

Monitoring, modeling, and forecasting long-term changes in coastal seawater quality due to climate change

Corresponding Author: Professor Chaohai Wei

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

Reviewer #2

(Remarks to the Author)

Extremely interesting manuscript, bringing valuable information to climate change metal and metalloid dynamics alterations
Minor revisions only.

Please alter "trace element" to "metals and metalloids" according to IUPAC recommendations throughout

Line 39 - Regarding "Acidification leads to changes in the phase distribution of trace elements, affects the functions of electron donors involved in cell respiration, and disrupts the promotion of enzymatic reactions (Richir, 2016)." - You must also indicate that acidification (pH) also alters metal and metalloid toxicity and bioavailability, due to altered redox state.

Lines 46-54 require references

Line 81 - Remove JW from the Erisman reference.

Line 159 - Indicate why Cr(VI) and not Cr total, due to the sampling station particularity. No other elements are analyzed per chemical species?

Line 166 - What are HMs? Heavy metals? Remove, replace by metals and metalloids

Better explain the meaning of environmental Kuznets curve for metal elements.

Line 169 - You cite e-waste but do not indicate Rare Earth Element contamination in your assessment.

Table 1- Define GB3838 - WHO, CMC

I missed a discussion on altering water currents due to climate change as drivers for metal and metalloid contamination

Line 217 - Remove fortunately, indicative of opinion

Line 246 - Change 3 categories to three categories

Line 251 - Cd, Hg, and Pb do not originate mainly from sulfur deposition, correct.

Line 228-240: " Based on our estimates, the dissolved phase concentrations of Fe, Cu, Pb, and Cr in 2099 will increase by 63.4±7.2 %, 63.1±5.7 %, 61.9±7.0 %, and 48.2±8.2 %, respectively, compared to 2020" Why only cite these three elements? What of the rest?

Line 345 - Define ORP.

Line - 364 - explain the term "non-biophilic". Not very uptake by living organisms? This is not very true for Cr. Not very bioavailable? Not true for both elements.

Line 411 - remove the capital from Marine organisms

Line 411-415 - you neglected to indicate that marine organisms will also be exposed to higher metal and metalloid concentrations in some cases, leading to altered metabolism and physiology.

Line 426 - You state you used Cr(VI), this becomes a problem as you have no speciation for other elements you analyzed. I recognize this limitation due to the samplings of the monitoring stations. However, you must at least acknowledge this and discuss speciation in your manuscript with regard to the other analyzed elements, as this directly affects metal and metalloid bioavailability. You have done so in lines 81-85 for Hg, although you neglected to indicate methylation processes as one of the drivers for Hg cycling.

Line 441 - Data points lower than the limit of detection (LOD) were replaced with half of the LOD - insert reference on this

approach, and report the LOD for each element. Is there more than one LD reported?

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

Review 449403_1

This manuscript explores the issue related to long-term changes in coastal seawater quality due to climate change. The authors used a huge dataset to make predictions about the changes in water quality in the coastal area of the Guangdong Province (China) for the period 2015-2022. The authors carried out a notable effort for modeling this dataset which may be useful to predict long-term changes in the water quality of the investigated coastal area. The general topic of the manuscript is undoubtedly original, interesting and useful.

The authors adequately responded to most of my comments and updated the text of the manuscript accordingly. In the revised version of the manuscript, the authors made a great effort to better prepare the main text and to better organise the Supplementary Material (SM). The SM is now reorganized in 4 sections and, in my opinion, more better prepared. In addition, the authors provided a better and more detailed explanation of the sampling strategy.

Please, excuse me for suggesting to place Materials and Methods before the section dedicated to the Results. Actually, I have not checked the general writing style of the Journal. This is my fault.

I retain that the manuscript may be accepted in the Journal Nature Communications, just following some minor revisions.

Please, consider the following minor general comments.

Regarding terminology, the authors modified “trace elements” with “trace metals”. Between the two, I suggest the authors to use the first, so “trace elements”. With my comment on the original version of the manuscript, I simply meant to specify which elements were taken into account (so Cd, Cr, Cu, Fe, Hg, mn, Pb and Zn). Just name them in the text.

I suggest the authors to avoid the use of “we” such as “we show” (line 28 in the abstract). What about: “The aim of this study is to show the nexus of re-exposure-eutrophication-emission reduction with human activities and climate feedback, cannot be ignored in the pursuit of effective environmental governanc”? or something like that.

Line 95 I am not sure about “Incompatible elements”. Could you just write: “elements such as Hg, Cd and Pb also enter....?”

Line 99 the term “uptake” usually refers to Hg bioaccumulation along the trophic chain. Please, consider the idea to write “the influx of Hg(0) at the water-atmosphere interface is...”. Indeed, the term “influx” referred to a flux of Hg(0) from the air to the water and it is more appropriate with respect to “uptake”.

Line 121 physico-chemical properties. Check also through the text

(Remarks on code availability)

Reviewer #2

(Remarks to the Author)

The authors have responded adequately to both my initial comments and the first reviewer’s assessments. I thus endorse this paper for publication as is.

(Remarks on code availability)

Reviewer #3

(Remarks to the Author)

This manuscript presents an 8-year long data set of trace metal measurements in the coastal area of the Guangdong Province (China). Based on this data set they create a receptor model and subsequently apply this to a future simulation made by a model in the CMIP6 archive. The data set they present is quite unique and a significant contribution in itself. The PCA analysis and receptor model also provide novel and interesting insight into trace metal dynamics in coastal systems under climate change. There is not much current literature on this topic so this work presents a valuable and original piece of work. The quality of the data appear very good and the methods are described in detail. The overall analysis and analytical approach are both robust and comprehensive. With the exception of some strange, to me at least, use of terminology and a general lack of definitions for the abbreviations used the paper is well written.

However, despite having now read the manuscript, and supplemental information, several times I struggle to identify what the key result is. Nature Communications publishes work that “aim to represent important advances in all areas of the biological, health, physical, chemical, Earth, social, mathematical, applied, and engineering sciences”. It is unfortunately not at all clear to me what important advance this work makes. Partly this is because the authors also have not written any conclusions, but the noteworthy results are also not apparent in the “results and discussion” section. I believe there is noteworthy insight in the presented data and would urge the authors to restructure and revise their manuscript to focus on the key findings and their impact. You have a story in here – you just need to tell it.

Like the previous reviewer #1 I would really like the materials and methods to be presented before the results and discussion. It is not easy to understand the presented results before first reading what has been done. I've read your responses to the previous reviewer and I understand your argumentation, but this structure is making it challenging for the reader to understand your work.

Specific comments:

Line 51: It is unclear what “carbonate atmospheres” means.

Line 52-53: The sentence “trace metals may become sensitive factors in response to changes in marine water quality” is unclear. What responds to what?

Line 56: I think “butterfly effect” is a poor choice of words here. Consider revising.

Line 65: Replace “tracers” by “influencing factors” to avoid confusion

Line 67: Add “organic” before “C, N, P, and S elements”

Line 74: It is unclear what “incompatible elements” are. I realize this term is in response to a previous review, but it is not made clear what the elements are incompatible with so the sentence is unclear.

Line 74-94: Beginning with “Incompatible elements” this should all be a separate paragraph. The paragraph also needs a proper topic sentence and better flow between the sentences. It now reads like a list of bullet points someone have put together but not linked. There is no introduction, no coherency.

Line 77 and 78: why are you defining nitrogen and mercury here? The abbreviation for both have been used previously, and you do not define the other metals and elements.

Line 78: what is “Mercury's(Hg) earth system model”? Please explain.

Line 85: replace “limits” with “limitation”

Line 88-89: I do not understand this sentence. Nitrogen removal and denitrification does happen in situ in the ocean. And even if it did not I do not follow why that would mean that “nutrients and trace metals are affected by thermohaline circulation”. I'm not questioning that both nutrients and trace metals are affected by the thermohaline circulation, I just do not follow the logic of the sentence and cannot understand what you are trying to communicate.

Line 95-96: please use “physico-chemical” throughout

Line 114-115: Please reiterate here what these three factors are

Line 118: It is unclear what you mean by “CMIP6 framework” here. You have, as I understand it, used output from one of the models contributing to CMIP6. If that is what you want to communicate here please state that, otherwise explain what the framework is.

Lines 128-129: The warming process cannot be controlled anywhere at the moment so the meaning of this final statement is unclear. Please revise.

Lines 153-154: Could you provide a reference for the environmental Kuznets curve?

Line 166: TN is not defined

Line 171: This sentence is unclear. Please revise for clarity.

Lines 246-247: I do not know what “energy substances” are

Line 314: it is not clear what “compatible elements” are. See also my comment above regarding “incompatible elements”

Line 322-323: This sentence is unclear. Please revise.

Line 331: The terminology “concentration levers” was commented on in the previous review and I find that an explanation is still lacking. Please add some detail.

Line 352: Do you mean “decreases”?

Lines 354-357: Why do you see such a dip and recovery pattern for Pb and Cr? Such a pattern is not seen in the other trace metals. A bit of additional discussion on this would have been nice.

Lines 366: The decrease in C/N ratio does not appear to be significant given the uncertainty range. That should be noted.

Line 367: Please explain what the “carbon sources” are. Same for line 395.

Line 375: add “coastal” before “ocean's”

Lines 394-398: This is interesting, but could use some elaboration. If properly phrased and referenced I believe this could be an impactful conclusion. But it is unclear whether it is a true result of the work, or just supposition.

Line 409: replace “which were” by “and”

Line 451: “... among variables ...” What variables are these?

Section 3.2:

Here it is apparent that the authors are not very familiar with CMIP6. That is perfectly fine. I'm suggesting some changes to the text to make it more “standard” and thus more easily understood.

Line 430 onwards: You could rewrite as “Here we use ScenarioMIP simulations from the Earth System model GISS-E2. The model has a spatial resolution of 2 degrees latitude by 2.5 degrees longitude, and we use the monthly output from the “r1i1p1f1” ensemble member.”

I am wondering here why you have used this particular model. Many of the models in CMIP6 have a higher spatial resolution (at least 1x1 degree) which would better resolve your study area.

Lines 433-434: Please use here the exact name of these variables in the model output. This will help readers to identify and the correct data.

You should repeat here which scenario you use even though you have mentioned this before in the text.

(Remarks on code availability)

Version 2:

Reviewer comments:

Reviewer #3

(Remarks to the Author)

In my first review I wrote that “The data set they present is quite unique and a significant contribution in itself. The PCA analysis and receptor model also provide novel and interesting insight into trace metal dynamics in coastal systems under climate change. There is not much current literature on this topic so this work presents a valuable and original piece of work. The quality of the data appear very good and the methods are described in detail. The overall analysis and analytical approach are both robust and comprehensive.” I stand by this assessment, and the authors have responded well to most of my concerns. However, based on the authors’ rebuttal to my first review I have identified a major issue with their methodology. To remedy this parts of the analysis needs to be redone, and the new results analyzed. This major issue is described below. In addition, there remains some minor issues left unanswered in the rebuttal and revised manuscript. These are described under “minor issues” below. The manuscript will require major revisions before it can be published.

Major issue

In my first review I noted that it is clear from the methods that the authors are not familiar with Earth System Models (ESMs) or CMIP6. I did not find any errors in their work, but requested the add additional detail in the methods to identify which variables they had used as well as more detail about their choice of model. The authors have not added any of this information in the revised manuscript, but they did provide additional detail in their rebuttal to me. Unfortunately, this additional information shows me very clearly that the authors have not done the analysis they think they did, or at least not the analysis they describe in the manuscript. In my original review I asked the authors to include the actual variable names from the model so that the reader can more easily identify what has been used (since all CMIP6 models use the same variable naming conventions). This the authors have not done, but they have included an appendix (Appendix 1) with detailed metadata for three variables they have extracted. To better highlight the issues I’ve identified I’ve removed much of the information from Appendix 1 of the rebuttal and only kept what is important to support this review.

On lines 446-447 it is stated that “water temperature, rainfall, CO2 levels (pCO2), and ocean acidification rates were extracted.” In Appendix 1 of the rebuttal the authors include detailed metadata for three variables under the headings “Surface sea temperature”, “pCO2”, and “Wet deposition”. The information in Appendix 1 shows that the variables extracted are “tasmin”, “co23D”, and “clwvi”.

“tasmin” is the variable extracted under the headline “Surface sea temperature”. However, this is not water temperature. Realm is given as “atmos” indicating that it is an atmospheric variable, and the long_name attribute states that this is “Daily Minimum Near-Surface Air Temperature”. So rather than using water temperature in their model the authors have used air temperature. And not even the actual air temperature, but the minimum of the near-surface (typically 2-meter) temperature. This variable is also extracted from a different model than that stated on line 442 (see below).

“co23D” is the variable extracted under the headline “pCO2”. This is also an atmospheric variable as indicated by the attribute realm (“atmos”). To me it would make more sense to use surface ocean pCO2, but it is not actually stated anywhere whether atmospheric or oceanic pCO2 is identified as one of the three drivers. Importantly though, the attribute long_name shows that this variable is a “3D field of transported CO2”. The authors have not made clear whether they have used the entire 3D field as input to their model, or whether they’ve used just one layer. Regardless, I believe the authors want to use atmospheric CO2 concentration here, which is part of the model forcing (the SSP3-7.0 scenario defines the atmospheric CO2 concentration for each timestep).

“clwvi” is the variable extracted under the heading “Wet deposition”. This is probably what the authors believe represents rainfall (one of the variables they claim on line 446 to have extracted). This is another atmospheric variable (realm = atmos), but that would in this case make sense. However, the attribute long_name shows that this is “Condensed Water Path” and the comment, as well as a quick check in the CMIP6 variable list, indicates that this variable represents the amount of liquid water and ice in clouds. So not related to precipitation. In addition, this variable is from a different ensemble run from that given on line 444 (see below).

On line 442 the authors write that output from the GISS-E2 model is used. This appears to be incorrect. The variable “tasmin” is retrieved from the Chinese FGOALS-g3 model (parent_source_id=FGOALS-g3). On line 444 it is stated that the ensemble member “r1i1p1f1” is used. However, for variable “clwvi” parent_variable_label=r1i1p3f1 is used. It is not advised to use two different ensemble members for different variables, and if done it needs to be stated clearly.

On lines 446-447 four variables are listed, but there are only three in Appendix 1. I therefore do not know what data, if any, has been extracted as “ocean acidification rates” since no information is given.

The information I’ve provided above clearly shows that the authors have not used the variables they say they have. I’m sorry that this is discovered so late in the review process, but this is why the actual variable names should always be provided. Had the authors included this information in the previous version it would have been clear to me, and probably also the other

reviewers, that something is wrong with the analysis. The authors will have to extract the correct variables (this list should be helpful in identifying which they are: <https://pcmdi.llnl.gov/mips/cmip3/variableList.html>) and redo their analysis for the future. The results and conclusions will then have to be modified based on the new results.

Finally, I want to note that in their rebuttal to my comment about why they have chosen the model they have the authors show their inexperience with Earth System Models and CMIP6. The ocean resolution in the models they've extracted data from is 1 by 1 degree, which is clear from the attributes listed in Appendix 1. The atmospheric resolution in Earth System models is always coarser than the ocean. I assumed in my initial review that they had extracted from the ocean, not the atmosphere. Given this obvious inexperience I would urge the authors to reach out to the contact person given for the CMIP6 models (the attribute contact gives an e-mail address) and ask for advice on how to best use their data, and which variables are the most appropriate.

Minor issues:

Line 24: "proved" is a very strong word to use here. Consider revising.

Line 38: Typo. Separate "crucialwater" into two words (crucial and water)

Line 53: The change to "acidification atmosphere" does not make any more sense than the original "carbonate atmosphere". It is true that the nearshore environment is affected by acidification, but the term "acidification atmosphere" is not terminology that is commonly used in the scientific literature. At the very least the authors need to add an explanation/definition to the text, but better would be to just say that the nearshore environment is affected by acidification (which is apparently what they are trying to communicate here).

Line 67: The common terminology is "sea surface temperature". Please revise.

Line 92-93: I commented on this sentence in my first review and it makes no more sense now. Nitrogen removal and denitrification does happen in situ in the ocean. The reference used here for the statement that these processes do not happen (Hutchins and Capone, 2022) does not in any way support such a statement. It is factually incorrect to state that "N removal and denitrification processes have been proven not to occur in situ", and the authors are also incorrectly referencing Hutchins and Capone (2022) to support the incorrect statement. This needs to be revised.

Line 120: Here you state that "sea surface temperature, dry and wet deposition amount, and pH" are the three factors being considered in your model. However, on lines 206, 228, and 297 as well as in Figure 1b you list the three components as "SST, deposition, and pCO₂" suggesting that pH is not one of the three components. But then on line 216 pH is mentioned as one of the three. Please correct the information in all these places to avoid confusion. In addition, you should indicate in all these places which type of deposition. I assume it is both wet and dry as initially stated, but this should be clear everywhere. Also, please define pCO₂ the first time it is used, not the last. Now "partial pressure of carbon dioxide" is used on line 297, but not before. You also need to clarify whether you've used atmospheric or oceanic pCO₂.

Line 124: In my original review I wrote here that "It is unclear what you mean by "CMIP6 framework" here. You have, as I understand it, used output from one of the models contributing to CMIP6. If that is what you want to communicate here please state that, otherwise explain what the framework is." The authors answered in their rebuttal that they would make a modification, and link to the Materials and Methods section but this has not been done. It is clear that the authors are not familiar with the use of Earth System Models and CMIP6 (as also explained above). They have not used the CMIP6 framework, which is a framework to allow the intercomparison of many different models, but rather output from one Earth System model which has contributed to the CMIP6 project. There is no model comparison, or model ensemble used in this work. So here it would be more appropriate to state "the CMIP6 version of the GISS-E2 model was used to predict ...". However, as explained above this is also not accurate since the authors have actually used data from two models.

Line 127-128: This paper does not simulate the SSP370 scenario. Output from Earth System Models that have simulated this scenario is used, but that is not the same thing. This needs revision for clarity.

Lines 134-135: The authors have revised this statement somewhat after my initial review. However, the statement is still unclear and my original comment that "the warming process cannot be controlled anywhere at the moment" is still valid. Adding "or reversed" does not change or clarify anything. I do not understand what the authors are trying to say so the sentence still needs revision.

Line 399-400: Here two sentences in a row begin with "However". That does not make sense so please revise.

