

Use of cold waters geochemistry as a geothermal prospecting tool for hidden hydrothermal systems in Réunion Island



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16th Aug 23

Dear Dr Benard,

Please allow us to sincerely apologise for the long delay in sending a decision on your manuscript titled "Cold waters geochemistry as a geothermal prospection tool of hidden hydrothermal systems (Réunion Island)". It has now been seen by 2 reviewers, whose comments are appended below. You will see that they find your work of some potential interest. However, they have raised quite substantial concerns that must be addressed. In light of these comments, we cannot accept the manuscript for publication, but would be interested in considering a revised version that fully addresses these serious concerns.

We hope you will find the reviewers' comments useful as you decide how to proceed. Should additional work allow you to address these criticisms, we would be happy to look at a substantially revised manuscript. If you choose to take up this option, please either highlight all changes in the manuscript text file, or provide a list of the changes to the manuscript with your responses to the reviewers. In particular, please ensure that the revised manuscript meets the following editorial thresholds:

- * Present a robust and compelling demonstration that cold groundwater geochemistry can be used to identify potential hydrothermal resources, including placing quantitative constraints on the model.
- * Fully describe and explain your methodology and data set including any analytical issues.
- * Place your findings firmly in the context of other work on ambient water geochemistry related to hydrothermal systems.

Please bear in mind that we will be reluctant to approach the reviewers again in the absence of substantial revisions.

If the revision process takes significantly longer than three months, we will 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 are committed to providing a fair and constructive peer-review process. Please do not 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 reviewers' comments with a list of your changes to the manuscript text (which should be in a separate document to any cover letter), a tracked-changes version of the manuscript (as a PDF file) and any completed checklist:

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Please do not hesitate to contact us if you have any questions or would like to discuss the required revisions further. Thank you for the opportunity to review your work.

Best regards,

Lucia Pappalardo, PhD

Editorial Board Member
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orcid.org/0000-0002-3598-587X

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

This paper uses the chemistry of cold water samples to elucidate the location of possible blind geothermal resources on Reunion Island. I support the methodology and the results of this paper in concept. Major recommendations (in order of significance) are as follows:

1) revisit the language and organization of the paper. For example, a) Lines 21-23 describe blind resources but 24-25 describe water with surface evidence of heat (so not blind). There are similar examples throughout. More consistency is needed to be clear of the point authors are trying to make, b) I find it peculiar that the Methods are the paper's last section, as more typically Methods precede Results. Unless this organization is called for by the journal, the authors may want to consider restructuring.

2) in two locations (line 145, 233) 'analytical issues' are mentioned, without any further explanation. More explanation of the "analytical issues" is necessary in order for readers to trust the validity of the rest of the data collected and presented.

3) Hawaii is the only other location mentioned where cold water samples have been analyzed for geothermal indicators, which, according to the authors, helps to constitute the novelty of this approach. Did the authors do a comprehensive literature search to ensure Hawaii is truly the only other location that has looked at the chemistry of ambient (not precisely cold) water?

4) A nice addition to the manuscript would be an explanation of the relevancy of this work to communities on Reunion and/or the planet, for example does Reunion have a renewable electricity or geothermal electricity generation goal? How much electricity is needed/used? what are current plans? Also, it would also be good to point out that the end goal of this work, if true, is in fact geothermal electricity generation, over other forms of geothermal, e.g. heat pump, direct use, etc.

Not a recommendation but to mention: I do not have the offhand expertise to evaluate the methodology presented from line 152, section "Highlighting the presence of a geothermal system"

I appreciate the effort put into this work and look forward to its ultimate publication.

Reviewer #2 (Remarks to the Author):

The manuscript "Cold water geochemistry as a geothermal prospection tool of hidden hydrothermal system (Reunion Island) by Bernard and coauthors discusses a very large dataset of the chemistry (major, minor components and isotopes) of cold groundwater for exploring the occurring of hydrothermal systems at depth. The aim of this study is timely, new tools for the exploration of geothermal resources are a worldwide common point.

