Peer Review File

Manuscript Title: Primary N2-He gas field formation in intracratonic sedimentary basins

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referee #1:

General assessment
This manuscript presents a new view on gas transport in the crust, by taking the possible formation of a gas phase driven by nitrogen (and thus unrelated to CO2 or hydrocarbons) into account. To my knowledge, this is a novel idea, which appears plausible and quite important as it may explain the presence of nitrogen-dominated gaes with a high helium content. Such gas reservoirs are interesting resources in times where helium is in short supply. There may also be implications for crustal hydrogen resources, although the manuscript does not further expore this possibility. Given the significance of the proposed scenario for our fundamental understanding of gases in the crust and the importance regarding potential helium resource development, the paper should be of broad interest and suitable for publication in Nature.
The manuscript is generally well-written and adequately referenced. Abstract, introduction and conclusions are appropriate in my opinion. The study makes use of a high-quality data set, which was previously published by a similar group of authors (ref. 7 in the manuscript). That earlier paper focussed on the diffusive helium flux and its modeling. The current manuscript adds the modeling of nitrogen and the related formation of a gas-phase to the previous work. This is novel, innovative and relevant, as outlined above.
The modeling and interpretation undertaken in this study appears to be valid and of high quality as far as I can judge, leading to robust results. The manuscript does not contain a lot of statistics and treatment of uncertainties, but I think that this is adequate for the type of results presented. The illustrative movie that is provided to show the modeled system evolution is a nice feature that helps grasp the idea of this study.
Despite the overall clear presentation, I think that some aspects would benefit from more detailed explanations, as outlined below. However, my remarks and suggestions for improvement are of a rather minor and technical nature. I can therefore recommend the paper for publication after moderate revision.

Remarks
I tried to follow and understand the calculations and modeling done by the authors in some detail. In general, this was well possible and I feel that everything is correct. In some cases, however, I think that a bit of additional information would be helpful for readers who are interested in the details or even in reproducing the modeling.

1) I found it difficult to fully understand the profiles for the nitrogen solubility limit in Figs. 2b and 3b. I was naively expecting a rather linear increase with depth and corresponding pressure, maybe modulated by changes in salinity. However, the increase of the solubility limit flattens with increasing depth. I can understand this as an effect of increasing temperature and thus decreasing

solubility, but the effect visible in the plots seems rather stronger than what I would have expected. Only after looking up the cited ref. 38 (Mao and Duan), I realised that the solubility limit also decreases with higher pressures. It might be a good idea to provide some information about pressure, temperature and salinity profiles as well as solubility dependence on these parameters for the interested reader in the methods section (in addition to the statement on lines 334-336).

2) This may only be a minor formulation problem, but as it stands I find it hard to understand the sentence on lines 181 – 183 about the effect of "reset scenarios" on model results. What results in the gas phase formation being delayed? The weak gas accumulation memory (which just seems to be explained here) or the resetting of gas concentrations? I think what is meant is that the reset scenarios only show a slight delay of gas phase formation, which may be called a weak gas accumulation memory.

3) Figure 2a and Appendix Figure 1: What exactly are the flushing scenarios indicated in the legend? I looked up the previous paper by Cheng et al., 2021 (ref. 7), where there are very similar figures included. However, I also did not find a detailed answer there, but at least a better idea that this refers to flushing scenarios for the shallower aquifers. This is potentially confusing as this flushing is different from the reset scenarios discussed above in remark 2, but in both cases the words flushing and scenario do occur. I think that an explanation should be given on the time ranges listed in these figures for the different aquifer flushing scenarios. What exactly happens in these time ranges (e.g., concentrations set to atmospheric solubility equilibrium)?

4) I also did not find it possible to verify the calculation behind the 20Ne solubility mentioned on line 289 (in my understanding, the value of 1.33E-7 cc/cc given here refers to an ASW concentration). There, Appendix Table 2 is referred to. However, there only the measured values of gas samples are listed, as far as I can see. The value given is plausible for some reasonable temperature and salinity values, but I don't see the corresponding information anywhere in the manuscript.