Below is a subset of the information provided in the rebuttal (Appendix 1):

Surface sea temperature

Global Attributes:

Conventions = 'CF-1.7 CMIP-6.2'

activity_id = 'ScenarioMIP'

contact = 'Lijuan Li (ljli@mail.iap.ac.cn)'

experiment = 'update of RCP8.5 based on SSP5'

experiment_id = 'ssp370'

further_info_url = 'https://furtherinfo.es-doc.org/CMIP6.CAS.FGOALSg3.ssp370.none.r1i1p1f1'

parent_activity_id = 'CMIP'

parent_experiment_id = 'historical'

parent_mip_era = 'CMIP6'
parent_source_id = 'FGOALS-g3'
parent_variant_label = 'r1i1p1f1'
realm = 'atmos'
variable_id = 'tasmin'
tasmin
Size: 180x80x60
Dimensions: lon,lat,time
Datatype: single
Attributes:
standard_name = 'air_temperature'
long_name = 'Daily Minimum Near-Surface Air Temperature'
comment = 'minimum near-surface (usually, 2 meter) air temperature (add cell_method attribute 'time: min')'
units = 'K'

pCO2
Global Attributes:
Conventions = 'CF-1.7 CMIP-6.2'
activity_id = 'ScenarioMIP'
contact = 'Kenneth Lo (cdkkl@giss.nasa.gov)'
experiment = 'update of RCP7.0 based on SSP3'
experiment_id = 'ssp370'
further_info_url = 'https://furtherinfo.es-doc.org/CMIP6.NASA-GISS.GISSE2-1-G-CC.ssp460.none.r10i1p1f1'
parent_activity_id = 'CMIP'
parent_experiment_id = 'historical'
parent_variant_label = 'r1i1p1f1'
parent_source_id = 'GISS-E2-1-G-CC'
realm = 'atmos'
variable_id = 'co23D'
co23D
Size: 144x90x40x432
Dimensions: lon,lat,lev,time
Datatype: single
Attributes:
standard_name = 'CO2_3D_tracer'
long_name = '3D field of transported CO2'
comment = 'report 3D field of model simulated atmospheric CO2 mass mixing ration on model levels'
units = 'kg kg-1'

Wet deposition
Global Attributes:
Conventions = 'CF-1.7 CMIP-6.2'
activity_id = 'ScenarioMIP AerChemMIP'
contact = 'Kenneth Lo (cdkkl@giss.nasa.gov)'
experiment = 'gap-filling scenario reaching 7.0 based on SSP3'
experiment_id = 'ssp370'
further_info_url = 'https://furtherinfo.es-doc.org/CMIP6.NASA-GISS.GISSE2-1-G.ssp370.none.r1i1p3f1'
parent_experiment_id = 'historical'
parent_experiment_rip = 'r1i1p3'
parent_mip_era = 'CMIP6'
parent_source_id = 'GISS-E2-1-G'
parent_variant_label = 'r1i1p3f1'
realm = 'atmos'
variable_id = 'clwvi'
clwvi
Size: 144x90x432
Dimensions: lon,lat,time
Datatype: single
Attributes:
standard_name = 'atmosphere_cloud_condensed_water_content'
long_name = 'Condensed Water Path'
comment = 'Mass of condensed (liquid + ice) water in the column divided by the area of the column (not just the area of the cloudy portion of the column). Includes precipitating hydrometeors ONLY if the precipitating hydrometeors affect the calculation of radiative transfer in model.'
units = 'kg m-2'

(Remarks on code availability)
I have not reviewed the code.

Version 3:

Reviewer comments:

Reviewer #3

(Remarks to the Author)

The authors have revised the manuscript very well after my last review. I have no further comments.

(Remarks on code availability)

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Response to Reviewers' Comment

Manuscript Number: NCOMMS-23-39043

Title: Monitoring, modeling, and forecasting long-term changes in coastal seawater quality due to climate change

Journal: Nature Communications

Dear Reviewers,

We sincerely appreciate the reviewers' time and efforts devoted in reviewing our manuscript and the insightful and pertinent suggestions provided for us to further improve our work. We have carefully revised the manuscript according to the reviewers' comments. Detailed responses are included below. All our responses are color-coded in **blue**. All revisions are highlighted in **red** in a marked version of the revised manuscript. The line and page numbers referred correspond to those in the revised version manuscript. We hope that the revised manuscript now meets the high standard of Nature Communications.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

This manuscript explores the issue related to long-term changes in coastal seawater quality due to climate change. The authors used an 8-year dataset to make predictions about the changes in water quality in the coastal area of the Guangdong Province (China) for the period 2015-2022. The authors carried out a notable effort for modeling a huge dataset which may be useful to predict long-term changes in the water quality of the investigated coastal area.

The general topic of the manuscript is undoubtedly original, interesting and useful.

However, the manuscript itself has many shortfalls, especially regarding its structure. Generally, too much information was provided (both in the main text and in the Supplementary Material, often repeating the same statements in both documents) without following a clear structure able to make the reader able to effectively understand which are the main aims of the study and the main results.

✓ **Response:** Thank you for your comments and suggestions. Following your suggestion, we have modified the paper in the following ways. First, the most important content, that is, the combination of the model, the measured data and the prediction of the future ecological risks, are retained in the Manuscript. At the same time, the causality of modeling, feature engineering, computational process, and other verifications are integrated into the Supplementary Materials (SM). We are giving the readers a new index in Figure 4 of the Materials and Methods. The SM is now composed of four parts: A. the collection and collation of measured data; B. the model dataset including source and output results; C. the establishment of receptor model including factor screening and construction; D. model verification including model comparison. The output results of source identification, meta-analysis, and machine learning which serve as auxiliary validations to reflect the reliability and

validity of the model are incorporated in the Materials and Methods as a necessary process. Noteworthy, these results are briefly referred to in the SM without directly impacting the discussion of environmental effects in this paper. In the Manuscript, we focus more on the purpose and significance of the research and the value orientation of the research findings.

The section Materials and Methods was placed before the section focused on the results. Why? In my opinion, this is confusing as well as part of the discussion about PTE behaviour in the Supplementary Material and not in the main text. The sampling strategy was also placed in the Supplementary Material and it is not clear what was actually performed since the text appears like a standard sampling protocol.

✓ **Response:** Thank you for the comment and suggestion. Notwithstanding, we're sorry to say that upon reviewing the general writing style of *Nat. Commun.* we have decided to not alter the presenting process as ours meets the requirements of *Nat. Commun.* and is suitable for the readers accustomed to reading the **2. Results and discussion** first. Specifically, the first part provides an overview of the experimental dataset. Taking into account the length constraints posed by the Materials and Methods, **Supplementary Material A** focuses on providing a detailed account of the actual sampling and analysis procedures involved in the experimental dataset. Still, we have followed your comments and seen to it that any redundancies or contradictions between the Manuscript and **Supplementary Material A** are properly addressed and rectified.

The manuscript is really too long and some statements are repeated in different sections of the manuscript and/or in the Supplementary Material. This is confusing for the reader. Actually, the Supplementary Material seems another manuscript (76 pages!!). It should be notably shortened. Some sections in the supplementary material should be summarised and moved in the full text avoiding repetition (sections A and B).

✓ **Response:** Thank you for the comments. We have reorganized the structure of the Supporting Materials (please refer to the first question for details) and refined the contents. The four Supporting Materials mainly contain descriptive and verifiable information, corresponding to the four aspects of the method and materials. Non-essential content, overlappings, redundant proofs, and confirmatory calculations are removed to ensure concise structures for the Supporting Materials for enhanced readability.

After we collated, the following is the length of each supporting material,

Supplementary Material A, 5 pages;

Supplementary Material B, 17 tables;

Supplementary Material C, 9 pages;

Supplementary Material D, 8 pages.

Generally, I suggest the authors to carefully revise the text (main text and Supplementary Material) to organise its different sections in a clearer way, avoiding repetition, selecting the discussion about PTE behaviour strictly related to the main aims/results of their study and better highlighting the most relevant results and take home messages. I suggest the authors to clearly state that the dataset employed for predictions refers to a specific coastal area,

providing a brief description of the investigated area and of the sampling strategy in the main text.

✓ **Response:** Thank you for your suggestion. We have discussed the structural adjustment strategies for the manuscripts and the Supplementary Materials according to your advice and optimized the writing structure while reducing the length. In addition, we have highlighted our research aim in the abstract to clarify its representativeness. In the research methods, we have improved the survey area and data sampling strategy. As for the sampling strategy, geographically, the dataset covers the monitoring sections of the main rivers entering the sea in Guangdong Province, PR China representing different degrees of disturbance of human activities, and temporally, samples were collected once a month, due to the significant seasonal differences in nutrients and trace metals. The land and sea location, hydrological status and economic development in the studied area have been described at the end of the Introduction. In addition, to ensure the random sampling of the dataset, the subsequent Meta-test is conducted. Through the heterogeneity test, the spatial difference and representativeness between the sections are determined, and the outliers and bias values are identified by the bias test.

The specific modifications are as follows:

Page 22 Line 513: The data selection process involved 2549 existing samples, which geographically covered the monitoring sections of the main rivers entering the sea in the province. In terms of time, due to the significant seasonal differences in nutrients and trace metals, the sampling frequency was one month.

Terminology should be also improved through the text, especially regarding the topic related to the phase partitioning of trace elements (or nutrients) between dissolved and solid fractions.

✓ **Response:** We have taken your suggestions into consideration and made the necessary changes according to your advice. In this study, our main focus is on the impact of climate change on the concentration change of dissolved trace metals and nutrients. We believe that this behavior is a result of phase transfer. The process of phase transfer is highly complex in complex systems. To better illustrate this process and incorporate the sampling process, we adopt a simplified phase partition. This means that we define two phases, dissolved and solid, using water and solid separated by a 200-mesh filter. As a result, both suspended matter and sediment are classified below the category of solid phase.

The authors often referred to “trace elements” through the text. I suggest to specify which are these trace elements in the abstract and at the beginning of the introduction.

✓ **Response:** We have uniformly modified "trace elements" to the more standardized term "trace metals". Moreover, several trace metals studied in this paper are described in detail in the abstract and introduction, and their representativeness is explained before the modeling process. It must be emphasized that the total dissolved concentration of each element is below 0.1 mg L⁻¹.

The specific modifications are as follows:

Page 2 Line 26: trace metals, such as Cd, Cr, Cu, Fe, Hg, Mn, Pb and Zn.

Page 3 Line 51: Acidification leads to changes in the phase distribution of trace metals (Cd, Cr, Cu, Fe, Hg, Mn, Pb and Zn) elements.

I am not a modelist, so I cannot provide a careful and specific revision in this sense.

I retain that this manuscript can be accepted for publication in the Journal Nature Communications following major revision. Please, consider the previous and following general comments as well as specific comments on the PDF version of the manuscript:

✓ **Response:** We are grateful for your consideration of our manuscript. In our study, we utilized both a traditional receptor model and a machine learning model to connect with a generic CMIP6 model. The results indicate that the additional changes induced by alterations in physical conditions are concentrated, implying that the regression patterns of nutrients, trace elements, and energy substances are similar. This suggests that it may be impractical to address pollution remediation separately, and changes in their proportions have a significant impact on the marine biogeochemical cycle. We appreciate your feedback and we have carefully considered your comments and made substantial revisions to our manuscript and supplementary materials.

Lines 19-29 (Abstract). I suggest the authors to better prepare the abstract clearly stating which are the aims of their research and briefly summarising their main results. Indeed, some statements in the abstract are not clearly explained. For instance, the authors stated that an 8-year dataset was employed to make predictions about changes in water quality in this offshore area... this is too general: what is this area? I think that this should be specified in the abstract. Then, the authors stated that “trace elements will be displaced by calcium and magnesium, leading to re-suspension in the water”. I do not understand what the authors meant with this sentence. They are talking about dissolved trace elements? Which elements? About trace element associated to suspended particles? About the partitioning behaviour of trace elements between dissolved and solid phases? Please, clarify.

✓ **Response:** Thanks for your suggestions. We have rewritten the unclear statements in the abstract and clarified the studied area and time range of the dataset. We specified trace elements including Cr, Cu, Fe and Pb in the abstract. "Re-suspension" was originally referred to as the process of trace elements transferring from the solid phase to the liquid phase in an acidic water environment. To avoid misunderstanding, we replace it with "dissolution".

The specific modifications are as follows:

Page 1 Line 19: As climate change affects the physicochemical properties of coastal water, the resulting element re-exposure may override the emission reductions achieved by human pollution control efforts. Combined with CMIP6 future model projections, an analysis of the water quality-climate effect over eight consecutive years from 2015 to 2022 along the South China coast was conducted. A data-driven model was utilized to predict the concentrations of trace metals and

nutrients over the next 80 years. It is proved that the acidification process carries the risk of triggering the ocean's buffering mechanisms. During this alkalinity replenishment process, trace metals, such as Cd, Cr, Cu, Fe, Hg, Mn, Pb, and Zn, in the sediment are released into the water phase, along with Ca^{2+} and Mg^{2+} . Here, we show that the nexus of re-exposure-eutrophication-emission reduction with human activities and climate feedback, cannot be ignored in the pursuit of effective environmental governance.

Line 51 What do the authors meant with “concentration lever” ?

✓ **Response:** There is a complex exchange of compounds during phase transfer. Due to various limitations of material and energy laws, such as charge conservation, the total charge of ion exchange remains consistent. To illustrate this, let's consider the simplified exchange of macro element A and trace element B. The total amount exchanged by both sides is equal. However, since the total concentration of element A in the water column is greater than that of element B, the relative change in concentration of element B is higher. Consequently, in the actual change of the water body, trace elements have a much greater influence than macro elements. We refer this phenomenon as a concentration lever.

However, we were well aware of this result from the experimental dataset, but could not capture this view in the simulation results. This is because the sensitivity of different trace metals to acidification is different, and it is affected by both surface water temperature and rainfall. Nonetheless, to address the contentious conclusions, we decided to abolish the use of "concentration lever".

Line 68 “metal elements”. Which elements? Heavy metals or potentially toxic elements? I suggest the authors to use “trace elements or potentially toxic elements” through the text.

✓ **Response:** We agree with your suggestion, which is a disunity in our expression. Here, we discuss more about the concentration effect, and the potential toxicity of metals should be discussed in the Introduction. Therefore, considering the views of different experts and our understanding of this article, we uniformly use "trace metals" throughout the text.

Lines 73-74 this affects the trace element behaviour at the sediment-water interface? Or the trace element phase partitioning between dissolved and the suspended particulate matter along the water column? This is not clear, please specify.

✓ **Response:** Thank you for your suggestions. In fact, in this case, dissolved oxygen affects the behavior of trace elements at the sediment-water interface by controlling the redox potential. The migration of metals from solid phase to aqueous phase is strongly affected by the redox process. Studies have shown that under the condition of good reduction, heavy metals will remain in the form of close binding, and a very small number of heavy metals will be released from the sediment. However, the increase and decrease of redox potential leads to the release of heavy metals ¹.

References:

- 1 Wong, J. W. C. & Yang, C. L. The effect of pH and redox potential on the release of nutrients and heavy metals from a contaminated marine sediment. *Toxicological & Environmental Chemistry*. **62**, 1-10 (1997).

Lines 76-77 What do the authors meant with “suspended elements”? elements associated to the suspended particulate matter?

✓ **Response:** Here, “suspended elements” refer to elements that are more inclined to migrate to the gas phase in their geochemical behavior. To avoid misunderstanding, we replace it with “incompatible elements” in geosciences.

The specific modifications are as follows:

Page 4 Line 95: **Incompatible elements, such as Hg, Cd, and Pb, also enter the ocean through this process.**

Line 80: Nitrogen (N) and S deposition has become one of the factors promoting coastal eutrophication?

✓ **Response:** As reported in literatures, the wet deposition of nitrate have brought abundant N to the coast. Here, you are right that the contribution of S deposition to coastal eutrophication is doubtful and can bring doubt to readers. Therefore, we delete the statement that S deposition has become one of the factors of coastal eutrophication.

The specific modifications are as follows:

Page 4 Line 98: **Nitrogen(N) deposition has become one of the factors behind the coastal eutrophication.**

Line 82: absorption or adsorption?

✓ **Response:** As in the reference study ¹, this refers to the absorption of Hg by surface water from the atmosphere. To better match the original meaning of the reference, we replace "absorption" with "uptake".

The specific modifications are as follows:

Page 4 Line 99: **Mercury's(Hg) earth system model shows that, for the modern open ocean, the dry and wet deposition flux of Hg(II) is 4600 mg yr⁻¹, the uptake of Hg(0) is 1700 mg yr⁻¹, and the confluence of rivers is 2050–5600 mg yr⁻¹.**

References:

- 1 Jiskra, M. et al. Mercury stable isotopes constrain atmospheric sources to the ocean. *Nature*. **597**, 678-682 (2021).

Line 85: “As the invading river water is anoxic”. In general? Or in the case of a specific river? Anoxia is quite uncommon in riverine environments. Please, clarify.

✓ **Response:** Thank you for your question. It is generally believed that there is widespread deoxygenation in coastal waters ¹. In particular, for this study area, the large area of seasonal hypoxia events in the Pearl River Estuary has expanded from summer to early autumn ². However, it is obvious that the increase of CO₂ in the atmosphere is the most important factor leading to ocean acidification, so here we delete the contribution of river input to ocean acidification.

References:

- 1 Zhang, W., Han, S., Zhang, D. & Shan, B. Recovery in Dissolved Oxygen Levels in Chinese Freshwater Ecosystems in the Past Three Decades. *Acs Es&T Water*. 2, 967-974 (2022).
- 2 Hu, J., Zhang, Z., Wang, B. & Huang, J. Long-term spatiotemporal variations in and expansion of low-oxygen conditions in the Pearl River estuary: a study synthesizing observations during 1976–2017. *Biogeosciences*. 18, 5247-5264 (2021).

Lines 97-99 To which elements the authors are referring to? It is not clear.

✓ **Response:** Thank you for your question. Here “three elements” means “Surface water temperature, atmospheric deposition, and pH”. As this expression may cause misunderstanding, we change it to “three factors (surface water temperature, atmospheric precipitation, and pH)”.

The specific modifications are as follows:

Page 5 Line 116: the above three factors (surface water temperature, atmospheric deposition, and pH) not only affect the exchange of elements but also respond to climate change.

Line 191 or declining trend, as predicted by the environmental Kuznets curve. Please, provide a brief explanation.

✓ **Response:** The environmental Kuznets curve may not be a very common concept, so we explained the meaning of the environmental Kuznets curve more clearly in the Introduction.

The specific modifications are as follows:

Page 7 Line 185: This means that metal consumption is undergoing a transition from a sharp increase to a stable trend.

Page 9 Line 229: that is, the concentration of pollutants in the region has reached its maximum value.

Lines 227-228 what the authors meant with “the initial field”? please, clarify.

✓ **Response:** "The initial field" is a more commonly used concept in the field of model construction. It refers to the initial value or state assumed at the beginning of the model construction. In the receptor model, each sub-region has a geological background value, which is determined as the initial field that can be used for the construction of the model and subsequent analysis and prediction.

