Editor of *Geoscientific Model Development*

Dear Editor,

we received the Reviewers' reports on our manuscript entitled "Spatio-temporal dynamics of mercury concentration in the Augusta Bay (southern Italy): matching model and data" by G. Denaro, D. Salvagio Manta, A. Borri, M. Bonsignore, D. Valenti, A. Cucco, B. Spagnolo, M. Sprovieri, A. De Gaetano.

We thank you and the Reviewers for the constructive comments and criticisms, which definitively helped us to improve the model and the manuscript. We took into account all the specific points raised by the referees and submit a revised version of the manuscript. Detailed discussions and corrections are listed below. Please note that we highlighted in red words and/or sentences which we removed from the old version, and in blue words and/or sentences inserted in the revised version.

Reviewer 1

A.General

1. **Reviewer** wrote:
   Introduction: I am missing a more comprehensive overview of Hg modelling performed on the scale of the Mediterranean Sea and its parts. The authors do not report any of the 2D and 3D models developed and applied before this study. These models, although not as complex as the presented HR3DHG, were also supported by a hydrodynamic model and performed quite well at the scale of the entire Mediterranean and at smaller scale (Gulf of Trieste, Adriatic Sea) with regard to both transport and transformations of two or three Hg species. My suggestion to the authors would be to investigate the article by Zhu et al. (https://doi.org/10.1016/j.scitotenv.2018.04.397) and the references therein, and to include the previously developed multi-dimensional models into the section Introduction. The same comment is valid for the chapter Discussion: 2D and 3D models were used before the HR3DHG model.

   Authors answer:
   The Introduction section has been modified according to the reviewer's suggestions. Specifically, we inserted the following paragraph at page 3, lines 49-67, of the new version of manuscript:
   "In general, the appropriate modelling to reproduce the spatial and temporal variability of Hg species in highly heterogeneous marine ecosystems, such as Augusta Harbour, requires the use of a hydrodynamics model integrated with a biogeochemical model (Zagar et al., 2007, 2014). To this aim, Zagar et al. (2007) introduced a PCFLOW3D model upgraded with the biogeochemical module for simulating simultaneously velocity field of marine currents, suspended particles transport and mercury biogeochemical transformations for the whole Mediterranean Sea. (...) Among these, Zhang et al. (2014) reproduced the $[Hg_T]$ in oceans and calculated a Hg mass balance by using a 3D ocean tracer model (OFFTRAC) coupled with a general circulation model (GEOS-Chem) (Zhang et al., 2014). Here, the sinking flux of Hg bound to POM was calculated exploiting the remote sensing data for net primary production (NPP) and chlorophyll concentration, which are associated to phytoplankton abundance."
   We also added the following two new sentences at page 26 of Discussion section:
   "For comparison, the different approach used in the WASP models did not allow to reproduce the dynamics of mercury concentration distribution at 3D high resolution in polluted sites characterized by elevated spatial heterogeneity. (...) In general, only few models (Rajar et al., 2007; Zagar et al., 2007; Canu et al., 2017) were able to make forecasts about the mercury depletion time in the sediment compartment of highly polluted sites, such as Augusta Bay."

2. **Reviewer** wrote:
   Sensitivity analysis confirms high significance of circulation (Line 501). Is therefore the constant-density approach correct? Non-stratified conditions are acceptable in winter months, while the temperature stratification in the summer may significantly influence the circulation and the fluxes through the picnocline. Whether to use stratified or non-stratified conditions depends on temporal resolution applied: with seasonal (or finer temporal resolution) stratified conditions should be taken into account.

Authors answer:

We followed a constant-density approach being the water circulation in the bay mainly ruled by the wind and tidal forcing only. Density gradients and the stratification were not strongly affecting the bay water circulation. This was suggested in previous modelling works of De Marchis et al. (2014) and Lisi et al. (2009). The latter, in particular, suggested that the water circulation in the Augusta bay is influenced mainly by tides and wind and the harbour can be investigated as a lagoon.

In the Section S3.2 of the new version of Supplement, some paragraphs have been inserted to clarify and improve this point.

3. **Reviewer** wrote:

To proceed with the same concern: the temporal and spatial dynamics of the simulations are unclear. Several questions arose during reading:

a) How were data from various seasons (Fig 1) taken into account (for calibration/validation)? Particularly when the constant water-density was accounted for in the model.

b) What is the temporal resolution of hydrodynamics (real-time = hourly, or any other resolution) used in transport simulations, and how often was the velocity field changed in a long-term simulation? Was a perpetual year used or did the conditions change (using any of the possible IPCC scenarios for changing climate conditions or anything similar)? A 250-year simulation would require an explanation of the applied parameters.

c) When adapting hydrodynamics from the SHYFEM model to the HR3DHG grid, were the velocities interpolated to the HG grid or integrated over the cells of the HG grid? When using real-time hydrodynamics, the correct transport can only be achieved by integration.

d) A table with temporal dynamics of each of the variables and (environmental) parameters would be useful. I.e. how often are the input parameters changed (annually/seasonally/weekly) and in which way the results were obtained (re-initialisation with experimental data/a single long simulation for 250 years?)

Authors answer:

About the various points:

a) The calibration procedure, together with the experimental data involved, is reported in the Section 3.3 of main text at pages 17-18, lines 463-487. The experimental findings involved in the validation of model results, are listed at the ending of the Section 3.3 of main text.

b) The temporal resolution of hydrodynamics is set to three hours (see page 22 of Section S3.2 of Supplement), as a consequence the velocity field changes 2920 times for each simulation year.

In our simulations, we used a perpetual year (see the ending of Section S3.2 of Supplement).

The most part of parameters does not depend on environmental variables. However, we are aware that some parameters (biological rate constants in seawater, methylation/de-methylation rate constants in sediments, desorption rate etc) are strictly connected with environmental variables (NPP, temperature etc.). Although we know that the model results could change significantly if the effects of climate changes on our parameterization were considered, currently we could not enough and robust information to simulate the mercury dynamics for different future scenarios. Following suggestions from the second reviewer, we also removed any reference to the climate changes.

c) The velocities obtained by the SHYFEM model were interpolated to the HR3DHG grid. Specifically, the velocity field used as the dataset for the interpolation was derived from a time average procedure of the SHYFEM model output produced at hourly frequency in order to have a three-hours residual velocity field. The obtained dataset consisting in three-hours time averaged velocity field was then used as input data for the interpolation. Considering the circulation within the bay is mostly homogeneous and the spatial discretization used by the HR3DHG comparable to the spatial scale of the flow variability, the adopted interpolation procedure was sufficiently accurate to reproduce the original hydrodynamic model dataset time and space variability.

The detailed interpolation procedure is reported at page 22, lines 561-566, of the Supplement.

d) The Table S2 (environmental variables used in the model) has been modified according to the reviewer's indications. The model results were obtained by running a single long simulation for 250 years (see pages 16-17, lines 443-446, of

the new version of manuscript).

4. **Reviewer** wrote:
Another question is the agreement of simulated and experimental results:
a) The complexity of the model requires thorough verification, calibration and validation. In order to confirm an "excellent agreement of the model with experimental data", validation of the model should have been performed with calibrated parameters. Was such a procedure done and if yes, on which temporal scale? The latest available experimental data are from 2017, and the modelling results for 2017 can be reproduced from the initial 2005, 2011 or 2012 experimental data. How different would be the modelling results for 2017 using the same set of calibrated parameters? In any case, it is very difficult to justify results of a 250-year long simulation even without the climate and other environmental changes that may occur in such a long time interval.
b) Several statistical methods for evaluation of model efficiency (Nash-Sutcliffe, Kling-Gupta, rmse) can be applied in order to quantify the agreement with experimental data. The results of these tests would give a better impression on the model performance than qualitative description by using excellent/good/poor based on visual agreement between figures.

Authors answer:
About the various points:
a) The validation of model results with calibrated parameters has been performed (see Section 4.1 of the new version of manuscript). Specifically, the theoretical results for $[MeHg_D]$, $[Hg_D]$ and $[Hg_T]$ were validated with the respective experimental data measured in seawater between May 2011 and October 2017, while all annual Hg fluxes obtained by modelling were validated with those estimated empirically by Salvagio Manta et al. (2016) for the sampling period May 2011- June 2012.
All model results have been obtained by using the same set of calibrated parameters.
About climate changes, we know that the model results could change significantly if the effect of environmental changes was considered, however we would need more information to simulate the mercury dynamics for the different future scenarios. See comments reported in the previous point.
b) The results of some statistical checks performed on $[MeHg_D]$, $[Hg_D]$ and $[Hg_T]$ are reported at pages 19-20 of the new version of manuscript. Specifically, a statistical analysis based on the χ^2 test is now introduced for $[MeHg]$, while a quantitative comparison between the model results and field observations for the $[Hg_D]$ and $[Hg_T]$ is performed on the basis of observed experimental error. In fact, we could not make the χ^2 test for $[Hg_D]$ and $[Hg_T]$ since their magnitude were below the detention limit in many sampling points. Moreover, the most part of the experimental data for $[Hg_D]$ and $[Hg_T]$ were acquired only in two/three sampling points for each station. For these reasons, we chose to make an alternative statistical check for $[Hg_D]$ and $[Hg_T]$.

5. **Reviewer** wrote:
Mass balance (Table S9 and Conclusions lines 544-545):
a) In the section Conclusions (line 544) the authors discuss the mass balance, which has never been established and presented. A mass balance should consist of quantities of the species under consideration (inventories) and fluxes, and in most cases, (see the references in Zhu et al.) such balances are presented in graphical form.
b) What is the inventory of (at least HgT) in the domain and in each of the compartments (water/sediment)? How do the fluxes affect the inventory? All that is evident from the numbers in the Table S9 is the constant decrease of the fluxes. Is the presented mass balance obtained solely from the results of the model or is it supported by experimental results? Furthermore, is the annual balance closed or open? With steadily decreasing fluxes and the deposition remaining more or less unchanged (term AD in Table S9), the inputs and the outputs should balance once in the future. When?

Authors answer:
About the various points:

a) The mass balance for Hg_T is introduced in the Section 3.1.1 of the new version of manuscript, while the scheme of transport processes (fluxes) at the boundaries of domain is reported in the new Figure 3 (see also the new caption of Figure 3). Moreover, we recall that the annual fluxes of $MeHg$ are estimated by our model, even if they cannot validated with experimental findings on the contrary of Hg_T. For this reason, the mass balance for MeHg has been not reported in the paper.

b) The Hg_T mass balance is performed only for seawater compartment.

The effects of the fluxes on the mercury concentration are described throughout the Section 4.1 of the new version of the manuscript. For example, we reported at page 20 the following assertion:

" It should be noted that the model results suggest that the benthic Hg_D fluxes are mainly generated by the diffusion process at the seawater-sediment interface and that the amount of Hg_D release from the re-suspended particulate matter is negligible."

Also, we inserted the following sentence at page 24 of the new version of manuscript:

" In general, the contribute of AD is negligible in the mercury mass balance of the Augusta Bay. Indeed, the simulations indicate that a strong increase of atmospheric mercury deposition caused by environmental changes (dust fall increase and/or rainfall increase), would not affect on numerical results of our model significantly."

About the presented mass balance, we recall that the annual mercury fluxes obtained by our model are compared with those obtained by Salvagio Manta et al. (2016) using the experimental data collected during the sampling period May 2011- June 2012 (see pages 21-23 of the new version of manuscript). The results of this comparison indicates a good agreement only for the mercury evasion flux (V) at atmospheric-seawater interface.

The Hg_T mass balance is open. According to this, we inserted three new paragraphs at page 24 of the new version of manuscript:

"In this work, the annual recycled mercury flux (D) is calculated by subtraction using the mass balance equation (18), (...). Here, values calculated by our model (2.50 $kmol\ y^{-1}$ for the year 2011 and 2.46 $kmol\ y^{-1}$ for the year 2012) are larger and probably more realistic than those estimated in Salvagio Manta et al. (2016) (0.84 $kmol\ y^{-1}$). (...)

In order to reproduce the effects induced by scavenging process on the mercury dynamics, our model calculates the annual sinking mercury flux, whose results are shown in Fig. 6d. Here, a significant gap between the recycled flux (2.50 $kmol\ y^{-1}$ for the year 2011) and the sinking flux (0.07 $kmol\ y^{-1}$ for the year 2011) is observed probably due to the underestimation of the amount of mercury captured by POM (see Eqs. (4)-(5)). (...)

On the contrary, very high values of the annual Hg_T accumulation rate in surface sediment layer (12.07 $kmol\ y^{-1}$ for the year 2011), respect to those of the annual recycled flux (2.50 $kmol\ y^{-1}$ for the year 2011), are obtained by our model. (...) In fact, the results obtained by the sediment transport model indicate a low average sedimentation rate for the Augusta Bay."

The benthic mercury flux decreases slowly at quasi-steady state (0.12 percent for $t_{max} = 250\ years$). Therefore, it will remain quite high for a very long time (more than 250 years). As a consequence, we aspect that the balance between the inputs and the outputs may occur only after several centuries.