Minor corrections

Line 131: the addition *of* a crystalline *basement nitrogen* flux

Line 134: observatio*n

Line 144: For example, *in* the model … concentrations … are reduced

Line 164: delete comma

Lines 171 – 173: incomplete sentence, who reports?

Line 193: delete first comma

Line 212: timing and location *of* helium-rich gas reservoir formation

Line 296: in the water-filled pore spaces *as* a function of time

Line 351: dissolve*d*

Lines 377, 387, appendix Table 1 and caption Figure 2: subscripts in Vg/Vl

Line 441: simulated from *the* diffusion-exsolution model

Reviewer signature
Review by Werner Aeschbach

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Referee #2:
Comments for Author

A. Summary of the key results

The Authors present a quantitative model for the distribution of N_2 and He in a range of sedimentary depositional systems leading the free gas occurrences.

Originality and significance: if not novel, please include reference

The approach is novel in that it gives a theoretical explanation (supported by observational data) leading to controls on conditions for free gas occurrences from crystalline basement fluxes of N_2 and He and is readily applicable to other gas components. Given the rather high N_2/He ratios from crystalline basement needed to form a free gas phase, I'm still unsure if these high ratios are realistic given the uncertainty in the source of N2

B. Data & methodology: validity of approach, quality of data, quality of presentation
Sound

C. Appropriate use of statistics and treatment of uncertainties

yes

D. Conclusions: robustness, validity, reliability

yes

E. Suggested improvements: experiments, data for possible revision

See accompanying edits/comments below

F. References: appropriate credit to previous work?

yes

G. Clarity and context: lucidity of abstract/summary, appropriateness of abstract, introduction and conclusions

Yes

Note: I leave it to others to decide if the paper meets the criteria for a Nature publication or better suited in Nature Geoscience.

Nitrogen is always a significant component in helium-rich natural gases [11,12]

Also other reference [13, 31-34]

Also what do the Authors consider as 'helium-rich natural gas'?

'to nitrogen which is radio/physio-chemically derived from minerals, has not been unambiguously resolved [12-15]'. What about N_2 from thermal decomposition of organic matter e.g., Krooss and others?

(typically >0.3% helium [add reference])

Multiple sources of nitrogen gas exist in sedimentary basins [12,13]

$N_2/^4He$ ratios in stratigraphic units distal from the crystalline basement ranging from hundreds to thousands [12,13,31-34]

Change from 20-50 to 20–50 [12]

Change from 6-59 to 6–59 (occurs twice in text)

The upper concentration limit of nitrogen in porewater in these calculations is controlled by its saturation solubility, which is a function of temperature, pressure and salinity [38] – Authors, do other gas major gases influence N_2 solubility e.g., common occurrence of N_2-CO_2 rich natural gases

As such, the calculated results are conservative, as gas phase formation would occur earlier if additional sedimentary-sourced nitrogen or other gas sources such as H_2, CH_4 or CO_2 are significant. Authors, can you give a qualitative estimate of the order of formation of free gas for the 4 gases

In our calculations, the original nitrogen concentrations in water-filled pore spaces are set to air saturated water values and are calculated iteratively as a function of depth and sediment formation with time. Authors need to explain if this is set for all formation units on deposition and give the proportion that the dissolved atmospheric N_2 contributes to the total N_2 considering the range in N_2/He ratios from crystalline basement input.

pore fluids proximal to the crystalline basement if the crystalline basement helium flux has a ^{4}~~He/N$_2$~~ $N_2/^{4}$He between 25-50 (Figure 2b).

observatio~~0~~n

0.05–1239

19% – 32% and N_2 concentrations are reduced by 2% – 50%

7.03–7.18% ^{4}He

basement helium flux of 0.8–1.6×10^{-6} mol ^{4}He/m^{2} yr and $N_2/^{4}$He of 25–50