In order for the readers to better understand this concept, we replace it with the more commonly used "the initial value of the forecast model".

The specific modifications are as follows:

Page 10 Line 269: Each of the three subareas has a geochemical background value, which is identified as the initial value of the forecast model.

Line 262 in this section the authors often refers to sensitive elements. I do not completely understand which are these sensitive elements. Please, clarify.

✓ **Response:** Thank you for your question. Here "sensitive elements" refer to the elements (dependent variables) that are highly responsive to the three factors (i.e., surface water temperature, atmospheric deposition and pH) (they are independent variables). Sensitivity analysis is used between the dependent variables and independent variables, and the sensitivity coefficient higher than 0.25 is identified as sensitive elements. For example, dependent variables such as BOD and COD are sensitive elements of SST. In order to improve the readability of the passage, we add the judgment basis and interpretation of sensitive elements, and improve the language expression.

The specific modifications are as follows:

Page 11 Line 292: In the sensitivity analysis, the elements with sensitivity coefficient greater than 0.25 are identified as sensitive elements, and they show sensitive response behavior to specific variables.

Page 12 Line 311: Direct human emissions have such an impact that different emission sources lead to high-throughput exposure to specific elements. However, the influence of the change of physical conditions of the water body is different, and the sensitive response of elements to specific factors leads to the overall change of the group of elements with similar chemical properties..

Lines 335-336 Dissolved Ca and Mg? I do not understand what do the authors meant with “will be replaced into trace elements”? In the water fraction or in the SPM? Please, clarify.

✓ **Response:** Thank you for the question. Calcium (Ca) and magnesium (Mg) belong to a group of elements known as compatible elements. They possess the characteristics of having a small radius and high charge when compared to the other two macroions, sodium (Na) and potassium (K). When more carbon dioxide (CO₂) is dissolved in water, Ca and Mg will dissolve in order to buffer the increase in H⁺ ions resulting from acidification. It is important to note that this dissolution buffering process is not limited to Ca and Mg alone, but also applies to other compatible elements studied, such as iron (Fe), manganese (Mn), chromium (Cr), copper (Cu), zinc (Zn), and so on. Therefore, here we refer to dissolution in the water body, not the migration from sediment to suspended matter.

The specific modifications are as follows:

Page 17 Line 393: The process of ocean acidification depletes the alkalinity of the water column and triggers the buffering mechanism of the ocean. This leads to the exchange of seawater with minerals on the seafloor, which regulates acidity and alkalinity. Acid dissolution results in a significant release of Ca²⁺ and Mg²⁺ from the sedimentary phase into the water. However, Ca²⁺ and

Mg²⁺ are compatible elements with small radii and low water solubility. Their substitution also occurs in other ions of elements with similar chemical properties, such as Fe, Mg, Cr, Cu, Zn, and so on.

Lines 346-348 not clear. In which environment and under what boundary conditions? The interaction between trace elements and sulfides is affected by several factors. Trace element precipitation along with sulfides usually occurs under specific boundary conditions (sulfate reduction, anoxia).

✓ **Response:** Thank you for your question. We were planning to explain to the readers that sulfides have the ability to regulate the REDOX potential, which in turn helps to mitigate the impact of future environmental changes in water ¹. This, however, adds uncertainty to the prediction of trace metal element behavior. Fortunately, after reviewing the dataset, we found that there was no significant negative relationship between sulfides and trace metals ($p > 0.05$). This suggests that our study, due to its limited duration, was unable to uncover the potential limiting effect of sulfur on metal dissolution. As a result, the limitations of the study can be clearly stated in the article.

References:

1 Scholz, F., Mcmanus, J., Mix, A. C., Hensen, C. & Schneider, R. R. The impact of ocean deoxygenation on iron release from continental margin sediments. *Nat. Geosci.* 7, 433-437 (2014).

Line 350 deposited where? Adsorbed on the surface of the SPM and accumulated in the sediment? Please, clarify.

✓ **Response:** We believe that predicting the specific direction of pollutants in different phases is a crucial issue. However, this study focuses on the exposure of pollutants in the water phase under the conditions of climate change. It examines the processes of dissolution and precipitation of trace elements and highlights the importance of understanding the trends that occur after separation from the water phase. This issue requires further attention in future research.

Lines 353-375 in this section the authors referred to the seasonal cycle of trace elements mainly from a biological point of view. However, the biogeochemical cycle of trace elements is also affected by other factors. Moreover, it is not clear if the authors referred to dissolved trace elements, trace elements associated with the suspended particles or both.

✓ **Response:** Thank you for your question. We mainly discuss the seasonal cycle of trace elements in two aspects, that is, we examine the essential elements from the biological point of view, and supplement the non-essential elements from the rainfall process. As mentioned above, we focus on the trace metals dissolved in the aqueous phase, so here we discuss the dissolved trace elements and the behavior of trace metals on suspended particles is not involved in this paper. It must be emphasized that the concentrations of these trace elements are below 0.1 mg L⁻¹.

Line 416 This section “Materials and Methods” should be placed before the discussion of the results. The same for the section “the establishment and accuracy of the model”.

✓ **Response:** Thank you for your suggestion, but honestly, we re-examined the general style of *Nat. Commun.* writing, and the results can be placed before the methods section, so our writing meets the requirements of *Nat. Commun.* And the "The establishment and accuracy of the model" section describes the method of building the model, which is part of the "Results and discussion".

SA. 1 The sampling description in supplementary material may be notably improved. In my opinion, it sounds like a standard sampling protocol since the authors often referred to operations that should be done for sampling (the ship should be selected, samplers should collect, it should be carried out). The authors should provide a description of the sampling operations which were actually performed and not simply list the operations that should be done for sampling. Moreover, in the section focused on special samples, the sampling strategy for petroleum samples was described although any data about this samples was reported in the main text. The authors also referred to “special storage containers” and “preservatives” for different analytes. Please, specify which containers and preservatives were used. Which analytical technique was employed for PTE determination? What about their LOD?

✓ **Response:** Thanks for your suggestion. We have rewritten **Supplementary Material A** to highlight the description of the actual sampling and analysis performed instead of the sampling instructions. And the content of oil sample sampling was removed, because the relevant data was not used in this study. According to your advice, we describe in detail the agents and analytical instruments used in the sampling, and for the analysis methods and LD of the metal samples, we give a detailed description of each metal.

SB. 1 This section is focused on “Coordinates and classification of sampling sites”. However, the behaviour of trace elements is discussed. Why the authors decided to place this text in the Supplementary Material and not in the main text?

✓ **Response:** Thank you for your question. The main purpose of the content described in the **Supplementary Material B** is to reduce the dimension of the data set through principal component analysis, extract the key factors in the process of water quality change under the influence of climate change, and construct the receptor model as independent variables. Since this step is the preparatory work for building a model, only its conclusion is needed, and placing it in the main text will take up a lot of space, so we decided to transfer the relevant content to **Supplementary Material C** based on the core purpose of the attachment and the requirement of reducing the space. Only the results highly related to the dimensionality reduction of the dataset are retained in the attachment.

Reviewer #2 (Remarks to the Author):

Extremely interesting manuscript, bringing valuable information to climate change metal and metalloid dynamics alterations. Minor revisions only.

Please alter "trace element" to "metals and metalloids" according to IUPAC recommendations throughout.

✓ **Response:** Thank you for your suggestion, but since the discussion of metal-like behavior is not involved in this study, and our work is focused on concentration change, and the explanation of concentration conditions is an essential task, we have changed "trace element" to "trace metals".

Line 39 - Regarding "Acidification leads to changes in the phase distribution of trace elements, affects the functions of electron donors involved in cell respiration, and disrupts the promotion of enzymatic reactions (Richir, 2016)." - You must also indicate that acidification (pH) also alters metal and metalloid toxicity and bioavailability, due to altered redox state.

✓ **Response:** Thank you for your suggestion. We highly agree upon the important impact of ocean acidification on biological physiological and biochemical processes by changing the valence of trace metals. Therefore, we supplement the relevant discussion here.

The specific amendments are as follows:

Page 3 Line 53: In addition, the increase in H^+ concentrations caused by ocean acidification alters the chemical form of metals, thus affecting their toxicity and bioavailability in seawater.

Lines 46-54 require references.

✓ **Response:** Thank you for your suggestion, and we have supplemented the corresponding references.

Line 81 - Remove JW from the Erisman reference.

✓ **Response:** Thank you for your suggestion and we have revised it accordingly.

Line 159 - Indicate why Cr(VI) and not Cr total, due to the sampling station particularity. No other elements are analyzed per chemical species?

✓ **Response:** We admit that because of the high toxicity of Cr(VI), we selected this highly representative chemical form for testing. We have described in detail the analytical forms of each element in the **Supplementary material A**.

The specific amendments are as follows:

Supplementary Material A Page 4 Line 96: Regarding the valence states tested, each valence state was measured for all metals except Cr, only VI valence of which was recorded.

Line 166 - What are HMs? Heavy metals? Remove, replace by metals and metalloids.

✓ **Response:** HMs refers to heavy metals. In order to regulate the use of terminology, we have revised HMs to "trace metals".

Better explain the meaning of environmental Kuznets curve for metal elements.

✓ **Response:** Thanks for your suggestion. The environmental Kuznets curve may not be a very common concept, so we have explained the meaning of the environmental Kuznets curve more explicitly in the Introduction, and provided a short explanation in section 2.1.

The specific modifications are as follows:

Page 7 Line 185: This means that metal consumption is undergoing a transition from a sharp increase to a stable trend.

Page 9 Line 229: that is, the concentration of pollutants in the region has reached its maximum value.

Line 169 - You cite e-waste but do not indicate Rare Earth Element contamination in your assessment.

✓ **Response:** Thank you for your suggestion. E-waste does bring rare earth element pollution, but the trace metals we discussed in this article do not involve rare earth elements, so we have excluded the relevant content of e-waste pollution.

Table 1- Define GB3838 - WHO, CMC.

✓ **Response:** Thank you for your suggestion. The four water quality standards cited in the table are explained below:

¹ GB3838-2002 limit value(III) (SEPA, 2002): the National Environmental Quality Standards for Surface Water (GB3838-2002) of PR China;

² WHO (2004) (World and WHO., 2004): the World Health Organization (WHO) Guidelines for Drinking-water Quality;

³ CMC, acute (EPA, 2009): The criteria maximum concentration (acute) in the national recommended water quality criteria – aquatic life criteria table;

⁴ CMC, chronic (EPA, 2009): The criteria maximum concentration (chronic) in the national recommended water quality criteria - aquatic life criteria table.

However, considering that the contents of the table are not closely related to the main conclusions of this study, we delete the table from the manuscript to simplify the structure.

I missed a discussion on altering water currents due to climate change as drivers for metal and metalloid contamination.

✓ **Response:** Thank you for your comment. We acknowledge that the change in water flow brought about by climate change is one of the drivers of metal pollution. However, in this study, we focus on the effects of physical and chemical properties of water bodies and microbial activities on the

behavior of trace metals under climate change. And the hydrological factors are two aspects of the effect of climate change on trace metals. It would be lengthy to discuss the role of both at the same time. In this regard, we have acknowledged in the last paragraph of the section "Results and discussions" the possible shortcomings caused by ignoring hydrological movements, and look forward to the follow-up research.

The specific modifications are as follows:

Page 21 Line 482: **In particular, the effect of hydrological changes caused by climate change on the behavior of trace metals has not been taken into account in our model.**

Line 217 - Remove fortunately, indicative of opinion.

✓ **Response:** Thank you for your suggestion. We have modified it accordingly.

Line 246 - Change 3 categories to three categories.

✓ **Response:** Thank you for your suggestion. We have modified it accordingly.

Line 251 - Cd, Hg, and Pb do not originate mainly from sulfur deposition, correct.

✓ **Response:** Thank you for your comment. This place should be wet deposition rather than S deposition, which is an important source of offshore Cd, Hg and Pb. For example, the atmospheric fluxes of Cd, Cu, Pb and Zn in the Bay of Biscay are of the same order of magnitude as those of river inputs ¹; in the Southeast Atlantic, the contribution of wet deposition to total lead flux can reach 92–99 % ². Therefore, we modify this part accordingly.

The specific modifications are as follows:

Page 12 Line 301: **As for elements sensitive to deposition, namely Cd, Hg, and Pb, wet deposition is an important way for them to enter the offshore area.**

References:

- 1 Maneux, E. et al. Temporal Patterns of the Wet Deposition of Zn, Cu, Ni, Cd and Pb: The Arcachon Lagoon (France). *Water, Air, and Soil Pollution*. **114**, 95-120 (1999).
- 2 Chance, R., Jickells, T. D. & Baker, A. R. Atmospheric trace metal concentrations, solubility and deposition fluxes in remote marine air over the south-east Atlantic. *Mar. Chem.* **177**, 45-56 (2015).

Line 228-240: " Based on our estimates, the dissolved phase concentrations of Fe, Cu, Pb, and Cr in 2099 will increase by 63.4±7.2 %, 63.1±5.7 %, 61.9±7.0 %, and 48.2±8.2 %, respectively, compared to 2020" Why only cite these three elements? What of the rest?

✓ **Response:** Thank you for reminding us of that. We have added the predicted results for other trace elements (i.e., Cd, Hg, Mn, and Zn) in the manuscript and have updated Figure 3 to ensure that the predicted results for all trace elements discussed in this paper are presented.

The specific modifications are as follows:

Page 17 Line 408: Based on our estimates, the dissolved phase concentrations of Cd, Cr, Cu, Fe, Hg, Mn, Pb, and Zn in 2099 will increase by 79.7%, 48.4%, 63.0%, 63.3%, 68.4%, 104.5%, 61.8%, and 83.2%, respectively, compared to 2020.

Line 345 - Define ORP.

✓ **Response:** Thank you for your suggestion. ORP refers to Oxidation-Reduction Potential, and we have given the full spelling of the abbreviation where ORP first appears.

The specific modifications are as follows:

Page 17 Line 416: Previous studies have suggested that sulfides may interfere with the redissolution of trace metals in low Oxidation-Reduction Potential (ORP) water in the future.

Line - 364 - explain the term "non-biophilic". Not very uptake by living organisms? This is not very true for Cr. Not very bioavailable? Not true for both elements.

✓ **Response:** Thank you for your question. Non-biophilic elements refer to elements that may have toxic or potential adverse effects in organisms. As the word is easy to be misunderstood by readers, we replace "biophilic element" and "non-biophilic element" with "essential element" and "non-essential element" respectively.

The specific modifications are as follows:

Page 14 Line 437: The essential elements, such as Fe and Cu, are primarily complexed with a higher level of COD during winter.

Page 14 Line 442: non-essential elements like Cr and Pb are influenced by deposition factors.

Line 411 - remove the capital from Marine organisms.

✓ **Response:** Thank you for your suggestion. We have modified it accordingly.

Line 411-415 - you neglected to indicate that marine organisms will also be exposed to higher metal and metalloid concentrations in some cases, leading to altered metabolism and physiology.

✓ **Response:** Thank you for your suggestion. We have made corresponding changes.

The specific modifications are as follows:

Page 17 Line 493: It is expected that marine organisms will have to work in an environment with high electron donors, low carbon sources, and high nutrient load, with their growth to be impacted by overheated sea temperature, and the ecological value and sink function of the ocean will be irreversibly weakened, while the rising concentration of trace metals increases the uncertainty of this process.

Line 426 - You state you used Cr(VI), this becomes a problem as you have no speciation for other elements you analyzed. I recognize this limitation due to the samplings of the monitoring stations. However, you must at least acknowledge this and discuss speciation in your manuscript with regard to the other analyzed elements, as this directly affects metal and metalloid bioavailability. You have done so in lines 81-85 for Hg, although you neglected to indicate methylation processes as one of the drivers for Hg cycling.

✓ **Response:** We believe that your tips deserve high attentions from us, and all the indicators included in the discussion are equally important and interesting. We should discuss the effects of each indicator in the context of temperature, rainfall, and pH on pollution indicators. This confirms that the scope of our study is limited and that these factors affect more pollutants. In the following examples, we should choose elements that are sensitive to pH response as representatives for prediction. Although chromium is a compatible element, it can exist in various valence states in natural water body. Therefore, we believe that the results may not be representative of other water body. Mercury ion has a large radius, a low melting /boiling point, and is easily released into the atmosphere. Methyl mercury also has the characteristic of sedimentation. Therefore, the process of water acidification does not significantly influence it.

Line 441 - Data points lower than the limit of detection (LOD) were replaced with half of the LOD - insert reference on this approach, and report the LOD for each element. Is there more than one LD reported?

✓ **Response:** Thanks for your advice. We have cited references for this data processing method and added an LD for each element in the **Supplementary Material A**, with only one LD for each element.

The specific modifications are as follows:

Supplementary Material A Page 4 Line 99: The detection limit was 0.06 µg/L for Cd, 0.7 µg/L for Cr, 0.8 µg/L for Cu, 0.05 µg/L for Hg, 0.01 mg/L for Mn, 0.03 mg/L for Fe, 0.09 µg/L for Pb, and 0.5 µg/L for Zn.

Review Comments

Reviewer #1 (Remarks to the Author):

This manuscript explores the issue related to long-term changes in coastal seawater quality due to climate change. The authors used a huge dataset to make predictions about the changes in water quality in the coastal area of the Guangdong Province (China) for the period 2015-2022. The authors carried out a notable effort for modeling this dataset which may be useful to predict long-term changes in the water quality of the investigated coastal area. The general topic of the manuscript is undoubtedly original, interesting and useful.

The authors adequately responded to most of my comments and updated the text of the manuscript accordingly. In the revised version of the manuscript, the authors made a great effort to better prepare the main text and to better organise the Supplementary Material (SM). The SM is now reorganized in 4 sections and, in my opinion, more better prepared. In addition, the authors provided a better and more detailed explanation of the sampling strategy.

[Thank the reviewers for affirming our first round of revision work.](#)

Please, excuse me for suggesting to place Materials and Methods before the section dedicated to the Results. Actually, I have not checked the general writing style of the Journal. This is my fault.

I retain that the manuscript may be accepted in the Journal Nature Communications, just following some minor revisions.

Please, consider the following minor general comments.

Regarding terminology, the authors modified “trace elements” with “trace metals”. Between the two, I suggest the authors to use the first, so “trace elements”. With my comment on the original version of the manuscript, I simply meant to specify which elements were taken into account (so Cd, Cr, Cu, Fe, Hg, Mn, Pb and Zn). Just name them in the text.

[We still believe that the opinions you raised in the first round have the necessity of modification.](#)

[Although earlier published articles have been reviewed, the term "trace metals" is not commonly used. However, since the research subjects of this article encompass both metals and metalloids, with a focus on changes in their water solubility, it may be more appropriate to use the term "metal" in our descriptions. We plan to introduce a specific abbreviation in the article: "TMs \(trace metals and metalloid elements\) " to enhance the clarity of our expression.](#)

I suggest the authors to avoid the use of “we” such as “we show” (line 28 in the abstract). What about: “The aim of this study is to show the nexus of re-exposure-eutrophication-emission reduction with human activities and climate feedback, cannot be ignored in the pursuit of effective environmental governanc”? or something like that.

