nature portfolio

Peer Review File

Abiotic and biotic-controlled nanomaterial formation pathways within the Earth's nanomaterial cycle

Corresponding Author: Dr Michael Schindler

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Version 0:

Decision Letter:

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Dear Dr Schindler,

Your manuscript titled "Abiotic and biotic-controlled nanomaterial formation pathways in the Earth's critical zone" has now been seen by 3 reviewers, and we 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.

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. In addition, we recommend that you: 1) expand your discussion and conclusion sections, further discussing the broader scientific impact this new classification scheme will have on the multidisciplinary study of nanomaterials' formation pathways, 2) expand the theoretical background of your field with previously published relevant literature and discuss your approach in relationship to similar approaches, and 3) provide further justification for including the discussion of the role of oceanic hydrothermal vents and the upper mantle.

Please submit your point-by-point responses as a separate file, distinct from your cover letter where you can add responses to the Editors' comments that you do not want to be made available to the reviewers. Word files are preferred.

Important: The response to reviewers must not include any figures, tables or graphs. If you wish to respond to the reviewer reports with additional data in one of these formats, please add them to the main article or Supplementary Information, and refer to them in the rebuttal. Due to current technical limitations, any figures, tables, or graphs embedded in your rebuttal will not be included in the peer review file, if published.

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), a tracked-changes version of the manuscript (as a PDF file) 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.

Please do not hesitate to contact us 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,

Carolina Ortiz Guerrero, Ph.D.
Associate Editor
Communications Earth & Environment

EDITORIAL POLICIES AND FORMATTING

We ask that you ensure your manuscript complies with our editorial policies. Please ensure that the following formatting requirements are met, and any checklist relevant to your research is completed and uploaded as a Related Manuscript file type with the revised article.

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Furthermore, please align your manuscript with our format requirements, which are summarized on the following checklist: Communications Earth & Environment formatting checklist

and also in our style and formatting guide Communications Earth & Environment formatting guide .

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All Communications Earth & Environment manuscripts must include a section titled "Data Availability" at the end of the Methods section or main text (if no Methods). More information on this policy, is available at http://www.nature.com/authors/policies/data/data-availability-statements-data-citations.pdf.

In particular, the Data availability statement should include:
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- Accession codes where appropriate
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- If a dataset has a Digital Object Identifier (DOI) as its unique identifier, we strongly encourage including this in the Reference list and citing the dataset in the Data Availability Statement.

DATA SOURCES: All new data associated with the paper should be placed in a persistent repository where they can be freely and enduringly accessed. We recommend submitting the data to discipline-specific, community-recognized repositories, where possible and a list of recommended repositories is provided at http://www.nature.com/sdata/policies/repositories.

If a community resource is unavailable, data can be submitted to generalist repositories such as figshare or Dryad Digital Repository. Please provide a unique identifier for the data (for example a DOI or a permanent URL) in the data availability statement, if possible. If the repository does not provide identifiers, we encourage authors to supply the search terms that will return the data. For data that have been obtained from publically available sources, please provide a URL and the specific data product name in the data availability statement. Data with a DOI should be further cited in the methods reference section.

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

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Schindler et al. present a new classification scheme for nanomaterial formation in the critical zone, with 4 schemes representing biotic and abiotic pathways coupled and decoupled from weathering processes. From these four major categories, 17 subcategories can also be identified based on changes to the nanomaterials or their surrounding environment. In my view, this communication is an important entry into our understanding of nanogeochemistry. The authors do an admirable job of explaining why examining these processes are necessary, putting the oft-overlooked nano-size regime in context of earth scale processes. The classification scheme put forth also is well explained and follows a logic that will set the foundation of thinking about nanogeochemical processes in subsequent work. There are however, 2 main points that should be addressed prior to acceptance for publication.