B. Details (manuscript):

6. **Reviewer** wrote:

Line 73: Rajar et al. (doi:10.1016/j.marchem.2006.10.001) and Zagar et al. (DOI 10.1007/s11356-013-2055-5) established two (annual) Hg mass balances in the Mediterranean Sea. There, the atmospheric deposition and the rivers contributions were found to be significantly more important than any of the point sources. In order to support the statement that "the Augusta Bay has a key role in Mediterranean Hg inventory" this role should be quantified and compared to the previously published values.

Authors answer:

According to the reviewer's indication, we inserted the following sentence at page 5, lines 121-123, of the new version of manuscript:

"The estimate of the Hg export from Augusta Bay to the open sea (0.54 $kmol\ y^{-1}$, Salvagio Manta et al., 2016), corresponds to about 4% of total input from coastal point/diffuse sources to the Mediterranean Sea (12.5 $kmol\ y^{-1}$, Rajar et al., 2007)."

7. **Reviewer** wrote:
Lines120-121: The sentence explaining why the results were unaffected by the chosen initial condition is not clear.

Authors answer:
The paragraph on the initial condition has been moved in the "Model and simulation setup" Section according to the second reviewer's indication. Moreover, we modified the sentence at page 16, lines 441-442, of the new version of manuscript, as follows:
" The numerical results were not affected by the chosen initial conditions, indeed the same spatial distribution of $[Hg]$ at nearly-steady state was obtained when higher initial Hg concentrations than detection limit were fixed."

8. **Reviewer** wrote:
Line 149: What is the temporal resolution of hydrodynamics? I.e., how many different velocity fields were used for computing transport?

Authors answer:
The temporal resolution of hydrodynamics is set to three hours (see page 22 of Section S3.2 of Supplement), as a consequence the velocity field changes 2920 times for each simulation year.

9. **Reviewer** wrote:
Lines 320-340: Were the results of the calibration procedure constant or variable (in time) input parameters? If temporally constant, for which period (set of measurements). Were the same constant coefficients used for another time interval between measurements? If variable, on what temporal scale?

Authors answer:
The input parameters obtained by the calibration procedure were temporally constant. Since the acquisitions of all experimental data were performed for time limited periods, we used for each calibrated parameter different set of measurements collected during different sampling periods. The same constant coefficients were used for the whole time simulation. Conversely, the photo-chemical and biological rate constants changed as a function of time due to the seasonal oscillations of RAD and NPP (see Section S1 of Supplement).

10. **Reviewer** wrote:
Figure 6: Shows a decreasing trend for all fluxes. The Hg inventory in the Bay is most probably decreasing as well (unfortunately the mass balance is not established in a way to either confirm or contradict this hypothesis). Were these results obtained by accounting for computed or measured deposition? There is a high discrepancy (factor2.5) between these two values. As reported by several previous modelling/mass balance studies (Zhu et al., and the references therein), deposition is a very important source of Hg in the Mediterranean. Including deposition into the performed sensitivity analysis would be very useful for clarification of this question.

Authors answer:
The reviewer is right. The Hg concentrations follow the same decreasing trend of all fluxes. The annual mass balance for mercury is now described in complete way throughout the Section 4.1 of new version of manuscript. In particular, we explain the reason of high discrepancy between our model result and that estimated in experimental work by Salvagio Manta et al. (2016), at page 22, lines 581-586, of new version of manuscript:
"This discrepancy is due to different calculation methods used in the two works. Specifically, in our model the AD is calculated by using both the atmospheric mercury concentrations and the average precipitations, measured for all months of the year. On the contrary, in Bagnato et al. (2013) the AD is calculated by averaging the experimental data acquired during a time limited sampling period (from 29^{th} August 2011 to 23^{th} April 2012), namely without considering the year period in which the amount of precipitation is very low. By this way, the AD obtained by Bagnato et al. (2013) is very higher than that of our model, even if it is probably overestimated due to calculation method used."
Moreover, a rough sensitivity analysis has been performed on the atmospheric deposition calculated in our model. According to this, we inserted at page 24, lines 586-589, of new version of manuscript the two following sentences:
" In general, the contribute of AD is negligible in the mercury mass balance of the Augusta Bay. Indeed, the simulations

indicate that a strong increase of atmospheric mercury deposition caused by environmental changes (dust fall increase and/or rainfall increase), would not affect on numerical results of our model significantly."

C. Details (Supplement):

11. **Reviewer** wrote:
References to Figs in the supplement should be noted as eg. Fig S5, not Fig 5.

    Authors answer:
    We modified the references to Figures in the Supplement.

12. **Reviewer** wrote:
In several equations the annual flux is debated. Were fluxes calculated also on seasonal (or finer) temporal scale?

    Authors answer:
    The fluxes were calculated for each node of 3D grid using a finer temporal scale. Specifically, the our code allowed to print all fluxes for each node of grid and for each month of year.

13. **Reviewer** wrote:
Equations S8 and S21: Where is the dry deposition, as the first term in line 59 and the most right-hand term in S21 only have the wet part, connected to precipitation P?

    Authors answer:
    Following the reviewer's suggestions, in new simulations we considered the dry deposition of Hg^{II} and $MeHg$ at atmosphere-seawater interface, while only the wet deposition is taken into account for Hg^0. According to this, we modified the text and the equations of Sections S1.1.1, S1.2.1 and S1.3.1 of Supplement.

14. **Reviewer** wrote:
Equations S23 (and S37): if tortuosity is not taken into account in neither Dw-in (or Dw-or) nor δ_w , please explain whether and how this is compensated in the equation.

    Authors answer:
    We better explained the relation between tortuosity and porosity at page 10, lines 230-231, of the new version of Supplement.

15. **Reviewer** wrote:
Tables S4, S10: The presented concentrations are given for 2011 and 2017. Any other comparison possible?

    Authors answer:
    No other comparison is possible.

Reviewer 2

1. **Reviewer** wrote:
L4-5: why is the sediment module presented as two different models?

    Authors answer:
    There is only one model for the sediment compartment. To clarify this point, we modified the sentence at page 1, lines 4-6, of Abstract as follows:
    "... an advection-diffusion-reaction model for the dissolved mercury in the seawater compartment coupled with a diffusion-reaction model for dissolved mercury in the pore water of sediments, in which the de-sorption process for the sediment total mercury is taken into account."

2. **Reviewer** wrote:
L5: an adsorption/desorption model would need an adsorption and desorption rate constants, while here do you have a desorption rate constant combined with the Kd. I am concerned about this approach, given that the Kd represents the ratio between adsorption and desorption rate constants at the equilibrium. Can you provide a theoretical background for the parameterization chosen?

Authors answer:
The purpose of diffusion-reaction model was to reproduce the spatio-temporal behaviour of dissolved mercury in the pore water of sediments in quasi-steady conditions (instead of the customary stationary conditions used in other models). To this aim, in the model differential equations we did need to consider both the sediment - pore water distribution coefficients and the desorption rate for the total mercury concentration in the sediment. The former described the ratio between adsorption and desorption rate constants at the steady state without considering pertubations induced by mercury concentration reduction in pore water. The latter reproduced the effects of these pertubations on the solid phase of the sediments. The sediment - pore water distribution coefficients were fixed by using the experimental data, while the desorption rate for the total mercury concentration in the sediment was calibrated to obtain the best fit with the experimental mercury concentrations in pore water.
According to this, we inserted three sentences at pages 3-4, lines 84-88, of the new version of manuscript.

3. **Reviewer** wrote:
L5-7 "the spatio-temporal variability of dissolved and total mercury concentration both in seawater ([HgD] and [HgT]) and first layers of bottom sediments ([HgsedD] and [HgsedT]), and the Hg fluxes at the boundaries of the 3D model domain have been theoretically reproduced, showing an excellent agreement with the experimental data". This sentence is not clear and misleading. It should be said what is simulated with the biogeochemical model (HgD) and what is estimated with other methods (HgT). Avoid claiming excellent agreement that is not supported by facts.

Authors answer:
To clarify this point, we modified the sentences at page 1, lines 5-10, of Abstract as follows:
"The spatio-temporal variability of mercury concentration both in seawater ($[Hg_D]$) and first layers of bottom sediments ($[Hg_D^{sed}]$ and $[Hg_T^{sed}]$), and the Hg fluxes at the boundaries of the 3D model domain have been theoretically reproduced, showing an acceptable agreement with the experimental data, collected in multiple field observations during six different oceanographic cruises. Also, the spatio-temporal dynamics of total mercury concentration in seawater have been obtained by using both model results and field observations." Moreover, the word "excellent" has been replaced by the word "acceptable" throughout the whole manuscript.

4. **Reviewer** wrote:
L8-9 "The mass-balance of the different Hg species in seawater has been calculated for the Augusta Harbour, improving previous estimations" I only found the budget for HgT, not for other Hg species.

Authors answer:
Only the budget for HgT is calculated. In accordance with the reviewer's comment, we modified the sentence as follows: "The mass-balance of the total Hg in seawater has been calculated for the Augusta Harbour, improving previous estimations."

5. **Reviewer** wrote:
L10-11 "The HR3DHG 10 model includes modules that can be implemented for specific and detailed exploration of the effects of climate change on the spatio-temporal distribution of Hg in highly contaminated coastal-marine areas." This is never shown or discussed in the manuscript, except for a couple of similar mentions at the end of the Introduction and Discussion, so I do not find it relevant nor true.

Authors answer:
The effects of climate changes were not discussed in the manuscript. We removed the sentence. Moreover, we modified the ending of the abstract as follows:

"The HR3DHG model could be used as an effective tool to predict the spatio-temporal distributions of dissolved and total mercury concentrations, while contributing to better assess the hazard for environment and therefore for human health in highly polluted areas."

6. **Reviewer** wrote:
L18: why sophisticated?

Authors answer:
We replaced the word "sophisticated" with the word "innovative".

7. **Reviewer** wrote:
L20: Among the references cited to support this sentence, only the work Melaku Canu et al., (2015) is based on the WASP model. Zhang et al., (2014) do not use a box model, but a 3D ocean tracer model (OFFTRAC) coupled with a general circulation model. Ciffroy et al., (2015) use the MERLIN-Expo model, as explained just below by the authors. Other works based on the WASP model are Canu and Rosati (2017) and Rosati et al., (2018). There are also other applications of biogeochemical models specific for mercury in water (and thus more relevant here than the MERLIN-Expo model) that are never mentioned. I suggest modifying the paragraph and correcting the references after carrying out a more systematic review.

Authors answer:
We correct the references throughout the paragraph. We also modified the paragraph as follows:
"Over the last few years some theoretical studies have offered innovative tools to reproduce the mass balance and the dynamics of $[Hg]$ in the marine environment by means of biogeochemical models based on interconnected zero dimensional boxes, representing water or sediment compartments: among these are the River MERLIN-Expo model (Ciffroy et al.,(2015)) and the WASP (Water Analysis Simulation Program) model (Melaku Canu et al., (2015), Canu and Rosati (2017) and Rosati et al., (2018)). (...) The WASP models have been used to simulate the Hg cycle within aquatic ecosystems characterized by well-mixed water layers and homogeneous sediment layers coupled through the boundary conditions at the water-sediment interface (Melaku Canu et al., 2015, Canu and Rosati, 2017, Rosati et al. 2018). In particular, a WASP model applied to a 1D domain and calibrated by using experimental data for dissolved Hg and $MeHg$, allowed to explore $[Hg]$ dynamics in the Black Sea (Rosati et al. 2018)."
Moreover, we inserted in page 3, lines 49-67, the following new paragraph:
"In general, the appropriate modelling to reproduce the spatial and temporal variability of Hg species in highly heterogeneous marine ecosystems, such as Augusta Harbour, requires the use of a hydrodynamics model integrated with a biogeochemical model (Zagar et al. (2007)). (...) Here, the sinking flux of Hg bound to POM was calculated exploiting the remote sensing data for net primary production (NPP) and chlorophyll concentration, which are associated to phytoplankton abundance."

8. **Reviewer** wrote:
L21: As before, the citation "Zhang et al., (2014)" is unrelated to the sentence.

Authors answer:
The reference has been removed. Moreover, we inserted the citation "Canu and Rosati (2017)".

9. **Reviewer** wrote:
L21: I suggest to change "WASP-based approach" with box model approach.

Authors answer:
We replaced " ‘WASP-based approach" with "box model approach".

10. **Reviewer** wrote:
L22-25: "Similarly, a box-model approach has been adopted by the River MERLIN Expo model (Ciffroy, 2015), which has been used to reproduce the spatio-temporal distribution of inorganic and organic contaminants in the abiotic compartments of rivers, and to calculate [Hg] mass balance for each of them." The reference here provided "Ciffroy et

al.,(2015)" seems to be the model documentation https://merlin-expo.eu/wpcontent/uploads/2015/10/Documentation-River-V2.1.pdf I could not find any case study for Hg in the reference provided.