[We will review the terms in the article and modify the unreasonable use of the word "we", including the following parts.](#)

Line 95 I am not sure about “Incompatible elements”. Could you just write: “elements such as Hg, Cd and Pb also enter....?”

[Incompatible elements are geological term. It describes the partitioning characteristics of trace elements between the solid phase and the melt phase during magma formation \(partial melting\) or](#)

crystallization. Therefore, according to the tendency of elements to preferentially enter the solid or melt phase under phase equilibrium conditions, that is, the magnitude of the D value, trace elements are classified as compatible elements and incompatible elements. The D value of incompatible elements is < 1 , and they tend to be enriched in silicate melts during the mineral crystallization process. Due to their low concentration, they cannot form independent mineral phases. Due to their ionic radius, charge and chemical bond limitations, it is difficult for them to enter the crystal structure of rock-forming minerals, and they are relatively enriched in residual magma or hydrothermal fluids. Their solid-liquid phase distribution coefficient is close to zero.

If the sentence you pointed out needs to be re-clarified, a concise and commonly used classification term is required to describe metals like Hg and Cd. Based on your suggestion, we add "elements with larger radii" to describe the characteristics of these elements.

Line 99 the term “uptake” usually refers to Hg bioaccumulation along the trophic chain. Please, consider the idea to write “the influx of Hg(0) at the water-atmosphere interface is...”. Indeed, the term “influx” referred to a flux of Hg(0) from the air to the water and it is more appropriate with respect to “uptake”.

Thank you for your suggestion. We will correct it.

Line 121 physico-chemical properties. Check also through the text

We have checked the above expression errors throughout the full text. There are a total of four such errors and they have been changed to "physico-chemical properties". Respectively on line 96, 122, 189 and 192.

Reviewer #2 (Remarks to the Author):

The authors have responded adequately to both my initial comments and the first reviewer's assessments. I thus endorse this paper for publication as is.

Thank the reviewers for giving affirmative opinions on the first round of manuscript revision.

Reviewer #3 (Remarks to the Author):

This manuscript presents an 8-year long data set of trace metal measurements in the coastal area of the Guangdong Province (China). Based on this data set they create a receptor model and subsequently apply this to a future simulation made by a model in the CMIP6 archive. The data set they present is quite unique and a significant contribution in itself. The PCA analysis and receptor model also provide novel and interesting insight into trace metal dynamics in coastal systems under climate change. There is not much current literature on this topic so this work presents a valuable and original piece of work. The quality of the data appear very good and the methods are described in detail. The overall analysis and analytical approach are both robust and comprehensive. With the exception of some strange, to me at least, use of terminology and a general lack of definitions for the abbreviations used the paper is well written.

Thank you for your recognition and support of this study.

However, despite having now read the manuscript, and supplemental information, several times I struggle to identify what the key result is. Nature Communications publishes work that “aim to represent important advances in all areas of the biological, health, physical, chemical, Earth, social,

mathematical, applied, and engineering sciences”. It is unfortunately not at all clear to me what important advance this work makes. Partly this is because the authors also have not written any conclusions, but the noteworthy results are also not apparent in the “results and discussion” section. I believe there is noteworthy insight in the presented data and would urge the authors to restructure and revise their manuscript to focus on the key findings and their impact. You have a story in here – you just need to tell it.

This article aims to describe the pollution exposure caused by acidification of the coastal environment through measured data and model data, which belongs to the progress in the field of geochemistry. The core finding of our research work is that it reveals that the enhanced exposure of heavy metal elements in coastal areas caused by climate change will affect the ecological health of the water environment on the earth's surface.

At the end of the article, we restate the conclusion and significance of this paper.

However, we are able to analyze and foresee the significant changes in element cycles brought about by physical environmental changes within a regional dataset. In the context of hypoxia and acidification, the changes in the valence bonds and ion binding modes of trace metals (TMs) will lead to irreversible changes in their phase partition coefficients. The nitrification reaction caused by eutrophication consumes the alkalinity of the water body and enhances the above effect. The increase in the exposure of metal elements in the nearshore aqueous phase is collective and irreversible. Marine organisms may have to survive in an environment with high electron donors, low carbon sources, and high nutrient loads. Their growth will be affected by overheated sea temperatures, and the ecological value and sink capacity of the ocean will be irreversibly weakened.

Like the previous reviewer #1 I would really like the materials and methods to be presented before the results and discussion. It is not easy to understand the presented results before first reading what has been done. I’ve read your responses to the previous reviewer and I understand your argumentation, but this structure is making it challenging for the reader to understand your work.

Placing the Materials and Methods section at the end is a common format for Nature Communications. This study further optimizes the logical structure of the article based on this framework.

Specific comments:

Line 51: It is unclear what “carbonate atmospheres” means.

The nearshore environment is affected by acidification. The term "acidification atmosphere" should be used.

Line 52-53: The sentence “trace metals may become sensitive factors in response to changes in marine water quality” is unclear. What responds to what?

Trace metal exposure is one of the manifestations of water quality changes. Therefore, it should be changed to potential risk here.

Line 56: I think “butterfly effect” is a poor choice of words here. Consider revising.

Here, we intend to describe the environmental effects of climate change on the offshore area from the perspective of systems science. Therefore, we consider using "critical" or "irreversible" to elaborate the intention of this research.

Line 65: Replace “tracers” by “influencing factors” to avoid confusion.

The modification has been made. Thank you for your correction.

Line 67: Add “organic” before “C, N, P, and S elements”

The participation of the processes of bio-utilization, release, circulation, and remineralization involves the transformation between organic matter and inorganic matter. Pointing out the organic part alone may lack evidence.

Line 74: It is unclear what “incompatible elements” are. I realize this term is in response to a previous review, but it is not made clear what the elements are incompatible with so the sentence is unclear.

Incompatible elements are a classification of elements in geoscience. Without any explanation, it might cause confusion for readers from other academic backgrounds. We will further explain this in the following sentences.

Incompatible elements are geological term. It describes the partitioning characteristics of trace elements between the solid phase and the melt phase during magma formation (partial melting) or crystallization. Therefore, according to the tendency of elements to preferentially enter the solid or melt phase under phase equilibrium conditions, that is, the magnitude of the D value, trace elements are classified as compatible elements and incompatible elements. The D value of incompatible elements is < 1 , and they tend to be enriched in silicate melts during the mineral crystallization process. Due to their low concentration, they cannot form independent mineral phases. Due to their ionic radius, charge and chemical bond limitations, it is difficult for them to enter the crystal structure of rock-forming minerals, and they are relatively enriched in residual magma or hydrothermal fluids. Their solid-liquid phase distribution coefficient is close to zero.

Line 74-94: Beginning with “Incompatible elements” this should all be a separate paragraph. The paragraph also needs a proper topic sentence and better flow between the sentences. It now reads like a list of bullet points someone have put together but not linked. There is no introduction, no coherency.

We have reorganized the paragraphs. The discussion ideas are the three major factors affecting element distribution: SST, sedimentation, and pH. Each is summarized into a separate paragraph.

Line 77 and 78: why are you defining nitrogen and mercury here? The abbreviation for both have been used previously, and you do not define the other metals and elements.

Thank you for your reminder. We have deleted the explanatory information in this sentence.

Line 78: what is “Mercury’s(Hg) earth system model”? Please explain.

The expression here is incorrect. It should be "Accounting of mercury's cross-media flux."

Line 85: replace “limits” with “limitation”

We have made the revisions at the corresponding places of the manuscript. There are two modifications in this paper.

Line 88-89: I do not understand this sentence. Nitrogen removal and denitrification does happen in

situ in the ocean. And even if it did not I do not follow why that would mean that “nutrients and trace metals are affected by thermohaline circulation”. I’m not questioning that both nutrients and trace metals are affected by the thermohaline circulation, I just do not follow the logic of the sentence and cannot understand what you are trying to communicate.

There is an error in the expression here. It should be changed to “meaning that nutrients and TMs will further affect the open sea through ocean currents.”

Line 114-115: Please reiterate here what these three factors are.

We have listed it clearly here. Rewritten as: To accurately model these fluctuations, we intend to establish a ternary receptor model with background values. This model will consider the influence of three factors, including sea surface temperature, dry and wet deposition amount and pH, on the migration tendencies of different elements.

Line 118: It is unclear what you mean by “CMIP6 framework” here. You have, as I understand it, used output from one of the models contributing to CMIP6. If that is what you want to communicate here please state that, otherwise explain what the framework is.

This paper is inaccurate in this aspect. This paper uses the model data set of three variables in CMIP6 and the prediction data set of the climate change model SSP3-7.0. We will make the modification and provide guidance, with a link to the relevant introduction in the Materials and Methods section.

Lines 128-129: The warming process cannot be controlled anywhere at the moment so the meaning of this final statement is unclear. Please revise.

In order to avoid ambiguity, the sentence is changed to the following:

This is particularly relevant to the cases in which the warming process cannot be effectively controlled or reversed at present.

Lines 153-154: Could you provide a reference for the environmental Kuznets curve?

We have added the citation in the next sentence. Both sentences are from the 11th cited literature. Therefore, citations should be made once after each of these two sentences.

Line 166: TN is not defined

The full names and abbreviations are added in the text.

Line 171: This sentence is unclear. Please revise for clarity.

It has been modified to: Rivers play a notable role in transporting nutrients, with freshwater culture contributing a staggering 20% (N) and 53% (P) of compounds.

Lines 246-247: I do not know what “energy substances” are

Carbon-containing organic compounds show an energy-supplying role in the environment and are usually called Energy substances.

Line 314: it is not clear what “compatible elements” are. See also my comment above regarding “incompatible elements”

Compatible elements belong to the geological concept and are corresponding to incompatible

elements.

The same metal group will have a more similar fate pattern, which is necessary in discussing the metal fate process. Since no more commonly used alternative conceptual description has been found, we tend to add corresponding explanations later.

Line 314: it is not clear what “compatible elements” are. See also my comment above regarding “incompatible elements”

Compatible elements belong to the geological concept and are corresponding to incompatible elements. Corresponding to the sentence with "incompatible" that appeared in the previous text, delete "Compatible" in this sentence.

Line 322-323: This sentence is unclear. Please revise.

“The outcome depends on elements sensitive to pH (Figure 1-b)” This sentence refers to calculating the sensitivity of the concentration of metal elements to the pH variable based on the sensitivity calculation (3.4.2).

The original text is revised as follows: The outcome depends on the sensitivity of component concentration to pH (Figure 1-b). The sensitivity calculation adopts the calculation method in 3.4.2 of Materials and Methods.

Line 331: The terminology “concentration levers” was commented on in the previous review and I find that an explanation is still lacking. Please add some detail.

The previous review was like this:

There is a complex exchange of compounds during the phase transfer process. Due to various limitations of material and energy laws, such as charge conservation, the total charge of ion exchange remains consistent. To illustrate this, let's consider the simplified exchange of macro element A and trace element B. The total amount exchanged by both sides is equal. However, since the total concentration of element A in the water column is greater than that of element B, the relative change in concentration of element B is higher. Consequently, in the actual change of the water body, trace elements have a much greater influence than macro elements. We refer to this phenomenon as a concentration lever.

We saw this result from the experimental data set. The reason is that different trace metals have different sensitivities to acidification. We added a brief supplement at the end of the sentence: That is, when alkalinity is released from sediments to the aqueous phase, TMs with a lower water/sediment partition coefficient have a higher migration probability.

Line 352: Do you mean “decreases”?

There are two "decrease" in this paragraph. According to the meaning, it would be more reasonable to use "amplitude reduction".

Lines 354-357: Why do you see such a dip and recovery pattern for Pb and Cr? Such a pattern is not seen in the other trace metals. A bit of additional discussion on this would have been nice.

The downward trend of Pb and Cr may be caused by changes in emissions in Guangdong Province (the study area). The total concentration of Cr reached its peak from 2018 to 2019. The combustion of coal and oil is one of the main sources. In recent years, due to the increase or upgrade of air

pollution control devices for combustion sources, the capture of HMs is more effective ^[1]. In addition, the removal rate of Pb in the coal industry has been improved, resulting in a relatively decreased atmospheric exposure of Pb. With China's increasingly strict supervision of e-waste treatment activities, the urban agglomeration in the eastern wing of the coastal economic belt, as the region with the most intensive e-waste recycling activities in China, shows a year-on-year downward trend in the pollution degree of various heavy metals ^[2,3]. Therefore, the industrial structure is one of the important factors affecting the migration of heavy metal elements.

Lines 366: The decrease in C/N ratio does not appear to be significant given the uncertainty range. That should be noted.

In Figure 3b, each node of C/N is divided by year, the data size is represented by the annual average value, and the error bars are represented by the standard deviation of each month within the year. The large range of error bars is caused by the different nitrogen leaching effects in the rainy and dry seasons. The large seasonal difference may mask the long-term change. We conducted a paired sample t-test for the data set every 10 years respectively, and the results are shown in **Table 1**. The annual variations of each month are as shown in **Figure 1**.

Table 1 The paired factor t-test results of two sets of data from 2020 to 2029 and 2090 to 2099 in the C/N ratio (predicted data set).

Paired sample statistics								
		avg	data volume	std	Mean of standard error			
Pairing	2020-2029	5.16	120	0.12	0.04			
	2090-2099	5.08	120	0.09	0.03			

Correlation of paired samples				
		data volume	Correlation	significance
Pairing	2020-2029 & 2090-2099	120	0.600	0.039

Paired samples test								
Pairing difference								
				95% confidence interval				Sig. (2-tailed)
		avg	std	mean	lower limit	upper limit	t	
Pairing	2020-2029 & 2090-2099	0.08	0.10	0.03	0.01	0.14	2.72	0.02

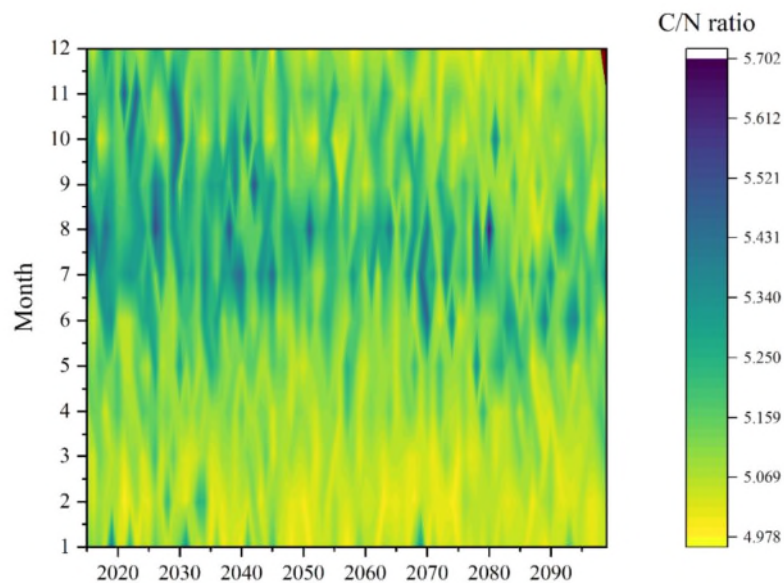


Figure 1 The paired factor t-test results of two sets of data from 2020 to 2029 and 2090 to 2099 in the C/N ratio (predicted data set).

From the results in Table 1, although there is a seasonal error, we use the paired factor t-test to perform paired comparisons at various time nodes within the year. The statistical results show ($\text{sig} < 0.05$), proving that the conclusion of the decline is significant. In addition, the results in Figure 1 show the change pattern of 1020 time nodes and are compared according to the division of year and month. For the C/N ratio of the data set (including measured and predicted parts), the horizontal coordinate represents the change of years, and the vertical coordinate represents the change of seasons. On the one hand, there is a higher C/N ratio in the rainy season. On the other hand, after considering the seasonal changes, the proportion of the dark color gradation of the C/N ratio is reduced.

Line 367: Please explain what the “carbon sources” are. Same for line 395.

Carbon source is used to form carbon in microbial cells and metabolites. It is an important substance in water treatment engineering and also a necessary substance for the natural purification of rivers. In this paper, the carbon source appears three times, at lines 256, 377 and 405 respectively. In order to enable readers to clearly understand its meaning, we add an explanation at the first place.

Line 375: add “coastal” before “ocean’s”

As mentioned in the text. Thank you for your reminder.

Lines 394-398: This is interesting, but could use some elaboration. If properly phrased and referenced I believe this could be an impactful conclusion. But it is unclear whether it is a true result of the work, or just supposition.

This is a supposition. The expression here is not appropriate enough. It should be rewritten in the form of a supposition. Change “expected” to “possible”.

Line 409: replace “which were” by “and”

We have made modifications.

Line 451: “... among variables ...” What variables are these?

What is introduced here is a general method. Before performing PCA, the attributes of the components are undetermined and a posteriori. After extracting the principal components, one or two variables are used as tracer variables according to the load presented by all variables participating in the analysis to identify the label of the component. In order to express this clearly here, we have decided to expand this paragraph.

Section 3.2:

Here it is apparent that the authors are not very familiar with CMIP6. That is perfectly fine. I’m suggesting some changes to the text to make it more “standard” and thus more easily understood.

Line 430 onwards: You could rewrite as “Here we use ScenarioMIP simulations from the Earth System model GISS-E2. The model has a spatial resolution of 2 degrees latitude by 2.5 degrees longitude, and we use the monthly output from the “r1i1p1f1” ensemble member.”

Thank you for your suggestion. We will revise according to the above opinion.

I am wondering here why you have used this particular model. Many of the models in CMIP6 have a higher spatial resolution (at least 1x1 degree) which would better resolve your study area.

Lines 433-434: Please use here the exact name of these variables in the model output. This will help readers to identify and the correct data.

The advantage of CMIP6 is that it can not only provide the values of different sections of each month that match our measured data set, but also provide the prediction results up to 2099, which is highly consistent with the methods and goals of this study.

Low resolution is indeed an aspect that needs improvement in this research model. When looking for variables with the help of CMIP6, screening is first carried out according to labels such as ssp370 and r1i1p1f1. At this time, the search results show multiple resolutions. To ensure the consistency of the resolutions of the three variables, we decided to uniformly adopt a resolution of 250 km × 200 km. The output results of the data source are exported through the ncdisp instruction of Matlab. For specific content, please refer to **Appendix 1**.

You should repeat here which scenario you use even though you have mentioned this before in the text.

We will supplement the corresponding information.