-The writing in general of this manuscript is a little weak. There are some instances where the authors rely on certain words or phrases to the point of distraction. As an example, Page 4 and 5 contain multiple variations of "this review" or "in this review" that I believe are unwarranted.
-The table, though comprehensive, is difficult to follow in it's current format. It might be worth considering break the table into multiple difference tables or otherwise reorganizing the information in a way that makes more sense. Perhaps flipping parent phases and location so that the table has a more logical flow.
-The 'critical zone' as described on page 4 (lines 81-89) should be moved toward the beginning of this section
-The concluding paragraph of page 13 should be expanded on. The points made here salient and can be reworked within the context of the proposed classification scheme to speak to its importance.

Overall, I'd recommend this manuscript be considered for publication after minor edits detailed above.

Reviewer #2 (Remarks to the Author):

Schindler et al. discuss abiotic and biotic-controlled nanomaterial formation pathways within the Earth's critical zone. While this topic has been extensively explored and documented in numerous review articles, the authors' categorization into 17 subcategories is novel and significant, providing new insights into the role and function of nanomaterials (NMs) in the upper soil and aquifers. I recommend the paper for publication following major revisions.

The paper's primary focus is the critical zone. However, the final sentence of the abstract shifts the discussion to oceanic hydrothermal vents and the upper mantle, which is inconsistent. I strongly recommend maintaining a focus solely on the critical zone. Additionally, the paper should further emphasize the ecosystem services of the critical zone and the influence of NMs on these services. A thorough exploration of this aspect is preferable to expanding the scope to include the mantle or hydrothermal vents. Otherwise, the paper, its title, and its focus would need substantial adjustments.

In this context, I suggest refining the definition of the critical zone. Lines 83 to 85 state, “This zone is defined as the terrestrial portion of the surface of the Earth that extends from the top of the vegetation canopy down through soil and rock layers where meteoric water circulates.” This definition potentially extends down to magma, which is well beyond the critical zone. According to the National Research Council, the critical zone is the near-surface environment that "regulates the natural habitat and determines the availability of life-sustaining resources," typically encompassing the first 100 to 200 meters. While I don’t want to stimulate a detailed discussion on defining the critical zone, I advise against titling the paper "Critical Zone” in the current form. Instead, for the current version, consider a title such as "Nanomaterials in Earth's Cycles”. This would clarify the paper's objectives. Or focus on the top 100 meters and its functions, focus on the critical zone and its services only. This second option would be my preferred suggestion. Otherwise, it is unclear how this paper adds new insights compared to the reviews by Hochella et al., which have been previously published.

Line 81 and following: It is not accurate to claim that this paper is the first to review the principles of biotic and abiotic-controlled formation pathways of nanomaterials (NMs). A substantial body of literature on this topic exists, including contributions from the authors themselves and many others. These findings are not new. Please revise this section to acknowledge and cite the extensive and excellent literature that has been published on this topic over the years (and decades).

However, the discussion starting at line 90 is particularly interesting and represents a key aspect of this paper—the development of these subcategories. I suggest deleting the statement at line 81 and instead describing the significant body of existing literature. Define the aim of this paper as developing the 17 subcategories based on the existing literature.

Line 133 and following: Please elaborate on and cite the extensive literature regarding homo- and hetero-aggregation, emphasizing their significance for the formation pathways. The brief mention of these processes does not suffice considering their importance and the substantial amount of research in these fields. Combining aggregation and surface attachment in a single sentence overlooks the depth of these major research areas.

Line 263: It may be crucial to discuss the formation of nanomaterials in surface water flocks. Several publications in 2023 and 2024 have explored this topic, recognizing flocks as dynamic reactors for nanomaterial formation. Flocks serve as habitats for microorganisms, potentially passively trapping nanomaterials through extracellular polymeric substances (EPS) and providing nutrients to microbiota. The paper lacks consideration of flock formation and function, which is significant in understanding nanomaterial dynamics.

Line 319 and following: The assertion made here is fundamental mineralogical knowledge. It is widely understood that all bulk mineral phases originate as nanomaterials and may develop into ore deposits or materials. Concluding the paper with

such basic information, akin to introductory content in Earth Sciences 101, may be redundant for the intended readership. Instead, I recommend delving deeper into the potential contributions of the new classification to the scientific community, how it can inspire further research, and the novel insights derived from the delineation of the 17 subgroups.

