The bubbles aren't highly pressurized, correct? There is also a lot of seismic activity in the area and yet few explosions. What evidence is there that the two large explosions over the past 13 ky were related to CO2 bubbles vs. pressure build up under an impermeable hydrothermal cap?

This should be toned down / removed or supported with evidence."

- ➔ *As noted above, we are grateful for the opportunity to clarify the possible relationship between our observations and hydrothermal explosions in Yellowstone Lake. Two, distinct types of hydrothermal explosion deposits are found in Yellowstone Lake: 'large' explosions triggered by vaporization of subsurface fluids, and 'small' explosions likely triggered by the episodic release of CO_2. It was not our intention to imply that the bubble nucleation process we observed is linked to the large, catastrophic explosions. Rather, we believe our results pertain to the small, pockmark forming, explosions that occur in the lake. Thank you for pointing out this discrepancy, as it can be a sensitive topic. We have revised our discussion of this topic to clarify our interpretation. These bulk of these revisions are found in lines 142-154 of the revised manuscript.*

Line 9: A volcanic reference that would be appropriate here is:
Lyons, J.J., Haney, M.M., Fee, D. et al. Infrasound from giant bubbles during explosive submarine eruptions. Nat. Geosci. 12, 952–958 (2019). https://doi.org/10.1038/s41561-019-0461-0

- ➔ *We have now added this reference*

Line 24-25: While this is still somewhat true for CO2, the advancements in both ground-based and satellite sensors make this statement read as a bit out of touch with the current state of the art for other gas species, like SO2. e.g., Carn, S., Fioletov, V., McLinden, C. et al. A decade of global volcanic SO2 emissions measured from space. Sci Rep 7, 44095 (2017). https://doi.org/10.1038/srep44095

- ➔ *We have toned this down: 'Gas fluxes* ***can be*** *difficult…'*

Line 43: field -> suggest replacing with a different or more descriptive term since 'field' is being used to at least 4 times in the previous few lines to describe a thermal field.

- ➔ *Corrected*

Line 43: pockmark -> pockmarked

- ➔ *Corrected*

Line 46: When did the Mary Bay explosion occur?

- ➔ *13,400 years ago. We have added the reference*

Line 50: potentially explosive -> this statement needs to be backed up with references, particularly since it is stated in the same sentence that the exsolution process is poorly understood.

- ➔ *We refer to the seminal papers of Thiéry and co-authors*

Figure 1. Maps are a bit blurry / low-fi in the PDF.

➔ *Corrected*

Line 66: am isn't necessary since you are using a 24 hr convention above. However, it should read ~02.30.

➔ *Corrected*

Line 67: pm -> see previous comment. Please insure all time references follow the same convention throughout the paper.

➔ *Corrected*

Line 68-69: What evidence is there that these signals represent bubble nucleation? Given that the rest of the interpretation hinges on this, it is surprising that no evidence is presented. If this is your interpretation, it needs to be written as such, rather than a statement of fact, which is how it currently reads.

➔ ***R1**: Based on our analyses, bubble nucleation is the only plausible source mechanism for the acoustic signals. However, we recognize that we did not adequately explain our reasoning for this conclusion in the manuscript. We have revised the manuscript to address this issue, and to also include more details about the conditions required to nucleate CO_2 bubble in saturated aqueous solutions. One outcome of the additional research we did on this topic is that the only way to nucleate CO_2 bubbles in fluids with the relatively low supersaturation values associated with the fluids at our study site and the relatively low amplitude of the pressure fluctuations that appear to be triggering nucleation, is for there to be pre-existing microbubbles in the fluids. This information is now included in the discussion. We discuss the fact that the bubbles are most likely composed dominantly of CO_2 in lines 106-181 of the revised text. We discuss the process model for CO_2 bubble nucleation in lines 127-135.*

Line 73: Please add that the gas is assumed to be solely CO2 e.g., a 'spherical bubble of CO2'

➔ *Corrected*

Line 76-77: For the non-experts, would you explain briefly (or add an appropriate reference) why a vapor bubble would be expected to condense?

