Reviewer 2:

General

This paper provides a technical note on application of causal inference to the effects of solar radiation and water temperature on dissolved gaseous mercury (DGM). This research is really interesting, instrumental, and insightful.

This research showcases a wonderful collaboration between experimental scientists and causal inference scholars.

What a Wonderful World is this interdisciplinary field.

This paper is expected to embrace a wide range of readers, including those who know or have a good commend of causal inference already and also those who are lay people, well-trained in experimental sciences, yet knowing little about causal inference and how to use and apply it in their experimental science areas. The present reviewer is among the latter group. Hence, this review will focus on two aspects: (1) experimental and (2) how to help and guide the latter group of readers to follow, understand, and learn how to use causal inference by means of the case study provided by this paper. Some readers, if not many, may share the same or similar feedback as presented in this review.

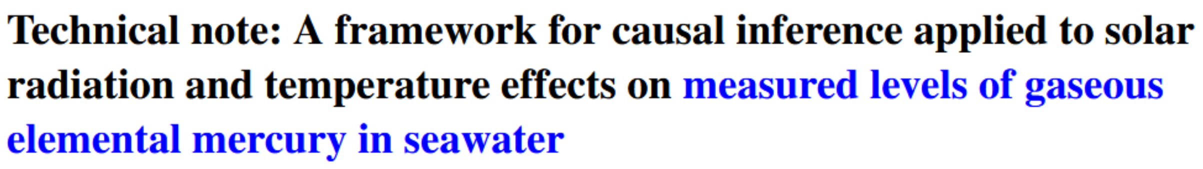
- *We thank the reviewer for the kind words about our paper. Indeed, one of the aims of the paper was to demonstrate successful collaboration between two different research disciplines; something the research community needs more of. The paper is hopefully useful to a wide range of readers and aims to inform, demonstrate and inspire future research collaborations between research fields.*

Specific

1. **Paper title**

The paper title uses the word "on dissolved gaseous mercury". Perhaps, this term in the context of this research is kind of vague and could be more specific, say, on levels of DGM, or generation process and mechanism of DGM, or speciation of Hg, etc. So it's a bit unclear what is exactly the effect (effect of solar radiation on what exactly, DGM level, dynamics, production?), since DGM itself is only a particular species of aquatic Hg.

- *We are thankful for the nice suggestions to make the title more informative and clearer. We suggest a change of the title to "Technical note: A framework for casual inference applied to solar radiation and temperature effects on measured levels of gaseous elemental mercury in seawater." The title in the paper has been changed accordingly, see below:*



Technical note: A framework for causal inference applied to solar radiation and temperature effects on measured levels of gaseous elemental mercury in seawater

2. **Experimental**

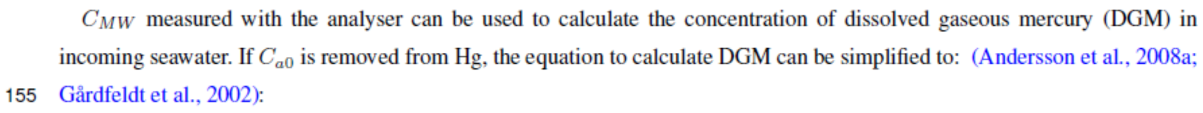
Regarding the in-situ field measurement of DGM, a number of questions arise:

First, the citation for this method seems to use a less relevant paper (by Andersson ME et al., 2008b; see L140 in the present paper). I checked on this and found the relevant references probably would be:

- A description of an automatic continuous equilibrium system for measurement of dissolved gaseous mercury. By Andersson, Gardfeldt, and Wangberg, Anal. Bioanal. Chem. 391, 2277-2282, 2008a
- Seasonal and spatial evasion of mercury from the western Mediterranean Sea by Nerentorp Mastromonaco, Gardfeldt, and Wangberg, 2017 (L896-897 in the present paper).

- *We agree with the reviewer that the two suggested references fit better to L140 than the previous added reference. The suggested Ref#1 has now replaced the previous reference on L140 in the revised paper. Also, another reference has been added: "Gårdfeldt et al., 2002.: "Comparison of procedures for measurements of dissolved gaseous mercury in seawater performed on a Mediterranean cruise." Analytical and Bioanalytical chemistry 374.6 (2002): 1002-1008.", which further describes how the equations used to calculate DGM, using the same continuous system as used in this study, were derived and verified.*

- *Section 2.1 #154-155:*



C_{MW} measured with the analyser can be used to calculate the concentration of dissolved gaseous mercury (DGM) in incoming seawater. If C_{a0} is removed from Hg, the equation to calculate DGM can be simplified to: (Andersson et al., 2008a;
155 Gårdfeldt et al., 2002):

Second, with limited time available, I consulted Ref. #2 above and had some findings as detailed below.

Ref. #2 shows that the researchers also used another manual method, i.e., purge-and-trap method, instead of the in-situ auto-method, to determine the DGM. For this manual method, first, the Hg(0) in a water sample of a certain volume is completely purged out of the water sample using zero air (or pure Ar or N2) and then collected on a Hg trap to analyze the total Hg(0) purged out of the water sample. By measuring the volume of the water sample and the total Hg(0) purged from the water sample and collected on the Hg trap, the DGM can thus be calculated to be DGM = (total Hg(0) purged)/(volume of water sample). This method gives a clear determination of the DGM for the water sample without confusion or misunderstanding.

- *We thank the reviewer for opening this interesting discussion. We believe that it is indeed true that the discrete manual purge and trap method earlier has been the most common method for analysing DGM in water samples. The reviewer is right, when using the manual method, the approach is to completely purge the sample. DGM is then calculated by dividing the amount of purged Hg by the sample* *volume. We do agree with the reviewer that this method is more straight-forward and leaves less confusion regarding the experimental performance. However, discrete sampling would not have been an appropriate method to use for our study. The number of data points needs to be large for the statistical significance of our model, and a high time resolution of Hg analyses is crucial to match fast changes in solar radiation. Using a manual method would require an immense workload and would result in an insufficient time resolution that is needed for this study.*

Moreover, Ref. #2 also mentioned that they compared the DGM results from the auto-method and the manual method and found “a good correlation” between the two method results. This means that the DGM calculated using Eq. 1 and the DGM obtained by the manual method differ, although correlated, that is, one may not replace the other, but one can be obtained from another using the correlation.

- *We agree with the reviewer that Ref#2 (also Ref #1and Gårdfeldt et al., 2002) compared DGM results from the automated- and manual methods with good correlations. Although this is a nice discussion point, it was not part of our paper to compare manual and automated methods to measure DGM in surface water.*

However, Ref. #2 does not mention or indicate if they used the correlation (or calibration) to get the DGM corresponding to the actual DGM (calibrated by the manual method), or they simply took the DGM results calculated using the equation of DGM = Ca(1/H + ra/rw) (L141 Eq. 1 in the present paper). This missing detail is a highly important technical detail, which is connected to the credibility of this auto-method, and subsequent causal inference operations and outcomes.

- *We understand the reviewer’s concern regarding the confusion. As mentioned before, a rigorous comparison between the automated and manual sampling was performed in Ref#1. To our knowledge, Ref#2 compared the two methods at five measurement points, showing some agreement between the methods. The comparison was performed to check and compare the two methods, not to calibrate them against each other. Ref #2 used equation 1 to calculate DGM concentrations in their paper, an equation that has been used to calculate DGM in many other papers, without the necessity to re-calculate the values using calibration against the manual method.*

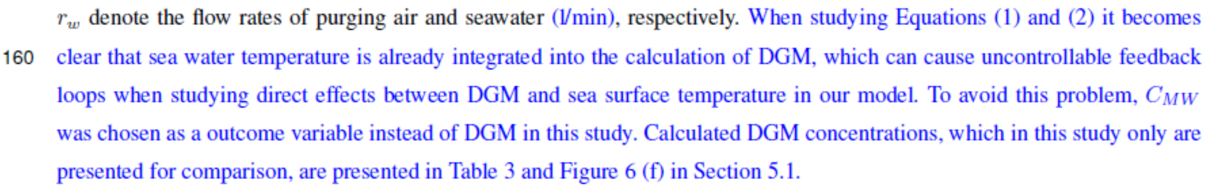
I’d think the correlation (equation) should be reported and used to get the real DGM as calibrated using the correlation, rather than just using the DGM results directly from the calculations using Eq. 1, for the reasons given below. By the way, it’s understandable there is a need to have an in-situ auto method to continuously measure DGM in the field.

But, it remains unclear for the present paper under review, all the DGM results used for the causal inference are those directly from the calculation using Eq. 1, or those after processing using the correlation between the auto and manual methods (calibration of the auto-method by the manual method). This important technical detail needs to be clarified.

- *We agree with the reviewer that the automated method would benefit from being compared to other field methods to measure and calculate DGM concentrations in seawater (although, to our knowledge, not many other methods exist yet). However, in our study, the calculated DGM concentrations were not used in the causal model. For the model we used measured (not calculated) Hg concentrations (CMW) for comparing with solar radiation and temperature. The reason for this choice was because Henry's law constant, which is used in equation 1 to calculate DGM, is temperature dependent (see equation 2) and therefore, calculated DGM cannot be used in the causal model when comparing DGM to temperature since it would cause an uncontrollable feedback loop. Although we believe equation 1 to be correct for calculating DGM, we only used this equation to calculate DGM values used for demonstration and comparison in Table 3 and Figure 6.*

  We are sorry for the confusion. Throughout the text we have now tried to be clearer that we are studying measured gaseous elemental Hg (CMW) rather than DGM. In Section 2.1 we also added an explanation why we made this choice, see below:

  Section 2.1 #159-163:



r_w denote the flow rates of purging air and seawater (l/min), respectively. When studying Equations (1) and (2) it becomes clear that sea water temperature is already integrated into the calculation of DGM, which can cause uncontrollable feedback loops when studying direct effects between DGM and sea surface temperature in our model. To avoid this problem, C_{MW} was chosen as a outcome variable instead of DGM in this study. Calculated DGM concentrations, which in this study only are presented for comparison, are presented in Table 3 and Figure 6 (f) in Section 5.1.