Figure 1 is partly adapted from Donald Sparks, Ruben Kretschmar in the work form Chorover et al. 2007 in Elements, isn't it?

Reviewer #3 (Remarks to the Author):

The manuscript entitled Abiotic and biotic-controlled nanomaterial formation pathways in the Earth's critical zone by Schindler et al. reviews the main pathways for nanoparticle formation, by establishing a novel classification into several categories and subcategories of mechanisms and providing significant examples of environmental processes relevant to each category. It is very well written, and easy to follow.
It is an extremely interesting manuscript, representing a key reference for the field as (i) it allows identifying potential nanoparticle formation mechanisms as it outlines key characteristic of each pathway, and (ii) it provides basic understanding on the mechanism underlaying the formation of bulk materials, as nanomaterials are commonly precursors to them after their growth or aggregation. At such, it represents an unvaluable piece of work for researchers on this and related topics, and given my impression on the manuscript, I recommend it acceptance in its present form.

Version 1:

Decision Letter:

** Please ensure you delete the link to your author home page in this e-mail if you wish to forward it to your coauthors **

Dear Dr Schindler,

Your revised manuscript titled "Abiotic and biotic-controlled nanomaterial formation pathways within the Earth's nanomaterial cycle" has now been seen by our editors. We are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment, as a REVIEW ARTICLE.

We therefore invite you to edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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

Best regards,

Carolina Ortiz Guerrero, Ph.D.
Associate Editor
Communications Earth & Environment

Responses to the reviewer comments (in bold)

Schindler et al. present a new classification scheme for nanomaterial formation in the critical zone, with 4 schemes representing biotic and abiotic pathways coupled and decoupled from weathering processes. From these four major categories, 17 subcategories can also be identified based on changes to the nanomaterials or their surrounding environment. In my view, this communication is an important entry into our understanding of nanogeochemistry. The authors do an admirable job of explaining why examining these processes are necessary, putting the oft-overlooked nano-size regime in context of earth scale processes. The classification scheme put forth also is well explained and follows a logic that will set the foundation of thinking about nanogeochemical processes in subsequent work. There are however, 2 main points that should be addressed prior to acceptance for publication.

-The writing in general of this manuscript is a little weak. There are some instances where the authors rely on certain words or phrases to the point of distraction. As an example, Page 4 and 5 contain multiple variations of "this review" or "in this review" that I believe are unwarranted.
Thank you for your comment.
We agree with the reviewer and have changed our wording. We also think that our paper should not be called a "Review", but an "Article", in that something new is created from the data in the literature that we collected. That is, the collection of data is mined out of the current literature and then put together to build a classification system. This is similar to phylogeny in the biological sciences. The product of our data mining is a new classification scheme and ties the many types of NM formation into a coherent system. Any system of classification allows one to form a coherent framework which forms the foundation of that particular field of science, in this case, naturally occurring Earth-based NMs.

-The table, though comprehensive, is difficult to follow in it's current format. It might be worth considering break the table into multiple difference tables or otherwise reorganizing the information in a way that makes more sense. Perhaps flipping parent phases and location so that the table has a more logical flow.

We agree with the reviewer. We flipped parent phase and location and transferred the Table into image files, where we use different colours, height of rows and letter sizes to differentiate between headings for major categories, subcategories and examples.

-The 'critical zone' as described on page 4 (lines 81-89) should be moved toward the beginning of this section
-The concluding paragraph of page 13 should be expanded on. The points made here salient and can be reworked within the context of the proposed classification scheme to speak to its importance.

We expanded the concluding paragraph from lines 298-327 to lines 372-406 and emphasized at the end of the paper the importance of our classification scheme. The new wording is as follows:

Finally, as mentioned above, the discovery and delineation of all of the chemical/physical/biological-based pathways of NM formation will result in better understanding the NM cycle of the Earth as a fully connected and evolving system[1]. There is also the possibility (likelihood) that completely new and/or novel NM formation pathways will be discovered. Such conceptual frameworks have been shown repeatedly to have great value, e.g. the rock cycle, the water cycle, and the many chemical cycles of the Earth. Ultimately, the nano-reactants that appear through this cycle, including those NMs that only exist for very short times, consequently impact the entire Earth system (atmosphere, hydrosphere, and terrestrial Earth) in highly consequential ways as has been recently reviewed[1].