➔ *To be stable, a vapor bubble must be in thermal equilibrium with the surrounding fluid. When the ambient fluid is at a temperature of 4°C a vapor bubble will quickly condense. We feel it is appropriate to assume this level of basic thermodynamic knowledge for readers of Nature Communications Earth & Environment, but if the Editor disagrees we can add a few sentences to explain it.*

Figure 2. Not quite publication ready. 1) Y-axis labels are varying font size, some are cut-off and not fully legible, and all should be aligned to the left. 2) The scales of the wavelet transform figures would show better if they spanned the height of the wavelet figure, and were all in the same location relative to the wavelet image. 3) X-axis labels are cut off at the bottom. 4) vertical dotted line between left and right panels is unnecessary. 5) Why have two different time scales (sec vs ms) for the plots in the left and right columns? 6) White space to left and right of right column waveforms should be eliminated.

➔ *Thanks, we have modified the figure according to the points listed by the reviewer*

Line 116: why is it that these fluxes are only 'apparently much larger'? Have they not been measured? Is this just based on observations from the ROV?

➔ *We have clarified our discussion in lines 127-135.*

Line 136: The idea that a 'catastrophic hydrothermal explosion' could be initiated based on the nucleation of the small bubbles in this study seems like conjecture without more supporting evidence. How would a larger bubble result in a 'catastrophic explosion'? The bubbles aren't highly pressurized, correct? There is also a lot of seismic activity in the area and yet few explosions. What evidence is there that the two large explosions over the past 13 ky were related to CO2 bubbles vs. pressure build up under an impermeable hydrothermal cap? This should be toned down / removed or supported with evidence.

- ➔ *Two, distinct types of hydrothermal explosion deposits are found in Yellowstone Lake: 'large' explosions triggered by vaporization of subsurface fluids, and 'small' explosions likely triggered by the episodic release of CO_2. It was not our intention to imply that the bubble nucleation process we observed is linked to the large, catastrophic explosions. Rather, we believe our results pertain to the small, pockmark forming, explosions that occur in the lake. Thank you for pointing out this discrepancy, as it can be a sensitive topic. We have revised our discussion of this topic to clarify our interpretation. These bulk of these revisions are found in lines 142-154 of the revised manuscript.*

Line 149: What was the height of the hydrophone off the lake floor?

- ➔ *~30 cm. We have added this information in the Methods section*

Line 163: Are the hydrothermal fluids all cold / lake temp in the study area?

- ➔ *No. The mean exit-fluid temperature of 49 active vents was 132°C, with a range of 74-174°C. We have added this information in paragraph 2 (introduction), lines 40-41.*

Reviewer #2

In general, the manuscript should be “adjusted” to broader the implications of the key and important results from this study; this can be done by comparing/discussing findings from this study case in the context of other known works on degassing and venting dynamics from underwater environments. Concerning the figures, please double check consistency between units discussed in the text and those reported in the graphs, as well make sure that no labels are missing from the x-y-axis of graphs (see also more comments below).

➔ *Thanks, we have paid much attention to this in the new version*

In the Summary paragraph:
Line 1: expand a bit more on the effect of gases, for instance: "...including CO2, which can affect the surrounding environments, and eventually threaten health and life of humans and animals, as well as damage crops and property. Therefore, the understanding of gas fluxes remains a...”. This will strengthen more the importance of the study.

➔ *Thanks, we have followed this suggestion*

Line 11: Would be good to specify in the sentence where the instabilities occur during the rising of hydrothermal fluids. "... within sediments close to the lake floor...?

➔ *Yes, this has been added*

Line 19: To broaden the impact of this novel observation, it would be worthy to briefly mention how the findings from this study can improve the understanding of similar processes from other "intra-caldera lakes" (e.g. lake Rotorua in NZ) or lake sitting within actively degassing craters (e.g. Rotomahana lake in NZ, or Albano Lake in Italy). See also comment below.

➔ *Good point, we have added:*
“The observation of CO_2 bubbles nucleating in hydrothermal fluids due to small pressure perturbations informs our understanding of hydrothermal explosions in Yellowstone Lake, demonstrates that acoustic data in underwater environments can provide insight into the stability of gas-rich systems, as well as gas fluxes.”

• In Main – Introduction:
Line 24: Another good example could be the deadly effect of gas observed after the phreatic eruption within the Diëng plateau (Le Guern et al. 1982).

➔ *Suggestion followed*

Line 26: The study from Aiuppa et al. (2019) can be also another good example of CO2 flux estimation.