The auto-method appears not quite straightforward in conjunction with Eq. 1. By the auto-method, a given water volume is first pumped into the inner cylinder. Then (or simultaneously) zero air is used to purge the Hg(0) in the given water to the headspace of the inner cylinder. Then the air concentration of Hg(0) in that headspace is measured by Lumex (or Tekran 2537A). By the way, the efficiency of the purging is not mentioned or discussed in this paper. The efficiency of purging is certainly critical for the manual method. Incomplete purging of the DGM can cause under-estimation of real DGM level.

<y curious why Eq. 1 is used to calculate the real DGM of the sea water, instead of using the same approach as the manual method to get the total Hg purged out of the water left in the cylinder headspace and then the DGM thus determined. It is also highly curious why the DGM is the Hg(0) concentration in the water of the cylinder supposedly at equilibrium with the Hg(0) purged out of the same water then present in the headspace measured by Lumex. Intuitively, this is quite confusing and not revealing. The key point here is why the equilibrium of Hg(0) distribution between air and water gets involved in the DGM determination? In any context, it is the real DGM of interest, not the equilibrium DGM.

It is very hard to see and understand how this so-calculated equilibrium Hg(0) concentration can represent the real DGM in the water sample. First of all, the real DGM should be the one at the equilibrium with the ambient air Hg(0) above the sea, rather than with the Hg(0) purged out of the water sample in the cylinder headspace, unless coincidentally, the Hg(0) in the ambient air has the same concentration as the purged Hg(0) in the headspace. It is very hard to see the materialization of such a coincidence, consistently occurring all the time. Or was this coincidence confirmed experimentally?

Using the Henry's law method to get DGM only gives the Hg(0) concentration in the water at the equilibrium, while as known, water is commonly saturated or often over-saturated with Hg(0), i.e., DGM at equilibrium < or << DGM-real.

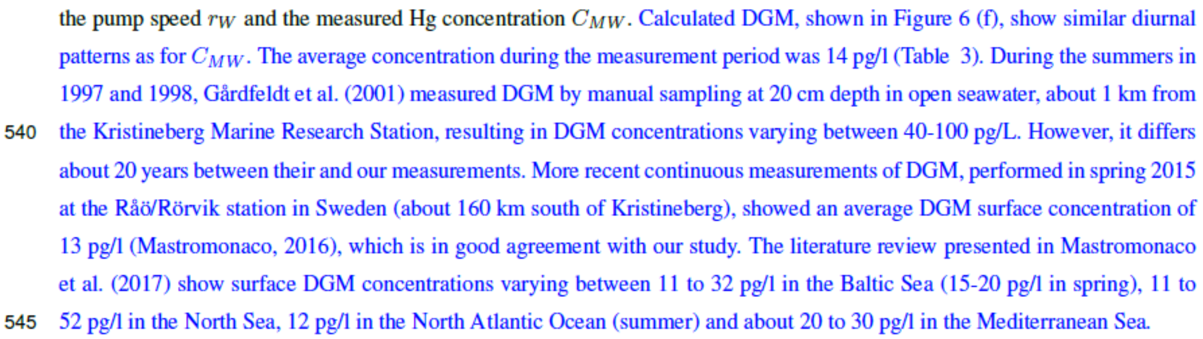
- *We understand that this confusion remained, but we hope that we now have explained our choice of using equation 1 to calculate DGM concentrations used for demonstration and comparison with other studies. In manual sampling, Henry's law is not needed, as it is needed in Equation 1 when using the automated method. Henry's law constant, that shows how much a gas dissolves in water at equilibrium, is used in equation 1 to compensate for the choice of measuring equilibrium concentrations rather than purging the total amount of Hg in the sample, as in manual methods. This approach of measuring equilibrium concentrations is also used for measuring other gases in water, such as CO2, using similar methods and equations, using Henry's law constant for CO2 in seawater. Hence, the theory behind this method is not original but well studied. See for example "Wanninkof, R. and K. Thoning (1993) Measurement of fugacity of CO2 in surface water using continuous and discrete sampling methods. Mar. Chem. 44: 189- 204".*

Table 3 and Fig. 6f all show quite low levels of DGM, as compared to many studies that reported higher DGM levels for various waters. This suspected underestimation of the DGM might be due to that the calculated DGM is only for the equilibrium condition as calculated using Henry's law.

The unclarity and confusion regarding the meaning and credibility of the DGM calculated using Eq. 1 need to be resolved in the first place before readers go further to see any causal inference using the DGM results.

- *We thank the reviewer for demonstrating that a comparison with literature is missing in our paper. The calculated DGM concentrations (that are in this paper only presented for comparison and not used in the analysis) showed an average concentration of 14 (5-28) pg/L. For comparison, surface DGM was measured using an in situ purging system in mars/April 2015, also at the Swedish west coast at Råö/Rörvik station, about 160 km south of Kristineberg. Here, the average DGM concentration was 13 pg/L, which is in good agreement with our results (Nerentorp Mastromonaco PhD thesis, 2016). A literature review has now been added in section 5.1, see below:*

- *Section 5.1: #537-545:*



the pump speed r_W and the measured Hg concentration C_{MW}. Calculated DGM, shown in Figure 6 (f), show similar diurnal patterns as for C_{MW}. The average concentration during the measurement period was 14 pg/l (Table 3). During the summers in 1997 and 1998, Gårdfeldt et al. (2001) measured DGM by manual sampling at 20 cm depth in open seawater, about 1 km from the Kristineberg Marine Research Station, resulting in DGM concentrations varying between 40-100 pg/L. However, it differs about 20 years between their and our measurements. More recent continuous measurements of DGM, performed in spring 2015 at the Råö/Rörvik station in Sweden (about 160 km south of Kristineberg), showed an average DGM surface concentration of 13 pg/l (Mastromonaco, 2016), which is in good agreement with our study. The literature review presented in Mastromonaco et al. (2017) show surface DGM concentrations varying between 11 to 32 pg/l in the Baltic Sea (15-20 pg/l in spring), 11 to 52 pg/l in the North Sea, 12 pg/l in the North Atlantic Ocean (summer) and about 20 to 30 pg/l in the Mediterranean Sea.

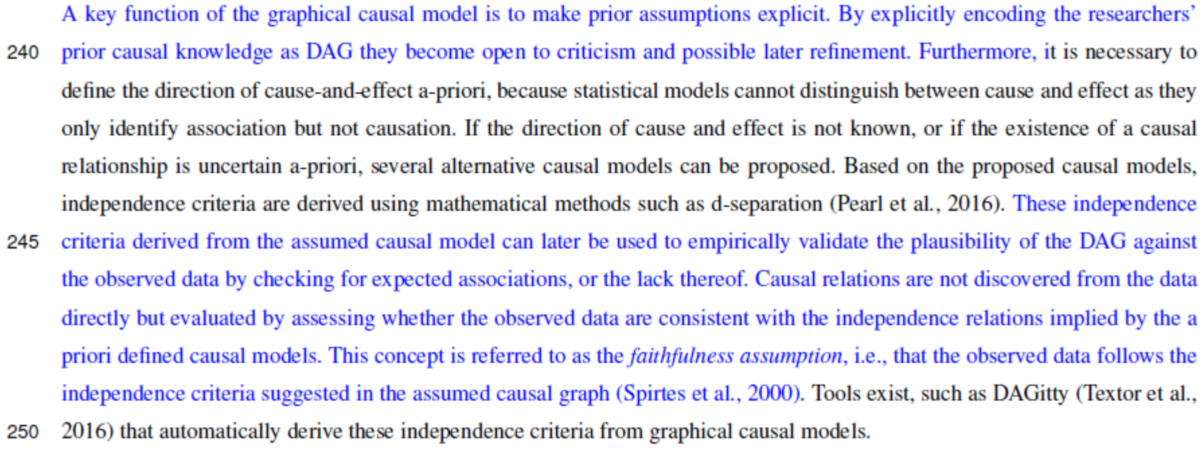
3. **Causal inference general**

Before and during reading this paper for a while, I always thought this causal inference model or operation can determine if two factors given are actually indeed causally related, instead of simply correlated. In other words, the expectation was that by running the causal inference (going through the entire framework and running the causal inference operations or models), it can be determined if one factor is causally related to another, followed further by the effect size.

But, the more I read through, the more I thought or realized (maybe I'm still wrong or doesn't get it) that actually, it seems that to begin the causal inference, one needs to assume, in the first place, the two factors are indeed causally related, and then running the causal inference through the framework would provide more knowledge about the relationship between the two factors, like the effect size, this percentage for this factor, or that percentage for that factor, etc.

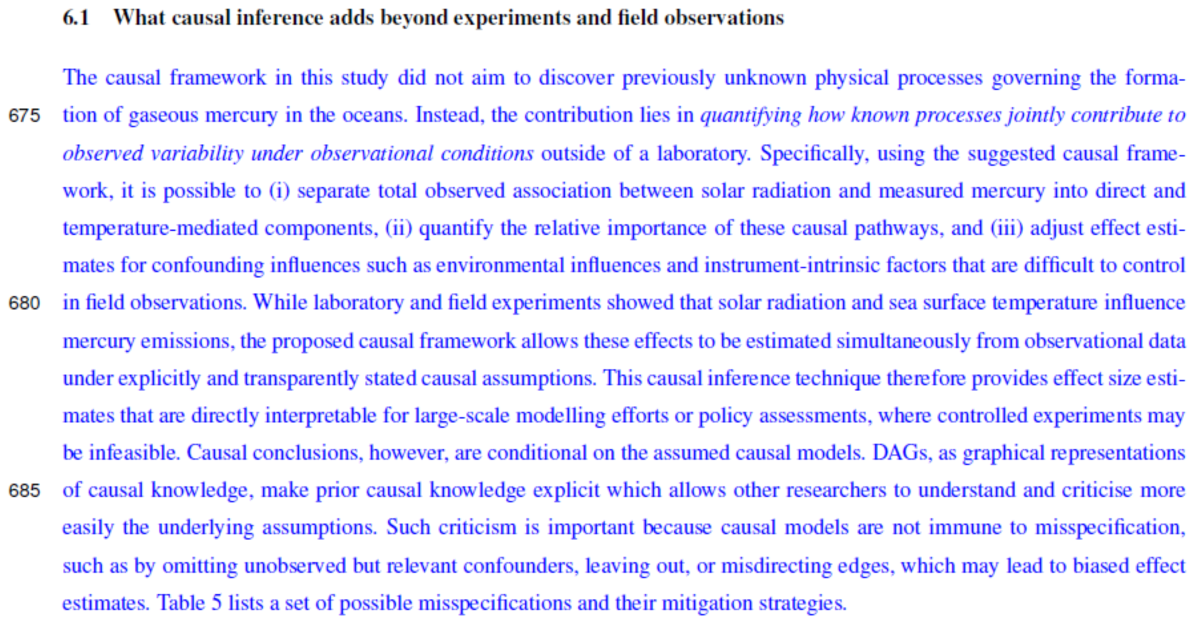
- *We thank the reviewer for giving this insightful comment about the lacking information regarding how to interpret and understand how the causal model works. It is indeed true that for the model to work you need to first have an idea how and if two factors are related. That's why it is important to draw your DAGs correct before running the model and interpret the results. That is what we describe in the paper to be prior scientific knowledge. We have improved the description of what causal models can do and what they cannot do throughout the manuscript.*