Reviewer #2 (Remarks to the Author):

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Thank you very much for these stimulating comments. Indeed, we initially focused on the critical zone, but soon realized that similar pathways of nanomaterial formation occur also in other compartments of the Earth. It is our opinion that one should not ignore the processes in the latter compartments as they are important for our understanding of ore forming processes and element cycling. Hence, we decided to cover all compartments of the Earth for which nanomaterial formation processes have been reported. We do not agree with the reviewer that this expansion of pathways has been previously published by Hochella et al. as these papers focused on the properties, abundance and cycling of nanomaterials between compartments and did not depict specific pathways of nanomaterial formation. We made the following changes to include nanomaterial formations for the different Earth compartments:
We changed the title to

Abiotic and biotic-controlled nanomaterial formation pathways within the Earth's nanomaterial cycle

As we still focus on processes in the Earth's critical zone (following partially the recommendation of the reviewer), we also now look at other Earth compartments. We introduce this on pages 4 and 5, lines 90-102, as follows:

This present article concerning the group recognition and current understanding of abiotic- and biotic-controlled formation pathways is the first of its type. Pathways of NM formation can occur in highly diverse environments, for example in the presence of aqueous fluids in soils, sediments, and the biological environment therein, all the way to within the gases or magmas in volcanic systems. We will focus primarily on formation pathways of NM in aqueous fluids as those occur in the Earth's critical zone (CZ). Within this zone, humans interact directly or indirectly with NMs as they impact the form and function of living and non-living things. [4] However, we will also show that pathways of NM formation similar to those observed in the CZ also occur in hydrothermal systems, as well as in gas-dominated systems such as volcanic fumaroles, wildfires, and particle formation in the stratosphere and lower troposphere. Addressing pathways of NM formation outside the CZ is important as they also play fundamental roles, for example, in the cycling of elements, the formation of ore deposits, and the radiation budget of the atmosphere.

These lines also provide a good definition of the critical zone.

Line 81 and following: It is not accurate to claim that this paper is the first to review the principles of biotic and abiotic-controlled formation pathways of nanomaterials (NMs). A substantial body of literature on this topic exists, including contributions from the authors themselves and many others. These findings are not new. Please revise this section to acknowledge and cite the extensive and excellent literature that has been published on this topic over the years (and decades).

However, the discussion starting at line 90 is particularly interesting and represents a key aspect of this paper—the development of these subcategories. I suggest deleting the statement at line 81 and instead describing the significant body of existing literature. Define the aim of this paper as developing the 17 subcategories based on the existing literature.

There are indeed many review papers on nanomaterials and now we cite many of them at lines 84-89:

Formation, properties, cycling and budgets of NM's in Earth compartments (lithosphere, atmosphere and hydrosphere) have been reviewed numerous times [4,5] with these papers also focusing on specific types of NMs [6,7] and their role in a vast array of Earth and environmental processes [8]. However, this field is still in its infancy with new and often unexpected discoveries constantly reported. In addition, research challenges are great due to chemical and physical complexities of nano-processes, and the exceptionally small scales that must be navigated.

However, our paper still differs from those as we list different formation pathways and the reviewer will agree that the pathways listed in Tables 1 and 2 have not previously published. Hence, the statement at the lines 90-91 is valid:
This present article concerning the group recognition and current understanding of abiotic- and biotic-controlled formation pathways is the first of its type.

Line 133 and following: Please elaborate on and cite the extensive literature regarding homo- and hetero-aggregation, emphasizing their significance for the formation pathways. The brief mention of these processes does not suffice considering their importance and the substantial amount of research in these fields. Combining aggregation and surface attachment in a single sentence overlooks the depth of these major research areas.

As this paper is not on growth mechanisms, we only added one more sentence and wrote in lines 146 to 150 the following:
The latter mechanism is termed crystallization through particle attachment (CPA) [12], where the attachment of the NM can occur in a random or orientated fashion. For random attachment, structural re-organization is required for incorporation of the NM into the bulk crystal, whereas orientated attachment requires the rotation of the NM upon attachment [13].