Reference

- [1] Zhong Z, Li J, Ma Y, et al. The adsorption mechanism of heavy metals from coal combustion by modified kaolin: Experimental and theoretical studies. *Journal of Hazardous Materials*, 2021,418:126256.
- [2] Li W, Achal V. Environmental and health impacts due to e-waste disposal in China – A review. *Science of The Total Environment*, 2020,737:139745.
- [3] Zhang K, Schnoor J L, Zeng E Y. E-Waste Recycling: Where Does It Go from Here?. *Environmental Science & Technology*, 2012,46(20):10861-10867.

Appendix 1

Surface sea temperature

Global Attributes:

Conventions	= 'CF-1.7 CMIP-6.2'
activity_id	= 'ScenarioMIP'
branch_method	= 'standard'
branch_time_in_child	= 0
branch_time_in_parent	= 60225
contact	= 'Lijuan Li (ljli@mail.iap.ac.cn)'
creation_date	= '2019-08-18T13:08:18Z'
data_specs_version	= '01.00.31'
experiment	= 'update of RCP8.5 based on SSP5'
experiment_id	= 'ssp370'
external_variables	= 'areacella'
forcing_index	= 1
frequency	= 'mon'
further_info_url	= 'https://furtherinfo.es-doc.org/CMIP6.CAS.FGOALS-g3.ssp370.none.r1i1p1f1'
grid	= 'native atmosphere area-weighted latxlon grid (80x180 latxlon)'
grid_label	= 'gn'
history	= '2019-08-18T13:04:31Z ;rewrote data to be consistent with ScenarioMIP for variable cl found in table Amon.'
initialization_index	= 1
institution	= 'Chinese Academy of Sciences, Beijing 100029, China'
institution_id	= 'CAS'
mip_era	= 'CMIP6'
nominal_resolution	= '250 km'
parent_activity_id	= 'CMIP'
parent_experiment_id	= 'historical'
parent_mip_era	= 'CMIP6'
parent_source_id	= 'FGOALS-g3'
parent_time_units	= 'days since 1850-01-01'
parent_variant_label	= 'r1i1p1f1'
physics_index	= 1
product	= 'model-output'
realization_index	= 1
realm	= 'atmos'
run_variant	= '3rd realization'
source	= 'FGOALS-g3 (2017): aerosol: none atmos: GAMIL2 (180 x 90 longitude/latitude; 26 levels; top level 2.19hPa) atmosChem: none

land: CLM4.0
landIce: none
ocean: LICOM3.0 (LICOM3.0, tripolar primarily 1deg;
360 x 218 longitude/latitude; 30 levels; top grid cell 0-10 m)
ocnBgchem: none
seaIce: CICE4.0'

source_id = 'FGOALS-g3'
source_type = 'AOGCM'
sub_experiment = 'none'
sub_experiment_id = 'none'
table_id = 'Amon'
table_info = 'Creation Date:(24 July 2019)
MD5:3039b0071259358b3c55557c5f3d21bf'

title = 'FGOALS-g3 output prepared for CMIP6'
tracking_id = 'hdl:21.14100/d24fc1ce-8066-43ac-a858-b095d1fc82be'
variable_id = 'tasmin'
variant_label = 'r1i1p1f1'
license = 'CMIP6 model data produced by Lawrence Livermore

PCMDI is licensed under a Creative Commons Attribution ShareAlike 4.0 International License (<https://creativecommons.org/licenses>). Consult <https://pcmdi.llnl.gov/CMIP6/TermsOfUse> for terms of use governing CMIP6 output, including citation requirements and proper acknowledgment. Further information about this data, including some limitations, can be found via the further_info_url (recorded as a global attribute in this file) and at <https://pcmdi.llnl.gov/>. The data producers and data providers make no warranty, either express or implied, including, but not limited to, warranties of merchantability and fitness for a particular purpose. All liabilities arising from the supply of the information (including any liability arising in negligence) are excluded to the fullest extent permitted by law.'

cmor_version = '3.5.0'

Dimensions:

time = 60 (UNLIMITED)
lat = 80
lon = 180
bnds = 2

Variables:

time

Size: 60x1

Dimensions: time

Datatype: double

Attributes:

bounds = 'time_bnds'
units = 'days since 2015-01-01 00:00:00'
calendar = '365_day'
axis = 'T'
long_name = 'time'

```

                                standard_name = 'time'
time_bnds
    Size:          2x60
    Dimensions: bnds,time
    Datatype:      double
lat
    Size:          80x1
    Dimensions: lat
    Datatype:      double
    Attributes:
        bounds      = 'lat_bnds'
        units       = 'degrees_north'
        axis        = 'Y'
        long_name    = 'Latitude'
        standard_name = 'latitude'
lat_bnds
    Size:          2x80
    Dimensions: bnds,lat
    Datatype:      double
lon
    Size:          180x1
    Dimensions: lon
    Datatype:      double
    Attributes:
        bounds      = 'lon_bnds'
        units       = 'degrees_east'
        axis        = 'X'
        long_name    = 'Longitude'
        standard_name = 'longitude'
lon_bnds
    Size:          2x180
    Dimensions: bnds,lon
    Datatype:      double
height
    Size:          1x1
    Dimensions:
    Datatype:      double
    Attributes:
        units       = 'm'
        axis        = 'Z'
        positive     = 'up'
        long_name    = 'height'
        standard_name = 'height'
tasmin

```

Size: 180x80x60

Dimensions: lon,lat,time

Datatype: single

Attributes:

```
standard_name = 'air_temperature'
long_name      = 'Daily Minimum Near-Surface Air Temperature'
comment        = 'minimum near-surface (usually, 2 meter) air
temperature (add cell_method attribute 'time: min')'
units          = 'K'
cell_methods   = 'area: mean time: minimum (interval: 10 minutes)
within days time: mean over days'
cell_measures  = 'area: areacella'
history        = '2019-08-18T13:04:31Z altered by CMOR: Treated
scalar dimension: 'height'. 2019-08-18T13:08:18Z altered by CMOR: Converted type from 'd' to 'f.'
coordinates    = 'height'
missing_value  = 1.000000020040877e+20
_FillValue     = 1.000000020040877e+20
```

[Note] The above data set consists of 9 files, which describe the data of each time period from 2015 - 2020, 2020 - 2030..., 2090 - 2099 respectively. The difference between these file sets lies only in the change of time, and the rest remains unchanged. Due to space limitations, only the first file is shown here.

pCO₂

Global Attributes:

```
Conventions      = 'CF-1.7 CMIP-6.2'
activity_id      = 'ScenarioMIP'
branch_method    = 'standard'
branch_time_in_child = 0
branch_time_in_parent = 0
contact          = 'Kenneth Lo (cdkkl@giss.nasa.gov)'
creation_date    = '2022-01-28T17:58:03Z'
data_specs_version = '01.00.23'
experiment       = 'update of RCP7.0 based on SSP3'
experiment_id    = 'ssp370'
external_variables = 'areacella'
forcing_index    = 1
frequency        = 'mon'
further_info_url = 'https://furtherinfo.es-doc.org/CMIP6.NASA-GISS.GISS-
E2-1-G-CC.ssp460.none.r10i1p1f1'
grid             = 'atmospheric grid: 144x90, ocean grid: 288x180'
grid_label       = 'gn'
history          = '2022-01-28T17:58:03Z ; CMOR rewrote data to be
consistent with CMIP6, CF-1.7 CMIP-6.2 and CF standards.'
initialization_index = 1
```

USA'	institution	= 'Goddard Institute for Space Studies, New York, NY 10025,
	institution_id	= 'NASA-GISS'
	mip_era	= 'CMIP6'
	model_id	= 'Eargcc_460emis'
	nominal_resolution	= '250 km'
	parent_activity_id	= 'CMIP'
	parent_experiment_id	= 'historical'
	parent_experiment_rip	= 'r1i1p1f1'
	parent_mip_era	= 'CMIP6'
	parent_source_id	= 'GISS-E2-1-G-CC'
	parent_time_units	= 'days since 2015-1-1'
	parent_variant_label	= 'r1i1p1f1'
	physics_index	= 1
	product	= 'model-output'
	realization_index	= 101
	realm	= 'atmos'
	references	= 'https://data.giss.nasa.gov/modelE/cmip6'
	source	= 'GISS-E2-1-G-CC (2019):
		aerosol: varies with physics-version (p==1 none, p==3
OMA, p==4 TOMAS, p==5 MATRIX)		atmos: GISS-E2.1 (2 x 2.5 degrees; 144 x 90
		longitude/latitude; 40 levels; top level 0.1 hPa)
		atmosChem: varies with physics-version (p==1 Non-
interactive, p>1 GPUCCINI)		land: GISS LSM
		landIce: Fixed
		ocean: GISS Ocean (1 deg; 360 x 180 longitude/latitude;
40 levels; top grid cell 0-10m)		ocnBgchem: NOBM (NASA Ocean Biogeochemistry
Model; same grid as ocean)		seaIce: GISS SI (same grid as ocean)'
	source_id	= 'GISS-E2-1-G-CC'
	source_type	= 'AOGCM'
	sub_experiment	= 'none'
	sub_experiment_id	= 'none'
	table_id	= 'Emon'
	table_info	= 'Creation Date:(21 March 2018)
MD5:3087ac46757d524f149af7ac233b0a95'		
	title	= 'GISS-E2-1-G-CC output prepared for CMIP6'
	tracking_id	= 'hdl:21.14100/45890a11-f13c-4860-bebb-3a63f2eb43bf'
	variable_id	= 'co23D'
	variant_label	= 'r10i1p1f1'
	license	= 'CMIP6 model data produced by NASA Goddard Institute

for Space Studies is released with a Creative Commons Zero Universal (CC0 1.0, <https://creativecommons.org/publicdomain/zero/1.0/>) and has no restrictions on its use. Consult <https://pcmdi.llnl.gov/CMIP6/TermsOfUse> for more general terms of use governing CMIP6 output, including citation requirements and proper acknowledgment. Further information about this data can be found via the further_info_url (recorded as a global attribute in this file) and at <https://pcmdi.llnl.gov/>. The data producers and data providers make no warranty, either express or implied, including, but not limited to, warranties of merchantability and fitness for a particular purpose. All liabilities arising from the supply of the information (including any liability arising in negligence) are excluded to the fullest extent permitted by law.'

cmor_version = '3.3.2'

Dimensions:

lev = 40
bnds = 2
time = 432 (UNLIMITED)
lat = 90
lon = 144

Variables:

a

Size: 40x1
Dimensions: lev
Datatype: double
Attributes:
long_name = 'vertical coordinate formula term: a(k)'

a_bnds

Size: 2x40
Dimensions: bnds,lev
Datatype: double
Attributes:
long_name = 'vertical coordinate formula term: a(k+1/2)'

b

Size: 40x1
Dimensions: lev
Datatype: double
Attributes:
long_name = 'vertical coordinate formula term: b(k)'

b_bnds

Size: 2x40
Dimensions: bnds,lev
Datatype: double
Attributes:
long_name = 'vertical coordinate formula term: b(k+1/2)'

co23D

Size: 144x90x40x432
Dimensions: lon,lat,lev,time

```

Datatype:  single
Attributes:
    standard_name = 'CO2_3D_tracer'
    long_name      = '3D field of transported CO2'
    comment        = 'report 3D field of model simulated atmospheric
CO2 mass mixing ration on model levels'
    units          = 'kg kg-1'
    cell_methods   = 'area: time: mean'
    cell_measures  = 'area: areacella'
    history        = '2022-01-28T17:58:03Z altered by CMOR: replaced
missing value flag (-1e+30) with standard missing value (1e+20).'
    missing_value  = 1.000000020040877e+20
    _FillValue     = 1.000000020040877e+20

lat
    Size:          90x1
    Dimensions:    lat
    Datatype:      double
    Attributes:
        bounds      = 'lat_bnds'
        units       = 'degrees_north'
        axis        = 'Y'
        long_name    = 'latitude'
        standard_name = 'latitude'

lat_bnds
    Size:          2x90
    Dimensions:    bnds,lat
    Datatype:      double

lev
    Size:          40x1
    Dimensions:    lev
    Datatype:      double
    Attributes:
        bounds      = 'lev_bnds'
        units       = '1'
        axis        = 'Z'
        positive     = 'down'
        long_name    = 'hybrid sigma pressure coordinate'
        standard_name = 'atmosphere_hybrid_sigma_pressure_coordinate'
        formula      = 'p = a*p0 + b*ps'
        formula_terms = 'p0: p0 a: a b: b ps: ps'

lev_bnds
    Size:          2x40
    Dimensions:    bnds,lev
    Datatype:      double

```

```

Attributes:
    formula      = 'p = a*p0 + b*ps'
    standard_name = 'atmosphere_hybrid_sigma_pressure_coordinate'
    units        = '1'
    formula_terms = 'p0: p0 a: a_bnds b: b_bnds ps: ps'

lon
    Size:      144x1
    Dimensions: lon
    Datatype:  double
    Attributes:
        bounds      = 'lon_bnds'
        units       = 'degrees_east'
        axis        = 'X'
        long_name    = 'longitude'
        standard_name = 'longitude'

lon_bnds
    Size:      2x144
    Dimensions: bnds,lon
    Datatype:  double

p0
    Size:      1x1
    Dimensions:
    Datatype:  double
    Attributes:
        long_name = 'vertical coordinate formula term: reference pressure'
        units     = 'Pa'

ps
    Size:      144x90x432
    Dimensions: lon,lat,time
    Datatype:  single
    Attributes:
        long_name = 'Surface Air Pressure'
        units     = 'Pa'

time
    Size:      432x1
    Dimensions: time
    Datatype:  double
    Attributes:
        bounds      = 'time_bnds'
        units       = 'days since 2015-1-1'
        calendar    = '365_day'
        axis        = 'T'
        long_name    = 'time'
        standard_name = 'time'

```

time_bnds
Size: 2x432
Dimensions: bnds,time
Datatype: double

Wet deposition

Global Attributes:

Conventions = 'CF-1.7 CMIP-6.2'
activity_id = 'ScenarioMIP AerChemMIP'
branch_method = 'standard'
branch_time_in_child = 0
branch_time_in_parent = 0
contact = 'Kenneth Lo (cdkkl@giss.nasa.gov)'
creation_date = '2020-01-29T20:00:30Z'
data_specs_version = '01.00.23'
experiment = 'gap-filling scenario reaching 7.0 based on SSP3'
experiment_id = 'ssp370'
external_variables = 'areacella'
forcing_index = 1
frequency = 'mon'
further_info_url = 'https://furtherinfo.es-doc.org/CMIP6.NASA-GISS.GISS-E2-1-G.ssp370.none.r1i1p3f1'
grid = 'atmospheric grid: 144x90, ocean grid: 288x180'
grid_label = 'gn'
history = '2020-01-29T20:00:30Z ; CMOR rewrote data to be consistent with CMIP6, CF-1.7 CMIP-6.2 and CF standards.'
initialization_index = 1
institution = 'Goddard Institute for Space Studies, New York, NY 10025, USA'
institution_id = 'NASA-GISS'
mip_era = 'CMIP6'
model_id = 'E212TomaSSP370aF40oQ40'
nominal_resolution = '250 km'
parent_activity_id = 'CMIP'
parent_experiment_id = 'historical'
parent_experiment_rip = 'r1i1p3'
parent_mip_era = 'CMIP6'
parent_source_id = 'GISS-E2-1-G'
parent_time_units = 'days since 2015-1-1'
parent_variant_label = 'r1i1p3f1'
physics_index = 3
product = 'model-output'
realization_index = 1
realm = 'atmos'

```

references      = 'https://data.giss.nasa.gov/modelE/cmip6'
source         = 'GISS-E2.1G (2019):
                  aerosol: Varies with physics-version (p==1 none, p==3
OMA, p==4 TOMAS, p==5 MATRIX)
                  atmos:  GISS-E2.1  (2.5x2  degree;  144  x  90
longitude/latitude; 40 levels; top level 0.1 hPa)
                  atmosChem: Varies with physics-version (p==1 Non-
interactive, p>1 GPUCCINI)
                  land: GISS LSM
                  landIce: none
                  ocean: GISS Ocean (GO1, 1 degree; 360 x 180
longitude/latitude; 40 levels; top grid cell 0-10 m)
                  ocnBgchem: none
                  seaIce: GISS SI'

source_id      = 'GISS-E2-1-G'
source_type    = 'AOGCM'
sub_experiment = 'none'
sub_experiment_id = 'none'
table_id       = 'Amon'
table_info     = 'Creation Date:(21 March 2018)
MD5:c93735846d66458966fc81f390b2d714'

title          = 'GISS-E2-1-G output prepared for CMIP6'
tracking_id    = 'hdl:21.14100/45371886-0135-4055-acf5-fea017b21574'
variable_id    = 'clwvi'
variant_label  = 'r1i1p3f1'
license        = 'CMIP6 model data produced by NASA Goddard Institute
for Space Studies is licensed under a Creative Commons Attribution ShareAlike 4.0 International
License (https://creativecommons.org/licenses). Consult
https://pcmdi.llnl.gov/CMIP6/TermsOfUse for terms of use governing CMIP6 output, including
citation requirements and proper acknowledgment. Further information about this data, including
some limitations, can be found via the further_info_url (recorded as a global attribute in this file)
and at https://pcmdi.llnl.gov/. The data producers and data providers make no warranty, either
express or implied, including, but not limited to, warranties of merchantability and fitness for a
particular purpose. All liabilities arising from the supply of the information (including any liability
arising in negligence) are excluded to the fullest extent permitted by law.'

cmor_version   = '3.3.2'

Dimensions:
time = 432  (UNLIMITED)
lat  = 90
lon  = 144
bnds = 2

Variables:
time
Size:      432x1

```

Dimensions: time
Datatype: double
Attributes:
 bounds = 'time_bnds'
 units = 'days since 2015-1-1'
 calendar = '365_day'
 axis = 'T'
 long_name = 'time'
 standard_name = 'time'

time_bnds
 Size: 2x432
 Dimensions: bnds,time
 Datatype: double

lat
 Size: 90x1
 Dimensions: lat
 Datatype: double
 Attributes:
 bounds = 'lat_bnds'
 units = 'degrees_north'
 axis = 'Y'
 long_name = 'latitude'
 standard_name = 'latitude'

lat_bnds
 Size: 2x90
 Dimensions: bnds,lat
 Datatype: double

lon
 Size: 144x1
 Dimensions: lon
 Datatype: double
 Attributes:
 bounds = 'lon_bnds'
 units = 'degrees_east'
 axis = 'X'
 long_name = 'longitude'
 standard_name = 'longitude'

lon_bnds
 Size: 2x144
 Dimensions: bnds,lon
 Datatype: double

clwvi
 Size: 144x90x432
 Dimensions: lon,lat,time

Datatype: single

Attributes:

```
standard_name = 'atmosphere_cloud_condensed_water_content'  
long_name     = 'Condensed Water Path'  
comment       = 'Mass of condensed (liquid + ice) water in the  
column divided by the area of the column (not just the area of the cloudy portion of the column).  
Includes precipitating hydrometeors ONLY if the precipitating hydrometeors affect the calculation  
of radiative transfer in model.'  
units         = 'kg m-2'  
cell_methods  = 'area: time: mean'  
cell_measures = 'area: areacella'  
history       = '2020-01-29T20:00:30Z altered by CMOR: replaced  
missing value flag (-1e+30) with standard missing value (1e+20).'  
missing_value = 1.000000020040877e+20  
_FillValue    = 1.000000020040877e+20
```

REVIEWER COMMENTS

Reviewer #3 (Remarks to the Author):

In my first review I wrote that “The data set they present is quite unique and a significant contribution in itself. The PCA analysis and receptor model also provide novel and interesting insight into trace metal dynamics in coastal systems under climate change. There is not much current literature on this topic so this work presents a valuable and original piece of work. The quality of the data appear very good and the methods are described in detail. The overall analysis and analytical approach are both robust and comprehensive.” I stand by this assessment, and the authors have responded well to most of my concerns. However, based on the authors’ rebuttal to my first review I have identified a major issue with their methodology. To remedy this parts of the analysis needs to be redone, and the new results analyzed. This major issue is described below. In addition, there remains some minor issues left unanswered in the rebuttal and revised manuscript. These are described under “minor issues” below. The manuscript will require major revisions before it can be published.