- *Section 3: #239-240 and 245-249:*



A key function of the graphical causal model is to make prior assumptions explicit. By explicitly encoding the researchers' prior causal knowledge as DAG they become open to criticism and possible later refinement. Furthermore, it is necessary to define the direction of cause-and-effect a-priori, because statistical models cannot distinguish between cause and effect as they only identify association but not causation. If the direction of cause and effect is not known, or if the existence of a causal relationship is uncertain a-priori, several alternative causal models can be proposed. Based on the proposed causal models, independence criteria are derived using mathematical methods such as d-separation (Pearl et al., 2016). These independence criteria derived from the assumed causal model can later be used to empirically validate the plausibility of the DAG against the observed data by checking for expected associations, or the lack thereof. Causal relations are not discovered from the data directly but evaluated by assessing whether the observed data are consistent with the independence relations implied by the a priori defined causal models. This concept is referred to as the *faithfulness assumption*, i.e., that the observed data follows the independence criteria suggested in the assumed causal graph (Spirtes et al., 2000). Tools exist, such as DAGitty (Textor et al., 2016) that automatically derive these independence criteria from graphical causal models.

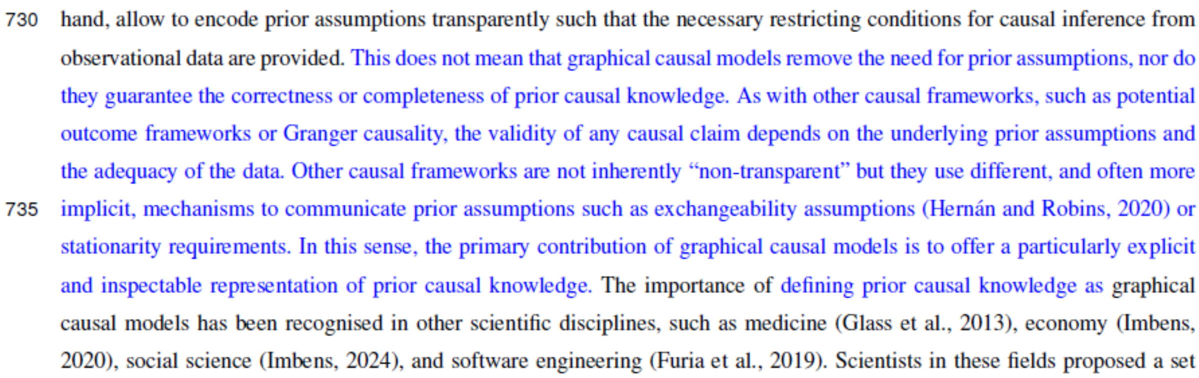
- *New Section 6.1 #680-695:*



6.1 What causal inference adds beyond experiments and field observations

The causal framework in this study did not aim to discover previously unknown physical processes governing the formation of gaseous mercury in the oceans. Instead, the contribution lies in *quantifying how known processes jointly contribute to observed variability under observational conditions* outside of a laboratory. Specifically, using the suggested causal framework, it is possible to (i) separate total observed association between solar radiation and measured mercury into direct and temperature-mediated components, (ii) quantify the relative importance of these causal pathways, and (iii) adjust effect estimates for confounding influences such as environmental influences and instrument-intrinsic factors that are difficult to control in field observations. While laboratory and field experiments showed that solar radiation and sea surface temperature influence mercury emissions, the proposed causal framework allows these effects to be estimated simultaneously from observational data under explicitly and transparently stated causal assumptions. This causal inference technique therefore provides effect size estimates that are directly interpretable for large-scale modelling efforts or policy assessments, where controlled experiments may be infeasible. Causal conclusions, however, are conditional on the assumed causal models. DAGs, as graphical representations of causal knowledge, make prior causal knowledge explicit which allows other researchers to understand and criticise more easily the underlying assumptions. Such criticism is important because causal models are not immune to misspecification, such as by omitting unobserved but relevant confounders, leaving out, or misdirecting edges, which may lead to biased effect estimates. Table 5 lists a set of possible misspecifications and their mitigation strategies.

- *Section 6.3 #731-737:*



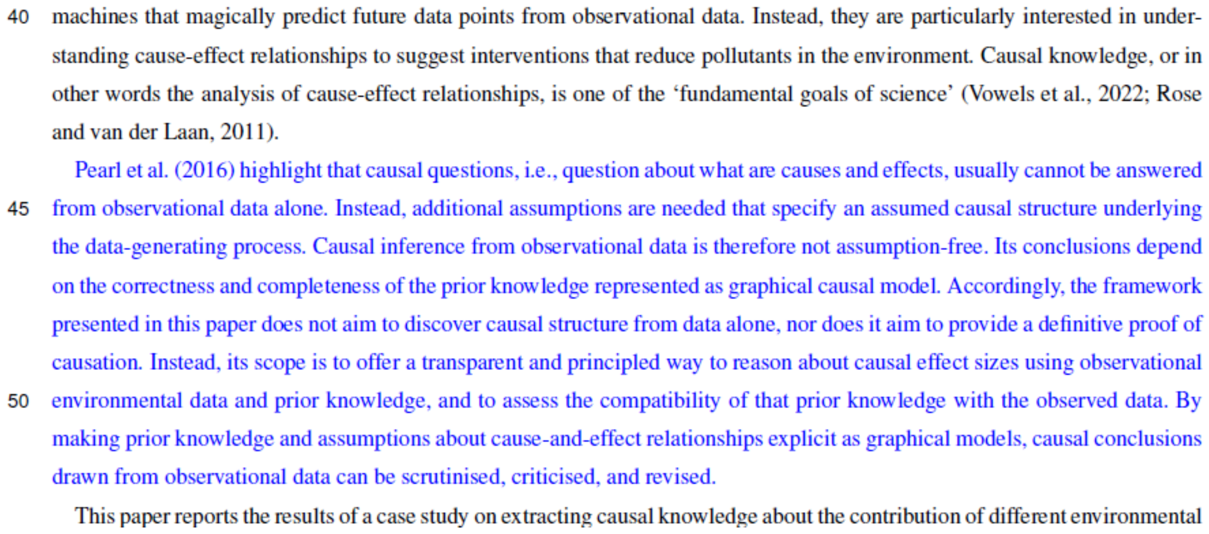
hand, allow to encode prior assumptions transparently such that the necessary restricting conditions for causal inference from observational data are provided. This does not mean that graphical causal models remove the need for prior assumptions, nor do they guarantee the correctness or completeness of prior causal knowledge. As with other causal frameworks, such as potential outcome frameworks or Granger causality, the validity of any causal claim depends on the underlying prior assumptions and the adequacy of the data. Other causal frameworks are not inherently "non-transparent" but they use different, and often more implicit, mechanisms to communicate prior assumptions such as exchangeability assumptions (Hernán and Robins, 2020) or stationarity requirements. In this sense, the primary contribution of graphical causal models is to offer a particularly explicit and inspectable representation of prior causal knowledge. The importance of defining prior causal knowledge as graphical causal models has been recognised in other scientific disciplines, such as medicine (Glass et al., 2013), economy (Imbens, 2020), social science (Imbens, 2024), and software engineering (Furia et al., 2019). Scientists in these fields proposed a set

So, top front, it would be very helpful to provide a general description of the causal inference, it's goal, logic assumption and framework, approach, what the causal inference is and can or could do, what we can or could expect the causal inference to offer, and moreover, what the causal inference cannot offer or do. This general introduction is much needed. Or, readers, like me, would be struggling in the confusion about if the causal inference can settle the case to determine the causality, or instead, only can provide more inference about the relationship between two or more factors and the effect size of each factor, beyond simple correlation analysis.

So, if the causal inference cannot determine if two or more given factors are indeed causally related, and which is the cause of which (or otherwise), then this nature of the causal inference needs to be stated/indicated clearly in the very beginning. This would help and benefit many readers, like me, who, inference-via-scientific-experiments oriented, probably first time encounter a detailed case like the one provided by this paper. For example, a lot has been known about how solar radiation can causally induce and enhance DGM generation via photochemical reactions by means of well-controlled manipulative experiments (with only one factor tested in variation and other factors fixed to logically satisfy both necessity and sufficiency requirements for causal-effect relationship determination).