Line 263: It may be crucial to discuss the formation of nanomaterials in surface water flocks. Several publications in 2023 and 2024 have explored this topic, recognizing flocks as dynamic reactors for nanomaterial formation. Flocks serve as habitats for microorganisms, potentially passively trapping nanomaterials through extracellular polymeric substances (EPS) and providing nutrients to microbiota. The paper lacks consideration of flock formation and function, which is significant in understanding nanomaterial dynamics.
Thank you for this information. We added the pathway via EPS in lines 298-306 and wrote:

Biological EPS matrices may facilitate new exopolymer NM formation out of the pre-existing polymeric components (IIId, Table 1) [71]. They may also concentrate mobile metal species through chelation and biosorption [72,73], which occasionally lead to NM formation [74,75]. We note that the category IVc in Table 1 is inclusive of decoupled, EPS-mediated NM

formation processes as naturally occurring microbial communities, by default, exist as biofilms with EPS matrices. Although EPS of biological flocculates have been previously reported as effective traps and stabilizers for pre-existing, mostly engineered nanoparticles in the environment, the focus of this paper is on the formation pathways of naturally occurring NMs, and thus we will not elaborate on this aspect.

Line 319 and following: The assertion made here is fundamental mineralogical knowledge. It is widely understood that all bulk mineral phases originate as nanomaterials and may develop into ore deposits or materials. Concluding the paper with such basic information, akin to introductory content in Earth Sciences 101, may be redundant for the intended readership. Instead, I recommend delving deeper into the potential contributions of the new classification to the scientific community, how it can inspire further research, and the novel insights derived from the delineation of the 17 subgroups.

We agree with this comment, and we removed the sentence:

"*All bulk materials (in this case, minerals and other organics/inorganic amorphous matter) must start as a nanomaterial that may, given proper conditions, grow into a bulk phase, obviously a key part of Earth element distribution and cycling."*

and focused instead on future research and wrote new lines 401 to 409:

Finally, as mentioned above, the discovery and delineation of all of the chemical/physical/biological-based pathways of NM formation will result in better understanding the NM cycle of the Earth as a fully connected and evolving system[1]. There is also the possibility (likelihood) that completely new and/or novel NM formation pathways will be discovered. Such conceptual frameworks have been shown repeatedly to have great value, e.g. the rock cycle, the water cycle, and the many chemical cycles of the Earth. Ultimately, the nano-reactants that appear through this cycle, including those NMs that only exist for very short times, consequently impact the entire Earth system (atmosphere, hydrosphere, and terrestrial Earth) in highly consequential ways as has been recently reviewed[1].

Figure 1 is partly adapted from Donald Sparks and Ruben Kretschmar in the work form Chorover et al. 2007 in Elements.
We believe that we changed the Figure significantly and thus do not violate any copyrights. However, we acknowledge Chorover et al. in the Figure caption and wrote:

Fig. 1. Schematic overview of various pathways of nanomaterial formation in aqueous fluids within the critical zone (CZ) (the sketch of the CZ is adapted from [101]). Note that the upper half of the schematic is for biotic-controlled pathways, and the lower half for abiotic-controlled pathways.

Reviewer #3 (Remarks to the Author):

The manuscript entitled Abiotic and biotic-controlled nanomaterial formation pathways in the Earth's critical zone by Schindler et al. reviews the main pathways for nanoparticle formation, by establishing a novel classification into several categories and subcategories of mechanisms and providing significant examples of environmental processes relevant to each category. It is very well written, and easy to follow.
It is an extremely interesting manuscript, representing a key reference for the field as (i) it allows identifying potential nanoparticle formation mechanisms as it outlines key characteristic of each

pathway, and (ii) it provides basic understanding on the mechanism underlaying the formation of bulk materials, as nanomaterials are commonly precursors to them after their growth or aggregation. At such, it represents an unvaluable piece of work for researchers on this and related topics, and given my impression on the manuscript, I recommend it acceptance in its present form.

Thank you for these kind comments.