➔ *Suggestion followed*

Line 47: Mary Bay is more a cluster of craters or a crater field. …115 m crater field…

➔ *Mary Bay has a nested structure of craters, with many small ones superposed on the largest one. Our text in the paper is correct. The smaller craters formed after the large explosion.*

Line 50: I think here is worthy to mention the work of Thiéry et al. (2010), which discusses the decompression of aqueous solutions and their explosive potential.

➔ *We completely agree. This is a very important, and unfortunately poorly known, work.*

• In Results:
Line 57: citation of Figure S1 and S2 are missing. I suggest to add the first one here: …acoustic hydrophone (Figure S1) on the floor…

- ➔ *Suggestion followed. We are now citing Figure S2 too (l. 74))*

Lines 63-67: I recommend to label each panel in the Figure 2 - a, c, e on the left panels, and b, d, f the right panel. Then you can cite (Figure 2 a,c,e) after oscillation, and put the reference (Figure 2 b,d, f) at the end of the next sentence. This would make it easier for the reader to follow text and figure. To be also noted that in panel c labels on the y-axis are cut and/or missing (e.g. -20 in the right panel of c).
Maybe I don't get it correctly, but by looking at the figure, it seems that the largest amplitude signals last 10 ms, while the overall duration is 50 ms? if not, please try to explain better the sentence.

- ➔ *Corrected. We have not followed the figure referencing as both left and right panels are showing the same spectral features*
- ➔ *And we have modified this sentence*

Line 67: change time in 13:00 for consistency with the time format given before

- ➔ *Corrected*

Line 72: … bubble 20 (see Methods)…

- ➔ *Corrected*

Line 74: The estimated volume here should be referenced to Figure S3: in the figure the volume on the y-axis should be changed in mL for consistency with the reported median volume in the text (or vice versa text changed in L).

- ➔ *Corrected*

Line 75: The authors stated that the video observations are consistent with the estimated radius. However, from the video (.MP4 file) it is hard to have an idea of the surrounding elements sizes (e.g. rocks on the lake floor, ROV arm).
I recommend to add a figure/panel that shows one (or more) frame(s) of the video with bubbles, and including a reference scale (size of the ROV arm for example?) to which to compare the bubbles size. Such a Figure might be implemented as a panel in Figure 1, for instance.

- ➔ *We have tried to do this, but the frame rate of the video was too slow to capture the bubbles. They show up as blurs in the still images with sizes that cannot be measured accurately, unfortunately.*
  In the video, the outer diameter of the semi-transparent snorkel at the end of the sampling device is ~5 cm. We have added this information.

Line 76: Somewhere before, or in the Methods it is important to specify P-depth of the investigated site, as well as the temperature (T-profile if measured with probes at the investigated site) since these are key parameters controlling the fluids behaviour and the bubble nucleation process. Moreover, the pressure is used in the Minnaert's relation used to calculate the bubble radius.

- ➔ *We have added a sentence regarding vent fluid temperatures in the introduction and mentioned the pressure used to derive bubble radii.*

Lines 74-78: The paragraph is too long. I recommend to split it at least in two to make it easier to read and follow.

- ➔ *Suggestion followed*

Lines 90-97: Generally, this whole paragraph is not well explained by referencing to Figure 3, where: i) units of y-axis are missing, and reported values do not match what reported in the text (for instance, the frequencies are reported and referenced as Hz in the text - not in the Figure), ii) decompression rate

is in Pa/s, not reported in the figure y-axis.
Please, modify the text or provide a new Figure 3

- ➔ *Corrected*

Line 97: It might be worthy to clarify the meaning of cavitation:
…consistent with cavitation (i.e. formation of vapor-filled bubbles at low pressures) in a flowing fluid…

- ➔ *We mean hydrodynamic cavitation and have clarified this in the text, lines 192-196.*

Line 104:...interaction with the hosting sediments could...