Here, the Principal Component Analysis (PCA) is combined with some classic uses of the fluid geochemistry and it is presented a model about the presence of hydrothermal components in the geological and structural framework of the study area.

This study is well structured, the manuscript is also well organized and the figures are clear. However, it needs a deeper data elaboration to support a more robust discussion of the results and better constrain the model. Often the discussion is too qualitative or based on assumptions and connections with previous studies. For instance, 1) the use of the classical geothermometers and 2) a robust discussion of the relationship between the amount of carbon dissolved into the groundwater and the relative carbon isotopes (TDIC vs $\delta^{13}\text{C}_{\text{TDIC}}$) can contribute to well constrain the results. I understand that it could create problem with the manuscript length (number of words), so considering this and the rules of the journal I suggest to re-evaluate the choice of the journal.

The manuscript needs major revisions to be considered for a publication in a scientific journal.

Comments

Line 50, "...high temperature....". I understand that the evidence of high temperature water-rock interaction is due to the presence of traces of magmatic fluids in an active volcanic system. This "evidence" should be better discussed, furthermore considering the big database the authors produced they have data for applying at least the classical geothermometers to assess the temperature of the geothermal system at depth. This could help understand more about a high temperature system and a range of temperature.

Lines 74-75, too general this sentence, furthermore it is quite normal that the thermal water are hot.

Lines 75-80. It is not clear the discussion of these data in contrast with the previous sentence.

In fig 1B it is difficult to observe a clear sea water contamination trend, because of the low number of samples along this supposed trend. I suggest to review this explanation.

Line 96, I suggest to clarify what the authors assume as "hydrothermal activity". This will help the readers and avoid misunderstanding with the presented results.

Line 95-96, this sentence presents an apparent contradiction. I suggest to rewrite it or better explain it. Furthermore, what is the rule of CO_2 in the water-rock interaction?

Line 107, The authors did not use the HCO_3 contents in the PCA analysis, however successively they used it together with the carbon CO_2 isotopes investigate the presence of possible hidden hydrothermal system at Reunion island. Do the results of the PCA analysis change by adding the HCO_3 data?

Line 130-131, What does it mean “differentiated rock”? Does it fit with the geology at local scale? Or it is a simple hypothesis?

Line 142, Basaltic rock..... so it means that there is an fluid-rock interaction with differentiated rock and with basaltic rock too? What is a model constrained by geological data that support these results?

Line 163, so the authors do not consider Li as possible indicator of a hydrothermal input because of the low amounts and a possible similitude with a previous study. Hence nothing is new!

Lines 166-169, The authors should add more information to explain how the B isotopes are useful tools to explore the hydrothermal contributions. Furthermore, the discussion of the B isotopes is too generic and it results that the conclusions are not robust. I suggest to rewrite this discussion.

Lines 197-214, Here again the discussion of the C amount and its isotopic composition is too generic. Table 1 contains the value of HCO_3 and pH, and the authors computed the total dissolved inorganic carbon (TDIC). In section Methods it is discussed. A deeper elaboration of this dataset strongly contributes to figure out the origin of fluids and the processes at depth, so helping to decipherer the hydrothermal system at depth.

There is a large literature produced in the last ten years about the use of the C amounts (TDIC) coupled to the relative isotopes in groundwater (main authors, e.g., Chiodini, Capasso, Buttitta). It is not clear in Table and Methods if the isotopic signature in Table 1 is referred to the TDIC. I suppose it is the case, but I suggest to clarify it in the manuscript in order to avoid misunderstanding.

Line 220, “high temperature”, it is too generic. A range should be indicated. I suggest again to try with the geothermometers to understand if a range of temperature can be assessed.

Line 227, sea water contamination or sea water aerosol?