Authors answer:
The cited paper "Ciffroy et al.,(2015)" have no case study for mercury. However, the River Model reproduces the 1-D distribution of inorganic and organic contaminants and calculates the mass balance for each of them. According to this, we modified the sentence as follows:
"The River MERLIN-Expo model (Ciffroy et al.,(2015)) has been used to reproduce the spatio-temporal distribution of inorganic and organic contaminants in the 1D domain of rivers, and to calculate the mass balance for each of them."
The sentence has been moved at the beginning of the paragraph.

11. **Reviewer** wrote:
L25-27: "Although the River model is able to describe many of the physical and chemical processes involved in freshwater and sediment, corresponding this model specifically targets environments characterized by (i) nearly-homogeneous water bodies and (ii) limited variations in landscape geometry." The Introduction begins stating the importance of "biogeochemical dynamics of Hg species in the marine environment", and then it goes on with the WASP model that is specific for Hg. Why does it now go back to a general model for contaminants in freshwater and sediment and neglect other existing models?

Authors answer:
The sentence has been moved at the beginning of the paragraph. Other existing models are presented now in the Introduction.

12. **Reviewer** wrote:
L27-28: "In general, models based on zero dimensional boxes do not deliver reliable concentration values of contaminants in highly heterogeneous environments." I suggest rewording this sentence. Zero dimensional boxes models can provide reliable concentrations as much as other models. Box models can be less or more accurate than other models depending on the parameterizations used and on the spatial resolution, which however could be very fine.

Authors answer:
The sentence has been modified as follows:
"In general, models based on zero dimensional boxes do not deliver reliable concentration values of contaminants in highly heterogeneous environments unless they provide high spatial resolution and a proper parameterization of the biogeochemical system."

13. **Reviewer** wrote:
L28-34: "[...] in more recent works (Yakushev et al., 2017; Pakhomova et al., 2018) the biochemistry of Hg in aquatic ecosystems has been studied using a 1D advection reaction-diffusion model: the Bottom RedOx Model (BROM) has been used to reproduce the vertical dynamics of the total dissolved Hg and MeHg in the marine coastal areas of the Etang de Berre lagoon (France) (Pakhomova et al., 2018). However, even the BROM includes some criticalities in the estimation of mercury dynamics [...]" Why should a1D model be better than a box model for highly heterogeneous environments? Also, the reference "Yakushev et al., 2017" refers to the original BROM model formulation for water and sediment biogeochemistry but not Hg, I think it doesn't need to be here.

Authors answer:
In accordance with the reviewer's comment, the paragraph has been modified as follows:
"For this reason, in a recent work (Pakhomova et al., 2018) the biochemistry of Hg in aquatic ecosystems has been studied using a high resolution (HR) 1D advection-reaction-diffusion model, in which a mercury module has been integrated with the Bottom RedOx Model (BROM) (Yakushev et al., 2017) to reproduce the vertical dynamics of the total dissolved Hg and $MeHg$ in the marine coastal areas of the Etang de Berre lagoon (France) (Pakhomova et al., 2018). However, even this model includes some criticalities in the estimation of mercury dynamics. For example, the temporal variations of mercury benthic fluxes, due to reaction and diffusion processes which involve mercury species present in sediments, are not taken into account in the boundary conditions of this model. On the other hand, the sediment

chemistry and diffusion were investigated recently by Soerensen et al. (2016), who implemented a high resolution 1D model for Hg species in the water and sediments of the Baltic Sea. In both HR models, however, the strong impact of the horizontal velocity field on the spatio-temporal distribution of $[Hg]$ could not be considered since the 1D modelling was used."

14. **Reviewer** wrote:
L35-38: "All these approaches forego the complete representation of the spatial variability by approximating the model domain as a set of interconnected boxes or by detailing only the vertical dynamics of the investigated chemical species. In the present work we report on results obtained using a 3D advection-diffusion-reaction biogeochemical model for three Hg species in seawater (Hg0, HgII , and MeHg), coupled with a diffusion-reaction model in sediments and connected pore water.". This part needs rewording. No model can achieve a "complete representation of the spatial variability".

Authors answer:
To clarify, we modified the paragraph as follows:
"All these approaches do not allow a fine representation of the spatial variability by approximating the model domain as a set of interconnected boxes or by detailing only in seawater compartment the spatio-temporal dynamics of the investigated chemical species. For these reasons, we devised a new model to reproduce the spatio-temporal dynamics of $[Hg]$ in polluted marine sites characterized by very high spatial heterogeneity, such as the Augusta Harbour. In the present work we report on results obtained using a 3D advection-diffusion-reaction biogeochemical model for three Hg species in seawater (Hg^0, Hg^{II}, and $MeHg$), coupled with a diffusion-reaction model for dissolved mercury in the pore water of sediments."

15. **Reviewer** wrote:
L43: has the model been validated? Where is this explained in the text?

Authors answer:
The explanation of the calibration and validation is reported in Section 3.3.

16. **Reviewer** wrote:
L46: how exactly can this model be useful "to explore the effects of sorption-desorption dynamics"?

Authors answer:
The word "sorption" has been deleted by the sentence. Indeed, the equation for the dynamics of total mercury concentration in sediments takes into account only the de-sorption process, which mimics the "response" of solid particles to a perturbation of sorption/de-sorption equilibrium, triggered by a slowly decrease of dissolved mercury concentration in the pore water. For more details, please refer to answer n. 2.

17. **Reviewer** wrote:
L47: how can this model addresses the "role played by the spatio-temporal behavior of phytoplankton and the potential mechanism responsible for the uptake of Hg within the cells"?

Authors answer:
The sentence has been modified as follows:
"Moreover, the role played by the spatio-temporal behaviour of phytoplankton and the mechanisms responsible for the uptake of Hg within cells is taken into account as specific contribution to the scavenging process and the Hg release process by POM, respectively."

18. **Reviewer** wrote:
L58-61: here I would like to read something more informative such as the paper aim.

Authors answer:
To clarify the paper aim we inserted new sentences in the main text, at page 4, lines 96-104 of Introduction:
"The main objectives of the HR3DHG model can be synthesized as following: (i) to accurately reproduce and localize

the peaks of $[Hg]$ within the 3D domain, (ii) to estimate the Hg fluxes at domain boundaries, and (iii) to predict the evolution of mercury in sediment of polluted sites. Moreover, the HR3DHG model offers the possibility to describe the $MeHg$ and Hg^{II} partition between the dissolved phase (both seawater and pore water) and the particulate phase (suspended particulate matter and sediment particles).Specifically, in the dissolved phase the model describes the overall behaviour of Hg in ionic form and complexed with Dissolved Organic Carbon (DOC). Finally, the HR3DHG model can be a useful tool to predict and prevent the risks for the human health in marine areas close to industrial sites affected by Hg pollution extended for very long time intervals."

19. **Reviewer** wrote:
L88-95: I suggest to use this part to better explain the general architecture of the work, explaining with a figure how the various part of the model(s) interact with each other.

Authors answer:
According to reviewer's indication, at page 7 of manuscript, we inserted a new figure (Figure 2) where the basic structure of HR3DHG model is described.
Moreover, in order to explain the general architecture of the work, we inserted new sentences at page 6, lines 138-152 of manuscript:
"As well as the PCFLOW3D model of Zagar et al., the module of biogeochemical model for the seawater compartment is integrated with a hydrodynamics module. Specifically, the SHYFEM model is used to calculate the spatio-temporal behaviour of the horizontal components of the velocity field in the seawater compartment, fixing to zero the vertical velocity according to the experimental data (see Section S3 of the Supplement for details).(....) By using the curve of mean vertical profile obtained by Brunet et al. the picoeukaryotes abundances are converted into the chlorophyll concentration, which allows to reproduce the spatio-temporal distribution of NPP. This is used in our model to calculate both the biological rate constants and the sinking flux of Hg adsorbed by POM. (...) The two modules are coupled with the advection-diffusion-reaction sub-model in order to reproduce the spatio-temporal behaviour of the load of dissolved Hg released by dead picoeukaryotes cells in the seawater compartment."

20. **Reviewer** wrote:
L90 why eukaryotes? You mean phytoplankton here (see comment on S4)

Authors answer:
The word "eukaryotes" has been replaced by the word "picoeukaryotes" throughout the whole manuscript. In our work, we considered the picoeukaryotes community (i.e. phytoplanktonic eukaryotes with size less than 3 μm) since it represents the set of most representative populations of oligotrophic waters of Augusta Bay.
According to this, we modified the first sentence of Section 4 of the Supplement as follows:
"Our study includes the analysis of the abundance of picoeukaryotes community (i.e. phytoplanktonic eukaryotes with size less than 3 μm), which represents the set of most representative populations of the Augusta Bay."

21. **Reviewer** wrote:
L91-92: too many references

Authors answer:
We removed some useless references.

22. **Reviewer** wrote:
L93: what is this reference "Radomyski and Ciffroy 2015"?

Authors answer:
The reference "Radomyski and Ciffroy 2015" recalls the paper on the Phytoplankton MERLIN-Expo Model.

23. **Reviewer** wrote:
L93: here is the first mention to Supplement material, with no section specified, which after a long search I found to be S5, at the end of the document, and part of S1.3. This issue occurs throughout the manuscript and must be fixed.

Authors answer:
We specified in more detail all the sections of the Supplement throughout the manuscript.

24. **Reviewer** wrote:
L98: the reference "Melaku Canu et al., 2015" is not necessary here.

Authors answer:
We removed the reference.

25. **Reviewer** wrote:
L102-103: "By solving the model equations, we obtain the spatio-temporal distributions of Hg0(x,y,z, t), HgII (x,y,z, t), and MeHg(x,y,z, t)." Redundant with the previous lines.

Authors answer:
The sentence has been deleted. The second sentence of subsection 3.1. has been modified as follows:
"Specifically, the model equations are solved to obtain the behaviour of the three main Hg species in seawater, (...) with the reaction terms of the Partial Differential Equations (PDEs)."

26. **Reviewer** wrote:
L119: "As initial conditions, we assumed an uniformly distributed concentration of HgD and HgT, set to 1.9 ng/l corresponding to the experimental detection limit". This part should be moved to the "Model and simulation setup" section. I am not sure to understand it well, is it HgT(t0)=HgD(t0)=1.9? It should be specified "experimental detection limit for HgT in our dataset (reference)" or something similar, as 1.9 ng/l (about 9.5 pM) is very high as a detection limit. See for comparison (Mason et al. 1999; Horvat et al. 2003; Han et al. 2007; Monperrus et al. 2007; Hammerschmidt and Bowman 2012; Lamborg et al. 2012) who have detection limits well below 1 pM. What about initial conditions for MeHg?

Authors answer:
The sentence has been moved to the "Model and simulation setup" section, where also the initial conditions for all mercury species are defined now. Detection limit for Hg_T and Hg_D is 1.9 ng/l. This value is higher than that reported in cited previous works [1 pM versus our 9.5 pM] but it is near more than one order of magnitude lower with respect to the regulation level for Italian country and also this well captures the dynamic of Hg in the studied area.

27. **Reviewer** wrote:
L126-138: a crucial process such as biotic demethylation is missing. The description of photo demethylation is inconsistent with Figure 2.

Authors answer:
The biotic demethylation is now included in PDEs of our model. According to this modification, the paragraph at page 8, lines 192-197 of Model description has been reviewed as follows:
"The model includes three reaction terms regulated by first-order kinetics, which describe the photo-demethylation of $MeHg$, the methylation of Hg^{II} and the biotic demethylation of $MeHg$, respectively. The first is the amount of Hg^{II} produced by the $MeHg$ through photochemical reactions. The second is the amount of $MeHg$ obtained by the Hg^{II} through biotic and abiotic pathways in seawater. The third is the amount of Hg^{II} produced by the $MeHg$ through reductive demethylation processes caused by activity of bacteria in contaminated environments. The rate constants of three reaction terms are fixed according to previous works" Moreover, the Figure 2 of the old version of manuscript (now Figure 3) has been modified according to the reviewer's comment.

28. **Reviewer** wrote:
L132: the sentence "All data to estimate the rate constants of the redox reactions are derived from remote sensing (see Supplement)." should be moved to the Model setup section, along with similar sentences appearing below in the text (I listed some but not all of them) and the reference to the Supplement must be more specific.

Authors answer:
The sentence has been moved to page 17, lines 453-455 of the Model setup section, and modified as follows:
"Furthermore, the photochemical and biological rate constants of the redox reactions have been calculated by using both the outputs of NP model and the data coming from remote sensing (see Section S1 of the Supplement)."

29. **Reviewer** wrote:
L142-144: "The vertical turbulent diffusivity is calibrated according with experimental data, which indicate highly stratified water column conditions during the whole year". So is there oxygen depletion in the water column? If so, it should be discussed in the paper. The value given for vertical diffusivity in Table S1 is not representative of stratified conditions, and the reference provided in the table is missing in the bibliography. (This part should be moved to model setup section)

Authors answer:
On the basis of the experimental data, there is not oxygen depletion in the water column of Augusta Bay. In the new simulations, we re-calibrated the vertical turbulent diffusivity (see Table S1), which is now representative of weakly mixed layers. According to this, the sentence has been modified and moved at the "Model and simulation setup" section. Finally, the quotation Pham Thi et al. (2005) is replaced by Denman et al. 1983.