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Deadwood Formation

partition~~e~~s

Basement ^{4}He flux fitting the observed ^{4}He distribution is between 0.8 - 1.6×10^{-6} mol ^{4}He/m^{2} yr (available in [7])

Vg/V~~1~~l of 0.0478, 0.0474, 0.0422 and 0.0251

Appendix Table 3. *$N_2/^{4}$He or N_2/He ratios of sedimentary basin fluids sampled proximal to the basement and samples from the crystalline basement.*

Suggest Authors have another column for ‘inferred N_2 source’ where available

Li, L. *et al.* N_2

Karolytė, R. *et al.* The role of porosity in H_2/He

Hiyagon, H. & Kennedy, B. Noble gases in CH_4-rich

Mao, S. & Duan, Z... N_2–H_2O–NaCl

Thompson, J. (Williston Basin Symposium). Needs full reference

Author Rebuttals to Initial Comments:

Reply to reviewers comments

Primary N_2-He gas field formation in intracratonic sedimentary basins

Anran Cheng, Barbara Sherwood Lollar, Jon Gluyas and Chris J. Ballentine

Our point by point response is in red

We thanks both reviewers for their positive assessment, and the detail of their reviews which has enabled us to improved the clarity of the original manuscript.

Referee #1:

General assessment
This manuscript presents a new view on gas transport in the crust, by taking the possible formation of a gas phase driven by nitrogen (and thus unrelated to CO2 or hydrocarbons) into account. To my knowledge, this is a novel idea, which appears plausible and quite important as it may explain the presence of nitrogen-dominated gaes with a high helium content. Such gas reservoirs are interesting resources in times where helium is in short supply. There may also be implications for crustal hydrogen resources, although the manuscript does not further expore this possibility. Given the significance of the proposed scenario for our fundamental understanding of gases in the crust and the importance regarding potential helium resource development, the paper should be of broad interest and suitable for publication in Nature.
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Despite the overall clear presentation, I think that some aspects would benefit from more detailed explanations, as outlined below. However, my remarks and suggestions for improvement are of a rather minor and technical nature. I can therefore recommend the paper for publication after moderate revision.

We thank reviewer#1 (Werner Aeschbach) for the positive and insightful comments on our manuscript that highlight the novelty and importance of our work. We have addressed below, point by point, all the items that the reviewer raises.

Remarks
I tried to follow and understand the calculations and modeling done by the authors in some detail. In general, this was well possible and I feel that everything is correct. In some cases, however, I think that a bit of additional information would be helpful for readers who are interested in the details or even in reproducing the modeling.

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With thanks. We agree that the non-linearity of solubility response to temperature, pressure and salinity may not be intuitive. We have added a paragraph in the Method section, including the equations for calculating nitrogen solubility, to help make this more accessible (line 443 in the returned manuscript).

2) This may only be a minor formulation problem, but as it stands I find it hard to understand the sentence on lines 181 – 183 about the effect of "reset scenarios" on model results. What results in the gas phase formation being delayed? The weak gas accumulation memory (which just seems to be explained here) or the resetting of gas concentrations? I think what is meant is that the reset scenarios only show a slight delay of gas phase formation, which may be called a weak gas accumulation memory.

We appreciate the reviewer pointing this out. The reviewer is correct in the message we were trying to convey. We have edited this paragraph lightly (from line 184) to better communicate the concept of weak gas accumulation memory.

3) Figure 2a and Appendix Figure 1: What exactly are the flushing scenarios indicated in the legend? I looked up the previous paper by Cheng et al., 2021 (ref. 7), where there are very similar figures included. However, I also did not find a detailed answer there, but at least a better idea that this refers to flushing scenarios for the shallower aquifers. This is potentially confusing as this flushing is different from the reset scenarios discussed above in remark 2,

but in both cases the words flushing and scenario do occur. I think that an explanation should be given on the time ranges listed in these figures for the different aquifer flushing scenarios. What exactly happens in these time ranges (e.g., concentrations set to atmospheric solubility equilibrium)?