- ➔ *corrected*

Lines 112-114: Too long sentence. What about:
…The bubbles we detected are associated with relatively weak venting at a site that is ~100 m from the main discharge zone containing at least 20 active vents (Figure 1). Such vents exhibited continuous bubble fluxes during periods when our visual observations could be made with the remote operating vehicle…

- ➔ *This entire section has been revised to clarify our treatment of dissolved vs exsolved gas fluxes.*

Lines 115-116: How did you estimated/measured the 5x10-10 kg/s flux value? Eventually specify it here, and/or add the description in the Method
…"The estimated CO2 flux associated with bubbles in out acoustic data (ca. 5x10-10 kg/s; see Methods)"…

- ➔ *We have added this in the method and corrected the values*
  "We calculate the volume for each bubble, assuming they are all spherical (V=4/3πr3), then compute the cumulative volume. We then multiply by the density of CO_2 at these pressures (20.5 kg/m3) and divide by total time of bubble records (i.e., 3 hours). We estimate the flux to be $7.67*10^{-6}$ kg/s.

Lines 117-118: ...through the whole 'Deep Hole' area. Indeed, with a dissolved...

- ➔ *suggestion followed*

Figure 4: I really like this figure, simple and effective. I also think it might benefit from adding the produced frequency by the indicated process. For e.g. in the panel a, it can be added 1-40 Hz with a line/arrow pointing to the turbulent fluid-sediment interaction. Similar thing can be done in panels b-c.

In the Figure 4 caption: "flowpath constriction" can this be produced by a localized mineral precipitation-seal? Similar obstruction can be observed at discharging spots in sub-aerial environment (see comment below). Line 125: Maybe a brief explanation of breathing and dissolution when discussing the cavitation, should be briefly mentioned in the text within the "Results" text.

- ➔ *Yes we think it can be produced by a localized mineral precipitation-seal. The kinetics of these processes are poorly constrained so we can't elaborate too much on this. But the timescales presented in Kennedy et al. (2020) are compatible with our observations (i.e., vigorous bubbling then nothing). We have added this in lines 119-121.*
- ➔ *We have removed any reference to breathing to simplify the text. We are only referring to resonance and dissolution*

Lines 127-128: I think it is also worthy to briefly discuss the effect of pressure-depth at which the process was observed, and what expected at higher depth or shallower depth (pressures). In other words, I think it can be important to consider/discuss if there is a "depth-pressure limit" for unsteady flow to be an efficient process enhancing the heterogeneous bubble nucleation.

- ➔ *CO_2 solubility decreases in a nearly linear way for the pressure-temperature domain corresponding to Yellowstone (e.g., Duan and Sun (2003) in Chemical Geology). Hence, this process should be nearly independent of depth and we have decided not to discuss this aspect extensively.*

Moreover, the unsteady flows are explained to be the result of fluid-sediment interaction: another mechanism which might amplify-cause unsteady flow, favouring CO2 bubble nucleation, can be the precipitation of minerals from hydrothermal solutions (in analogy with sub-aerial environments) that can result in the formation of constriction just below – or at the lake floor level (for example cementing clasts as those shown in the video). Mineral precipitation and cementation processes have being already discussed in Shanks et al. (2005, 2007). This process can be briefly discussed here, considering known cases from Yellowstone (above mentioned Shanks studies), or from other volcanic and geothermal areas with active sub-aerial hydrothermal manifestations (e.g. Jones & Renaut, 2003; Kennedy et al., 2020).

- ➔ *See above*

Lines 128-130: This is a critical point, and a very important finding of this study.

Lines 134-135: It would be more impact-full here to make comparisons with other known cases (see next comment as well), to show that results – and derived processes – observed here by using the acoustic method for the first time, might have been overlooked in underwater environments elsewhere, where instead they can represent a key mechanism explaining lake floor deformation, collapses (sinkhole-like), or the occurrence of underwater explosions.

- ➔ *See below*

Lines 135-137: This statement “alone” seems to be too strong. It could be expanded a bit more with a short paragraph summarizing the trigger mechanism envisaged for mid-large eruptions at Yellowstone. For instance, Morgan et al. (2009) indicate that areas affected by cataclysmic explosion were (1) dominated by high heat flow, (2) had been repeatedly subjected to inflation and deflation of the caldera, and (3) had an active well-developed sub-lacustrine hydrothermal system with significant alteration and vein formation. Most importantly (and related to the key finding of bubbling process enhanced by small pressure perturbations) Morgan et al. suggest that a significant seismic event related to formation of the sublacustrine Lake Hotel graben may have helped trigger the explosions within and around the lake.
In this, the presence of large amount of dissolved CO2 in hydrothermal fluids within sediments, and the “fast” nucleation following larger P-perturbation, might have strongly contributed in over-pressurizing the system prior to, and/or during the explosive events – for the effect of CO2 in enhancing explosivity is also worthy to consider the works of (Thiéry and Mercury 2009; Hurwitz et al. 2016).