Lines 232-235, It is a generic discussion

We address all of the reviewers' comments point by point in this document.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

This paper uses the chemistry of cold water samples to elucidate the location of possible blind geothermal resources on Reunion Island. I support the methodology and the results of this paper in concept. Major recommendations (in order of significance) are as follows:

1) revisit the language and organization of the paper. For example, a) Lines 21-23 describe blind resources but 24-25 describe water with surface evidence of heat (so not blind). There are similar examples throughout. More consistency is needed to be clear of the point authors are trying to make, b) I find it peculiar that the Methods are the paper's last section, as more typically Methods precede Results. Unless this organization is called for by the journal, the authors may want to consider restructuring.

We made a significant effort to improve the reading experience and rephrased a large portion of the manuscript.

The methods section is the last section as required by the journal submission guidelines.

2) in two locations (line 145, 233) 'analytical issues' are mentioned, without any further explanation. More explanation of the "analytical issues" is necessary in order for readers to trust the validity of the rest of the data collected and presented.

We have addressed this second comment in our revised version (lines 329-338) and specified the analytical issues encountered. The issue was about only on one the analyzed parameters (HCO_3) and the samples on which we encountered the issue were removed from the dataset.

3) Hawaii is the only other location mentioned where cold water samples have been analyzed for geothermal indicators, which, according to the authors, helps to constitute the novelty of this approach. Did the authors do a comprehensive literature search to ensure Hawaii is truly the only other location that has looked at the chemistry of ambient (not precisely cold) water?

Other studies have explored the input of hydrothermal fluids to rivers and springs, in particular in volcanic contexts, and notably to assess the share of chemical weathering that was imputable to high temperature weathering compared to low-temperature weathering, but the Hawai'i study is the only one that used geochemical tracers in "cold" waters with the aim to reveal hidden or blind hydrothermal systems. We added this information and the references for these studies in the text (lines 33-38).

4) A nice addition to the manuscript would be an explanation of the relevancy of this work to communities on Reunion and/or the planet, for example does Reunion have a renewable electricity or geothermal electricity generation goal? How much electricity is needed/used? what are current plans?

Also, it would also be good to point out that the end goal of this work, if true, is in fact geothermal electricity generation, over other forms of geothermal, e.g. heat pump, direct use, etc.

Discussing consensual goals at the scale of the island is difficult. There is no geothermal use so far, although different actors are trying to develop it. Our study was funded by the Communauté d'Agglomération du Sud de La Réunion (CASUD), which is a group of neighboring cities. They wish to develop geothermal use in all possible forms (heat, electricity) on their territory as long as it aligns with the needs of the inhabitants. However, they lost the right to pursue their research on part of the study area, as an exclusive research permit was given to a private company working with other institutional partners. Given this complex political situation, we chose not to elaborate on the context and focus on the scientific meaning of these results.

Not a recommendation but to mention: I do not have the offhand expertise to evaluate the methodology presented from line 152, section "Highlighting the presence of a geothermal system"

I appreciate the effort put into this work and look forward to its ultimate publication.

We thank the reviewer for the insightful remarks.

Reviewer #2 (Remarks to the Author):

The manuscript "Cold water geochemistry as a geothermal prospection tool of hidden hydrothermal system (Reunion Island) by Bernard and coauthors discusses a very large dataset of the chemistry (major, minor components and isotopes) of cold groundwater for exploring the occurring of hydrothermal systems at depth. The aim of this study is timely, new tools for the exploration of geothermal resources are a worldwide common point.

Here, the Principal Component Analysis (PCA) is combined with some classic uses of the fluid geochemistry and it is presented a model about the presence of hydrothermal components in the geological and structural framework of the study area.

This study is well structured, the manuscript is also well organized and the figures are clear. However, it needs a deeper data elaboration to support a more robust discussion of the results and better constrain the model. Often the discussion is too qualitative or based on assumptions and connections with previous studies. For instance, 1) the use of the classical geothermometers and 2) a robust discussion of the relationship between the amount of carbon dissolved into the groundwater and the relative carbon isotopes (TDIC vs $\delta^{13}\text{C}_{\text{TDIC}}$) can contribute to well constrain the results. I understand that it could create problem with the manuscript length (number of words), so considering this and the rules of the journal I suggest to re-evaluate the choice of the journal.