We would like to express our gratitude to the reviewer for the positive comments on our article. Following your comments and suggestions, we have updated the model variables as required, rebuilt the model, and made corresponding changes to the results, descriptions, discussions and figures in the article.

Major issue

In my first review I noted that it is clear from the methods that the authors are not familiar with Earth System Models (ESMs) or CMIP6. I did not find any errors in their work, but requested the add additional detail in the methods to identify which variables they had used as well as more detail about their choice of model. The authors have not added any of this information in the revised manuscript, but they did provide additional detail in their rebuttal to me. Unfortunately, this additional information shows me very clearly that the authors have not done the analysis they think they did, or at least not the analysis they describe in the manuscript. In my original review I asked the authors to include the actual variable names from the model so that the reader can more easily identify what has been used (since all CMIP6 models use the same variable naming conventions). This the authors have not done, but they have included an appendix (Appendix 1) with detailed metadata for three variables they have extracted. To better highlight the issues, I’ve identified I’ve removed much of the information from Appendix 1 of the rebuttal and only kept what is important to support this review.

We realize that appropriate Realms and variables from the same institution should be adopted. In terms of variable selection, this study entirely used the data source of CanESM5, and the Nominal Resolution was uniformly set to 1×1 degree. For the sea surface temperature, we adopted the "Ocean" Realm, and the CF Standard Name was "sea surface temperature"; for the rainfall, we adopted the "atmosphere" Realm, and the CF Standard Name was "rainfall flux", with the long name being "Rainfall Flux where Ice Free Ocean over Sea". For the partial pressure of carbon dioxide, we adopted the "ocnBgchem" Realm, and the CF Standard Name was "abiotic surface aqueous partial pressure of CO₂", with the long name being "Abiotic Surface Aqueous Partial Pressure of CO₂". For the dataset used in this revision, please refer to Appendix I in this file.

The modifications of data and text resulting from the changes in the modeling variables are presented in the manuscript and the Supplementary Material. This includes the figures in the manuscript that show the prediction results, which have spatial differences due to the improvement

in resolution. The descriptions in the results are modified accordingly for the calculation results in the corresponding figures. In addition, we have provide a brand-new Supplementary Material B to display the calculation results, and corresponding modifications will also be made in the creation of the receptor model (Supplementary Material C) and Model verification (Supplementary Material D).

On lines 446-447 it is stated that “water temperature, rainfall, CO₂ levels ($p\text{CO}_2$), and ocean acidification rates were extracted.” In Appendix 1 of the rebuttal the authors include detailed metadata for three variables under the headings “Surface sea temperature”, “ $p\text{CO}_2$ ”, and “Wet deposition”. The information in Appendix 1 shows that the variables extracted are “tasmin”, “co23D”, and “clwvi”.

“tasmin” is the variable extracted under the headline “Surface sea temperature”. However, this is not water temperature. Realm is given as “atmos” indicating that it is an atmospheric variable, and the long_name attribute states that this is “Daily Minimum Near-Surface Air Temperature”. So rather than using water temperature in their model the authors have used air temperature. And not even the actual air temperature, but the minimum of the near-surface (typically 2-meter) temperature. This variable is also extracted from a different model than that stated on line 442 (see below).

“co23D” is the variable extracted under the headline “ $p\text{CO}_2$ ”. This is also an atmospheric variable as indicated by the attribute realm (“atmos”). To me it would make more sense to use surface ocean $p\text{CO}_2$, but it is not actually stated anywhere whether atmospheric or oceanic $p\text{CO}_2$ is identified as one of the three drivers. Importantly though, the attribute long_name shows that this variable is a “3D field of transported CO₂”. The authors have not made clear whether they have used the entire 3D field as input to their model, or whether they’ve used just one layer. Regardless, I believe the authors want to use atmospheric CO₂ concentration here, which is part of the model forcing (the SSP3-7.0 scenario defines the atmospheric CO₂ concentration for each timestep).

“clwvi” is the variable extracted under the heading “Wet deposition”. This is probably what the authors believe represents rainfall (one of the variables they claim on line 446 to have extracted). This is another atmospheric variable (realm = atmos), but that would in this case make sense. However, the attribute long_name shows that this is “Condensed Water Path” and the comment, as well as a quick check in the CMIP6 variable list, indicates that this variable represents the amount of liquid water and ice in clouds. So not related to precipitation. In addition, this variable is from a different ensemble run from that given on line 444 (see below).

Thank the reviewers for their detailed comments. The corresponding paragraph for the description of the methods has been revised. The following is the rewritten version:

CMIP6 is a project initiated under the World Climate Research Program to compare coupled models and enhance our understanding of the Earth's climate system. Its goal is to promote model development and facilitate scientific research by openly providing multi-model output in a standardized format. Here we use ScenarioMIP simulations from the Earth System model CanESM5. The model has a spatial resolution of 1 degrees latitude by 1 degrees longitude, and we use the monthly output from the “r1i1p1f1. The climate change model is set as ssp3-7.0. To extract data from the model from 2015–2099, R-based ncdf4 and raster toolkits were used. Variables such as surface sea temperature, rainfall flux, and abiotic surface aqueous partial pressure of CO₂ ($p\text{CO}_2$). Using a receptor model and machine learning techniques, the changes in nutrients and TMs levels in the water area over the next 80 years were simulated and predicted. By analyzing the extracted variables, the monthly change trend of the concentration of nutrients and TMs by the end of this

century could be projected.

On line 442 the authors write that output from the GISS-E2 model is used. This appears to be incorrect. The variable “tasmin” is retrieved from the Chinese FGOALS-g3 model (parent_source_id=FGOALS-g3).

The expression error has been corrected to "abiotic surface aqueous partial pressure of CO₂".

The corrected text of line 442 is: The variables include surface sea temperature, rainfall flux, and abiotic surface aqueous partial pressure of CO₂ ($p\text{CO}_2$). It appears on page 18, line 434 of the revised article.

On line 444 it is stated that the ensemble member “r11p1f1” is used. However, for variable “clwvi” parent_variable_label=r11p3f1 is used. It is not advised to use two different ensemble members for different variables, and if done it needs to be stated clearly.

The ensemble member for the newly adopted variables in this study is uniformly "r11p1f1". You can refer to Appendix I. The relevant content can be found on pages 10, 14 and 19 of this document.

On lines 446-447 four variables are listed, but there are only three in Appendix 1. I therefore do not know what data, if any, has been extracted as “ocean acidification rates” since no information is given.

Throughout the text, we have unified the names of the three variables as: surface sea temperature, rainfall flux, and partial pressure of carbon dioxide ($p\text{CO}_2$).

The information I’ve provided above clearly shows that the authors have not used the variables they say they have. I’m sorry that this is discovered so late in the review process, but this is why the actual variable names should always be provided. Had the authors included this information in the previous version it would have been clear to me, and probably also the other reviewers, that something is wrong with the analysis. The authors will have to extract the correct variables (this list should be helpful in identifying which they are: <https://pcmdi.llnl.gov/mips/cmip3/variableList.html>) and redo their analysis for the future. The results and conclusions will then have to be modified based on the new results.

We are extremely grateful to the reviewer for providing such detailed and valuable suggestions. We used the variable "tos" for the sea surface temperature. The partial pressure of carbon dioxide belongs to the category of ocnBgchem. In terms of rainfall, in the annotation, the "standard_name" is 'rainfall_flux', and the "long_name" is 'Rainfall Flux where Ice Free Ocean over Sea'.

Finally, I want to note that in their rebuttal to my comment about why they have chosen the model they have the authors show their inexperience with Earth System Models and CMIP6. The ocean resolution in the models they’ve extracted data from is 1 by 1 degree, which is clear from the attributes listed in Appendix 1. The atmospheric resolution in Earth System models is always coarser than the ocean. I assumed in my initial review that they had extracted from the ocean, not the atmosphere. Given this obvious inexperience I would urge the authors to reach out to the contact person given for the CMIP6 models (the attribute contact gives an e-mail address) and ask for advice on how to best use their data, and which variables are the most appropriate.

Thank the reviewer for the guidance provided in variable screening. In this study, variables have been reselected, and the contact person has been consulted via email regarding the specific details of the variable descriptions to ensure that the selected variables are consistent with what is described in the article.

Minor issues:

Line 24: “proved” is a very strong word to use here. Consider revising.

The word "suggested" was used in the original sentence instead of "proved". The sentence was modified as: It is suggested that the acidification process may carry the risk of triggering the ocean's buffering mechanisms.

Line 38: Typo. Separate "crucialwater" into two words (crucial and water)

The original sentence was modified as "However, this crucial water sector is suffering from potential impacts from climate change."

Line 53: The change to "acidification atmosphere" does not make any more sense than the original "carbonate atmosphere". It is true that the nearshore environment is affected by acidification, but the term "acidification atmosphere" is not terminology that is commonly used in the scientific literature. At the very least the authors need to add an explanation/definition to the text, but better would be to just say that the nearshore environment is affected by acidification (which is apparently what they are trying to communicate here).

We added notes to the original part to supplement the meaning of "acidification atmospheres". The sentence was changed to: We expect future marine ecology to be exposed to oceans with high water temperatures, low pH, and acidic atmospheres (a result of the elevation in the partial pressure of CO₂).

Line 67: The common terminology is "sea surface temperature". Please revise.

The sentence have been revised as follows: Sea surface temperature, atmospheric deposition and pH are considered to be the influencing factors of these transmission routes. The corresponding reference is also targeted at surface sea temperature. According to the search, there are also five other places in the subsequent part of this article where the same words appear, and we have made the revisions accordingly.

Line 92-93: I commented on this sentence in my first review and it makes no more sense now. Nitrogen removal and denitrification does happen in situ in the ocean. The reference used here for the statement that these processes do not happen (Hutchins and Capone, 2022) does not in any way support such a statement. It is factually incorrect to state that "N removal and denitrification processes have been proven not to occur in situ", and the authors are also incorrectly referencing Hutchins and Capone (2022) to support the incorrect statement. This needs to be revised.

We have rewritten the sentence and removed the original reference, replacing it with two new references. The sentences are revised as follows: The metals in the acid-soluble and reducible fractions migrate in response to the variation of marine water quality ²⁵. The stratification of ocean temperature and density modifies the nutrient transport ²⁶. Consequently, both nutrients and trace metals (TMs) can further influence the open sea via ocean currents.

Reference

25. Liu, M., Chen, J. B., Sun, X. S., Hu, Z. Z. & Fan, D. J. Accumulation and transformation of heavy metals in surface sediments from the Yangtze River estuary to the East China Sea shelf. *Environmental Pollution* 245, 111-121, doi:10.1016/j.envpol.2018.10.128 (2019).

26. Dai, Y. H. et al. Coastal phytoplankton blooms expand and intensify in the 21st century. *Nature* 615, 280+, doi:10.1038/s41586-023-05760-y (2023).

Line 120: Here you state that "sea surface temperature, dry and wet deposition amount, and pH" are the three factors being considered in your model. However, on lines 206, 228, and 297 as well as in Figure 1b you list the three components as "SST, deposition, and pCO₂" suggesting that pH is not one of the three components. But then on line 216 pH is mentioned as one of the three. Please correct the information in all these places to avoid confusion. In addition, you should indicate in all these

places which type of deposition. I assume it is both wet and dry as initially stated, but this should be clear everywhere. Also, please define pCO₂ the first time it is used, not the last. Now “partial pressure of carbon dioxide” is used on line 297, but not before. You also need to clarify whether you’ve used atmospheric or oceanic pCO₂.

To avoid disputes over wording, this paper determines that the three influencing variables of the model are sea surface temperature, precipitation and partial pressure of carbon dioxide. During the revision, the cited references 14, 15 and 16 have been changed. All the figures in the main text have been redrawn due to the change of data. They are in Appendix II.

Reference

14. Zhang, L. *et al.* Spatial and seasonal characteristics of dissolved heavy metals in the east and west Guangdong coastal waters, South China. *Marine Pollution Bulletin* **95**, 419-426, doi:10.1016/j.marpolbul.2015.03.035 (2015).

15. Zhai, F. G. *et al.* Satellite-Observed Interannual Variations in Sea Surface Chlorophyll-a Concentration in the Yellow Sea Over the Past Two Decades. *Journal of Geophysical Research-Oceans* **128**, doi:10.1029/2022jc019528 (2023).

16. Ardelan, M. V., Steinnes, E., Lierhagen, S. & Linde, S. O. Effects of experimental CO₂ leakage on solubility and transport of seven trace metals in seawater and sediment. *Science of the Total Environment* **407**, 6255-6266, doi:10.1016/j.scitotenv.2009.09.004 (2009).

Line 124: In my original review I wrote here that “It is unclear what you mean by “CMIP6 framework” here. You have, as I understand it, used output from one of the models contributing to CMIP6. If that is what you want to communicate here please state that, otherwise explain what the framework is.” The authors answered in their rebuttal that they would make a modification, and link to the Materials and Methods section but this has not been done. It is clear that the authors are not familiar with the use of Earth System Models and CMIP6 (as also explained above). They have not used the CMIP6 framework, which is a framework to allow the intercomparison of many different models, but rather output from one Earth System model which has contributed to the CMIP6 project. There is no model comparison, or model ensemble used in this work. So here it would be more appropriate to state “the CMIP6 version of the GISS-E2 model was used to predict ...”. However, as explained above this is also not accurate since the authors have actually used data from two models.

Following the reviewer's suggestions, the replaced data source (CanESM5), and the fact that the data source now comes from a unified origin, this sentence are changed into "The CMIP6 version of the CanESM5 model".

The sentence was changed to: Once a reliable receptor model had been established, the CMIP6 version of the CanESM5 model was employed to predict the future water quality.

Line 127-128: This paper does not simulate the SSP370 scenario. Output from Earth System Models that have simulated this scenario is used, but that is not the same thing. This needs revision for clarity.

This sentence does not clarify the specific way the model is used and have been changed into:

The data of three variables output by the experiment ID of SSP3-7.0 were used to fit the measured data, and a receptor model was constructed to predict nutrients and TMs.

Lines 134-135: The authors have revised this statement somewhat after my initial review. However, the statement is still unclear and my original comment that “the warming process cannot be controlled anywhere at the moment” is still valid. Adding “or reversed” does not change or clarify

anything. I do not understand what the authors are trying to say so the sentence still needs revision. The warming process cannot be controlled anywhere at the moment so the meaning of this final statement is unclear. Please revise.

We apologize for the confusion caused, we have rewritten the sentence as follows:

It reveals the pollution exposure risks of the increasingly acidified oceans, especially in the estuarine environment with high nutrients and high carbon dioxide.

Line 399-400: Here two sentences in a row begin with “However”. That does not make sense so please revise.

We are sorry for the narrative errors here. The sentence is changed into:

However, we are able to get a glimpse of the dramatic changes in the cycle of elements brought about by changes in the physical environment. We are able to further analyze and foresee the significant changes in element cycles brought about by physical environmental changes within a regional dataset.

Reviewer #3 (Remarks on code availability):

I have not reviewed the code.

No changes were made to the code in the last modification, so it wasn't uploaded. We have re-uploaded the code files this time. Please refer to the “Readme” .

In addition, we have provided the training set, which is placed in the package of "code and training data".