30. **Reviewer** wrote:
L148: there are eight references for one model implementation, is confusing.

Authors answer:
We removed some less crucial references following the reviewer's comment.

31. **Reviewer** wrote:
L152: it is not clear to me how are "the dynamics of the dissolved HgII and MeHg species [estimated], considering effects due to (i) the adsorption by SPM (scavenging process)". Your equations 2 and 3 contain the terms for Hg and MeHg scavenging (S^{II}_{SPM} e S^{MeHg}_{SPM}). These terms, adopted from the work of Zhang (2014) and described in the Supplemental do not include SPM:

$$S^{II}_{SPM} = \quad -\frac{\partial}{\partial z}\left[NPP\cdot(peratio)\cdot\left(\frac{z}{z_0}\right)^{-0.9}\cdot\left(\frac{k_D}{f_{org}}\right)\cdot Hg^{II}(z)\right], \tag{1}$$

The equation should be moved to the main text.

Authors answer:
To clarify the meaning of sentence we inserted at page 10, lines 245-257, of the main text the equations for the sinking flux of the SPM-bound mercury for Hg^{II}_D and $MeHg_D$, respectively. Moreover, the contribute of silt to the scavenging process is taken into account in new simulations.

32. **Reviewer** wrote:
L153-154: How is "the scavenging process for both Hg_D species regulated by the gradient of mercury concentration along the water column"?

Authors answer:
The sentence is modified as follows:
"The scavenging process for both Hg_D species is regulated by the sinking flux of particle-bound mercury along the water column (see Section 1.3 of the Supplement)."

33. **Reviewer** wrote:
L155: this is a repetition of L90-92, please mention explicitly which parameter or variable you extract from which model and where it has been used in the Hg model, choose for each parameter one or two references that actually refer to the model implementation you used.

Authors answer:
To specify the variables extracted by each module to be used in the Hg model, we inserted the following sentences at

pag 8, lines 205-212, of the of Model description section:
"Specifically, the NP model provides the spatio-temporal distribution of picoeukaryotes abundance, which is used to get the chlorophyll concentration and the net primary production through suitable conversion functions (see Sections S1.1 and S4 of the Supplement). These two variables are then exploited to calculate the contribute of the sinking flux for POM-bound Hg within the suspended particulate matter (see Sections S1.2 and S1.3 of the Supplement). The Phytoplankton MERLIN-Expo Model gives the spatio-temporal dynamics of the Hg^{II} and $MeHg$ contents within the picoeukaryotes cells. These two variables are then used, together with the picoeukaryotes abundance, to get the amount of Hg^{II} and $MeHg$ released by the dead picoeukaryotes cells (see Sections S1.2 and S1.3 of the Supplement)."

34. **Reviewer** wrote:
L172-176: I don't think there is any wet deposition for Hg0, please provide one reference for each process and parameterization selected/excluded. It should be stated explicitly that dry deposition and MeHg deposition are neglected. Somewhere should be also mentioned that Me2Hg is not explicitly considered.

Authors answer:
In accordance with previous works (see Zagar et al. 2007 and Zagar et al. 2014) the wet deposition forHg^0 is considered in our model, even if this contribute is negligible with respect to that of the Hg^{II} . In particular, the Hg^0 wet deposition flux is about $1/20000$ of the total wet deposition flux in the Augusta Harbour.
According to reviewer's comment, we inserted at least one reference for each process and parameterization selected/excluded. In new simulations, we also include both the dry deposition of Hg^{II} and total $MeHg$ deposition. According to this, the mentioned paragraph at pages 11-12, lines 288-297, has been modified as follows:
"Specifically, we take into account for the three mercury species: i) the evasion and the deposition of Hg^0 at the water-atmosphere interface; ii) the lack of Hg^0 diffusion at the water-sediment interface; iii) the wet and dry deposition of Hg^{II} at the water-atmosphere interface; iv) the wet and dry deposition of $MeHg$ at the water-atmosphere interface; v) the diffusion of Hg^{II} and $MeHg$ at the water-sediment interface; vi) the constant fixed value of $[Hg_D]$ out of Augusta Bay (Ionian Sea); vii) the exchange of the elemental mercury, Hg^{II} and $MeHg$ between the Augusta basin and the Ionian Sea through the two inlets."
Moreover, we inserted a new sentence at page 6, lines 159-160, of the Model description section, to mention the fact that Me2Hg is not explicitly considered:
"Since the experimental data indicate that Me_2Hg concentration is very low in the Augusta Harbour, the behaviour of this Hg species is not reproduced in our model."

35. **Reviewer** wrote:
L175-176: the boundary conditions describing the diffusion of HgII and MeHg at the sediment-water interface are very important in your model, as they represent the only connection between the water and the sediment, but they are not explained. Saying that you simulate "the exchange of HgII and MeHg at the seawater-sediment interface due to particulate matter deposition and re-suspension mechanisms" is extremely misleading because the reader will likely understand that you simulate deposition and resuspension fluxes of particulate Hg species, while this is not the case. I suggest to separate these equations from atmospheric boundary conditions and spend more words to explain well how they are implemented.

Authors answer:
Initially, we attempted to calculate also the amount of dissolved mercury embedded/released by the pore water due to the particulate matter deposition/re-suspension mechanisms at the sediment-water interface. However, the sediment transport module did not reproduce correctly the sedimentation rate experimentally observed in the Augusta Harbour. Therefore, we removed this module by our model and, according to experimental data, we fixed a constant sedimentation rate for the whole Augusta Bay and the whole period investigated. As a consequence, the mentioned sentence has been removed by the manuscript as well as the terms in Eqs. (11)-(12) which describe the effects of particulate matter deposition/re-suspension mechanisms. Finally, the equations for the boundary conditions are better explained in the Section S1 of Supplement.

36. **Reviewer** wrote:
L182-196: maybe the equations can be distributed within the text describing them, or at least they must be recalled in the text when being described.

Authors answer:
The equations are now recalled in the manuscript when they are described.

37. **Reviewer** wrote:
L207: references?

Authors answer:
The references have been removed.

38. **Reviewer** wrote:
L212-214: "the mass transfer coefficients ($MTC^{II}_{sed-water}$ and $MTC^{MM}_{sed-water}$) at the water-sediment interface are calculated in order to fit the experimental findings and according to previous works (Schulz and Zabel,2006; Ciffroy,2015)(see Supplement)." I think that the equations for these MTC should stay in the main text. The fact that they are adjusted to fit experimental findings should be said in the Model setup section. The reference "Schultz and Zabel 2006" is incomplete, and so is "Ciffroy 2015".

Authors answer:
The description of the mass transfer coefficients is very long (see Sections S1.2.2 and S1.3.2 of the Supplement). Therefore, it cannot stay in the main text due to the reduced size imposed by the journal. The sentence at page 13, lines 329-335, is modified as follows:
"The spatio-temporal dynamics of pore water mercury concentrations (...), while the mass transfer coefficients ($MTC^{II}_{sed-water}$ and $MTC^{MM}_{sed-water}$) at the water-sediment interface are calculated by sediment porosity, molecular diffusion coefficient, boundary layer thickness above and below sediment according to previous works."
Moreover, we described the setup of the parameters for the mass transfer coefficients ($MTC^{II}_{sed-water}$ and $MTC^{MM}_{sed-water}$) at the water-sediment interface in the Model setup section, where the followings sentences have been added at page 17:
"Before to calculate the mass transfer coefficients at the water-sediment interface, the *boundary layer thickness above the sediment* was optimized to better reproduce the spatial distribution of mercury benthic flux observed experimentally. Unlike the *boundary layer thickness above the sediment*, the other parameters used to obtain $MTC^{II}_{sed-water}$ and $MTC^{MM}_{sed-water}$ were not calibrated. In fact, the *boundary layer thickness below the sediment* was estimated by using the relationship between this parameter and the average velocity of marine currents defined by Sorensen (2001), while the spatial distribution of the sediment porosity within Augusta Harbour was reproduced, according to previous works, by exploiting the measurements on the sediment samples performed by $ICRAM$ in 2005. Also, the molecular diffusion coefficient was that reported by Schulz and Zabel (2006)."
According to reviewer's comment, some references have been added.

39. **Reviewer** wrote:
L214-218: "The dynamics of the mercury benthic fluxes (phi_res) caused by particulate matter deposition and resuspension mechanisms (Neumeier et al., 2008; Ferrarin et al., 2008) is obtained by considering both the spatial distribution of sediment porosity and the spatio-temporal behaviour of removed/settled sediment thickness at the seawater-sediment interface. The sediment exchanges at the water-bottom interface are obtained from the application of the hydrodynamic model, which accounts for sediment transport processes induced by currents (see Supplement)." This part seems very confusing to me. The equation for $phi-res$ and Er should be defined in the main text and should be probably given different names. Moreover, the hydrodynamic SHYFEM model can not provide fluxes of sediment, if the SEDTRANS model or any other Sediment model has been run with SHYFEM to obtain sediment fluxes in the Augusta Bay,it should be described in detail in the Supplement (including the parameters used). Also, if you had run a sediment model, why did you adopt experimentally measured SPM concentrations for your eq. 13? If you had not run a sediment model, explain where the sediment fluxes come from and make it more evident throughout the text that they are not transporting particulate mercury. In Figure 2 there are thick arrows with an "R" that looks like re-suspension, which is misleading.

Authors answer:
This paragraph has been removed by the manuscript. In our work no sediment model is now used to describe the particulate matter deposition/re-suspension mechanisms at the sediment-water interface. In the new Figure 3 (ex Figure 2), as well as in the whole manuscript, the benthic flux from sediments to seawater is now named "B".

40. **Reviewer** wrote:
L219-220: repetition

Authors answer:
The reviewer is right. The sentence has been removed.

41. **Reviewer** wrote:
L221: this should appear before in the section

Authors answer:
The paragraph has been moved at page 11 (lines 277-286).

42. **Reviewer** wrote:
L223: same KD for Hg and MeHg? No units for the terms of this equation?

Authors answer:
No experimental data has been collected for the K_D of Hg^{II} and $MeHg$. On the contrary, the seawater-SPM partition coefficient (K_D) has been measured for the total Hg_D. According to this, we chose to use the same experimental K_D for both dissolved Hg species (i.e. Hg^{II} and $MeHg$).
The units for the terms of the equation are now inserted in the text.

43. **Reviewer** wrote:
L225-227: please change "in very good agreement with" with "within the range of"

Authors answer:
The sentence has been deleted, and the whole paragraph has been modified (see page 11 of the new version of manuscript).

44. **Reviewer** wrote:
L228-229: is then SPM kept constant in time? If so, it should be state

Authors answer:
The spatial distribution of SPM is constant in time. The sentence has been modified as follows:
"The spatial distribution of SPM was set according to the experimental information collected during the oceanographic cruise of October 2017, and assumed constant for the whole simulation time."

45. **Reviewer** wrote:
L230-242: This part should be elsewhere, is not part of the model.

Authors answer:
We inserted this paragraph in a new subsection of the Model description (section 3.1.1).

46. **Reviewer** wrote:
L242: Figure 2 should be mentioned before in the text, and used to help the reader in the model description. The caption does not explain the figure.

Authors answer:
The ex figure 2 (now figure 3) has been moved at the beginning of the Model description section. In the new Figure 3 caption, we inserted the following description for the scheme:
"The scheme describes the transformation processes (k_1- photo-oxidation , k_2- photo-reduction, k_3-biological oxidation, k_4-biological reduction, k_{Ph-de}-photo-demethylation, k_{deme}-demethylation, k_{me}-methylation, K_{demeth}-demethylation, K_{meth}-methylation) and the main transport processes (A-anthropogenic input, AD-atmospheric deposition, B-benthic

flux, D-net flux due to particulate deposition and settling, O-net outflow from basin, V-atmospheric evasion) which involve the dissolved and particulate-bound Hg species in seawater (Hg_D^{II}, $MeHg_D$, Hg_D^0, Hg_P^{II}, $MeHg_P$) and sediments (Hg_{pw}^{II}, $MeHg_{pw}$, Hg_T^{sed})."

47. **Reviewer** wrote:
L245: please recall the equations where needed, not in this aggregated form.

Authors answer:
The recalling to equations has been removed.

48. **Reviewer** wrote:
L249: Please motivate this choice. This is a strong assumption that I have never seen before.

Authors answer:
We attempted to model the mercury concentration by adopting a different order kinetic for Eq.(22), however the best agreement between theoretical results and experimental data was obtained when a first-order kinetic was used. According to this, the sentence is modified as follows:
"In order to better reproduce the experimental findings, we describe mercury desorption using an exponential equation, which accounts, in the absence of external sources, for the loss of mercury through the desorption mechanism. Since the mercury desorption has to depend on its instantaneous concentration, the mechanism is regulated by a first-order kinetic."