The second reviewer recommended the use of quantitative approaches and of geothermometers. These classic approaches in geothermal studies are based on the study of the chemical equilibriums of the waters of geothermal systems. Therefore, to be valid, they can only be performed on thermal waters whose chemical composition are representative of the geothermal system at depth. These thermal waters sometimes reach the surface as hot springs, which by definition is not the case for blind systems, which are the ones targeted by our study. These thermal waters can also be accessed via drilling, but for that, the existence of the geothermal system and its location have to be already known,

which defeats the purpose of this study. Geothermometers and modeling can also be attempted with great cautions on thermal waters mixed with a small amount of cold water (Bénard et al., 2023) but cannot reasonably be applied to cold waters. Here, we attempt to find the existence of a small hydrothermal component in cold waters, merely to testify of the existence of hydrothermal activity, as part of a preliminary exploration phase. We do not aim at providing quantitative information on the potential for electricity generation.

Therefore, it is neither possible nor relevant to follow this recommendation in the frame of our work. We understand that in our first submission, we did not give sufficient explanation about the context for the readers to fully understand the purpose of our study and we better explain the approach in the revised version.

The manuscript needs major revisions to be considered for a publication in a scientific journal.

We thank the reviewer for his comments which helped us understand how to improve the manuscript.

Comments

Line 50, "...high temperature.... ". I understand that the evidence of high temperature water-rock interaction is due to the presence of traces of magmatic fluids in an active volcanic system. This "evidence" should be better discussed, furthermore considering the big database the authors produced they have data for applying at least the classical geothermometers to assess the temperature of the geothermal system at depth. This could help understand more about a high temperature system and a range of temperature.

As explained in the answer to the reviewers #2' general comments on the manuscript, geothermometers cannot be used in our study as the requirements for their application are not met. For instance, the chemical equilibriums of the sampled waters of Piton de la Fournaise are not representative of the chemical equilibriums in the geothermal system at depth.

Lines 74-75, too general this sentence, furthermore it is quite normal that the thermal water are hot. Lines 75-80. It is not clear the discussion of these data in contrast with the previous sentence.

Following the reviewer's recommendations, we rephrased the paragraph (lines 92-98) to better explain our point, which is that the waters of Piton de la Fournaise are not thermal waters. However, we kept the sentence considered as generic by the reviewer as it defines what we consider to be hydrothermal activity: "From their interaction with hot rocks, hot fluids, and magmatic gases, thermal waters are hot and/or acidic and/or with a high total dissolved solid content."

In fig 1B it is difficult to observe a clear sea water contamination trend, because of the low number of samples along this supposed trend. I suggest to review this explanation.

Seawater contamination resulting in an enrichment in Cl compared to Na is already described in Piton de la Fournaise groundwaters by Join et al, 1997, which we cite in this paragraph. Our results are consistent with former observations. To strengthen our statement, we rephrased the paragraph and we added the name of the datapoints following this trend on the figure. These datapoints correspond

to boreholes close to the coast and thus vulnerable to seawater contamination, as explained in the text (line 106-108).

Line 96, I suggest to clarify what the authors assume as “hydrothermal activity”. This will help the readers and avoid misunderstanding with the presented results.

As explained in a previous comment, we defined thermal waters as such: “From their interaction with hot rocks, hot fluids, and magmatic gases, thermal waters are hot and/or acid and/or with a high total dissolved solid content” in the text line 93. This definition extends to hydrothermal activity, as thermal waters are the result of hydrothermal activity. In the revised version, we believe the link between paragraphs will make it easy for the reader to relate to this definition.

Line 95-96, this sentence presents an apparent contradiction. I suggest to rewrite it or better explain it. Furthermore, what is the rule of CO₂ in the water-rock interaction?

This paragraph serves as a global description of the geochemical profile of Piton de la Fournaise waters. The specific role of CO₂ in water interaction is discussed further in the text (lines 228-248), where we address in detail the possibility of the presence of a hydrothermal component. Following the reviewer’s recommendations, we rephrased the paragraph to better convey its message.