Appendix I Details of Variables Used for Modeling

(1) Variables used for sea surface temperature

Global Attributes:

Conventions	= 'CF-1.7 CMIP-6.2'
activity_id	= 'ScenarioMIP AerChemMIP'
branch_method	= 'Standard'
branch_time_in_child	= 0
branch_time_in_parent	= 2015
contact	= 'Dr. Xinyao Rong (rongxy@cma.gov.cn)'
creation_date	= '2019-06-11T05:58:01Z'
data_specs_version	= '01.00.30'
description	= 'SSP3-7.0'
experiment	= 'gap-filling scenario reaching 7.0 based on SSP3'
experiment_id	= 'ssp370'
external_variables	= 'areacello'
forcing_index	= 1
frequency	= 'mon'
further_info_url	= 'https://furtherinfo.es-doc.org/CMIP6.CAMS.CAMS- CSM1-0.ssp370.none.r1i1p1f1'
grid	= 'native ocean tri-polar grid'
grid_label	= 'gn'
history	= '2019-06-11T05:58:01Z ;rewrote data to be consistent with ScenarioMIP for variable tos found in table Omon.; Output from CAMS-CSM1-0'
initialization_index	= 1
institution	= 'Chinese Academy of Meteorological Sciences, Beijing 100081, China'
institution_id	= 'CAMS'
mip_era	= 'CMIP6'
nominal_resolution	= '100 km'
parent_activity_id	= 'CMIP'
parent_experiment_id	= 'historical'
parent_mip_era	= 'CMIP6'
parent_source_id	= 'CAMS-CSM1-0'
parent_time_units	= 'days since 2015-01-01'
parent_variant_label	= 'r1i1p1f1'
physics_index	= 1
product	= 'model-output'
realization_index	= 1
realm	= 'ocean'
references	= 'Model described by Rong et al (J. Meteor. Res., 2019, 576-591).'
run_variant	= 'forcing: greenhouse gases,aerosol,solar constant,volcano,ozone'

source = 'CAM5_CSM 1.0 (2016):
aerosol: none
atmos: ECHAM5_CAMS (T106; 320 x 160
longitude/latitude; 31 levels; top level 10 mb)
atmosChem: none
land: CoLM 1.0
landIce: none
ocean: MOM4 (tripolar; 360 x 200 longitude/latitude,
primarily 1deg latitude/longitude, down to 1/3deg within 30deg of the equatorial tropics; 50 levels;
top grid cell 0-10 m)

ocnBgchem: none
seaIce: SIS 1.0'
source_id = 'CAM5-CSM1-0'
source_type = 'AOGCM'
sub_experiment = 'none'
sub_experiment_id = 'none'
table_id = 'Omon'
table_info = 'Creation Date:(30 April 2019)

MD5:cc2ae51c23960ce127b4d68ab3d8ac87'

title = 'CAM5-CSM1-0 output prepared for CMIP6'
tracking_id = 'hdl:21.14100/3b33f3fc-9159-424a-9502-3dbe2461eea7'
variable_id = 'tos'
variant_label = 'r1i1p1f1'
license = 'CMIP6 model data produced by Lawrence Livermore

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cmor_version = '3.4.0'

Dimensions:

time = 1020 (UNLIMITED)
j = 200
i = 360
bnds = 2
vertices = 4

Variables:

time
Size: 1020x1
Dimensions: time

Datatype: double
Attributes:
 bounds = 'time_bnds'
 units = 'days since 2015-1-1 00:00'
 calendar = '365_day'
 axis = 'T'
 long_name = 'time'
 standard_name = 'time'

time_bnds

Size: 2x1020
Dimensions: bnds,time
Datatype: double

j

Size: 200x1
Dimensions: j
Datatype: int32
Attributes:
 units = '1'
 long_name = 'cell index along second dimension'

i

Size: 360x1
Dimensions: i
Datatype: int32
Attributes:
 units = '1'
 long_name = 'cell index along first dimension'

latitude

Size: 360x200
Dimensions: i,j
Datatype: single
Attributes:
 standard_name = 'latitude'
 long_name = 'latitude'
 units = 'degrees_north'
 missing_value = 1.000000020040877e+20
 _FillValue = 1.000000020040877e+20
 bounds = 'vertices_latitude'

longitude

Size: 360x200
Dimensions: i,j
Datatype: single
Attributes:
 standard_name = 'longitude'
 long_name = 'longitude'

```

units          = 'degrees_east'
missing_value = 1.000000020040877e+20
_FillValue     = 1.000000020040877e+20
bounds        = 'vertices_longitude'

vertices_latitude
Size:         4x360x200
Dimensions: vertices,i,j
Datatype:    single
Attributes:
units          = 'degrees_north'
missing_value = 1.000000020040877e+20
_FillValue     = 1.000000020040877e+20

vertices_longitude
Size:         4x360x200
Dimensions: vertices,i,j
Datatype:    single
Attributes:
units          = 'degrees_east'
missing_value = 1.000000020040877e+20
_FillValue     = 1.000000020040877e+20

tos
Size:         360x200x1020
Dimensions: i,j,time
Datatype:    single
Attributes:
standard_name = 'sea_surface_temperature'
long_name     = 'Sea Surface Temperature'
comment       = 'Temperature of upper boundary of the liquid
ocean, including temperatures below sea-ice and floating ice shelves.'
units         = 'degC'
original_name = 'temp'
cell_methods  = 'area: mean where sea time: mean'
cell_measures = 'area: areacello'
history       = '2019-06-11T05:57:56Z altered by CMOR: replaced
missing value flag (-1e+10) with standard missing value (1e+20).'
missing_value = 1.000000020040877e+20
_FillValue     = 1.000000020040877e+20
coordinates    = 'latitude longitude'

```

(2) Variables used for wet deposition

Global Attributes:

```
CCCma_model_hash =
'1f91f92cb6d607391f44831504025d32fc44faa1'
CCCma_parent_runid = 'rc3.1-his01'
CCCma_pycmor_hash =
'33c30511acc319a98240633965a04ca99c26427e'
CCCma_runid = 'rc3.1-s7001'
Conventions = 'CF-1.7 CMIP-6.2'
YMDH_branch_time_in_child = '2015:01:01:00'
YMDH_branch_time_in_parent = '2015:01:01:00'
activity_id = 'ScenarioMIP AerChemMIP'
branch_method = 'Spin-up documentation'
branch_time_in_child = 60225
branch_time_in_parent = 60225
contact = 'ec.cccma.info-info.ccmac.ec@canada.ca'
creation_date = '2019-05-02T03:14:41Z'
data_specs_version = '01.00.29'
experiment = 'gap-filling scenario reaching 7.0 based on SSP3'
experiment_id = 'ssp370'
external_variables = 'areacello'
forcing_index = 1
frequency = 'mon'
further_info_url = 'https://furtherinfo.es-
doc.org/CMIP6.CCCma.CanESM5.ssp370.none.r1i1p1f1'
grid = 'ORCA1 tripolar grid, 1 deg with refinement to 1/3
deg within 20 degrees of the equator; 361 x 290 longitude/latitude; 45 vertical levels; top grid cell
0-6.19 m'
grid_label = 'gn'
history = '2019-05-02T03:14:41Z ;rewrote data to be
consistent with ScenarioMIP for variable prra found in table Omon.;
```

Output from \$runid'

```
initialization_index = 1
institution = 'Canadian Centre for Climate Modelling and Analysis,
Environment and Climate Change Canada, Victoria, BC V8P 5C2, Canada'
institution_id = 'CCCma'
mip_era = 'CMIP6'
nominal_resolution = '100 km'
parent_activity_id = 'CMIP'
parent_experiment_id = 'historical'
parent_mip_era = 'CMIP6'
parent_source_id = 'CanESM5'
parent_time_units = 'days since 1850-01-01 0:0:0.0'
parent_variant_label = 'r1i1p1f1'
```

physics_index	= 1
product	= 'model-output'
realization_index	= 1
realm	= 'atmos'
references	= 'Geophysical Model Development Special issue on CanESM5 (https://www.geosci-model-dev.net/special_issues.html)'
source	= 'CanESM5 (2019): aerosol: interactive atmos: CanAM5 (T63L49 native atmosphere, T63 Linear Gaussian Grid; 128 x 64 longitude/latitude; 49 levels; top level 1 hPa) atmosChem: specified oxidants for aerosols land: CLASS3.6/CTEM1.2 landIce: specified ice sheets ocean: NEMO3.4.1 (ORCA1 tripolar grid, 1 deg with refinement to 1/3 deg within 20 degrees of the equator; 361 x 290 longitude/latitude; 45 vertical levels; top grid cell 0-6.19 m) ocnBgchem: Canadian Model of Ocean Carbon (CMOC); NPZD ecosystem with OMIP prescribed carbonate chemistry seaIce: LIM2'
source_id	= 'CanESM5'
source_type	= 'AOGCM'
sub_experiment	= 'none'
sub_experiment_id	= 'none'
table_id	= 'Omon'
table_info	= 'Creation Date:(20 February 2019) MD5:374fbe5a2bcca535c40f7f23da271e49'
title	= 'CanESM5 output prepared for CMIP6'
variable_id	= 'prrra'
variant_label	= 'r1i1p1f1'
version	= 'v20190429'
license	= 'CMIP6 model data produced by The Government of Canada (Canadian Centre for Climate Modelling and Analysis, Environment and Climate Change Canada) is licensed under a Creative Commons Attribution ShareAlike 4.0 International License (https://creativecommons.org/licenses). Consult https://pcmdi.llnl.gov/CMIP6/TermsOfUse for terms of use governing CMIP6 output, including citation requirements and proper acknowledgment. Further information about this data, including some limitations, can be found via the further_info_url (recorded as a global attribute in this file) and at https://pcmdi.llnl.gov/ . The data producers and data providers make no warranty, either express or implied, including, but not limited to, warranties of merchantability and fitness for a particular purpose. All liabilities arising from the supply of the information (including any liability arising in negligence) are excluded to the fullest extent permitted by law.'
cmor_version	= '3.4.0'
tracking_id	= 'hdl:21.14100/5a1e98a9-aa21-4f43-9b04- 4b8bad7aa563'

Dimensions:

time = 1032 (UNLIMITED)
bnds = 2
j = 291
i = 360
vertices = 4

Variables:

time

Size: 1032x1
Dimensions: time
Datatype: double
Attributes:
 bounds = 'time_bnds'
 units = 'days since 1850-01-01 0:0:0.0'
 calendar = '365_day'
 axis = 'T'
 long_name = 'time'
 standard_name = 'time'

time_bnds

Size: 2x1032
Dimensions: bnds,time
Datatype: double

j

Size: 291x1
Dimensions: j
Datatype: int32
Attributes:
 units = '1'
 long_name = 'cell index along second dimension'

i

Size: 360x1
Dimensions: i
Datatype: int32
Attributes:
 units = '1'
 long_name = 'cell index along first dimension'

latitude

Size: 360x291
Dimensions: i,j
Datatype: double
Attributes:
 standard_name = 'latitude'
 long_name = 'latitude'
 units = 'degrees_north'

```

        missing_value = 1e+20
        _FillValue     = 1e+20
        bounds         = 'vertices_latitude'

longitude
    Size:          360x291
    Dimensions:    i,j
    Datatype:      double
    Attributes:
        standard_name = 'longitude'
        long_name      = 'longitude'
        units          = 'degrees_east'
        missing_value = 1e+20
        _FillValue     = 1e+20
        bounds         = 'vertices_longitude'

vertices_latitude
    Size:          4x360x291
    Dimensions:    vertices,i,j
    Datatype:      double
    Attributes:
        units          = 'degrees_north'
        missing_value = 1e+20
        _FillValue     = 1e+20

vertices_longitude
    Size:          4x360x291
    Dimensions:    vertices,i,j
    Datatype:      double
    Attributes:
        units          = 'degrees_east'
        missing_value = 1e+20
        _FillValue     = 1e+20

prra
    Size:          360x291x1032
    Dimensions:    i,j,time
    Datatype:      single
    Attributes:
        standard_name = 'rainfall_flux'
        long_name      = 'Rainfall Flux where Ice Free Ocean over Sea'
        comment        = 'CanNEMO employs a virtual salt flux, but water
flux is used in the free surface formulation, and hence useful to report. This field includes rain falling
on sea ice as seen by CanNEMO., ScenarioMIP_table_comment: In accordance with common usage
in geophysical disciplines, 'flux' implies per unit area, called 'flux density' in physics.'
        units          = 'kg m-2 s-1'
        original_name = 'prlq'
        cell_methods   = 'area: mean where ice_free_sea over sea time: mean'

```

```
cell_measures = 'area: areacello'  
missing_value = 1.000000020040877e+20  
_FillValue     = 1.000000020040877e+20  
coordinates    = 'latitude longitude'
```

(3) Variables used for partial pressure of carbon dioxide

Global Attributes:

```
CCCma_model_hash =
'1f91f92cb6d607391f44831504025d32fc44faa1'
CCCma_parent_runid = 'rc3.1-his01'
CCCma_pycmor_hash =
'33c30511acc319a98240633965a04ca99c26427e'
CCCma_runid = 'rc3.1-s7001'
Conventions = 'CF-1.7 CMIP-6.2'
YMDH_branch_time_in_child = '2015:01:01:00'
YMDH_branch_time_in_parent = '2015:01:01:00'
activity_id = 'ScenarioMIP AerChemMIP'
branch_method = 'Spin-up documentation'
branch_time_in_child = 60225
branch_time_in_parent = 60225
contact = 'ec.cccma.info-info.ccmac.ec@canada.ca'
creation_date = '2019-05-02T03:15:11Z'
data_specs_version = '01.00.29'
experiment = 'gap-filling scenario reaching 7.0 based on SSP3'
experiment_id = 'ssp370'
external_variables = 'areacello'
forcing_index = 1
frequency = 'mon'
further_info_url = 'https://furtherinfo.es-
doc.org/CMIP6.CCCma.CanESM5.ssp370.none.r1i1p1f1'
grid = 'ORCA1 tripolar grid, 1 deg with refinement to 1/3
deg within 20 degrees of the equator; 361 x 290 longitude/latitude; 45 vertical levels; top grid cell
0-6.19 m'
grid_label = 'gn'
history = '2019-05-02T03:15:11Z ;rewrote data to be
consistent with ScenarioMIP for variable spco2abio found in table Omon.;
Output from $runid'
initialization_index = 1
institution = 'Canadian Centre for Climate Modelling and Analysis,
Environment and Climate Change Canada, Victoria, BC V8P 5C2, Canada'
institution_id = 'CCCma'
mip_era = 'CMIP6'
nominal_resolution = '100 km'
parent_activity_id = 'CMIP'
parent_experiment_id = 'historical'
parent_mip_era = 'CMIP6'
parent_source_id = 'CanESM5'
parent_time_units = 'days since 1850-01-01 0:0:0.0'
parent_variant_label = 'r1i1p1f1'
```

physics_index	= 1
product	= 'model-output'
realization_index	= 1
realm	= 'ocnBgchem'
references	= 'Geophysical Model Development Special issue on CanESM5 (https://www.geosci-model-dev.net/special_issues.html)'
source	= 'CanESM5 (2019): aerosol: interactive atmos: CanAM5 (T63L49 native atmosphere, T63 Linear Gaussian Grid; 128 x 64 longitude/latitude; 49 levels; top level 1 hPa) atmosChem: specified oxidants for aerosols land: CLASS3.6/CTEM1.2 landIce: specified ice sheets ocean: NEMO3.4.1 (ORCA1 tripolar grid, 1 deg with refinement to 1/3 deg within 20 degrees of the equator; 361 x 290 longitude/latitude; 45 vertical levels; top grid cell 0-6.19 m) ocnBgchem: Canadian Model of Ocean Carbon (CMOC); NPZD ecosystem with OMIP prescribed carbonate chemistry seaIce: LIM2'
source_id	= 'CanESM5'
source_type	= 'AOGCM'
sub_experiment	= 'none'
sub_experiment_id	= 'none'
table_id	= 'Omon'
table_info	= 'Creation Date:(20 February 2019) MD5:374fbe5a2bcca535c40f7f23da271e49'
title	= 'CanESM5 output prepared for CMIP6'
variable_id	= 'spco2abio'
variant_label	= 'r1i1p1f1'
version	= 'v20190429'
license	= 'CMIP6 model data produced by The Government of Canada (Canadian Centre for Climate Modelling and Analysis, Environment and Climate Change Canada) is licensed under a Creative Commons Attribution ShareAlike 4.0 International License (https://creativecommons.org/licenses). Consult https://pcmdi.llnl.gov/CMIP6/TermsOfUse for terms of use governing CMIP6 output, including citation requirements and proper acknowledgment. Further information about this data, including some limitations, can be found via the further_info_url (recorded as a global attribute in this file) and at https://pcmdi.llnl.gov/ . The data producers and data providers make no warranty, either express or implied, including, but not limited to, warranties of merchantability and fitness for a particular purpose. All liabilities arising from the supply of the information (including any liability arising in negligence) are excluded to the fullest extent permitted by law.'
cmor_version	= '3.4.0'
tracking_id	= 'hdl:21.14100/5f30979d-9927-4f27-9bdd- 655f40af474a'

Dimensions:

time = 1032 (UNLIMITED)
bnds = 2
j = 291
i = 360
vertices = 4

Variables:

time

Size: 1032x1
Dimensions: time
Datatype: double
Attributes:
 bounds = 'time_bnds'
 units = 'days since 1850-01-01 0:0:0.0'
 calendar = '365_day'
 axis = 'T'
 long_name = 'time'
 standard_name = 'time'

time_bnds

Size: 2x1032
Dimensions: bnds,time
Datatype: double

j

Size: 291x1
Dimensions: j
Datatype: int32
Attributes:
 units = '1'
 long_name = 'cell index along second dimension'

i

Size: 360x1
Dimensions: i
Datatype: int32
Attributes:
 units = '1'
 long_name = 'cell index along first dimension'

latitude

Size: 360x291
Dimensions: i,j
Datatype: double
Attributes:
 standard_name = 'latitude'
 long_name = 'latitude'
 units = 'degrees_north'

```

missing_value = 1e+20
_FillValue     = 1e+20
bounds         = 'vertices_latitude'

longitude
  Size:          360x291
  Dimensions:    i,j
  Datatype:      double
  Attributes:
    standard_name = 'longitude'
    long_name     = 'longitude'
    units         = 'degrees_east'
    missing_value = 1e+20
    _FillValue    = 1e+20
    bounds       = 'vertices_longitude'

vertices_latitude
  Size:          4x360x291
  Dimensions:    vertices,i,j
  Datatype:      double
  Attributes:
    units         = 'degrees_north'
    missing_value = 1e+20
    _FillValue    = 1e+20

vertices_longitude
  Size:          4x360x291
  Dimensions:    vertices,i,j
  Datatype:      double
  Attributes:
    units         = 'degrees_east'
    missing_value = 1e+20
    _FillValue    = 1e+20

depth
  Size:          1x1
  Dimensions:
  Datatype:      double
  Attributes:
    units         = 'm'
    axis          = 'Z'
    positive      = 'down'
    long_name     = 'depth'
    standard_name = 'depth'

spco2abio
  Size:          360x291x1032
  Dimensions:    i,j,time
  Datatype:      single

```

Attributes:

```
standard_name =  
'surface_carbon_dioxide_abiotic_analogue_partial_pressure_difference_between_sea_water_and_  
air'  
  
long_name = 'Abiotic Surface Aqueous Partial Pressure of CO2'  
comment = 'The surface called 'surface' means the lower  
boundary of the atmosphere. The chemical formula for carbon dioxide is CO2. In ocean  
biogeochemistry models, an 'abiotic analogue' is used to simulate the effect on a modelled variable  
when biological effects on ocean carbon concentration and alkalinity are ignored. The partial  
pressure of a gaseous constituent of air is the pressure which it alone would exert with unchanged  
temperature and number of moles per unit volume. The partial pressure of a dissolved gas in sea  
water is the partial pressure in air with which it would be in equilibrium. The partial pressure  
difference between sea water and air is positive when the partial pressure of the dissolved gas in sea  
water is greater than the partial pressure in air.'  
  
units = 'Pa'  
original_name = 'spco2a'  
history = 'mltby0.101325 2019-05-02T03:15:11Z altered by  
CMOR: Treated scalar dimension: 'depth'.'  
  
cell_methods = 'area: mean where sea time: mean'  
cell_measures = 'area: areacello'  
coordinates = 'depth latitude longitude'  
missing_value = 1.000000020040877e+20  
_FillValue = 1.000000020040877e+20
```