We thank the reviewer for highlighting the importance of clearly communicating the difference between the previously identified recent and shallow aquifer flushing events described in our previous paper (ref. 7) and the theoretical full system 'reset' we use in this new paper for an entirely different purpose, that is, as a model sensitivity test. The different possible mechanisms for the shallow acquifer are explained in section 5.3.3 of the Cheng et al paper (reference 7). We refer the reader now to this with some additional text (from line 184) to help clearly solidify the distinction. We have further added an additional sentence to the Figure 2 caption. We think the additional text and edits carefully explains our model sensitivity test involving the theoretical reseting of background water concentrations to 'air-saturated water levels' and addresses the useful clarification requested by the reviewer. We hope this careful distinction addresses the important issue the reviewer raised.

4) I also did not find it possible to verify the calculation behind the 20Ne solubility mentioned on line 289 (in my understanding, the value of 1.33E-7 cc/cc given here refers to an ASW concentration). There, Appendix Table 2 is referred to. However, there only the measured values of gas samples are listed, as far as I can see. The value given is plausible for some reasonable temperature and salinity values, but I don't see the corresponding information anywhere in the manuscript.

We thank the reviewer for this point of clarity. ^{20}Ne concentration is calculated assuming the water is seawater (salinity 35‰) at 298K and in equilibration with the atmosphere at one-atmosphere pressure. We have added this information (line 393 in the returned maunscript).

Minor corrections

Line 131: the addition *of* a crystalline *basement nitrogen* flux

Corrected in the manuscript

Line 134: observatio*n

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Reviewer signature
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Referee #2:
The Authors present a quantitative model for the distribution of N2 and He in a range of sedimentary depositional systems leading the free gas occurrences.

The approach is novel in that it gives a theoretical explanation (supported by observational data) leading to controls on conditions for free gas occurrences from crystalline basement fluxes of N2 and He and is readily applicable to other gas components. Given the rather high N2/He ratios from crystalline basement needed to form a free gas phase, I'm still unsure if these high ratios are realistic given the uncertainty in the source of N2.

We appreciate the reviewer comments related to N_2. Our model provides an independent approach that explores basement $N_2/^4He$ flux ratios comparable to ratios summarised by other authors studying He-related N_2 from the crystalline basement. The convergence of the range of literature N_2/He values, observed in-place ratios, and our model values provides

confidence in the new model. It is particularly important to note, as does the reviewer later, that our new model presents a conservative approach (by neglecting the role of other gases) and a new understanding of the role of slow deep gas fluxes and their role in deep gas-phase/field formation.

Nitrogen is always a significant component in helium-rich natural gases [11,12]
Also other reference [13, 31-34]

Existing references were added as recommended by the reviewer.

Also what do the Authors consider as 'helium-rich natural gas'?

We thank the reviewer for pointing this out. Here we us ‘Helium-rich natural gas’ to refer to concentrations that may be economically viable, typically with helium concentrations greater than 0.1% (Danabalan 2022). We have updated the text in the manuscript and reference this literature example (revised manunscript, line 64).

'to nitrogen which is radio/physio-chemically derived from minerals, has not been unambiguously resolved 12-15'. What about N2 from thermal decomposition of organic matter e.g., Krooss and others?

The references here are for background information and include discussion of thermal nitrogen release from clays, metamorphic-sourced nitrogen and radioltytic release of nitrogen from crystalline rocks. In general though we are discussing nitrogen sources from the crystalline basement which will not typically contain a high organic matter content. For this reason we have chosen not to add any further references but have edited lightly to emphasise the crystalline crust source.