Line 107, The authors did not use the HCO₃ contents in the PCA analysis, however successively they used it together with the carbon CO₂ isotopes investigate the presence of possible hidden hydrothermal system at Reunion island. Do the results of the PCA analysis change by adding the HCO₃ data?

The mathematical execution of Principle Component Analysis (PCA) requires a homogeneous dataset, meaning that the set of parameters has to be the same for each sample considered in the analysis. As some HCO₃ analyses could not be performed on some samples due to the aforementioned analytical issues (response to reviewer #1 comment), this parameter could not be used for the PCA. We understand that it raises questions for the reader, and therefore added this justification in the text line 136.

Line 130-131, What does it mean “differentiated rock”? Does it fit with the geology at local scale? Or it is a simple hypothesis?

It fits with the geology at a local scale. We understand that our manuscript was missing geological context and we added a sentence line 83 in the paragraph presenting the study area: Lavas are transitional between the tholeiitic and alkaline fields and have a range of compositions from picrite to trachyandesite, with a large majority of aphyric basalt.

Line 142, Basaltic rock..... so it means that there is an fluid-rock interaction with differentiated rock and with basaltic rock too? What is a model constrained by geological data that support these results?

As both types of rocks exist in the edifice, groundwaters are known to interact with one, two or both, depending on their pathways. This has been described in Join et al., 2016, which is cited in the text line 168.

Line 163, so the authors do not consider Li as possible indicator of a hydrothermal input because of the low amounts and a possible similitude with a previous study. Hence nothing is new!

This is not what the paragraph says. We say we do find a possible hydrothermal contribution, because some samples display significantly higher amounts of Li compared to the others. We add, as not to conceal data, that isotopic analyses were performed in another study but not on the samples which we consider to have the hydrothermal contribution, therefore, it didn't show a hydrothermal isotopic signature. With that, regarding the "nothing is new" comment, this paragraph does in fact serve a purpose because it shows Li is an indicator of a possible hydrothermal activity. Since the reviewer did not understand the paragraph, we rephrased it for clarity (lines 174-191).

Lines 166-169, The authors should add more information to explain how the B isotopes are useful tools to explore the hydrothermal contributions. Furthermore, the discussion of the B isotopes is too generic and it results that the conclusions are not robust. I suggest to rewrite this discussion.

We disagree with this comment. We do not think the discussion is generic. This paragraph serves its one and only purpose which is to demonstrate that some samples show a hydrothermal contribution according to their isotopic signature, and it gives the reader the right amount of information that they need to understand the reasoning.

At the beginning of the paragraph, we list the hydrothermal processes associated with Boron enrichment in natural waters and the isotopic signature associated. Then we detail the other non-hydrothermal processes that can be associated with an enrichment in Boron and the isotopic signature associated. From that, we explain that hydrothermal processes and non-hydrothermal processes give distinct isotopic signatures, hereby showing that Boron is a useful tool to identify a hydrothermal component.

Boron enrichment in hydrothermal fluids and its isotopic signature close to that of the bedrock during high temperature water-rock interaction has been demonstrated and documented in many studies and is not new to the scientific community. Fifteen references about Boron geochemistry support all statements made in this paragraph. Every conclusion we draw is based on facts enunciated in the text, and therefore robust.

The boron isotope paragraph has nonetheless been reformulated in parts and we hope that the new version is much clearer to the reader, as it details more the B sources and the water-rock processes at low and high temperature, the associated B isotopic fractionations and resulting $\delta^{11}\text{B}$ signatures.

Lines 197-214, Here again the discussion of the C amount and its isotopic composition is too generic.

Again, and for the same reasons, we disagree with this comment.

Table 1 contains the value of HCO₃ and pH, and the authors computed the total dissolved inorganic carbon (TDIC). In section Methods it is discussed. A deeper elaboration of this dataset strongly contributes to figure out the origin of fluids and the processes at depth, so helping to decipher the hydrothermal system at depth.