49. **Reviewer** wrote:
L259-260: This is already mentioned in the section for the water model. I think features are common to the water and sediment modules for Hg might be explained just once at the beginning of section 3 (same for L265-266, L278-279).

Authors answer:
The manuscript has been modified in section 3 according to the reviewer's suggestion.

50. **Reviewer** wrote:
L284-285: diffusion coefficients are not constant in time in Melaku Canu et al., (2015)

Authors answer:
The reference Melaku Canu et al. (2015) has been removed by the sentence.

51. **Reviewer** wrote:
L290, L291: while I understand the use of MeHg fraction (k_{MeHg}) in setting the initial conditions, I do not understand why it is used in the partial difference equations that also include Hg methylation and demethylation (eq.16 and 17). Can you explain the parameterization in the text? I suggest changing k_{MeHg} with f_{MeHg} so it is more intuitive.

Authors answer:
The fraction of methyl-mercury is used as parameter in one of terms of the partial differential equations since it is necessary to calculate the amount of $MeHg$ and Hg^{II} released to pore waters during the desorption process of mercury bound to sediment particles. k_{MeHg} is replaced by f_{MeHg} throughout the manuscript.

52. **Reviewer** wrote:
L296 and L301: I've never seen this approach based on a desorption rate () in mercury modeling. In this way, your sediment HgT decrease with time by assumption, as there are no source terms. By adopting a reductio ad absurdum logic, if your simulation would start in 1900 or before, which initial conditions HgT(0) would you need to achieve observed HgT concentrations for 2005? Can you provide any theoretical background for this parameterization? Also, you state "The desorption rate is fixed to a low value to fit the slow mercury release from the sediment particles to pore water according to experimental observations" but in Table S1 "no data" is given as a reference for the parameter.

Authors answer:
The choice of this modelling for the total mercury in sediments is connected to the industrial history of the Augusta Bay.

Specifically, it is necessary to recall that an important chlor-alkali plant discharged (legally and illegally) a great amount of mercury at the southernmost part of Augusta Bay until 2005, namely when the plant has been closed. Soon after the chlor-alkali plant closure, the ICRAM performed several samplings in the sediments of the Augusta Bay by providing a 3D spatial distribution of total mercury concentration in the surface sediment layer (from 0.1 to 1.9 m depth), which constitute the initial condition for our model. Since no mercury source was anymore present in Augusta Bay after the chlor-alkali plant closure (2005), except for the release by buried sediments, we assumed that the mechanism responsible of the high mercury concentration measured in seawater and pore water until last sampling (2017) was the desorption process of particle-bound mercury. The same approach could not be used before the ending of mercury discharge in the Augusta Bay when other mercury sources were still present within the basin. In conclusion, our model is valid since it reproduces the spatio-temporal dynamics of mercury after the plant closure, by fixing initial conditions for the sediments equal to the total mercury concentration measured in 2005. Analogously, the model can reproduce the mercury distribution in all basins where the industrial pollution is ceased recently.

About the sentence mentioned, the reviewer is right. In fact, it is mistaken. For this reason, we moved the sentence at page 17 of "Model and simulation setup" Section and rewrote it as follows:

"In the Eq. (22), the desorption rate α was calibrated to obtain the best fit between the theoretical results and experimental observations for $[Hg]$ in pore water"

Since the desorption rate has been calibrated in the model by using the experimental data of mercury concentration in the pore water, no reference cannot given for this parameter.

53. **Reviewer** wrote:
L298 "The spatial distribution of the fraction of methylmercury in the sediments is that obtained by field observations, while the two sediment-pore water distribution coefficients are calibrated, according to previous work (Oliveri et al., 2016)" move to model setup, provide a reference for the data/publication from where you derived the fraction of methylmercury and mention how much is this fraction.

Authors answer:
The sentence has been moved at page 17 of "Model and simulation setup" Section, and modified as follows:
"Specifically, in Eqs. (20)-(21) the sediment-pore water distribution coefficients were calibrated to guarantee the best theoretical $[Hg]$ in pore water in agreement with the value ranges experimentally observed in a previous work (Oliveri et a. 2016), whereas the fraction of methyl-mercury in sediments for the whole spatial domain was set to that obtained by field observations during the oceanographic survey of October 2017 (see Table S1)."

54. **Reviewer** wrote:
L306 why is the MTC parameterization not included in the boundary conditions for sediment? Are you assuming only one-way fluxes toward the water?

Authors answer:
Yes, in our model we assume only one-way fluxes toward the water. This assumption is in agreement with the experimental data (not reported in the manuscript) which indicate a negative gradient for dissolved mercury concentration from the pore water to seawater.

55. **Reviewer** wrote:
L338-339, L342: this is different from what is said in L142-144.

Authors answer:
The sentences on the turbulent diffusivities have been modified in the Model setup section. The two sentences in section 3.1 have been removed.

56. **Reviewer** wrote:
L338-344: please do not go back and forth from vertical to horizontal diffusivity, discuss one at a time and select a reference that is consistent with that given in Table S1.

Authors answer:
According to the reviewer's indication, the paragraph has been modified, as follows:
"The vertical turbulent diffusivity was calibrated according to experimental data, (...) during the whole year. Specifically, the vertical turbulent diffusivity was set (...) at the surface layer of the water column. The calibrated vertical diffusivity was in good agreement with (...) under the condition of weakly mixed waters. The horizontal turbulent diffusivity was assumed (...) the values obtained in Massel (1999). In particular, the horizontal turbulent diffusivities were optimized (...) the observed mercury evasion flux. The calibrated horizontal diffusivities were (...) to those of the Augusta Bay."

57. **Reviewer** wrote:
L351: it is not clear to me why you chose to run 250 years of simulation without varying the forcings.

Authors answer:
To clarify the sentence has been modified as follows:
"Finally, the calibrated model has been run by considering the seasonal oscillations of the environmental data (water currents, wind etc.) provided by hydrodynamic modelling (see Section S4 of the Supplement)."

58. **Reviewer** wrote:
L357-361: this part should not be in the Results section, Table S1 must be introduced much before in the text.

Authors answer:
According to the reviewer's indication, the most part of this paragraph has been moved in Model setup section.

59. **Reviewer** wrote:
Figure 3: you must show modeled vertical profiles against observations when available. I do not find useful or interesting what is currently shown in Figure 3.

Authors answer:
We moved the Figure 3 of the old version of manuscript in the Supplement. Afterwards, we inserted a new Figure 3 in the manuscript in which the comparison between the theoretical results and experimental data for dissolved mercury concentration is shown.

60. **Reviewer** wrote:
L364: please provide the values of these ratios and values for comparison to prove this excellent agreement (this applies to most of the Results section, show the numbers).

Authors answer:
The average concentration ratios among the three mercury species dissolved are now provided in the main text (see page 18). Moreover, a new table with the numerical values of the three species has been inserted in the Supplement (see Table S5).

61. **Reviewer** wrote:
L368-369: "In general, the mercury concentration is maximal at the seawater-sediment interface, where the main sources of HgII and MeHg are localized.". Do you mean in general in your model? While from your budget I see that diffusion flux appears to be the first HgT source, what supports this statement for MeHg? Especially in this model configuration without biological demethylation in the water column, I would expect water column methylation to be significant. Is the increase of Hg species at the bottom of the water column in agreement with your dataset? Please show and discuss it.

Authors answer:
According to the reviewer's indication, the paragraph has been modified as follows:
"The model results indicate that the dissolved mercury concentration is usually maximal at the seawater-sediment interface (see Fig.4), where the main sources of Hg^{II} and $MeHg$ are localized. These numerical results are in reasonable agreement with the field observations (see Tables S6-S7 of the Supplement). Moreover, taking into account the redox conditions of sediments in the area, we speculate that maxima in $MeHg$ production be confined to the seawater/sediments interface."

Moreover, the points raised by the reviewer are clarified both in other parts of this letter and in the revised version of the Introduction.

62. **Reviewer** wrote:
L369: I don't think (x,y) is useful here.

Authors answer:
(x,y) has been removed by text.

63. **Reviewer** wrote:
L370-371: "[...] peaks of mercury concentration occur at mid-depth of the water column possibly due to the effects of the velocity field of marine currents and the bathymetric features of Augusta basin.". I do not understand this explanation.

Authors answer:
The sentence has been modified as follows:
" (...) we observe that the peaks of mercury concentration occur at mid-depth of the water column possibly due to the distribution of marine currents velocity field within Augusta basin, which determines sometimes the presence of a $[Hg]$ maximum in the intermediate layers of seawater."

64. **Reviewer** wrote:
L372: specify "in our model"

Authors answer:
We specified "in our model" at the beginning of the sentence.

65. **Reviewer** wrote:
L375-377: please support this "good agreement" with statistical and visual tools. There is no comparison of modeled and observed vertical profiles of Hg and MeHg, which would be interesting to see and discuss, especially considering the emphasis put in the 3D model domain.

Authors answer:
A statistical analysis based on the χ^2 test for $[MeHg]$ is now present. In general, it is worth to point out that no experimental data for $[Hg^{II}]$ and $[Hg^0]$ were collected in the Augusta Bay during the oceanographic surveys. In fact, the most part of the experimental data concerns $[Hg_D]$ and $[Hg_T]$, which were measured only in two/three sampling points for each station. Moreover, the magnitude of $[Hg_D]$ and $[Hg_T]$ is below the detention limit in many sampling points. For these reasons, in the first version of the paper we chose to make neither graphs nor statistical checks for comparing theoretical results and experimental data.
The sentence at pages 19-20 of manuscript is modified as follows:
"A quantitative analysis, based on the reduced χ^2 test, indicates a good agreement between the model results and experimental findings for $[MeHg]$ in stations $A3$ ($\tilde{\chi}^2 = 0.0005$) and $A7$ ($\tilde{\chi}^2 = 0.0005$), while differences can be observed in the stations $A9$ ($\tilde{\chi}^2 = 0.0955$) and $A11$ ($\tilde{\chi}^2 = 0.1065$), where the theoretical concentrations appear overestimated at the bottom layer (see Table S6 of the Supplement)."

66. **Reviewer** wrote:
L377-378: "This result is probably due to the overestimation of the $MeHg$ benthic fluxes in these two stations." Can you support this statement? Other reasons would be possible as well.

Authors answer:
We have no experimental data on the $MeHg$ benthic fluxes to support this statement. However, the two stations ($A9$ and $A11$) are localized in sites where the theoretical $MeHg$ benthic fluxes are very high, and the effects of transport mechanisms on the $MeHg$ concentration are negligible respect to diffusion process from sediments.

67. **Reviewer** wrote:
L379-81: I think it must be highlighted that HgD is modeled while HgT is estimated assuming a linear correlation with

modeled HgD concentrations and SPM through equation 13 (which might be not representative of processes occurring in the field).

Authors answer:
According to the reviewer's indication, the sentence at page19 has been modified as follows:
"On the other hand, the dynamics of the spatial distribution of the $[Hg_T]$ is estimated according to Eq. (9), assuming a linear correlation between the modeled $[Hg_D]$ and the experimental SPM concentrations."

68. **Reviewer** wrote:
L383-384: same comment as for L375-377

Authors answer:
Please, see the previous answer for $[MeHg]$. To clarify the magnitude of the difference between theoretical results and experimental data, we added the following sentences at page 20:
"Specifically, the difference between the model result and field observation for the $[Hg_D]$ is less than the experimental error ($\sigma = 3.2\ ng/l$) in 59% of sampling points, while it exceeds 2σ in only 17% of sampling sites. As a conclusion, the comparison between experimental data and theoretical results for the $[Hg_D]$ shows mostly small discrepancies except in some of the most contaminated areas, where concentration hot spots are hard to capture due to the resolution grid used in the present work."

69. **Reviewer** wrote:
L387: same comment as for L375-377 and 383-384

Authors answer:
Please, see the previous answer for $[MeHg]$ and$[Hg_D]$. To clarify the magnitude of the difference between theoretical results and experimental data, we added the following sentence at page 20:
"As a whole, the discrepancy for the $[Hg_T]$ is less than σ in 44% of cases, while it exceeds 2σ in 32% of sampling sites."

70. **Reviewer** wrote:
L391-395: the reader knows nothing about your modeled concentrations and spatial distribution of SPM since they are not shown.

Authors answer:
The modeled concentrations are shown in the Supplement, while the experimental data of SPM (acquired recently and never published) can be put in a repository if needed.

71. **Reviewer** wrote:
L399-400: "The model reliably reproduces the high benthic mercury fluxes also in the part of the south-east sector close to the inlets of the Augusta Bay, where intensive ship traffic and the relatively high velocity field of the marine currents cause sediment resuspension and intensive transport of SPM." is the high sediment resuspension induced by ship traffic considered in your model? How does it relate to your modeled benthic fluxes? From figure 4, I find modeled fluxes to be one order of magnitude higher than fluxes reported by (Salvagio Manta et al. 2016). Please explain what I am missing or what is going on.