- ➔ *Thanks again for your comment. We have added a paragraph at the end of the discussion in lines 155-165.*

• In the Concluding Paragraph:
As mentioned in the comments above, and in order to broaden the impact of the interesting findings from this study, I recommend the authors to discuss the potential key role of enhanced bubbling nucleation in the formation of pockmark fields (or large sinkholes), which have also been observed at other caldera-hosted lakes, as in NZ's Rotorua (https://www.gns.cri.nz/Home/News-and-Events/Media-Releases-and-News/Scientists-and-Navy; Pearson, 2007), Italy's Albano Lake (Colli Albani District - Anzidei et al., 2008), Azores Archipelago (Andrade et al. 2019), and at other crater-hosted lakes with strongly degassing activity as at Rotomahana (de Ronde et al. 2016). At such sites, the formation of large sinkholes, and high the fluxes of CO2 from the floor are recognized at specific

geological sites (e.g. fractured zones following main structural elements, vents of previous eruptions, etc.).

➔ *Again, we have followed your suggestion, many thank (lines 166-174)*

• In Methods:
Line 151: Please define STA/LTA (Short-Term to Long-Term moving averages)

➔ *done*

Lines 155-156: Might be useful a reference for these methods (Morlets-Fourier).

➔ *Done*

Lines 162-163: Pressure (or depth) at the investigated site should be indicated here in the Methods, or earlier in the introduction (see also comment above). This is a key parameter used in the calculation of the bubble radius.

➔ *done*

Reviewer #3

Major issues:
1. Line 12-13: “The resonant frequency of the bubbles (100-200 Hz) is consistent with diameters of ~3-6 mm, in agreement with visual observations.”
According to Minnaert’s equation and the given values through the text (P = 882kPa, \gamma = 1.3, \rho=1000 kg/m3, bubbles with diameters of 3~6 mm, water depth 100-115 m), I obtain the resonant frequency of the bubbles as 3.5-7 kHz, which is not the described 100-200 Hz in the text. This indicates that the authors have incorrect assumption of the acoustic regime.

- ➔ *Thanks for pointing this out. We indeed made a mistake, and the estimated radii were therefore wrong. These calculations have been corrected, but it doesn't change our interpretation.*

2. Line 17-18: “The nucleation of CO2 bubbles in hydrothermal fluids due to small pressure perturbations is a novel observation … “
There is no theory to support the claim of “the nucleation of CO2 bubbles” was existing “in hydrothermal fluids”.

- ➔ *We recognize that our discussion of CO_2 bubble nucleation was insufficient. Fortunately, there is an extensive literature regarding the nucleation of CO_2 bubbles in saturated aqueous solutions due to the beverage industry. The key point, that we now include in our discussion, concerns the necessity of there being microbubbles in the fluid, as we describe in our response to the Reviewer's 1 comment, **R1**, above. Our interpretation of CO_2 bubble nucleation is a logical deduction based on the available data, rather than an unequivocal fact, but if we are correct then the observation is indeed novel, and our data place hard constraints on the magnitude of the pressure drop required to nucleate CO_2 bubbles at the lake floor conditions, which has important implications for the stability of the hydrothermal system and how it may respond to pressure perturbations.*

3. Line 60 “The system was deployed in an area of relatively weak discharge, ~100 m north of the main vent field.”
Recent Research have shown that the acoustic signals from individual bubble film oscillation can only be detected in a few meters due to the attenuation and the low signal to noise ratio (SNR) in the field. The authors deployed only one acoustic hydrophone to monitor the acoustic signal, which is limited to improve the SNR. At 100m range, the authors won’t be able to measure sounds from individual “bubble events”, but a spectral frequency band if the leakage was strong. However, the authors measures some “bubble acoustic-like” acoustic waveforms (e.g., 1 ms at line 63 and top panels in Figure 2), which can only be possible that there were a number of weak gas seeps close to the hydrophone.