- *We are very grateful for this concrete suggestion to improve the paper. We have added clarifications to the Introduction (Section 1) and to the outline of the proposed framework in Section 3. In particular, we highlight that our suggested framework does not establish causality from observational data alone. Instead, causal inference means, in this context, estimating direct and indirect effect sizes conditional on explicitly stated causal assumptions encoded as graphical causal models following the methodology outlined by Pearl et al. (2016). We hope that the additional information helps set the reader's expectation early and to clarify how the proposed framework goes beyond usual correlation analysis while remaining conditional on the correctness and completeness of prior scientific knowledge. For example, we knew before writing this paper that solar radiation can induce DGM generation by photochemical processes. We also knew that temperature affect DGM in some way, but what we did not know was how it was all connected. Is it rather that solar radiation affects the measured mercury concentration (C_{MW}) indirectly by temperature increase alone? This we early realized by running the model that this is not true. Then arose the question, how much of the C_{MW} generation is affected by only solar radiation and only temperature? In this model we could "turn off/lock" the effect of one factor to see how much the other factor affected C_{MW} and vice versa. With the help of the model we could apply lab experiments on real measured data in the field. And this is the strength of this framework.*

- *Section 1 #44-52:*

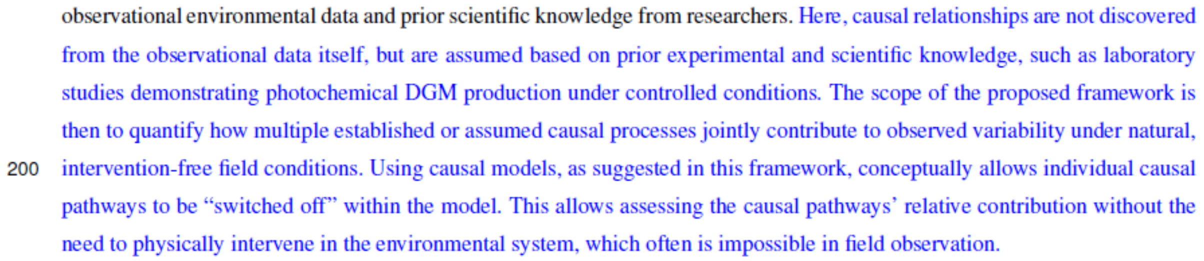


machines that magically predict future data points from observational data. Instead, they are particularly interested in understanding cause-effect relationships to suggest interventions that reduce pollutants in the environment. Causal knowledge, or in other words the analysis of cause-effect relationships, is one of the 'fundamental goals of science' (Vowels et al., 2022; Rose and van der Laan, 2011).

Pearl et al. (2016) highlight that causal questions, i.e., question about what are causes and effects, usually cannot be answered from observational data alone. Instead, additional assumptions are needed that specify an assumed causal structure underlying the data-generating process. Causal inference from observational data is therefore not assumption-free. Its conclusions depend on the correctness and completeness of the prior knowledge represented as graphical causal model. Accordingly, the framework presented in this paper does not aim to discover causal structure from data alone, nor does it aim to provide a definitive proof of causation. Instead, its scope is to offer a transparent and principled way to reason about causal effect sizes using observational environmental data and prior knowledge, and to assess the compatibility of that prior knowledge with the observed data. By making prior knowledge and assumptions about cause-and-effect relationships explicit as graphical models, causal conclusions drawn from observational data can be scrutinised, criticised, and revised.

This paper reports the results of a case study on extracting causal knowledge about the contribution of different environmental

- *Section 3 #195-202:*

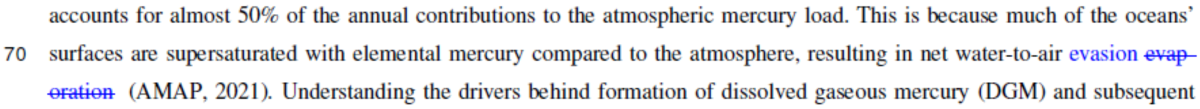


observational environmental data and prior scientific knowledge from researchers. Here, causal relationships are not discovered from the observational data itself, but are assumed based on prior experimental and scientific knowledge, such as laboratory studies demonstrating photochemical DGM production under controlled conditions. The scope of the proposed framework is then to quantify how multiple established or assumed causal processes jointly contribute to observed variability under natural, intervention-free field conditions. Using causal models, as suggested in this framework, conceptually allows individual causal pathways to be "switched off" within the model. This allows assessing the causal pathways' relative contribution without the need to physically intervene in the environmental system, which often is impossible in field observation.

III. Comments and thoughts

Line 62 (L62), “Hg…water-to-air evaporation”, evaporation refers to the escape of molecules of the liquid from liquid phase of that particular molecule to gas phase (e.g., pure water evaporation), but here, there is no liquid Hg involved, only dissolved gaseous Hg or Hg atoms as the solute in water (the solvent), the liquid is water. So rigorously, Hg evasion or emission, not evaporation, is more appropriate or accurate.

- *We thank the reviewer for noting this mistake and suggesting improvements. We have changed the word “evaporation” to “evasion” instead in the paper. See for example #70-71:*

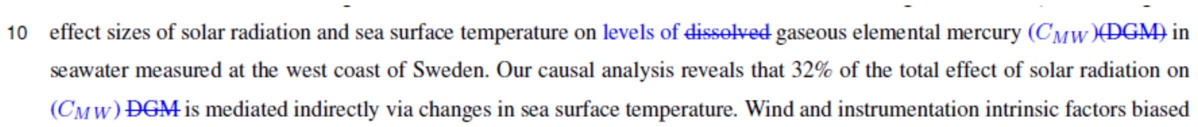


accounts for almost 50% of the annual contributions to the atmospheric mercury load. This is because much of the oceans' surfaces are supersaturated with elemental mercury compared to the atmosphere, resulting in net water-to-air evasion ~~evaporation~~ (AMAP, 2021). Understanding the drivers behind formation of dissolved gaseous mercury (DGM) and subsequent

By the way, as mentioned before, three issues are involved here: DGM generation, DGM emission or evasion, and DGM concentrations or levels. The title and the paper use “…causal inference applied to solar radiation and temperature effects on DGM. Then, exactly, which factor we are looking at? The DGM generation or emission, or concentration, which are the factors under consideration or treatment with the causal inference? This is unclear, another potential confusion point.

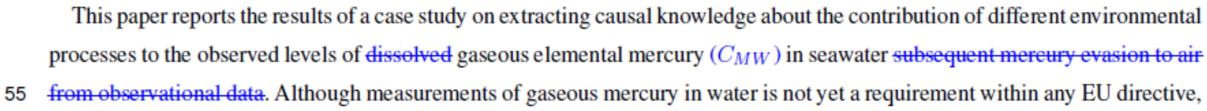
- *We are grateful that the reviewer pointed out this confusion point. We have taken the suggestion (stated in the beginning of this review) from the reviewer to change the title of the paper to make it clearer. The title is now “Technical note: A framework for casual inference applied to solar radiation and temperature effects on measured levels of gaseous elemental mercury in seawater.” We have also replaced “DGM” with gaseous elemental mercury (CMW), where appropriate in the text. Examples of changes in the paper are presented below:*

- *Abstract #10 and #12:*



effect sizes of solar radiation and sea surface temperature on levels of ~~dissolved~~ gaseous elemental mercury (C_{MW})~~(DGM)~~ in seawater measured at the west coast of Sweden. Our causal analysis reveals that 32% of the total effect of solar radiation on (C_{MW}) ~~DGM~~ is mediated indirectly via changes in sea surface temperature. Wind and instrumentation intrinsic factors biased

- *Introduction #54-55:*

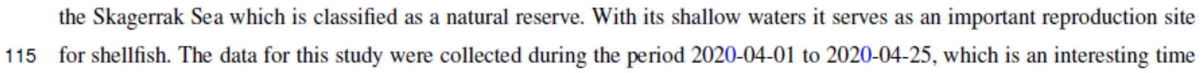


This paper reports the results of a case study on extracting causal knowledge about the contribution of different environmental processes to the observed levels of ~~dissolved~~ gaseous elemental mercury (C_{MW}) in seawater ~~subsequent mercury evasion to air from observational data~~. Although measurements of gaseous mercury in water is not yet a requirement within any EU directive,

L103-107, the campaign was 2019-2020, but the data used for this study was from 2024 April 1 to April 25. This is another potential confusion point. Which data were used? If the latter, why mentioning the 2019-2020 campaign?

- *We thank the reviewer for noticing this error that simply was a typing mistake. The real period for the measurement period is “2020-04-01 to 2020-04-25”. This has been changed accordingly in the paper, see below:*

- *Section 2 #115:*



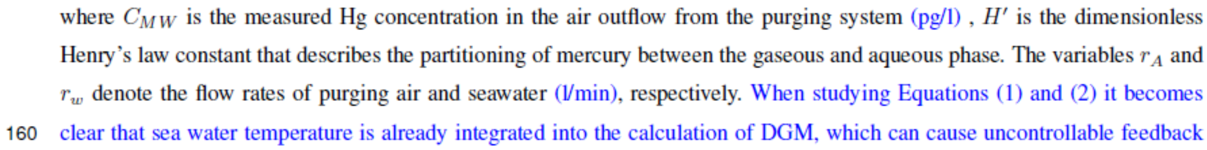
the Skagerrak Sea which is classified as a natural reserve. With its shallow waters it serves as an important reproduction site

115 for shellfish. The data for this study were collected during the period 2020-04-01 to 2020-04-25, which is an interesting time

L140-148, all parameters or quantities should be given together with their individual units, if any.

- *The reviewer is right, and we are grateful for pointing this out. We have added units for the factors presented in the equations, accordingly, see below:*

- *Section 2.1 #157 and #159:*



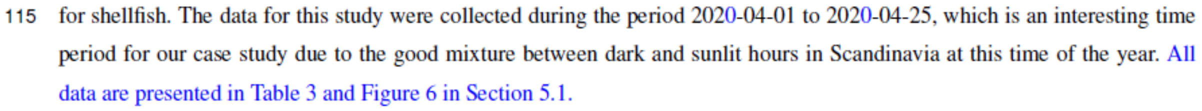
where C_{MW} is the measured Hg concentration in the air outflow from the purging system (pg/l) , H' is the dimensionless Henry's law constant that describes the partitioning of mercury between the gaseous and aqueous phase. The variables r_A and r_w denote the flow rates of purging air and seawater (l/min), respectively. When studying Equations (1) and (2) it becomes

160 clear that sea water temperature is already integrated into the calculation of DGM, which can cause uncontrollable feedback

Here, it may be helpful to mention the DGM, Solar, and T data are given or summarized in Table 3 and Fig. 6. At any rate, the data used for this study need to be presented clearly top front, rather than later. We need to know in the first place clearly what are the measurement data used for this study. This data can help readers to see or inspect, now, before the causal inference, the potential causal relationship, intuitively, or based on previous research experiences, independent of the causal inference.