Authors answer:
The resuspension induced by ship traffic is not considered in our model. Therefore, there is no relation between the modeled benthic fluxes and the sediment resuspension induced by ship traffic. According to this, we removed the second part of the sentence at page 20, lines 548-549, of the main text.
Since the experimental findings on benthic fluxes have been acquired in different sites than those mentioned by referee, the comparison between the theoretical results and experimental data cannot performed in these highly polluted sites. In general, it is possible to assert only that very high benthic fluxes modeled in some sites of Augusta Harbour are strictly connected to the high total mercury concentrations measured in sediments of the same sites. According to this, we added the following sentence at page 20, lines 553-555, of the new version of manuscript:

"Moreover, the model results confirm that the spatial heterogeneity of benthic fluxes observed experimentally is strictly connected to that of Hg_T concentration in sediments."

72. **Reviewer** wrote:
Figure 4: probably you don't need to show fluxes for both inorganic Hg and HgT, as they look the same. Why available data are not shown on the map,or in another figure?

Authors answer:
The maps on the inorganic flux are now removed by the Figure 5 (ex Figure 4). A map of the experimental total benthic fluxes cannot to be reproduced due to the reduced number of measurements (three for sampling) performed in the two periods investigated. Experimental data are reported in Table S9 of Supplement for a direct comparison.

73. **Reviewer** wrote:
L407-409: "In general, the theoretical distribution of the mercury evasion fluxes is in a very good agreement with the experimental results for the investigated periods". Same comment as for L375-377,383-384 and 387. From figure 5, I see that modeled evasion (up to 100 ug/m2d) is about 100 times as high as evasion modeled by Bagnato et al. 2013.

Authors answer:
In this case, no significant comparison between theoretical results and experimental data is possible to make on the basis of statistical tests. Indeed, only three experimental measurements (in three different sites) have been acquired for each sampling period. Too much few to perform a significant statistical check.
Moreover, the assertion "I see that modeled evasion (up to 100 ug/m2d) is about 100 times as high as evasion modeled by Bagnato et al. 2013" is not correct. In fact, also Bagnato et al. (2013) obtained high evasion fluxes $72\ \mu g \cdot m^{-2} \cdot d^{-1}$ in the coastal zones at the south-west of the Augusta Bay. Therefore, our model results are not so different than those obtained by Bagnato et al..
According to the reviewer's indication, the paragraph at the ending of page 20 has been modified as follows:
"In general, the theoretical distribution of the mercury evasion fluxes is in acceptable agreement with the experimental results for the investigated periods (see Table S10 of the Supplement). Specifically, small discrepancies are observed in the most part of the stations (four over six), while larger difference emerge in stations 3 (November 2011) and 5 (June 2012). From a qualitative point of view, (...)"

74. **Reviewer** wrote:
L416-419: "The modeled HgD benthic fluxes (...) are significantly larger than those estimated for both sampling periods on the basis of the field observations (...) (Salvagio Manta et al., 2016)." Why do you go back to benthic fluxes, which are already discussed from L396 to L404, contradicting your previous statements (L399 and 403)?

Authors answer:
No contradiction is present in this sentence. Indeed, in this part of section we discuss over the annual mass balance for the whole basin, while previous statements described the model results for benthic fluxes in each site. To clarify, we inserted at the beginning of the paragraph the following sentence:
"In this work, we make the annual mass balance of the Augusta Bay to study the fate of Hg coming from sediments, and to estimate the Hg outflows at the inlets of basin."

75. **Reviewer** wrote:
L419-420: "This probably depends on the limited number of sampling sites available in the experimental work with a consequent extremely coarse capacity to capture reliable estimates of benthic fluxes." Six sites for measuring benthic fluxes within the Augusta Bay is a lot. Other work of this kind I came across only had 2-3 sampling stations for larger study areas, I thus disagree with the statement that they provide an "extremely coarse representation".

Authors answer:
To clarify, we modified the sentence at page 22 as follows:
"This probably depends on (...) with a consequent limited capacity to capture reliable estimates of benthic fluxes within a basin, such as Augusta Bay, where the spatial distribution of sediment mercury is highly heterogeneous."

76. **Reviewer** wrote:
L421-423: I do not follow why this should ensure that your model results for a specific period are better than values experimentally detected under conditions that are representative of that period.

Authors answer:
The sentence has been removed.

77. **Reviewer** wrote:
L424-429: you have already discussed evasion fluxes before, so the two parts should go together. Be careful of the references.

Authors answer:
Please, see the previous answer for benthic fluxes. The two references have been removed.

78. **Reviewer** wrote:
L439-440: "The model results for the annual recycled mercury flux are shown in Fig.6d. In this case, values calculated (2.45 kmol y-1 for the year 2011 and 2.41 kmol y-1 for the year 2012) are larger and more realistic than those estimated in Salvagio Manta et al. (2016) by simple linear subtraction of the available fluxes in the mass-balance equation (0.84 kmol y-1).". How do you support this statement that your estimate is more realistic? Isn't the mercury recycling calculated by subtraction also in this work (equation 14)? How do you argue the significant gap that is shown in Figure 6d between the "Total recycled" and the "Scavenged recycled"? How large is your estimated HgT reservoir in the water and sediment of Augusta Bay?

Authors answer:
To answer the referee's questions, we modified the last paragraph of section 4.1 (see page 24) as follows:
" In this work, the annual recycled mercury flux is calculated by subtraction using the mass balance equation (18), as well as it was done in previous works on the Augusta Bay. The model results for the recycled mercury flux are shown in Fig.7d. Here, values calculated by our model (2.50 $kmol\ y^{-1}$ for the year 2011 and 2.46 $kmol\ y^{-1}$ for the year 2012) are larger and probably more realistic than those estimated in Salvagio Manta et al. (2016) (0.84 $kmol\ y^{-1}$). Indeed, the former are obtained by considering the seasonal oscillations of all other mercury fluxes during the year, while the latter are calculated without considering the seasonal changes of mercury fluxes.
In order to reproduce the effects induced by scavenging process on the mercury dynamics, our model calculates the annual sinking mercury flux, whose results are shown in Fig. 7d. Here, a significant gap between the recycled flux (2.50 $kmol\ y^{-1}$ for the year 2011) and the sinking flux (0.07 $kmol\ y^{-1}$ for the year 2011) is observed probably due to the underestimation of the amount of mercury captured by POM (see Eq. (4)-(5)). More specifically, this behaviour could be caused by the underestimation of NPP, which is calculated by using a conversion function calibrated for oceans rather than for coastal zones.
On the contrary, very high values of the annual Hg_T accumulation rate in surface sediment layer (12.07 $kmol\ y^{-1}$ for the year 2011), respect to those of the annual recycled flux (2.50 $kmol\ y^{-1}$ for the year 2011), are obtained by our model. This result is caused by the high sedimentation rate (11.7 $mm\ y^{-1}$) estimated experimentally and used in our calculations for annual Hg_T accumulation rate. However, the sedimentation rate could be overestimated due to sampling methods used. In fact, the results obtained by the sediment transport model indicate a low average sedimentation rate for the Augusta Bay."

79. **Reviewer** wrote:
L447: 50 cm is not shallow for sediment. Why did you choose to represent about 2 m of sediment with a coarse resolution (10 cm layers)?

Authors answer:
We recall that experimental findings in sediments constitute the initial conditions of model. According to this, in our work the choice of investigated sediment layer thickness is bound to the maximum depth of sediment samplings, while the grid resolution is imposed by the distance between the sampling points of total mercury concentration.
According to the Referee's indication, the sentence at page 25 has been modified as follows:

"(...) the vertical profiles of mercury concentration in the sediments (...) reach their maximum value within the surface layer of the sediments ($< 0.5\ m$ of depth)."

80. **Reviewer** wrote:
L459: you should make it clear which data are used for calibration and which are used for validation.

Authors answer:
To clarify, the section "Model and simulation setup" has been modified according to the reviewer's requests.

81. **Reviewer** wrote:
L465-466: "This "integrated" model, which allows to give a description of the mercury dynamics in the whole system (seawater, pore water, and particulate phase of the sediment), represents an absolute novelty in the landscape of the mathematical modelling of spatio-temporal dynamics in a biogeochemical context". This is not true, as you can realize with a thorough review of the mercury modelling at the state of the art. Besides, the parameterization chosen in your model for Hg species dynamics in water and sediment is overall questionable and not well supported by theoretical knowledge at the state of the art. Even correcting major oversights mentioned in other comments, this model still neglects the dynamics of Hg in the particulate phase. It thus does not provide a "description of the mercury dynamics in the whole system", and is probably less advanced than other existing models.

Authors answer:
According to the reviewer's indications, the sentence at page 26 has been modified as follows:
"This "integrated" model, which allows to give a description of the mercury dynamics in highly polluted marine sites, introduces some novelties in the landscape of the mathematical modelling of spatio-temporal dynamics in a biogeochemical context."

82. **Reviewer** wrote:
L480-482: "the different approach used in the WASP models and River MERLIN-Expo model allowed neither to reproduce the dynamics of the vertical profiles of mercury concentration in the seawater compartment, nor to obtain the spatio-temporal behaviour of mercury concentration in the sediments". Again, the River MERLIN model does not seem to be relevant here, and it is not true that the previous approaches could not reproduce the vertical dynamics nor the spatio-temporal evolution of Hg species. For example in Rosati et al.,(2018) the WASP model has been used to simulate the vertical profile of Hg and MeHg in the water column and sediments of the Black Sea; this has been done in 1D but it could have been done in 3D as well. Sorensen et al., (2016) also implemented a 1D model for Hg species in the water and sediments of the Arctic Sea. As for the spatio-temporal evolution of Hg and MeHg in water and sediment, an example can be found in Canu and Rosati (2017).

Authors answer:
According to the reviewer's comment, the sentence at page 26 has been modified as follows:
"For comparison, the different approach used in the WASP models did not allow to reproduce the dynamics of mercury concentration distribution at 3D high resolution in polluted sites characterized by elevated spatial heterogeneity. Similar criticalities came out from the study of HR-1D models (Soerensen et al., (2016), Pakhomova et al., (2018)), in which the effects of horizontal velocity field on the mercury dynamics could not be taken into account."
Moreover, the point raised by the reviewer is clarified in the revised version of the Introduction.

83. **Reviewer** wrote:
L484: "In general, no forecast about the mercury depletion time in the sediment compartment of Augusta Bay was possible by other models." See Canu and Rosati, 2017.

Authors answer:
According to the reviewer's indication, the sentence at page 26 has been modified as follows:
"In general, only few models (Rajar et al., (2007), Zagar et al. (2007), Canu and Rosati (2017)) were able to make forecasts about the mercury depletion time in the sediment compartment of highly polluted sites, such as Augusta Bay."

84. **Reviewer** wrote:
L486-490: "Finally, the biogeochemical models introduced in previous publications included neither the Nutrient-Phytoplankton model (Dutkiewicz et al., 2009; Morozov et al., 2010; Valenti et al., 2012; Denaro et al., 2013a, c, b; Valenti et al., 2015, 2016a, b, c, 2017) nor the Phytoplankton MERLIN-Expo model for the mercury content in eukaryotes cells (Pickhardt and Fischer, 2007; Radomyski and Ciffroy, 2015). All the aforementioned aspects are therefore an element of novelty in the context of 3D biogeochemical modelling." The model here presented do not include a plankton model, the two tools are used together but not integrated. Moreover, there has been previous work integrating phyto- and zooplankton in a biogeochemical model for marine Hg cycle (e.g. Soerensen et al., 2016).

Authors answer:
Actually both the Nutrient-Phytoplankton (NP) model and the Phytoplankton MERLIN-Expo (PME) model for the mercury content in eukaryotes cells are integrated with the biogeochemical (BG) model. Specifically, the NP and MPE model interact with the BG model through the load of Hg_D released by POM. Moreover, in the new version of the manuscript we inserted results obtained in the presence of a new interaction term (sinking fluxes of organic and inorganic mercury) between the BG and NP model, which makes "stronger" the integration among the three models. Specifically, in the new version of model the NPP coming from the NP model is used to calculate the sinking fluxes of the BG model. According to reviewer's indications and related model modifications, we replaced the first sentence with the following one: "Finally, the biogeochemical models introduced in previous works (Soerensen et al., 2016) provided neither the NPP coming from the Nutrient-Phytoplankton model (...), nor the load of POM-relseased Hg_D obtained using the Phytoplankton MERLIN-Expo model (...) (see Section 3.1)."

85. **Reviewer** wrote:
L491-492 "The HR3DHG model considers the effects of the seasonal changes of the environmental variables on the mercury out flows to wards the atmosphere and the open sea, and this also is a new feature in biogeochemical model." The seasonal effects are never discussed in the manuscript, and anyways this is clearly not a new feature.

Authors answer:
The seasonal changes of the environmental variables are discussed only in the Supplement. We deleted the second part of the sentence: "and this also is a new feature in biogeochemical model".