- ➔ *The hydrophone was deployed in close proximity to an active vent that was not inside the main discharge zone (see Figure 1B).*

4. Line 79-81: “These visual observations, combined with the fact that the hydrothermal fluids discharged at the Deep Hole field are rich in magmatic gases and are saturated in CO2, lead to the conclusion that the bubbles are dominantly composed of magmatic CO2, with possibly minor components of H2S.”
This is an ill-conditioned deduction. The authors won’t be able to have such a strong conclusion without enough chemical information from the seep site.

- ➔ *A comprehensive suite of fluid chemical data, including CO_2 concentrations, are available for our study site, as we reference in the manuscript (lines 125-135)*

5. Figure 2 “the resonance of ~200 Hz”
Again, 200 Hz could not be resonant frequency of bubbles at such depth.

- ➔ *With all due respect, we do not understand this comment. Why is it not possible? According to the Minnaert relationship, at the ambient pressure of the observations a resonance at 200 Hz is associated with a bubble having a radius of of ~2 cm, and this size is not inconsistent with those of bubbles observed to discharge from lake floor vents at the study site.*

6. Line 99, there is neither evidence nor theory to show there was “nucleation of non-condensable gas bubbles”.

- ➔ *Please see our reply to comment #2, above.*

7. Line 112 “relatively weak venting at a site that is ~100 m from the main discharge zone, which contains at least 20 active vents …”
Again, the hydrophone won’t be able to detect individual “bubble events” at 100 m. There have been a number of publications in the field research of underwater acoustic bubble to declare bubble acoustic detection threshold, either in the Mediterranean Sea or in the North Sea.

- ➔ *Please see our reply to comment #3, above.*

8. Line 138-141, Concluding paragraph: “Our data from Yellowstone Lake demonstrates the utility of acoustic methods for constraining gas fluxes in underwater environments, and provide new insight into bubble nucleation in hydrothermal fluids. “
Researchers have done a lot of theoretical and field work on gas flux quantification in underwater environments, which means that the former conclusion here is meaningless. There is neither theory nor evidence to support the nucleation of bubbles was exit in hydrothermal fluid.

- ➔ *We appreciate the fact that the reviewer was not convinced that the signals we detected represented the nucleation of CO_2 bubbles in the hydrothermal fluids. We hope that our revised text is more convincing (see response to Reviewer’s 1 comment* ***R1****). CO_2 bubbles discharge continuously from active vents in the main discharge zone, and they must come from somewhere. Our results suggest they migrate from the underlying vapor reservoir through the sediments as microbubbles that then nucleate into larger bubbles near the lake floor when they experience flow-induced pressure fluctuations. There is a large body of work regarding the nucleation of CO_2 bubbles in saturated aqueous solutions, and our interpretation is consistent with this literature.*

13th Jan 22

Dear Dr Caudron,

Please allow me to apologise for the delay in sending a decision on your manuscript titled "Turbulence-induced bubble nucleation in hydrothermal fluids beneath Yellowstone Lake". It has now been seen again by our reviewers, whose comments appear below. Reviewer #2 indicated to us that they were satisfied with the revision and provided no further comments. In light of the advice from our reviewers I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of Reviewer #3. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

EDITORIAL REQUESTS:

Please review our specific editorial comments and requests regarding your manuscript in the attached "Editorial Requests Table". Please outline your response to each request in the right hand column. Please upload the completed table with your manuscript files.

If you have any questions or concerns about any of our requests, please do not hesitate to contact me.

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We hope to hear from you within two weeks; please let us know if you need more time.

Best regards,

Joe Aslin
Senior Editor,
Communications Earth & Environment
https://www.nature.com/commsenv/
Twitter: @CommsEarth

Emma Liu
Editorial Board Member
Communications Earth & Environment

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have sufficiently addressed my previous comments. The significant re-write of sections of the paper has greatly improved how the paper reads. I look forward to seeing it published.

The only outstanding issue at this time is that the data availability is incomplete (no urls).

Reviewer #3 (Remarks to the Author):

1. The authors did not provide the ambient noise level as mentioned in the Line 47. How low is it? Please provide a spectral level of the ambient noise as that shown in Figure S2.
2. Not sure that Figure S2 shows the spectral level of “bubble sound radiation” or “ambient noise + bubble sound radiation”.
3. The authors do not know the exact location of the gas seeps relative to the location of the hydrophone. Considering the transmission loss in the acoustic channel, the distance could be no more than 5m (line 151-153). This indicates that the bubble acoustic waveforms shown in Figure 2 can only represent gas seeps in a small area with a radius of 5m, rather than those gas vents shown

in Figure 1. Is the small area representative of the whole region?