- *We thank the reviewer for pointing this out. Although we decided not to present any measurement results in the method part, we now added information in section 2 about where the data is presented so it will be easier for the reader to find it, see examples below:*

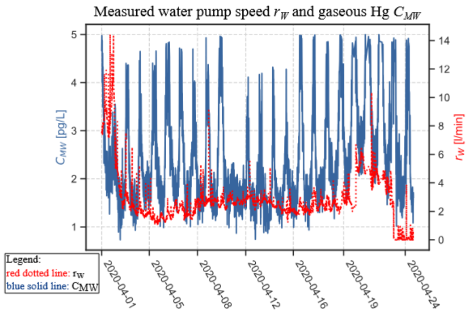
- *Section 2 #116-117:*



115 for shellfish. The data for this study were collected during the period 2020-04-01 to 2020-04-25, which is an interesting time period for our case study due to the good mixture between dark and sunlit hours in Scandinavia at this time of the year. All data are presented in Table 3 and Figure 6 in Section 5.1.

Fig. 6e has no legend, but it has two parameters, which is for which?

- *We thank the reviewer for this suggestion. We added a legend to Figure 6(e).*



(e)

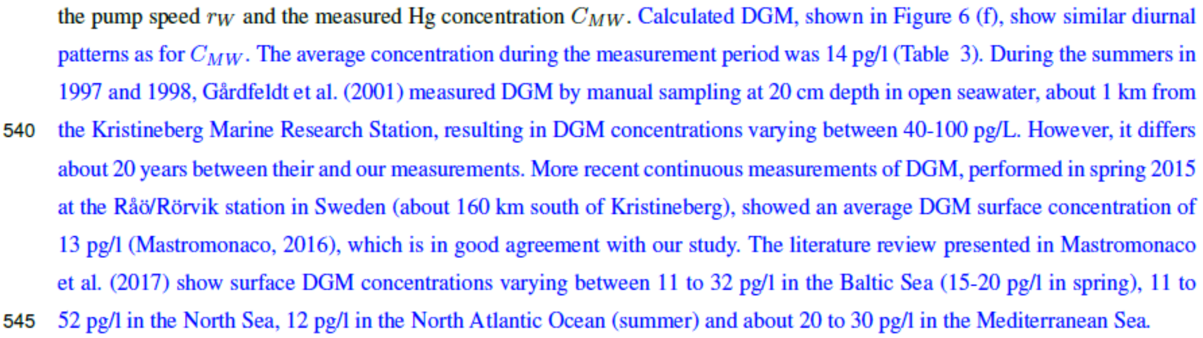
L141, from subsequent info, we know ra/rw < 1, this means for Eq. 1, DGM roughly = Ca/H, if so, why leave the item of ra/rw in the equation. This needs to be discussed. When the whole equation is needed, when the approximate, simplified one may be relevant in use. By the way, if the simplified equation is used, then the question regarding the meaning of the so calculated DGM arises, as discussed previously.

- *We agree with the reviewer that it's true that when rw is much bigger than ra, this term in the equation is small (in our case in average the factor would be 1.5/2.8 = 0.5). The reason why we feel it necessary to present the two factors in equation 1 is because we in our model discuss the influence of the water flow on measured CMW. The reviewer is again right with the point that the meaning of the calculated DGM is pointless for our study since we use CMW in the causal model. However, we think that many readers would find it interesting to compare calculated DGM concentrations in this study with other studies, since CMW is not a factor commonly reported.*

Table 3 and Fig. 6 show the DGM levels are quite low, as mentioned before. This is curious.

- *The reviewer is again pointing out a good point that no literature comparison was presented in the paper. Although the reviewer finds the DGM concentration rather low, we do believe that the calculated DGM concentrations are in good agreement with other studies. A comparison with literature has been added to section 5.1, see below.*

- *Section 5.1 #537-545:*

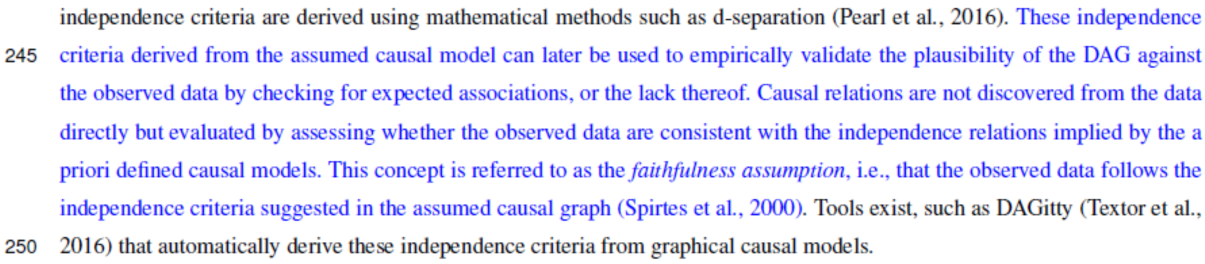


the pump speed r_W and the measured Hg concentration C_{MW}. Calculated DGM, shown in Figure 6 (f), show similar diurnal patterns as for C_{MW}. The average concentration during the measurement period was 14 pg/l (Table 3). During the summers in 1997 and 1998, Gårdfeldt et al. (2001) measured DGM by manual sampling at 20 cm depth in open seawater, about 1 km from the Kristineberg Marine Research Station, resulting in DGM concentrations varying between 40-100 pg/L. However, it differs about 20 years between their and our measurements. More recent continuous measurements of DGM, performed in spring 2015 at the Råö/Rörvik station in Sweden (about 160 km south of Kristineberg), showed an average DGM surface concentration of 13 pg/l (Mastromonaco, 2016), which is in good agreement with our study. The literature review presented in Mastromonaco et al. (2017) show surface DGM concentrations varying between 11 to 32 pg/l in the Baltic Sea (15-20 pg/l in spring), 11 to 52 pg/l in the North Sea, 12 pg/l in the North Atlantic Ocean (summer) and about 20 to 30 pg/l in the Mediterranean Sea.

L169, it is unclear which step in the framework will determine if the two or more factors are causally related, if the causal inference can determine that?

- *We thank the reviewer for this question. Causal inference itself cannot determine if two or more factors are causally related from observational data alone. It is necessary to define a priori causal assumptions in the form of causal models as suggested in our framework. However, it is possible to check if the a priori causal assumptions encoded in the causal model "fits" to the observed data. If two factors are causally not(!) related, they are also independent (except in very very very rare circumstances in which two opposing causal effects exactly cancel each other out). As the causal model provides (automatically) a set of independence criteria between variables, these criteria can be checked against* *the observed data. If they match, the model is said to be "faithful (see Spirtes et al., 2000) to the data. We clarify this concept now explicitly in Section 3:*

- *Section 3 #244-249:*

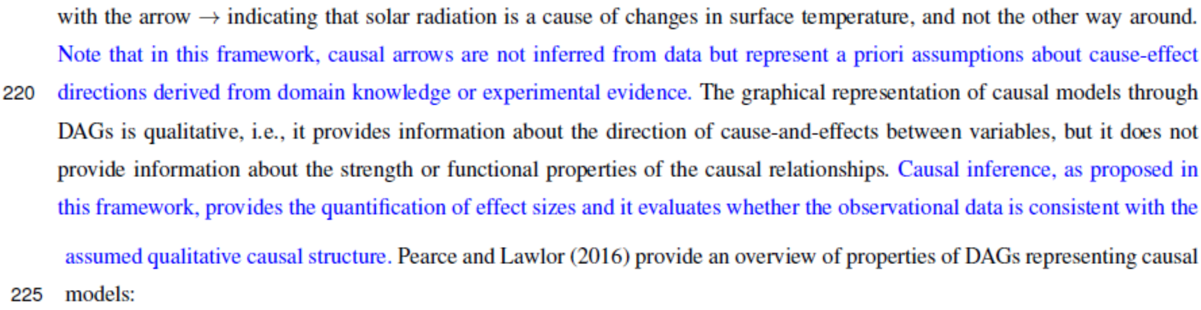


independence criteria are derived using mathematical methods such as d-separation (Pearl et al., 2016). These independence criteria derived from the assumed causal model can later be used to empirically validate the plausibility of the DAG against the observed data by checking for expected associations, or the lack thereof. Causal relations are not discovered from the data directly but evaluated by assessing whether the observed data are consistent with the independence relations implied by the a priori defined causal models. This concept is referred to as the *faithfulness assumption*, i.e., that the observed data follows the independence criteria suggested in the assumed causal graph (Spirtes et al., 2000). Tools exist, such as DAGitty (Textor et al., 2016) that automatically derive these independence criteria from graphical causal models.

L180-185, it appears that the causal arrow is what we assign or assume before the causal inference, rather than an outcome of the causal inference. This is, among others, what confuses me.

- *We thank the reviewer for this bringing up this point. The reviewer is right in that causal arrows in the proposed framework are specified a priori and are not an outcome of the causal inference itself because it is not possible to directly estimate the direction of cause-and-effect from data alone (a computer cannot distinguish associations from causations). In the proposed framework, the a priori causal models provide a qualitative specification of assumed cause-effect directions based on domain knowledge and experimental evidence. Causal inference, as suggested in our framework, then provides the quantitative effect sizes conditional on this assumed causal structure, and it evaluates, via the earlier discussed independence criteria, whether the observational data are consistent with the qualitative causal model. We have clarified this distinction now in Section 3 of the manuscript:*

- *Section 3 #219-220 and #222-224:*

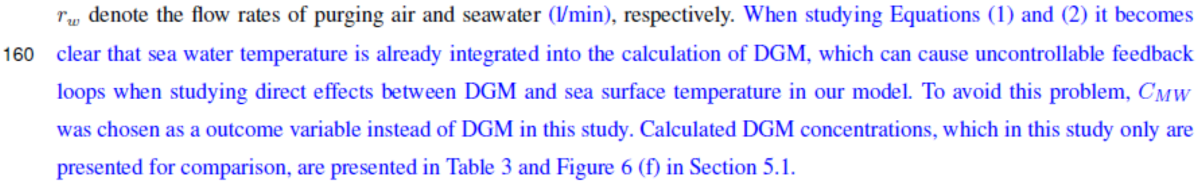


with the arrow → indicating that solar radiation is a cause of changes in surface temperature, and not the other way around. Note that in this framework, causal arrows are not inferred from data but represent a priori assumptions about cause-effect directions derived from domain knowledge or experimental evidence. The graphical representation of causal models through DAGs is qualitative, i.e., it provides information about the direction of cause-and-effects between variables, but it does not provide information about the strength or functional properties of the causal relationships. Causal inference, as proposed in this framework, provides the quantification of effect sizes and it evaluates whether the observational data is consistent with the assumed qualitative causal structure. Pearce and Lawlor (2016) provide an overview of properties of DAGs representing causal models:

From time to time, this becomes unclear: the casual inference is for solar and Ca or for solar and DGM?