There is a large literature produced in the last ten years about the use of the C amounts (TDIC) coupled to the relative isotopes in groundwater (main authors, e.g., Chiodini, Capasso, Buttitta).

The literature from these authors, as well as the study of deep processes using TDIC and $\delta^{13}\text{C}$ in general, is applicable only on waters whose geochemical composition are mainly driven by hydrothermal processes. Here, there is a biogenic component in most of the analyzed samples and the hydrothermal component is heavily diluted. Thus, the cold waters of Piton de la Fournaise waters cannot be considered representative of the hydrothermal system and therefore should not be used to attempt to characterize it. For these waters, only the possible hydrothermal/magmatic origin of the elements can be discussed, which is why we restrict our analysis to this discussion. This analysis is in agreement with the goal of the study, which is to discuss the existence of a hydrothermal system and not to characterize it. As explained in the introduction of this document, the characterization of the hydrothermal system in terms of potential and processes must come at later stages of the geothermal exploration of blind systems. In the case of the Piton de la Fournaise volcanic edifice, it will require drilling to gain access to the hydrothermal fluids, which can then be characterized further.

It is not clear in Table and Methods if the isotopic signature in Table 1 is referred to the TDIC. I suppose it is the case, but I suggest to clarify it in the manuscript in order to avoid misunderstanding.

Following the reviewer's recommendation, we clarified this point (lines 234 and 331).

Line 220, "high temperature", it is too generic. A range should be indicated. I suggest again to try with the geothermometers to understand if a range of temperature can be assessed.

This query has already been answered several times in previous comment in this document. It is not possible to apply geothermometers on cold waters.

Line 227, sea water contamination or sea water aerosol?

Seawater contamination. We understand the confusion of the reviewer regarding this point, and this should be understood better in the revised version as we modified the section discussing sea spray and seawater contamination (lines 99-108).

Lines 232-235, It is a generic discussion

Following the reviewer's recommendation, we rephrased this paragraph (lines 266-268) to make our point stand out better. However, we kept the paragraph, as we think the possible influence of dilution and the impossibility to correct it should be discussed.

24th Oct 23

Dear Dr Bénard,

Your manuscript titled "Cold waters geochemistry as a geothermal prospection tool of hidden hydrothermal systems (Réunion Island)" has now been seen by our reviewers. Reviewer #2 indicated that they recommend publication, Reviewer #1 has some further comments which appear below. In light of their advice we are 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 our reviewer. 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.

Please note that it may still be possible for your paper to be published before the end of 2023, but in order to do this we will need you to address these points as quickly as possible so that we can move forward with your paper.

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Editorial Board Member
Communications Earth & Environment
orcid.org/0000-0002-3598-587X

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

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

In my opinion the authors response to reviewer comments was sound. I still have some issues, in particular with the abstract and introduction.

In what follows, line numbers reference the pdf showing track changes.

- 1) For the title how about: "Use of cold water geochemistry as a prospecting tool for hidden hydrothermal systems on Réunion Island"
- 2) Regarding the abstract and intro: consider using "develop" or "development of" rather than "exploit" or "exploitation of" given the latter has a negative connotation for some.
- 3) Consider rephrasing 1st and 2nd sentences of abstract: "Most untapped high-enthalpy geothermal resources are blind, meaning lacking surface evidence of their existence. The first step in their discovery is to find evidence of hydrothermal activity. Here we apply a new approach based on the geochemistry of cold waters, which allowed us to identify we evidence of the existence and location of a hydrothermal system at Piton de la Fournaise volcano (Réunion Island).
- 4) Line 24: is the water dilute or the concentration of the ions dilute?

5) Revise first few sentences of Introduction, especially lines 33-34. Is it certain most non-blind systems been found? I would suggest wording this more carefully.

6) Suggest a careful read through and check of grammar on pg 12 to make sure scientific arguments are clear. e.g. 253-53 sea sprays or sea spray?

Nicole Lautze