4. Bubble oscillates at the instant of emitting from the sediment. The duration of the oscillation can be up to about 10 wavelengths at its resonant frequency, typically within 15ms. Figure 2 shows the two waveforms (black and blue) start at the same time, which means that the resonance and the cavitation start at the same time (or the low frequency resonance is bit earlier). However, Figure 4 (C) shows the cavitation (kHz, bubbles in micrometre) occurs earlier than the resonance (100-200Hz, bubbles in centimetre). Moreover, bubbles associated with the decompression nucleus form in the leakage pathways of the sediment as shown in the Figure 4 (B). Is there any acoustic radiation as the bubble was forming at this stage? The waveforms in Figure 2 do not show such an acoustic radiation before the resonance waveform.

Now, the questions come: If it is correct that the decompression nucleus occurs in the leakage pathway before resonance in the water column, why the waveforms in Figure 2 show the cavitation and the resonance start at the same time? Or the resonance (100-200Hz) occurred even a bit earlier than the cavitation (kHz)? And Why there is no kHz level acoustic radiation before the 100-200Hz acoustic radiation?

5. The authors have corrected their bubble radii calculation through Minnaert's equation to 1.6-3.6cm, which is compatible with the ROV observation (line 89-90) in a grade of cm. This can only say that there are cm-size bubbles emitted from the sediment, but is unable to give information about the exist of micrometre-size bubbles resulted from "catastrophic explosion".

Reviewer #1

The authors have sufficiently addressed my previous comments. The significant re-write of sections of the paper has greatly improved how the paper reads. I look forward to seeing it published.

The only outstanding issue at this time is that the data availability is incomplete (no urls).

- ➔ *This is fixed now.* @Rob

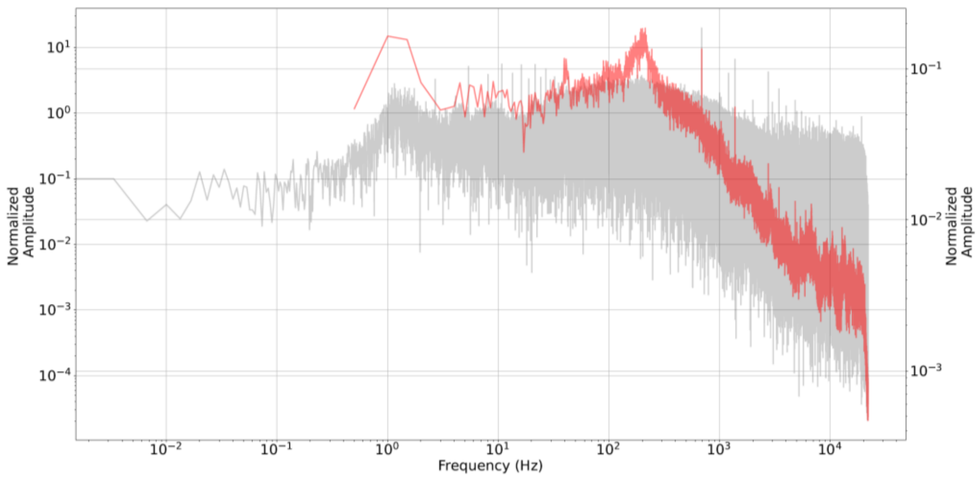
Reviewer #3

1. The authors did not provide the ambient noise level as mentioned in the Line 47. How low is it? Please provide a spectral level of the ambient noise as that shown in Figure S2.

- ➔ *We indeed did not provide the ambient noise level but did not claim so in the Line 47 "We used acoustic methods to monitor gas fluxes at the Deep Hole thermal field for 13 hours when ambient noise levels in the lake were low (overnight)."*
- ➔ *We now show the spectral level of ambient noise (see response 2 below, in grey) compared to bubbles (red)*

2. Not sure that Figure S2 shows the spectral level of "bubble sound radiation" or "ambient noise + bubble sound radiation".