- *The reviewer is right, and we thank the reviewer for pointing out this issue since this was causing confusion in our paper. We have now added information about our choice to use CMW instead of DGM in our model and have changed the text throughout the paper to be clear that we used and studied CMW, not DGM, in the model, see text below:*

- *Section 2.1 #159-163:*

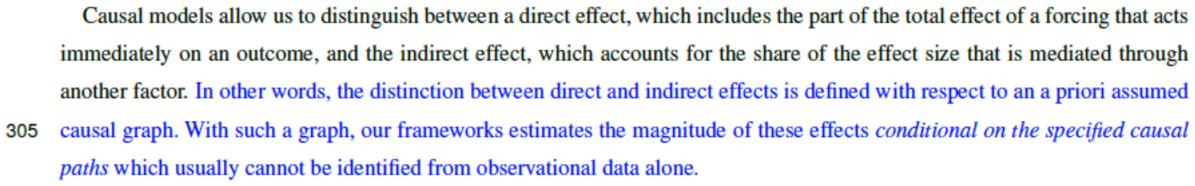


r_w denote the flow rates of purging air and seawater (l/min), respectively. When studying Equations (1) and (2) it becomes clear that sea water temperature is already integrated into the calculation of DGM, which can cause uncontrollable feedback loops when studying direct effects between DGM and sea surface temperature in our model. To avoid this problem, C_{MW} was chosen as a outcome variable instead of DGM in this study. Calculated DGM concentrations, which in this study only are presented for comparison, are presented in Table 3 and Figure 6 (f) in Section 5.1.

L250-251, regarding the nature of the effect, direct or indirect, again it seems that we need to pre-assign or assume it like the causal arrow, rather than an outcome of the causal inference.

- *We thank the reviewer for raising this point about the nature of direct and indirect effect and the role of causal models. We agree that the classification of effects as direct or indirect is not discovered automatically by the causal inference itself, but is rather defined a priori by the assumed causal model. In our proposed framework, the DAG specifies in a transparent way which causal paths are assumed to exist, and thereby also if an effect acts directly or indirectly on an outcome. Then, our with such an a priori assumed causal model, our frameworks allows to estimate the magnitude of the corresponding direct and indirect effects, conditional on the assumed causal model.*
  To clarify this potential ambiguity, we have added a clarification in Section 4.1:

- *Section 4.1 #304-306:*



Causal models allow us to distinguish between a direct effect, which includes the part of the total effect of a forcing that acts immediately on an outcome, and the indirect effect, which accounts for the share of the effect size that is mediated through another factor. In other words, the distinction between direct and indirect effects is defined with respect to an a priori assumed causal graph. With such a graph, our frameworks estimates the magnitude of these effects *conditional on the specified causal paths* which usually cannot be identified from observational data alone.

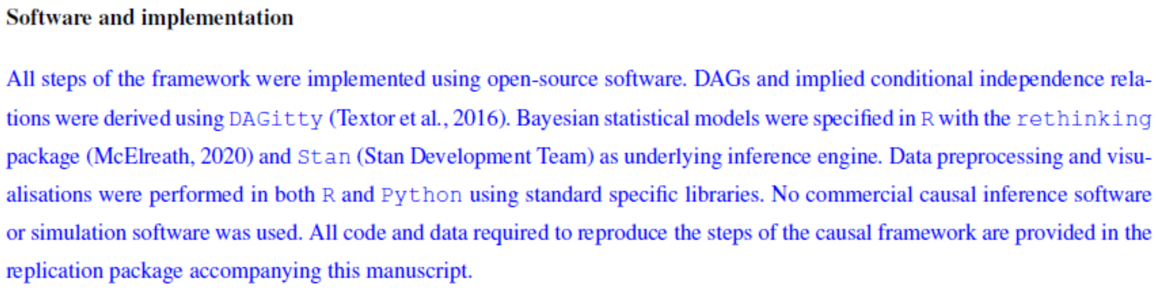
L306-308, how were the simulated data generated? From the data of Table 3 and Fig. 6, or from running the causal inference model? This is unclear. What software used to generate the simulated data?

A general comment, by the way, throughout this paper, it is always unclear if the causal inference was run or conducted by what software or causal inference model(s), any commercial software? If so, unless it is copyright or patent protected and thus cannot be disclosed, we need to know the brands or names of all the software and models used in this study, and which is used in which step to do what. This important info is missing and needs to disclosed in the early beginning as given by a list (like for experimental work, a list of chemicals and equipment used), like in a methodology section for the causal inference.

Furthermore, each time when a specific causal inference operation along the way going through the framework, we'd like to know what specific software or model(s) was used for this specific step or task or operation, with relevant references provided for more technical details.

- *We thank the reviewer for this important comment regarding the transparency about the used software and code. We have clarified in the manuscript that the simulated data were generated by forward-sampling from generative models that represent the assumed causal models. The simulations were used only to verify that the statistical models can recover known parameters and were not used as a substitute for inference from observational data later in the paper. We have clarified this in Section 4.4., including a statement listing the software packages used for creating the simulated data.*
  In addition, based on the reviewer's recommendation, we have added a dedicated paragraph at the beginning of Section 4 that lists the software and modelling tools used throughout the causal inference workflow. We emphasise also that the full software implementation, including simulation code, model specifications, diagnostics, and creation of the visualisations, are publicly accessible in the replication package which hopefully supports full reproducibility of our results.

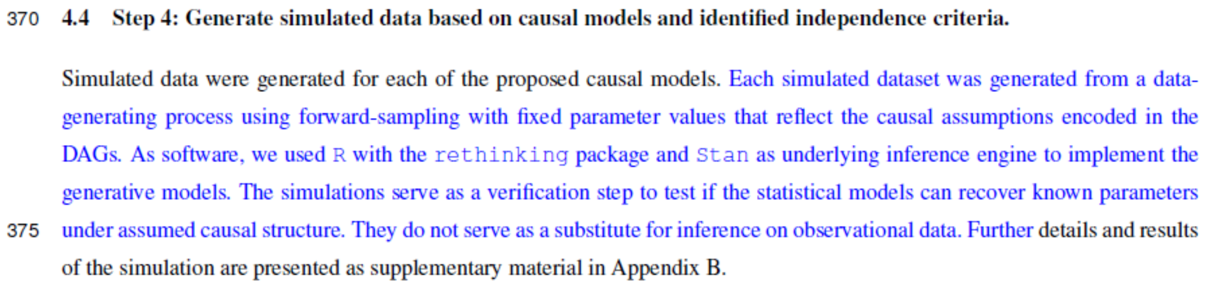
- *New paragraph in Section 4 #289-294:*



Software and implementation

All steps of the framework were implemented using open-source software. DAGs and implied conditional independence relations were derived using `DAGitty` (Textor et al., 2016). Bayesian statistical models were specified in `R` with the `rethinking` package (McElreath, 2020) and `Stan` (Stan Development Team) as underlying inference engine. Data preprocessing and visualisations were performed in both `R` and `Python` using standard specific libraries. No commercial causal inference software or simulation software was used. All code and data required to reproduce the steps of the causal framework are provided in the replication package accompanying this manuscript.

- *Section 4.4 #371-375:*



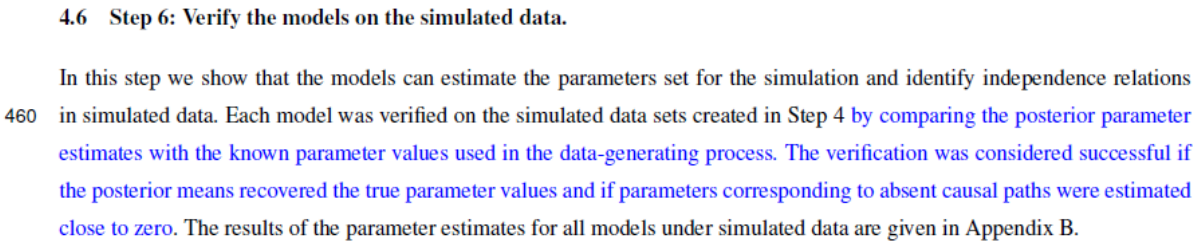
4.4 Step 4: Generate simulated data based on causal models and identified independence criteria.

Simulated data were generated for each of the proposed causal models. Each simulated dataset was generated from a data-generating process using forward-sampling with fixed parameter values that reflect the causal assumptions encoded in the DAGs. As software, we used `R` with the `rethinking` package and `Stan` as underlying inference engine to implement the generative models. The simulations serve as a verification step to test if the statistical models can recover known parameters under assumed causal structure. They do not serve as a substitute for inference on observational data. Further details and results of the simulation are presented as supplementary material in Appendix B.

L390, how to verify?

- *We thank the reviewer for this question. In Step 6, we added more details about the verification process, including when we considered verification to be successful.*

- *Section 4.6 #460-463:*



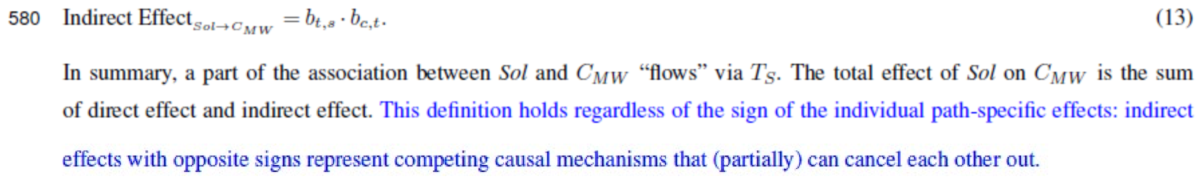
4.6 Step 6: Verify the models on the simulated data.