Appendix II The modified figures in manuscript

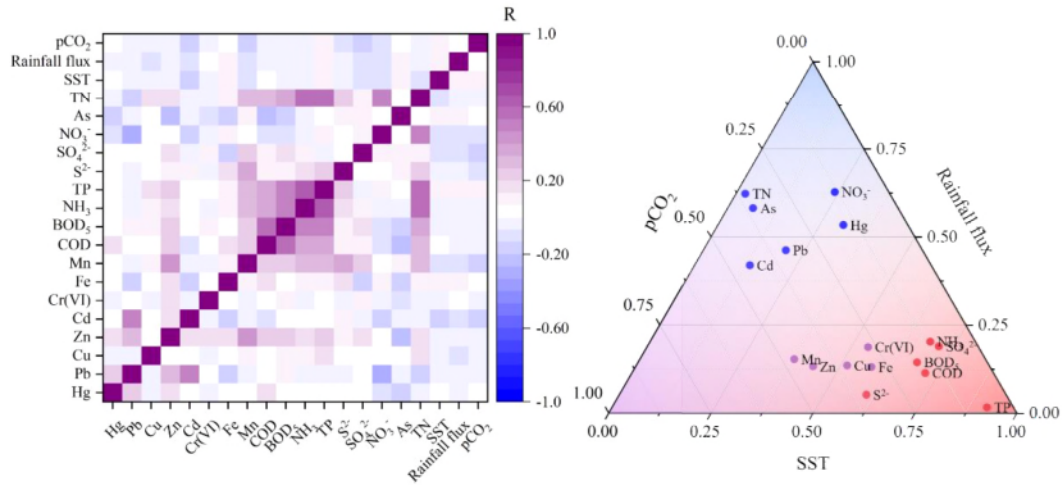
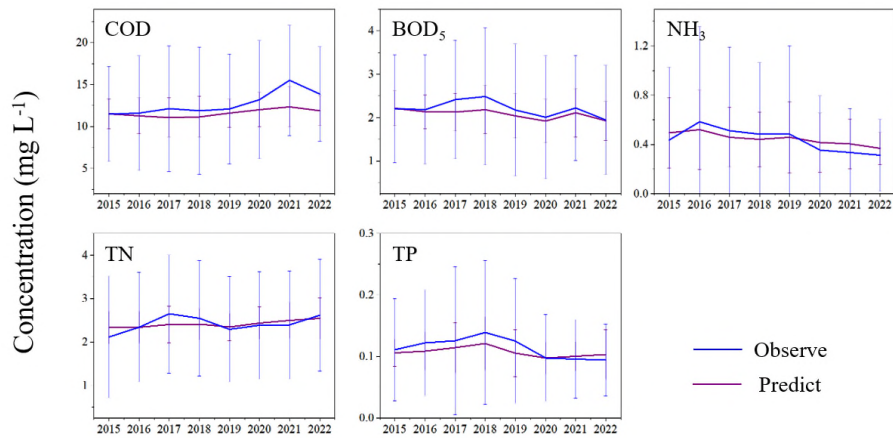


Fig. 1. (a) The correlation of each index to these three variables. (b) The feature importance of each index to these three variables. It depends on the use, chemical properties, and migration trend of the elements.

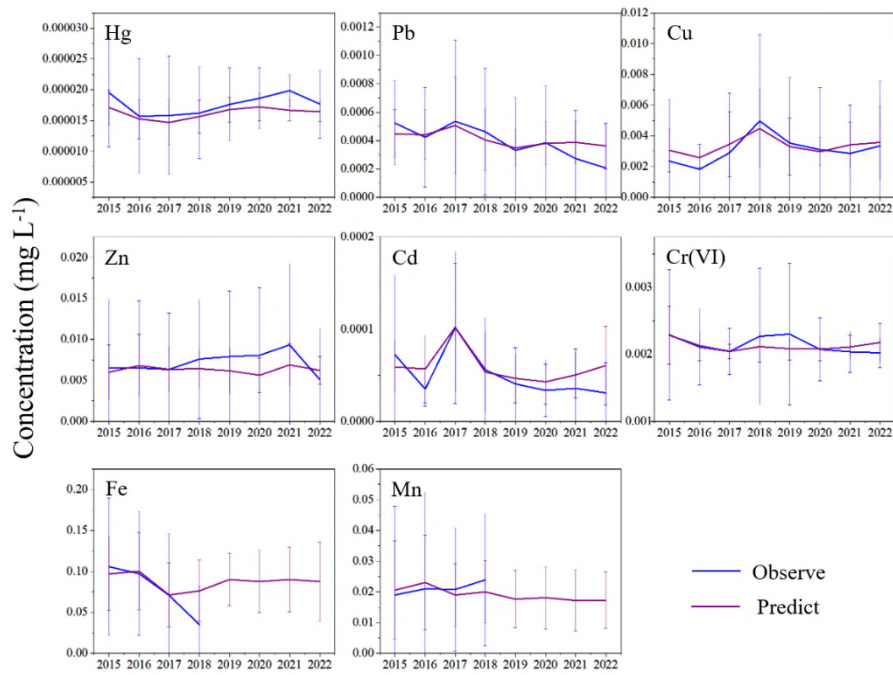
Note:

(a): Double correlation analysis of key response factors of climate change and elements in coastal water bodies. Positive correlations are indicated by purple, while negative correlations by blue. The intensity or darkness of the color represents the strength of the correlation, with darker colors signaling a stronger correlation;

(b): According to the difference in response to variables, elements are divided into three categories, in which red represents SST-sensitive elements, blue represents sedimentary-sensitive elements, and purple represents elements that play multiple roles in the geochemical process.



(a)



(b)

Fig. 2. Trends in simulated results (blue line) and measured values (purple line) of the receptor model (2015–2022). Error bars represent parallel samples of multiple sampling points. Part (a) represents nutrients, and part (b) represents TMs.

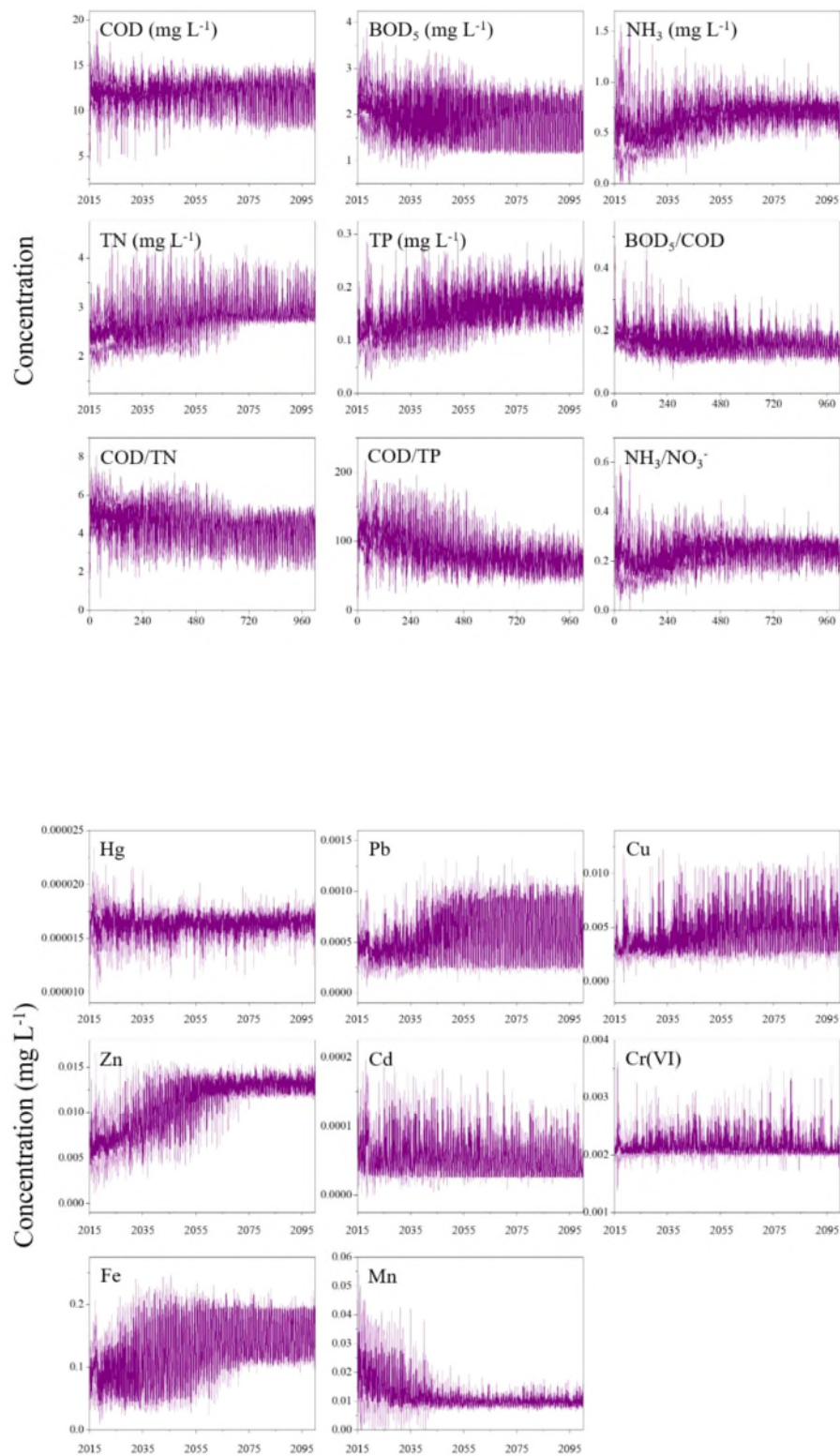


Fig. 3. The trend of dissolved phase concentration of some water quality indices in 2015–2099.
Error bars represent parallel samples of multiple sampling points.

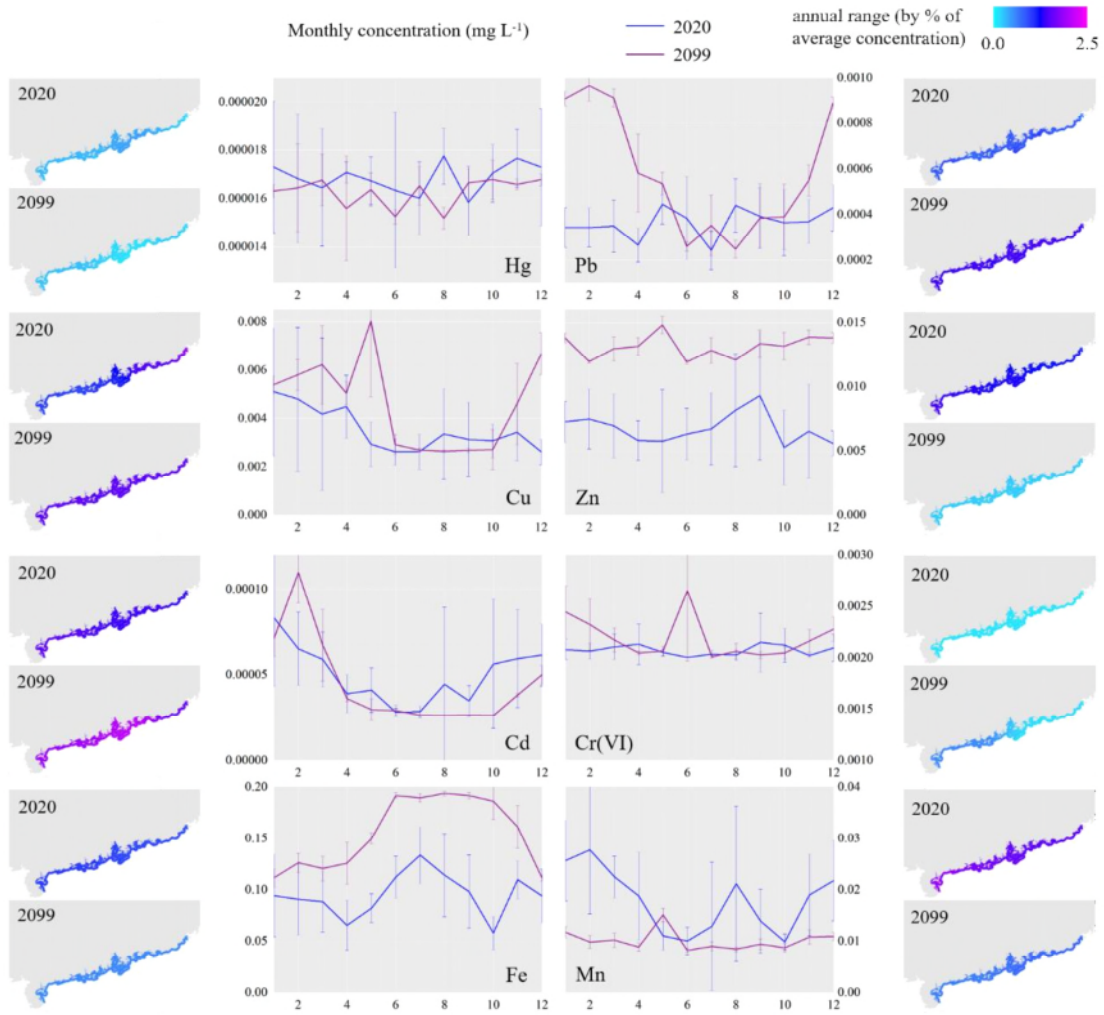


Fig. 4 The monthly variation of metal concentrations in the aqueous phase of the study area. The annual range of concentrations represents the ratio of the difference between the maximum and minimum months to the annual average concentration ($R = \frac{C_{max}-C_{min}}{C_{avg}}$).

Review 449403_0

This manuscript explores the issue related to long-term changes in coastal seawater quality due to climate change. The authors used an 8-year dataset to make predictions about the changes in water quality in the coastal area of the Guangdong Province (China) for the period 2015-2022. The authors carried out a notable effort for modeling a huge dataset which may be useful to predict long-term changes in the water quality of the investigated coastal area.

The general topic of the manuscript is undoubtedly original, interesting and useful.

However, the manuscript itself has many shortfalls, especially regarding its structure. Generally, too much information was provided (both in the main text and in the Supplementary Material, often repeating the same statements in both documents) without following a clear structure able to make the reader able to effectively understand which are the main aims of the study and the main results.

The section Materials and Methods was placed before the section focused on the results. Why? In my opinion, this is confusing as well as part of the discussion about PTE behaviour in the Supplementary Material and not in the main text. The sampling strategy was also placed in the Supplementary Material and it is not clear what was actually performed since the text appears like a standard sampling protocol.

The manuscript is really too long and some statements are repeated in different sections of the manuscript and/or in the Supplementary Material. This is confusing for the reader. Actually, the Supplementary Material seems another manuscript (76 pages!!). It should be notably shortened. Some sections in the supplementary material should be summarised and moved in the full text avoiding repetition (sections A and B).

Generally, I suggest the authors to carefully revise the text (main text and Supplementary Material) to organise its different sections in a clearer way, avoiding repetition, selecting the discussion about PTE behaviour strictly related to the main aims/results of their study and better highlighting the most relevant results and take home messages. I suggest the authors to clearly state that the dataset employed for predictions refers to a specific coastal area, providing a brief description of the investigated area and of the sampling strategy in the main text.

Terminology should be also improved through the text, especially regarding the topic related to the phase partitioning of trace elements (or nutrients) between dissolved and solid fractions.

The authors often referred to “trace elements” through the text. I suggest to specify which are these trace elements in the abstract and at the beginning of the introduction.

I am not a modelist, so I cannot provide a careful and specific revision in this sense.

I retain that this manuscript can be accepted for publication in the Journal Nature Communications following major revision. Please, consider the previous and following general comments as well as specific comments on the PDF version of the manuscript:

Lines 19-29 (Abstract). I suggest the authors to better prepare the abstract clearly stating which are the aims of their research and briefly summarising their main results. Indeed, some statements in the abstract are not clearly explained. For instance, the authors stated that an 8-year dataset was employed to make predictions about changes in water quality in this offshore area... this is too general: what is this area? I think that this should be specified in the abstract. Then, the authors stated that “trace elements will be displaced by calcium and magnesium, leading to re-suspension in the water”. I do not understand what the authors meant with this sentence. They are talking about dissolved trace elements? Which elements? About trace element associated to suspended particles? About the partitioning behaviour of trace elements between dissolved and solid phases? Please, clarify

Line 51 What do the authors meant with “concentration lever”?

Line 68 “metal elements”. Which elements? Heavy metals or potentially toxic elements? I suggest the authors to use “trace elements or potentially toxic elements” through the text.

Lines 73-74 this affects the trace element behaviour at the sediment-water interface? Or the trace element phase partitioning between dissolved and the suspended particulate matter along the water column? This is not clear, please specify.

Lines 76-77 What do the authors meant with “suspended elements”? elements associated to the suspended particulate matter?

Line 80: Nitrogen (N) and S deposition has become one of the factors promoting coastal eutrophication?

Line 82: absorption or adsorption?

Line 85: “As the invading river water is anoxic”. In general? Or in the case of a specific river? Anoxia is quite uncommon in riverine environments. Please, clarify.

Lines 97-99 To which elements the authors are referring to? It is not clear.

Line 191 or declining trend, as predicted by the environmental Kuznets curve. Please, provide a brief explanation.

Lines 227-228 what the authors meant with “the initial field”? please, clarify

Line 262 in this section the authors often refers to sensitive elements. I do not completely understand which are these sensitive elements. Please, clarify

Lines 335-336 Dissolved Ca and Mg? I do not understand what do the authors meant with “will be replaced into trace elements”? In the water fraction or in the SPM? Please, clarify

Lines 346-348 not clear. In which environment and under what boundary conditions? The interaction between trace elements and sulfides is affected by several factors. Trace element precipitation along with sulfides usually occurs under specific boundary conditions (sulfate reduction, anoxia).

Line 350 deposited where? Adsorbed on the surface of the SPM and accumulated in the sediment? Please, clarify

Lines 353-375 in this section the authors referred to the seasonal cycle of trace elements mainly from a biological point of view. However, the biogeochemical cycle of trace elements is also affected by other factors. Moreover, it is not clear if the authors referred to dissolved trace elements, trace elements associated with the suspended particles or both.

Line 416 This section “Materials and Methods” should be placed before the discussion of the results. The same for the section “the establishment and accuracy of the model”.

SA. 1 The sampling description in supplementary material may be notably improved. In my opinion, it sounds like a standard sampling protocol since the authors often referred to operations that should be done for sampling (the ship should be selected, samplers should collect, it should be carried out). The authors should provide a description of the sampling operations which were actually performed and not simply list the operations that should be done for sampling. Moreover, in the section focused on special samples, the sampling strategy for petroleum samples was described although any data about this samples was reported in the main text. The authors also referred to “special storage containers” and “preservatives” for different analytes. Please, specify which containers and preservatives were used. Which analytical technique was employed for PTE determination? What about their LOD?

SB. 1 This section is focused on “Coordinates and classification of sampling sites”. However, the behaviour of trace elements is discussed. Why the authors decided to place this text in the Supplementary Material and not in the main text?