86. **Reviewer** wrote:
L495 "Firstly, the mass transfer coefficients at the water-sediment interface are highly sensitive to the layer thickness above the sediment and their variation could cause significant changes of mercury benthic fluxes." Why is this relevant? Which are the environmental implications?

Authors answer:
To explain the environmental implications, we modified this paragraph as follows:
"Firstly, the mass transfer coefficients at the water-sediment interface are highly sensitive to the *layer thickness above the sediment*. Specifically, for each mercury species in sediments, a small decrease of this parameter causes a great increase of benthic fluxes, with a consequent strong enhancement of dissolved mercury concentration in seawater."

87. **Reviewer** wrote:
L497 "Sensitivity analysis performed on the sediment compartment indicates that the spatio-temporal dynamics of the benthic mercury flux strongly depends on the spatial distribution of the sediment porosity and of the initial total mercury concentration in the top-sediments". Sensitivity analysis is never discussed in the manuscript but should be.

Authors answer:
The sensitivity analysis has been not performed for the sediment porosity and the initial total mercury concentration in the top-sediments. These two spatial variables were fixed by using experimental findings. On the other hand, in the sentence we intended to stress only that the benthic mercury fluxes strongly depend on these two spatial variables. According to this, we modified the sentence as follows:
"The model framework for the sediment compartment causes that the spatio-temporal dynamics of the benthic mercury

flux strongly depends on the spatial distribution of the sediment porosity and of the initial total mercury concentration in the top-sediments, which were fixed using the experimental data."

88. **Reviewer** wrote:
L500 "Sensitivity analysis performed on the environmental parameters and variables used in the seawater compartment indicates that the spatio-temporal dynamics of [HgT] and [HgD] primarily depends on the velocity field of the marine currents obtained from the hydrodynamic model (Burchard and Petersen, 1999; Umgiesser et al., 2004; Umgiesser, 2009; Umgiesser et al., 2014; Ferrarin et al., 2014; Cucco et al., 2016a, b, 2019), even if the role played by the vertical and horizontal diffusivities (Pacanowski and Philander, 1981; Massel, 1999; Katz et al., 1979; Denman and Gargett, 1983; Peters et al.,1988; Valenti et al.,2015,2017)". How did you perform sensitivity analysis on HgT that is not modeled but assumed to be linearly correlated to HgD (eq.13)? From my experience, HgD and HgT dynamics are likely to have different drivers. If sensitivity analysis has been carried out, it should be described. There are too many references.

Authors answer:
We removed $[Hg_T]$ and some references from the sentence.
The sensitivity analysis is quite difficult to perform because of the variations of velocity field and/or diffusivities. The spatial distributions of the dissolved mercury indeed are strongly affected by these non-stationary conditions, which make hard to evaluate correctly the effects of a parameter and its variations.
In order to describe the performed sensitivity analysis , we inserted the following sentences at page 27:
"Specifically, the spatio-temporal behaviour of $[Hg_D]$ changed significantly when alternative velocity fields for the Augusta Bay were used in the biogeochemical module, confirming a feature already observed in previous models. Conversely, limited changes in the spatial distribution of $[Hg_D]$ were observed when different values of vertical and horizontal diffusivities were set in our model."

89. **Reviewer** wrote:
L508 if both data and model suggest Hg is mostly in the particulate phase, why is your model designed around the dynamics of dissolved Hg species?

Authors answer:
Our model has been designed to reproduce the dynamics of dissolved Hg species since we could not calibrate a sediment transport model with sufficient reliability to reproduce the spatio-temporal behaviour of suspended particulate matter.

90. **Reviewer** wrote:
L510 "the amount of mercury dissolved in pore water is negligible with respect to the total amount in the sediments" this is not a finding of this work.

Authors answer:
According to reviewer's indication and new simulations, we deleted the sentence at page 27 and modified the rest part of paragraph as follows:
"According to the available experimental data, the theoretical results obtained with the HR3DHG model suggest that the amount of mercury bound to the particulate matter is quite high in seawater compartment (about 47% of the Hg_T on average). Because of the exponential decay of $[Hg_T]$ in sediments, (...)".

91. **Reviewer** wrote:
L511-L512 "In general, the concentration of the three mercury species dissolved in seawater decreases slowly as a function of time, whereas their concentration ratios remain approximately constant" this happens due to the exponential decay of Hg parameterized for sediment and the absence of inputs in your system (except atmospheric deposition, which is set to be very low, about 3 times lower than observations in Bagnato et al. 2013).

Authors answer:
According the reviewer's comment, we modified the sentence at page 27 as follows:
"Because of the exponential decay of $[Hg_T]$ in sediments, the concentration of the three mercury species dissolved in seawater decreases slowly as a function of time, whereas their concentration ratios remain approximately constant."

92. **Reviewer** wrote:
L526-528 "the amount of mercury absorbed by the phytoplankton, and recycled in seawater, is negligible. In this last respect, it is however important to underscore that even a reduced amount of MeHg entering phytoplankton cells can be very dangerous for the health of human beings due to the bio-accumulation processes which occur throughout the food chain." this seems to be pointless in the discussion. Can you quantify the amount of Hg adsorbed and released?

Authors answer:
These theoretical findings are reported in the manuscript since they represent secondary results of model. In our simulations we could quantify only the Hg adsorbed by POM (see text at page 28) and the content of Hg^{II} (on average $0.000166\ mg/Kg$ in 2011) and $MeHg$ (on average $0.000021\ mg/Kg$ in 2011) within the picoeukaryotes cells. According to this, the paragraph at page 28 has been modified as follows:
" More specifically, in the quasi-stationary condition, the model results indicate that most of the recycled mercury returns to the sediments where is re-buried, and that the amount of mercury absorbed by the POM (0.008 $kmol\ y^{-1}$ for the year 2011), and recycled in seawater, is negligible. In this last respect, it is however important to underscore that even a reduced amount of $MeHg$ entering living phytoplankton cells can be very dangerous for the health of human beings due to the bio-accumulation processes which occur throughout the food chain."

93. **Reviewer** wrote:
L529 "The dynamics of the particulate matter deposition-resuspension process (Neumeier et al., 2008; Ferrarin et al., 2008) does not significantly modify the spatial distribution of the HgT recycled at the surface layer of the sediments" the dynamics of particulate matter are not included in the Hg model, and never presented in the manuscript, so where does this statement come from? (see comments on L214-218)

Authors answer:
This sentence has been removed by the manuscript. In our work no sediment model is now used to describe the dynamics of particulate matter deposition/re-suspension process at the sediment-water interface.

94. **Reviewer** wrote:
L530-533 "Moreover, the theoretical results show that the recycled mercury flux in the Augusta Bay can only partially be described by the scavenging process of organic particles, which however needs further experimental investigations. In fact, improved knowledge of the scavenging process would be necessary to obtain a better estimation of the HgT removed from the water column". Which are other processes affecting the "recycled mercury"? I argue that here, rather than more experimental investigation, you would need a model that can reproduce particulate Hg dynamics.

Authors answer:
According to reviewer's indications, we modified the paragraph at page 28 as follows:
"The theoretical results show that the recycled mercury flux in the Augusta Bay is only partially described by the scavenging process. In particular, an underestimation of the sinking flux for POM-bound mercury is observed when the NPP coming from the NP model is used in Eqs. (4)-(5). Probably, this behaviour is due to the $chl-a$ concentration conversion equation of Baines et al.(1994), which has been calibrated for oceans instead of coastal zones. For this reason, the NPP estimation would need further experimental and theoretical investigations. Moreover, a deeper knowledge of the scavenging process, which determines the particulate Hg dynamics, would be necessary, from a theoretical point of view, to obtain a better estimation of the Hg_T removed from the water column."

95. **Reviewer** wrote:
L537 I do not think is relevant talking about climate change here, at the very end of discussion.

Authors answer:
According to the reviewer's indication, we removed the second-last sentence of Discussion section. Moreover, we modified the last sentence of section as follows:
"Finally, for its features, the HR3DHG model may represent a useful tool to explore and predict the effects of environmental changes on the mercury dynamics for several possible forthcoming environmental scenarios."

96. **Reviewer** wrote:
L551 why should this model be a promising tool to explore and predict the effects of climate changes on Hg dynamics?

Authors answer:
We deleted the last sentence of Conclusion section.

Supplement material

97. **Reviewer** wrote:
S1.1 All the parts that are repetition of section 3.1 in the main text must be removed.

Authors answer:
All repetitions of section 3.1 in the main text have been removed.

98. **Reviewer** wrote:
L49 why is Net Primary Production obtained from satellite if you have a NP model run for the area?

Authors answer:
The Net Primary Production (NPP) is now calculated by using the NP model and conversion equation of Baines et al.(1994). According to this, the definition of NPP at page 3 of Supplement has been modified as follows:
"(...) where NPP is the net primary production obtained by the NP model, (...)"

99. **Reviewer** wrote:
L51-52: this seems to be a misunderstanding of the concept of euphotic layer. If equation S7 is not used must be removed.

Authors answer:
In our work, the euphotic zone depth ($z_0 = 75\ m$) is that arbitrarily fixed by Zhang et al.(2014) for equation S6. To clarify the concept of euphotic layer in this case, we modified the sentence at page 3 of Supplement as follows:
"Since the bathymetry of the Augusta Bay indicates that the water column depth in the whole basin is less than the theoretical euphotic zone depth ($z_0 = 75\ m$) fixed by Zhang et al.(2014), in our model we use only the equation for $z < z_0$"
Moreover, the equation S7 has been removed from the Supplement.

100. **Reviewer** wrote:
L54 why is chlorophyll concentrations obtained from measurements if you have a NP model run for the area? How should the reference Zhang be related to this sentence?

Authors answer:
The chlorophyll concentrations are now calculated by using the NP model and conversion curve of Brunet et al.(2007). To answer the reviewer's questions, the last paragraph of the Section S1.1 has been modified as follows:
"Here, the NPP is calculated by using the conversion equation for $chl\ a$ concentration, as follows:

$$log(NPP(x,y,z,t)) \quad = \quad 2.09 + 0.81 \cdot log(chl\ a(x,y,z,t)), \tag{2}$$

where $chl\ a$ is the chlorophyll concentration $[\mu g \cdot l^{-1}]$ obtained by the NP model (see Section S4).
On the other hand, the $peratio$ is obtained by using the following equation (Zhang et al.,2014):

$$peratio(x,y,z,t) \quad = \quad -0.0081 \cdot T + 0.0806 \ln chl\ a(x,y,z,t) + 0.426, \tag{3}$$

where T is the surface atmospheric temperature $[C]$ coming from remote sensing."

101. **Reviewer** wrote:
S1.1.1 $\phi_{dep} = \frac{Hg_{gas-atm} \cdot Pr}{\Delta t}$ is not dry+wet deposition of Hg, and is a questionable parameterization.

Authors answer:
We consider only the wet deposition for Hg^0. According to this, we modified the text of Supplement at page 4.

About parameterization, we used the equation of Bagnato et al. (2013) to calculate the wet deposition both for Hg^0 and Hg^{II}. Therefore, we do not understand why this parameterization is questionable.

102. **Reviewer** wrote:
S1.1.2 "The lateral fluxes for all variables are set up equal to zero at the boundaries of Augusta basin (Valenti et al., 2017) except where inlets, rivers and sewerage are localized." In this work, only inlets are considered while inputs from rivers and sewerage are assumed to be negligible, but this is not mentioned in the text (it can only be seen from the equations S13 and S14).

Authors answer:
To clarify this point, we modified the sentence at page 5, line 95, of Supplement as follows:
"Since the direct Hg^0 loads from rivers and sewerage are assumed to be negligible for the whole basin, we set:(...)"

103. **Reviewer** wrote:
S1.2 equation S17 is already in the main text (eq.2). Equation S18 should be moved in the main text. In the definition of PHg should be specified that it is estimated externally with the plankton model and is not coupled with the Hg model. It should be explained that is the nutrient recycling efficiency coefficient, which is assumed to be the same for mercury. Equation S19 should be also presented in the main text. The definition given for forg is inconsistent. How are the units for S^{II}_{SPM}? The name S^{II}_{SPM} is misleading, this is an estimate of Hg scavenged by phytoplankton, not SPM.

Authors answer:
According to reviewer's indications, the equations S18 has been moved in the main text, where PHg and λ are better defined. However, it is worth to stress that the Hg model and the Phytoplankton MERLIN-Expo Model are coupled. In fact, the PHg^{II} ($PMeHg$) is obtained by the Phytoplankton MERLIN-Expo Model using dissolved Hg^{II} ($MeHg$) concentration in the seawater (see Section 5 of the Supplement). At the same time, the Hg model uses the PHg^{II} ($PMeHg$) to calculate the load of the dissolved inorganic mercury released by the POM, which is necessary to reproduce the dynamics of dissolved Hg^{II} ($MeHg$) concentration in the seawater (see Section 3.1 of the main text).
The equation S19 has been modified according to reviewer's indications, and moved in the main text.
The S^{II}_{SPM} and S^{MM}_{SPM} have been redefined at page 10 of the revised manuscript. Moreover, all units of parameters and variables are now reported throughout the main text and the Supplement.