In this step we show that the models can estimate the parameters set for the simulation and identify independence relations in simulated data. Each model was verified on the simulated data sets created in Step 4 by comparing the posterior parameter estimates with the known parameter values used in the data-generating process. The verification was considered successful if the posterior means recovered the true parameter values and if parameters corresponding to absent causal paths were estimated close to zero. The results of the parameter estimates for all models under simulated data are given in Appendix B.

L498-499, total effect = direct effect + indirect effect, this is valid only for the cases where both effects are positive or negative, i.e., same direction. If one is positive and the other is negative, that total effect sum is not valid, or what is the meaning of that sum? For example, solar effect on T, two effects, one effect is that solar can enhance DGM generation, leading more DGM in water, while on the other hand, the other effect is that solar can increase water T, which in turn can lead to higher Henry's coefficient, and thus less DGM at the higher T, e.g., at Tw = 1 C, DGM at equilibrium = 7.2 pg/L, at 25 C, DGM = 3.8 pg/L. So, the two effects of solar radiation are opposite in direction. Then, how can these two opposite effects be additive in the causal inference? Or how the causal inference handles the opposite effects? Or the direction of the effect does not matter, since the cause inference tells if the effect is operative or not and in what extent?

- *We thank the reviewer for raising this important conceptual point. The total effect is the sum of direct and indirect effects. If some factors are negative and some positive, some of the effect would cancel each other out. The total effect would then be the sum that is left. This definition holds regardless of the sign of the individual causal paths. We have clarified this in Section 5.2.*
  We agree that calculated DGM is indeed negatively influenced by seawater temperature, as evident when studying Equation 1 and 2. However, we further clarify that the causal model in this study is specified at the level of measured mercury concentration C_{MW}, rather than an isolated subprocess such as equilibrium partitioning governed by Henry's law. Empirically, the inferred effect of seawater temperature on C_{MW} is positive in the observational data, indicating that temperature-related processes in this measurement context dominate the sub-mechanisms described by Henry's law. We have clarified this aswell in the manuscript under Section 5.2.

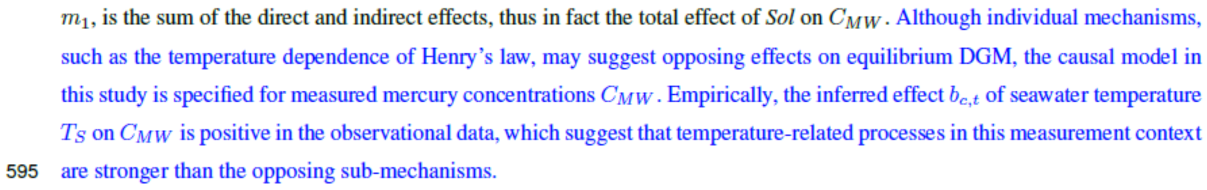
- *Section 5.2 #581-582:*



$$\text{Indirect Effect}_{Sol \rightarrow C_{MW}} = b_{t,s} \cdot b_{c,t}. \tag{13}$$

In summary, a part of the association between Sol and C_{MW} "flows" via T_S. The total effect of Sol on C_{MW} is the sum of direct effect and indirect effect. This definition holds regardless of the sign of the individual path-specific effects: indirect effects with opposite signs represent competing causal mechanisms that (partially) can cancel each other out.

#591-595:

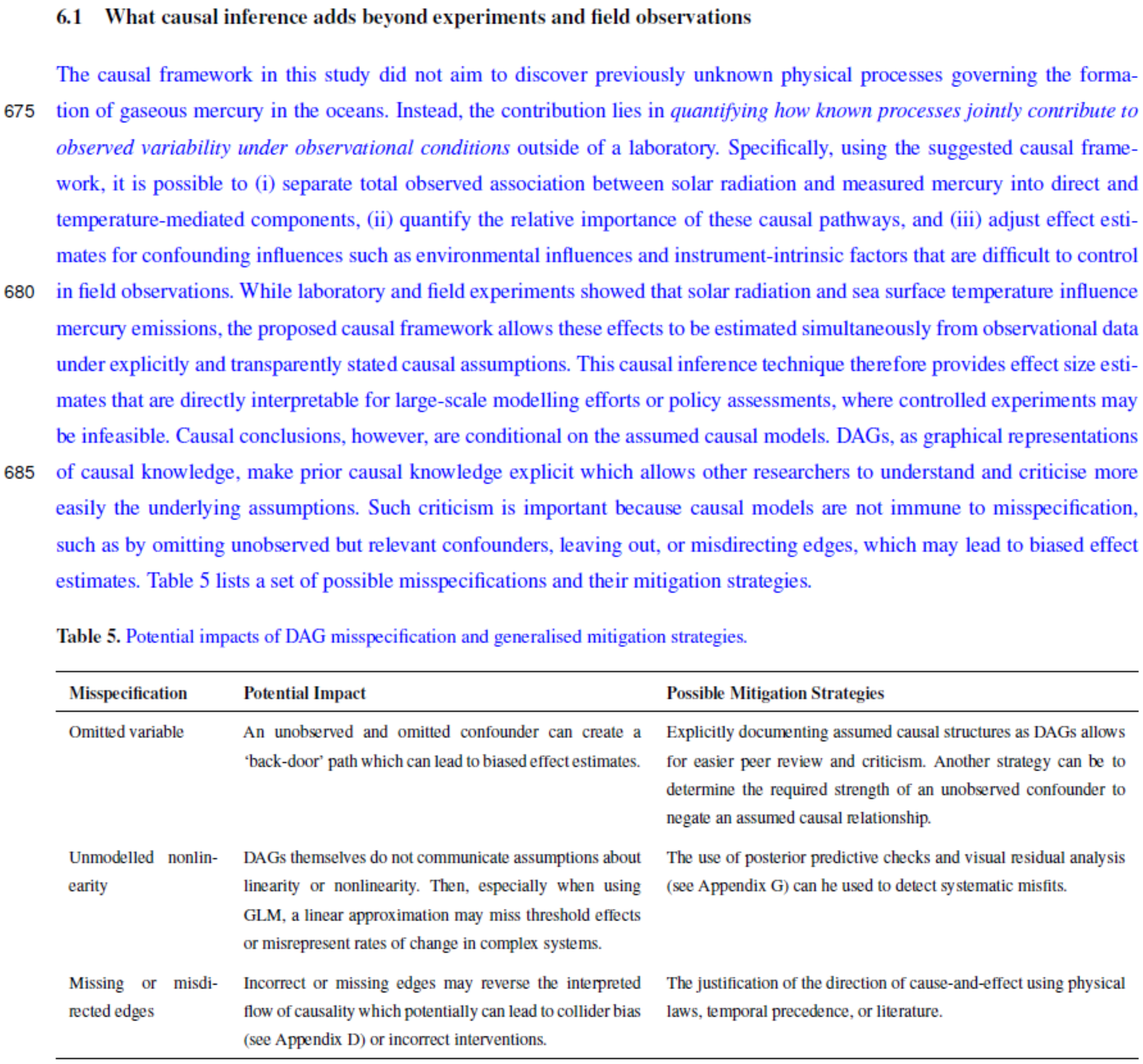


m_1, is the sum of the direct and indirect effects, thus in fact the total effect of Sol on C_{MW}. Although individual mechanisms, such as the temperature dependence of Henry's law, may suggest opposing effects on equilibrium DGM, the causal model in this study is specified for measured mercury concentrations C_{MW}. Empirically, the inferred effect $b_{c,t}$ of seawater temperature T_S on C_{MW} is positive in the observational data, which suggest that temperature-related processes in this measurement context are stronger than the opposing sub-mechanisms.

L582-583, What can the causal inference tell about the factors and their relationships that we still don't know, as from this particular study regarding DGM? In other words, what are new from the causal inference that has not been achieved by scientific experiments and field measurements?

- *We thank the reviewer for this important question. We have added a subsection in the Discussion outlining the novelty and contribution of causal inference within* *mercury emissions from oceans in particular and environmental research in general. We clarify in this subsection that the novelty of causal inference does not lie in identifying new physical mechanisms but in quantifying and decomposing the effect of already hypothesised, or in lab experiments discovered, physical drivers but using only observational and intervention-free data. The insight our proposed framework provide go beyond correlation analyses and complement therefore experimental studies by providing effect size estimates that are valid under explicitly, and transparently, stated causal assumptions. The new subsection also contain a discussion on the limitation*

- *New Section 6.1 #680-695:*



6.1 What causal inference adds beyond experiments and field observations

The causal framework in this study did not aim to discover previously unknown physical processes governing the formation of gaseous mercury in the oceans. Instead, the contribution lies in *quantifying how known processes jointly contribute to observed variability under observational conditions* outside of a laboratory. Specifically, using the suggested causal framework, it is possible to (i) separate total observed association between solar radiation and measured mercury into direct and temperature-mediated components, (ii) quantify the relative importance of these causal pathways, and (iii) adjust effect estimates for confounding influences such as environmental influences and instrument-intrinsic factors that are difficult to control in field observations. While laboratory and field experiments showed that solar radiation and sea surface temperature influence mercury emissions, the proposed causal framework allows these effects to be estimated simultaneously from observational data under explicitly and transparently stated causal assumptions. This causal inference technique therefore provides effect size estimates that are directly interpretable for large-scale modelling efforts or policy assessments, where controlled experiments may be infeasible. Causal conclusions, however, are conditional on the assumed causal models. DAGs, as graphical representations of causal knowledge, make prior causal knowledge explicit which allows other researchers to understand and criticise more easily the underlying assumptions. Such criticism is important because causal models are not immune to misspecification, such as by omitting unobserved but relevant confounders, leaving out, or misdirecting edges, which may lead to biased effect estimates. Table 5 lists a set of possible misspecifications and their mitigation strategies.