(typically >0.3% helium [add reference])

We have changed this to >0.1% helium and added the corresponding reference as mentioned above.

Multiple sources of nitrogen gas exist in sedimentary basins [12,13]

Existing references were added as recommended by the reviewer.

N2/4 He ratios in stratigraphic units distal from the crystalline basement ranging from hundreds to thousands [12,13,31-34]

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The upper concentration limit of nitrogen in porewater in these calculations is controlled by its saturation solubility, which is a function of temperature, pressure and salinity [38] – Authors, do other gas major gases influence N2 solubility e.g., common occurrence of N2-CO2 rich natural gases

See below for more on this important point.

As such, the calculated results are conservative, as gas phase formation would occur earlier if additional sedimentary-sourced nitrogen or other gas sources such as H2, CH4 or CO2 are significant. Authors, can you give a qualitative estimate of the order of formation of free gas for the 4 gases

We thank the reviewer for these two insightful comments. The interaction of gases and their effect on N_2 solubility is complicated by gas polarity, molecular size and also their relative abundance. Overall, the addition of gases other than N_2, including helium and hydrogen, will reduce the solubility of N_2 in water. We refer to this in lines 123-127 of the revised manunscript. In our paper we are presenting, for clarity, a simple model that considers only N_2 as the major gas phase to provide a conservative estimation (as the reviewer notes) of gas phase formation to clearly establish this new concept. In light of this new insight, presentation and discussion of many different scenarios and sources of gases H2, CH4 and CO2 (as well as sedimentary N2) will be possible in future work but is beyond the scope of this paper. This will certainly be the focus of future work, building on the new insights and concepts established here.

In our calculations, the original nitrogen concentrations in water-filled pore spaces are set to air saturated water values and are calculated iteratively as a function of depth and sediment formation with time. Authors need to explain if this is set for all formation units on deposition and give the proportion that the dissolved atmospheric N2 contributes to the total N2 considering the range in N2/He ratios from crystalline basement input.

We thank the reviewer for this point of clarity. The N_2 concentration of all units is set as air-saturated seawater value on deposition. The contribution of crustal N_2 increases with depth, input at the basal layer. The dissolved atmospheric contribution is $0.012 cm^3/cm^3/H_2O$ and insignificant compared with the crustal flux, which is evident from the surface value of Figure 2b. This has been added to the methods section on line 351. We have also lightly edited line 122-124.

pore fluids proximal to the crystalline basement if the crystalline basement helium flux has a ^{4}~~He/N$_2$~~ $N_2/\ ^4He$ between 25-50 (Figure 2b).

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19% – 32% and N_2 concentrations are reduced by 2% – 50%

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Vg/V1l of 0.0478, 0.0474, 0.0422 and 0.0251

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Appendix Table 3. $N_2/^4$He or N_2/He ratios of sedimentary basin fluids sampled proximal to the basement and samples from the crystalline basement.

Suggest Authors have another column for 'inferred N2 source' where available

We thank the reviewer for the suggestion. We agree that it is a very intriguing and important topic to understand the source of N_2. However, most referenced literature cannot unambiguously identify the nitrogen source. We don't think attempting to add the information

to the table is useful as without discussion or clarification this could potentially be misleading.

Li, L. et al. N_2

Corrected in the manuscript

Karolytė, R. et al. The role of porosity in H_2/He

Corrected in the manuscript

Hiyagon, H. & Kennedy, B. Noble gases in CH_4-rich

Corrected in the manuscript

Mao, S. & Duan, Z... N_2–H_2O–NaCl

Corrected in the manuscript and corrected the other similar formatting mistakes in the reference

Thompson, J. (Williston Basin Symposium). Needs full reference

Full reference added in the manuscript

Reviewer Reports on the First Revision:

Referee #1:

The authors have responded very well to the remarks of the reviewers. My rather minor concerns have been resolved. I can therefore recommend the revised manuscript for publication in Nature.