104. **Reviewer** wrote:
S1.2.1 equation S21 is only for wet deposition, differently from what is stated

Authors answer:
We modified the equation S21 according to the reviewer's indication. Moreover, the surface dry deposition flux of contaminated particles is now considered in the model. Therefore, we inserted the following sentence at the beginning of page 9 of Section S1.2.1:
"The $Drydep_{part}$ is set equal to that estimated by Rajar et al. (2007) for the whole Mediterranean basin."

105. **Reviewer** wrote:
S1.2.2 The definition for ϕ^{II}_{res} is misleading, why not keeping only the extended version of eq. S22 (which is eq. 7 in the main text)? Is not $Hg^{II}_{dis-water}$ the same as your modeled Hg^{II} with z=zb? Why using a different name?

Authors answer:
The sediment transport module is not included in our model anymore. As a consequence, the terms ϕ^{II}_{res} and ϕ^{MM}_{res} have been removed by Eqs. S22 and S36, respectively.
The Eqs. 7 and 8 of main text are recalled in the Supplement to better define mass transfer coefficients at the water-sediment interface, and to better describe the connections between benthic mercury fluxes and mercury concentrations in the pore water of sediment.
To clarify the terms used in Eq. S22, we replaced $Hg^{II}_{dis-water}$ with $Hg^{II}|_{z=z_b}$. A similar replacement has been done in Eq. S36 between $MeHg_{dis-water}$ and $MeHg|_{z=z_b}$.

106. **Reviewer** wrote:
L212 Why do the values of the "boundary layer thickness above sediment" for Hg and MeHg differ in Table S1? Why are their values so low (0.009 cm for HgII and 0.03 cm for MeHg)? And why are they lower than the "boundary layer thickness below sediment" that is 0.01 for both Hg and MeHg? Equations S22 and S25 is already given in the main text (eq. 16 and 18), refer to that. Equation S25 is not in agreement with Melaku Canu et al., 2015 as stated in L228.

Authors answer:
We recall that the *boundary layer thickness above sediment* for Hg^{II} and $MeHg$ are calibrated separately according to the procedure of Section 3.3, while the *boundary layer thickness below sediment* has been estimated for both mercury species by the average velocity of marine currents.
The Eqs. 16, 17 and 18 of main text are recalled in the Supplement to better describe the parameter setting of these equations. The reference Melaku Canu et al., (2015) in L228 has been removed.

107. **Reviewer** wrote:
S1.2.3 is already in the main text (eq.15).

Authors answer:
The Eq. 15 of main text is recalled in the Supplement to better describe the setting of $Hg_T^{sed}(0)$ coming from experimental data.

108. **Reviewer** wrote:
S1.2.5 eq. S27 is eq. 9 in the main text. Equations S28-S29 are the same as S13-S14, and S42-S43, I think they can be written in a more general formula without repeating them for each Hg species, especially considering that they are set to 0 for your implementation.

Authors answer:
According to reviewer's indications, we removed Sections S1.2.5 and S1.3.4 of the Supplement, and modified the last paragraph of Section S1.1.2 as follows:
"The same boundary conditions (lateral fluxes) are also valid for Hg^{II} , $MeHg$ and Hg_T. The annual net outflow of elemental mercury from (...) and for the whole year. Similarly, the annual net outflows of Hg^{II} and $MeHg$ are calculated.
In order to perform the mass balance for the Augusta Bay, we calculate the annual net outflow of total mercury (O) from the basin towards (...)."

109. **Reviewer** wrote:
S1.3 Equation S32 is already in the main text (eq.3).

Authors answer:
The whole text of Section S1.3 has been removed from the Supplement.

110. **Reviewer** wrote:
L295-296 why all these references?

Authors answer:
See previous answer of authors.

111. **Reviewer** wrote:
L304-307 the text is already in the main text.

Authors answer:
The text at page 13 lines 304-307 has been removed.

112. **Reviewer** wrote:
L308 "The rate constant for the methylation of inorganic mercury is fixed according to Monperrus et al. (2007) (Batrakova et al.,2014; Monperrus et al., 2007b)." Why are references reported in this way? There are other cases.

Authors answer:
The references are now reported in correct way throughout the manuscript.

113. **Reviewer** wrote:
L311-317 same comments as for S1.2 (and it can be explained just once for both Hg and MeHg) .

Authors answer:
The text at page 13 lines 311-317 has been removed.

114. **Reviewer** wrote:
S1.3 Almost the entire section is a repetition of information already given in the main text or in S1.2. The only difference with section S1.2 is the substitution of "HgII" with "MeHg", there must be a better way.

Authors answer:
All repetitions of Section 1.3 have been removed from the text.

115. **Reviewer** wrote:
S1.3.1 MeHg atmospheric deposition is > 0, although low (e.g. Mason et al., 2012).

Authors answer:
The boundary condition for MeHg atmospheric deposition has been changed according to Mason et al., (2012). As a consequence, the text of Section S1.3.1 has been rewritten as follows:
"According to Mason et al. (2012), the methyl-mercury flux at the water-atmosphere interface (z=0) is estimated to be 0.5 % of total Hg deposition flux ($\simeq 0.5\%$ of Hg^{II} deposition flux). Therefore, in our model we set: (...)
The annual atmospheric deposition of the methyl-mercury is calculated by integrating Eq. (S36) for the whole horizontal surface of the basin and for the whole year. The annual total atmospheric mercury deposition (AD) is equal to the sum of the amounts of elemental, inorganic and methyl mercury deposited on the surface of the Augusta basin in one year."

116. **Reviewer** wrote:
S2 "Since the direct loads of SPM (Ciffroy, 2015; Melaku Canu et al., 2015) for the Augusta basin were unknown, the SPM concentration dynamics could not be reproduced correctly. Therefore, we reproduced the spatial distribution of SPM concentration at the steady state by interpolating the experimental data observed in recent samplings (October 2017) performed in the site investigated." You obtained SPM values by interpolating observations and assume steady state conditions, thus why do you present a dynamic model for SPM that is not used (eq.S46)? What about the references? Besides, at a first and second reading is not clear why you need SPM if adsorption of particulate Hg species is based on NPP obtained from satellite data. At some point, I realized that this is only used in eq. 13 to estimate HgT.

Authors answer:
The equation for the SPM dynamics has been removed by Section S2, as well as the references. Moreover, to clarify why the experimental SPM concentrations need, we added at the ending of Section S2 the following sentences:
"The experimental SPM and POM concentrations were used to reproduce the spatial distribution of the fraction of suspended particulate matter as organic carbon (f_{oc}), which was necessary to obtain the sinking fluxes of Hg^{II} and $MeHg$. Afterwards, the SPM concentrations were used to calculate the $[Hg_T]$ in seawater (see Section 3.1)."

117. **Reviewer** wrote:
S2.1 the definition given for f_{org} "organic fraction of suspended particulate matter in dissolved-phase" is meaningless.

Authors answer:
We removed the words "in dissolved phase" from the f_{org} definition. Moreover, we defined the fraction of suspended particulate matter as organic carbon at the ending of Section S2.1, as follows:
"Since we assumed that 52% of organic matter was carbon, the fraction of suspended particulate matter as organic carbon was calculated by using the following equation: (...)"

118. **Reviewer** wrote:
S3 and S3.1 These sections should present the site-specific implementation of SHYFEM model for Augusta Bay, rather

than providing a general description of the model, which is available elsewhere in the literature. The reader should be able to understand how the hydrodynamic and sediment models have been run (add values used for parameters and coefficients, show the calibration). The title for S3.1 is incomplete.

Authors answer:
According to the reviewer's indications, we modified Section S3 of the Supplement. Specifically, a new paragraph entitled " Hydrodynamic model validation" has been added describing the comparison between the model results and ones obtained from previous application in the bay. See in the text from line 567 to line 611.

119. **Reviewer** wrote:
S4 Be aware that eukariotes are not a planktonic population, thus line 566 is meaningless. The term eukariotes is much broader than phytoplankton, which is what is actually simulated here; this must be corrected throughout the text.
The NP model is already described in many publications reported here and is not a central part of this work, so as for the hydrodynamic and sediment models I do not think the model theory should be explained in detail here. Probably a description of the implementation for the Augusta Bay (linked to Table S3) clarifying which are site specific parameters and which parameters are adopted from other areas, would be more useful. I would also like to see calibration, or a map of the output.

Authors answer:
The word "eukaryotes" has been replaced by the word "picoeukaryotes" in the Section S4. We recall that the picoeukaryotes domain is the set of most representative phytoplankton populations of oligotrophic waters of Mediterranean Sea.
To clarify which are site-specific parameters of NP model, we inserted at page 25, lines 661-664, the two following sentences:
"The half-saturation constants for growth of picoeukaryotes, used in the the Michaelis-Menten formulas, depend on the environmental conditions of investigated site. Since the *chlorophyll-a* concentrations, measured in the Augusta Bay, are those typical of oligotrophic waters of the Mediterranean Sea, the half-saturation constants are set equal to values previously obtained in the Southern Sicily by Valenti et al. (2017) adopting an accurate calibration procedure." Moreover, we modified the sentence at page 25, lines 664-667, as follows:
"All other parameters are set in accordance with the methods described in previous works, while the temporal behaviour of incident light intensity, $I_{in}(t)$, is obtained for the Augusta Bay by using the remote sensing data."

120. **Reviewer** wrote:
Table S1 all parameters where "no data" is reported are set to 0. Is it possible to make assumptions and estimate them?

Authors answer:
We cannot estimate the parameters where "no data" is reported. However, we can speculate that these parameters are negligible according to field observations.

121. **Reviewer** wrote:
Table S2 why is it separated from S1? The caption is the same. Do you have 0 porosity value?

Authors answer:
The caption is not the same. In fact, Table S1 describes the parameters of biogeochemical model, while Table S2 reports the range of the variables of model. Finally, the minimum value of porosity has been corrected.

122. **Reviewer** wrote:
Table S9 It should be explained in the main text why your modeled value of atmospheric deposition is 2.5 lower than the deposition measured at your study site in 2011-2012 (Bagnato et al. 2013; Salvagio Manta et al. 2016). How would affect your results the increase atmospheric deposition to the observed rate? It should be also explained in the main text why all inputs different from atmospheric deposition are set to 0.
Why is the term "MeHg released from sediment" in this HgT budget? Here only the total is needed. Why is MeHg budget not estimated?

Authors answer:
According to reviewer's indications, we inserted the following new paragraphs at the ending of page 22 of the main text: "This discrepancy is due to different calculation methods used in the two works. Specifically, in our model the AD is calculated by using both the atmospheric mercury concentrations and the average precipitations, measured for all months of the year. On the contrary, in Bagnato et al. (2013) the AD is calculated by averaging the experimental data acquired during a time limited sampling period (from 29^{th} August 2011 to 23^{th} April 2012), namely without considering the year period in which the amount of precipitation is very low. By this way, the AD obtained by Bagnato et al. (2013) is very higher than that of our model, even if it is probably overestimated due to calculation method used. In general, the contribute of AD is negligible in the mercury mass balance of the Augusta Bay. Indeed, the simulations indicate that a strong increase of atmospheric mercury deposition caused by environmental changes (dust fall increase and/or rainfall increase), would not affect on numerical results of our model significantly.
The annual net mercury inflows (A) from rivers and sewerage to basin are assumed to be negligible in agreement with field observations. Specifically, the flow rate of Marcellino river is equal to zero for the most part of year, while the inflow from the sewerage is low. Moreover, it is fair to speculate that the Hg concentration in fresh waters discharged in the Augusta Bay was decreased significantly after the chlor-alkali plant closure."
According to reviewer's indication, the "MeHg released from sediment" has been removed by Hg_T budget in Table S11. Moreover, it is worth to underline that the annual fluxes of MeHg are estimated by our model, even if they cannot validated with experimental findings. For this reason, the MeHg budget has been not reported in the paper.

123. **Reviewer** wrote:
Table S5-S6-S7-S8-S10. To improve readability, I would move the station/longitude/latitude information for data and model in a separate table and keep here only Station | Period | Depth | Hg.

Authors answer:
According to reviewer's indication, the station/longitude/latitude information for data have been moved in a separate table (see Table S4 of Supplement).

124. **Reviewer** wrote:
Figure S3. I don't see a good agreement here. I took the effort to copy your Hg_D data from Table S5 and found an average relative error of 86% (median 62%) between model and observations, this could even be fine if you acknowledge it and properly discuss it.

Authors answer:
The goodness of agreement between the theoretical results and experimental data is now discussed at pages 19-20 of the main text.

125. **Reviewer** wrote:
Figure S4. The scale (up to 140) is almost two time as high as the maximum value shown, so almost everything appears in blue.

Authors answer:
The color scale of Figure S4 has been changed to make comparable the experimental and theoretical data maps.

Best regards.

On behalf of all authors

Alessandro Borri