Table 5. Potential impacts of DAG misspecification and generalised mitigation strategies.

Misspecification	Potential Impact	Possible Mitigation Strategies
Omitted variable	An unobserved and omitted confounder can create a 'back-door' path which can lead to biased effect estimates.	Explicitly documenting assumed causal structures as DAGs allows for easier peer review and criticism. Another strategy can be to determine the required strength of an unobserved confounder to negate an assumed causal relationship.
Unmodelled nonlinearity	DAGs themselves do not communicate assumptions about linearity or nonlinearity. Then, especially when using GLM, a linear approximation may miss threshold effects or misrepresent rates of change in complex systems.	The use of posterior predictive checks and visual residual analysis (see Appendix G) can he used to detect systematic misfits.
Missing or misdirected edges	Incorrect or missing edges may reverse the interpreted flow of causality which potentially can lead to collider bias (see Appendix D) or incorrect interventions.	The justification of the direction of cause-and-effect using physical laws, temporal precedence, or literature.

L588-589, pump speed or water flow rate rw, L119 mentions that rw varied between 0 and 40 L/min. Then, first, if rw = 0, rA/rw is meaningless mathematically; if rw = 40, then rA/rw is 1/5/40 = 0.0375, very small, and so this item can be ignored, then DGMcal = Cmw/H. So this pump speed variation largely limits the accuracy of this auto-method. By the way, it remains hard to grasp or understand why DGM-real can be obtained by Cmw(1/H + rA/rw), how equilibrium gets there and why rA and rw got involved. The first item in Eq. 1 is about equilibrium and the second one is about the dynamics of the sampling flow, and then why DGM involves both equilibrium and dynamics?

- *We thank the reviewer for discussing this issue further. The second term in equation 1 is present due to the design of the system where the contact time between water and air is crucial to determine if the system is in steady state or not. This is important when calculating the efficiency of the system of how much mercury can be extracted from the system. The flow rates of air and water do affect the calculated DGM, as the reviewer also has noted with the above comment. Since equation 1 to calculate DGM only is used for demonstration in our study (and not used in the causal study), we advise the reviewer and the reader to further explore the derivation of the equation where it is originally explained in Andersson et al. 2008a.*

The pump speed involves measurement operational error or artifact, and so it is not a real physical effect for DGM like solar and/or Tw. Pump speed is not a direct effect, nor an indirect effect; it just has operational errors. One is about aquatic mechanisms and processes involving DGM generation kinetics and equilibrium and the other is about DGM measurement and measurement errors. Mixing the two in the causal inference is confusing.

- *We thank the reviewer for raising the importance of differentiating between real physical processes of mercury emissions and measurement-related artefacts. We agree that pump speed does not represent a physical process. To address this concern, we modified the terminology throughout the entire manuscript to consistently refer to pump speed as instrument-intrinsic factor. The causal analysis remains focused on disentangling the effect of environmental processes, such as solar radiation and sea surface temperature. However, the statistical models recognise the disturbing influence a varying pump speed.*

32% effect for solar radiation is due to indirect effect of water temperature. But, as mentioned before, the effects of solar and T on DGM are opposite. This result of 32% effect size seems to show that T has a positive effect just like solar radiation, higher solar higher DGM, but higher T, lower DGM based on equilibrium.

By the way, in many cases as shown by many field studies, the water T varied quite less during a day (as compared to solar radiation), only to a small extend as a result of very high specific heat of pure water (due to the Hydrogen bonding of the highly polar water molecules). But, 32% is almost 1/3, which means the effect of T is almost very strong.

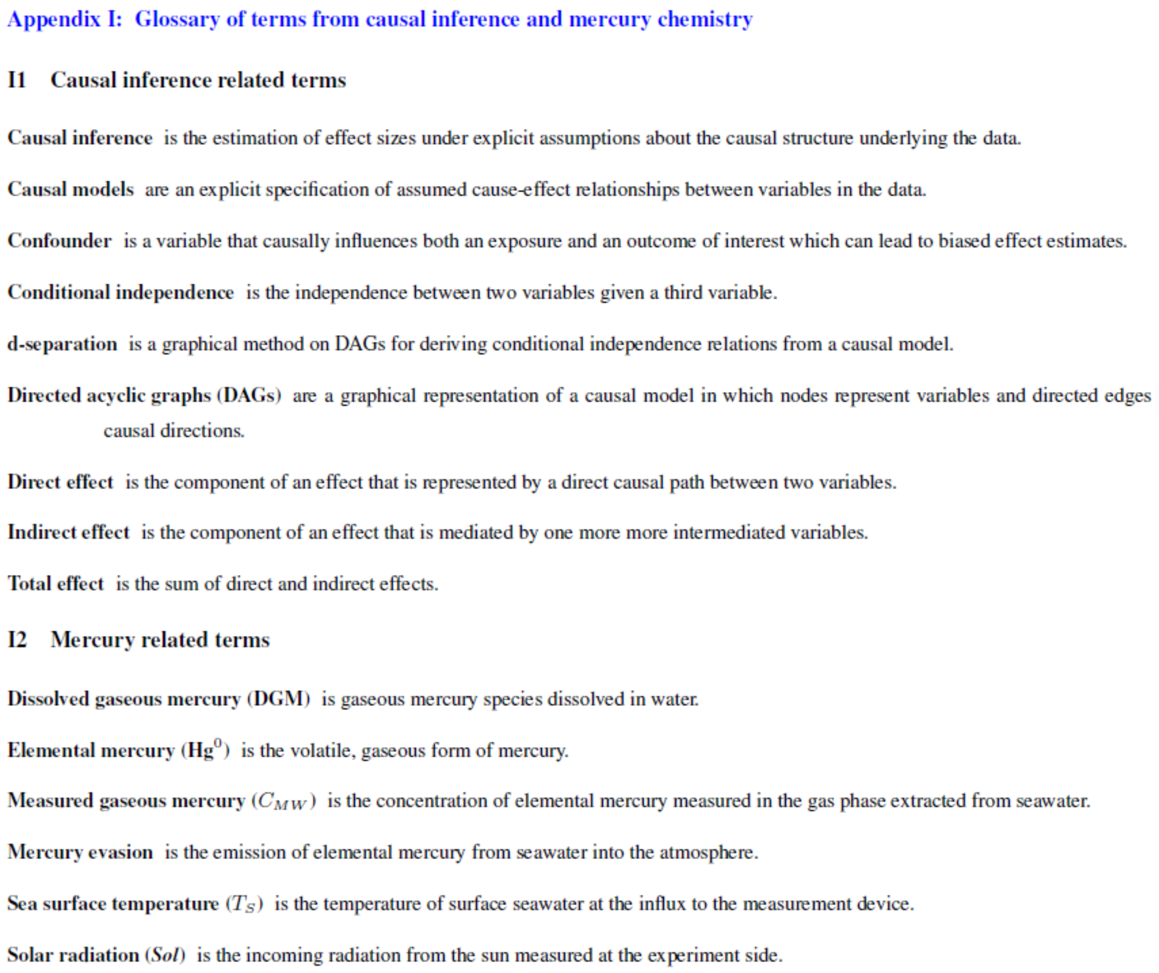
On the other hand, T can not only change Henry's constant and the Hg air/water distribution equilibrium (constant), but also can change the kinetic rate constants (and rates) of photochemical and/or thermal reduction of Hg(II) to Hg(0). This is another effect of water T. Then this effect is positive, enhancing DGM generation, like solar radiation. Thus, T has two opposite effects: positive to enhance the kinetics, and negative to increase H, then decrease DGM at equilibrium.

- *It is a very interesting point raised by the reviewer and we thank the reviewer for this nice discussion point. We agree that calculated DGM is negatively correlated* *with temperature, via the calculation of Henry's law coefficient, see equations 1 and 2. However, in our study, we removed this issue when choosing to instead study the measured Hg concentration (CMW). Our findings in this study were that our measured CMW showed a positive correlation to measured seawater temperature where an increase of 1K would lead to an increase in CMW of 0.156 pg/l (see Table 4). Our study also showed that the fraction of this temperature effect, that was associated with the indirect effect of solar radiation affecting the temperature, was 32%, which can be observed when looking at the standardised values of model m4 in Table 4: 0.186 (indirect effect of Sol mediated by sea surface temperature) versus 0.429 (the total effect of Ts) = 32%. This interestingly shows that the temperature effect on CMW can be explained by the indirect effect on solar radiation to only 32%. Other effects of temperature on CMW could be, as the reviewer mentioned, for example changing kinetic rates of abiotic, biotic and thermal reduction processes.*

Last but not the least, it would be helpful to provide a short glossary of the terms as an appendix, especially those involving causal inference.

- *We thank the reviewer for this suggestion. We have added a short glossary in appendix.*

  New Appendix I #976-994:



Appendix I: Glossary of terms from causal inference and mercury chemistry

I1 Causal inference related terms

Causal inference is the estimation of effect sizes under explicit assumptions about the causal structure underlying the data.

Causal models are an explicit specification of assumed cause-effect relationships between variables in the data.

Confounder is a variable that causally influences both an exposure and an outcome of interest which can lead to biased effect estimates.

Conditional independence is the independence between two variables given a third variable.

d-separation is a graphical method on DAGs for deriving conditional independence relations from a causal model.

Directed acyclic graphs (DAGs) are a graphical representation of a causal model in which nodes represent variables and directed edges causal directions.

Direct effect is the component of an effect that is represented by a direct causal path between two variables.

Indirect effect is the component of an effect that is mediated by one more more intermediated variables.

Total effect is the sum of direct and indirect effects.

I2 Mercury related terms

Dissolved gaseous mercury (DGM) is gaseous mercury species dissolved in water.

Elemental mercury (Hg^0) is the volatile, gaseous form of mercury.

Measured gaseous mercury (C_{MW}) is the concentration of elemental mercury measured in the gas phase extracted from seawater.

Mercury evasion is the emission of elemental mercury from seawater into the atmosphere.

Sea surface temperature (T_S) is the temperature of surface seawater at the influx to the measurement device.

Solar radiation (*Sol*) is the incoming radiation from the sun measured at the experiment side.