- ➔ *We were just showing the bubble sound radiation (stack of all the bubbles detected). We now show both the bubbles (red) and the ambient noise level*

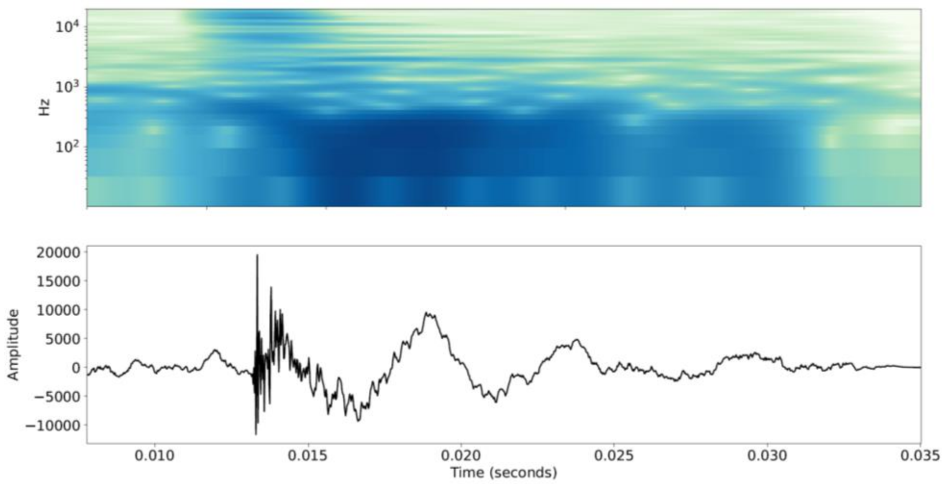


3. The authors do not know the exact location of the gas seeps relative to the location of the hydrophone. Considering the transmission loss in the acoustic channel, the distance could be no more than 5m (line 151-153). This indicates that the bubble acoustic waveforms shown in Figure 2 can only represent gas seeps in a small area with a radius of 5m, rather than those gas vents shown in Figure 1. Is the small area representative of the whole region?

➔ *The reviewer is right, we do not know the exact location. Yet, the hydrophone was located outside the main discharge but within 5-10 meters from a vent (Figure 1)*

4. Bubble oscillates at the instant of emitting from the sediment. The duration of the oscillation can be up to about 10 wavelengths at its resonant frequency, typically within 15ms. Figure 2 shows the two waveforms (black and blue) start at the same time, which means that the resonance and the cavitation start at the same time (or the low frequency resonance is bit earlier). However, Figure 4 (C) shows the cavitation (kHz, bubbles in micrometre) occurs earlier than the resonance (100-200Hz, bubbles in centimetre). Moreover, bubbles associated with the decompression nucleus form in the leakage pathways of the sediment as shown in the Figure 4 (B). Is there any acoustic radiation as the bubble was forming at this stage? The waveforms in Figure 2 do not show such an acoustic radiation before the resonance waveform.
Now, the questions come: If it is correct that the decompression nucleus occurs in the leakage pathway before resonance in the water column, why the waveforms in Figure 2 show the cavitation and the resonance start at the same time? Or the resonance (100-200Hz) occurred even a bit earlier than the cavitation (kHz)? And Why there is no kHz level acoustic radiation before the 100-200Hz acoustic radiation?

➔ *This relates to the wavelet technique. Finding the most relevant data processing tool is always challenging. We have chosen the wavelet as it better captures the resonance compared to the more conventional Fourier transform. Each event however initiates with energy in the kHz range followed by the resonance. This can be directly observed in the waveform but is more evident using spectral analyses. We have now added a figure in supplementary material (Figure S4). This does not change our interpretation and Figure 4.*



5. The authors have corrected their bubble radii calculation through Minnaert's equation to 1.6-3.6cm, which is compatible with the ROV observation (line 89-90) in a grade of cm. This can only say that there are cm-size bubbles emitted from the sediment, but is unable to give information about the exist of micrometre-size bubbles resulted from "catastrophic explosion".

➔ The reviewer is right, we have no idea regarding the size of bubbles required to trigger catastrophic explosion… and did not claim it. We have toned this down:

➔ *“Our results are ~~directly~~ relevant to the triggering of small explosions, which may be responsible for the pockmarks that cover the lake floor in hydrothermal areas, including the Deep Hole where our data were acquired.”*