However, there are still some minor technical details that should be corrected or at least checked by the authors. Below I note a few things that I detected when studying the revised manuscript and comparing it to the first version.

1) As a result of some changes of references made in response to reviewer 2 (e.g. additional refs. on line 49), the numbering of references is now not following the order of occurrence in the text anymore. This is a purely technical issue, of course.

2) There are 3 statements where the helium content of helium-rich gases / viable resources is quantified (lines 48, 64, 86), some of which have been changed in response to reviewer 2. I understand the reasoning of the reviewer and the authors in their reply, but I am not sure whether the adopted solution is ideal. Also, it is not fully clear whether the 0.3 % mentioned on line 86 for the Hugoton-Panhandle apply specifically for this field or generally (the latter would be in contradiction to the previous two occurrences). Maybe this can be optimized.

3) There are some grammatical mistakes (plural/singular of nouns and corresponding verb forms) in some edited sentences. This applies to the sentences on lines 48-51 and on lines 180-183.

4) Just a comment: The sentence referring to mass balance calculations and Figure 4 has been shifted backwards, from formerly lines 137-140 to now lines 171-175. This seems to be an improvement, among other things correcting the order of the references to Figs. 3 and 4 that I had apparently missed in the original manuscript.

5) I still do not understand the reference to Extended Data Table 2 in the context of the 20Ne concentration in air saturated water (methods, line 393), because this value does not occur in the table. Maybe the reference should just be moved one sentence up, as it may point to the 4He concentrations before degassing, which are listed in the table.

6) A formatting error with superfluous quotation marks seems to have occurred on lines 393, 407, and 459.

7) In Fig. 2b), the shaded area indicating the range where N2 solubility is exceeded seems to be shifted to the left from where it should be (old version).

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Author Rebuttals to First Revision:

Reply to reviewer comments

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Our point-by-point response is in red

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1) As a result of some changes of references made in response to reviewer 2 (e.g. additional refs. on line 49), the numbering of references is now not following the order of occurrence in the text anymore. This is a purely technical issue, of course.

We thank the reviewer for spotting this. We have reorganized the reference and updated the manuscript.

2) There are 3 statements where the helium content of helium-rich gases / viable resources is quantified (lines 48, 64, 86), some of which have been changed in response to reviewer 2. I understand the reasoning of the reviewer and the authors in their reply, but I am not sure whether the adopted solution is ideal. Also, it is not fully clear whether the 0.3 % mentioned on line 86 for the Hugoton-Panhandle apply specifically for this field or generally (the latter would be in contradiction to the previous two occurrences). Maybe this can be optimized.

We thank the reviewer for pointing this out and providing us with a chance to clarify this point. According to reference 29, natural gas fields with helium concentrations greater than 0.1% are referred to as helium-rich. Therefore, the Hugoton-Panhandle can also be classified as helium-rich, given that the samples have helium concentrations greater than 0.3%. We appreciate how this number mismatch could otherwise confuse the readers, and we removed '(>0.3)' on line 93.

3) There are some grammatical mistakes (plural/singular of nouns and corresponding verb forms) in some edited sentences. This applies to the sentences on lines 48-51 and on lines 180-183.
The grammatical mistakes have been corrected

4) Just a comment: The sentence referring to mass balance calculations and Figure 4 has been shifted backwards, from formerly lines 137-140 to now lines 171-175. This seems to be an improvement, among other things correcting the order of the references to Figs. 3 and 4 that I had apparently missed in the original manuscript.

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The reviewer is correct and we have moved the reference to the sentence above.

6) A formatting error with superfluous quotation marks seems to have occurred on lines 393, 407, and 459.

The quotation marks have all been checked and corrected

7) In Fig. 2b), the shaded area indicating the range where N2 solubility is exceeded seems to be shifted to the left from where it should be (old version).

We thank the reviewer for spotting this, and the figure is now corrected.