

Anonymous Referee #1 comment (1):

General comments:

The paper contributes some new understanding on possible impact of future climate change on the distribution and fate of POPs in the North Sea. The modeling methodology is adequate and the research questions addressed are relevant and interesting. The overall quality of the discussion of the results is poor since it does not relate to other publications in this field and does not discuss the significance of the climate change impact on contaminant fate. The explanations of the (however fairly small) observed differences between different climate conditions are generally clear and easy to follow. What is mainly lacking are scientifically interesting conclusions drawn from the results. The results are not put in a wider context. Results are mainly reported and explained, not really discussed.

Answer (1):

We are very grateful to the reviewer for all comments concerning our paper, which we consider to be thoughtful and helpful. We will incorporate their critiques and suggestions into the resubmitted version of the paper. We think this will improve the manuscript substantially.

We will improve the quality of the discussion of the results by relating to other publications in the field, while discussing the significance of the impact of climate change on contaminant fate.

We will put the results in a wider context while drawing scientifically interesting conclusions from the results.

Anonymous Referee #1 comment (2):

Specific comments:

Title

The title is a bit simplistic, should maybe extend to something like "The effects of climate change on levels/concentrations/distribution/fate of POPs in the North Sea"

Answer (2):

We will extend the title based on the revision and the suggestions of both anonymous referees.

Anonymous Referee #1 comment (3):

Abstract

The abstract is not complete. Only the ocean circulation model (HAMSOM) is mentioned by name, but not the POP fate and transport model (FANTOM). The language is a bit unclear, for example the use of "in situ" concentrations instead of "initial concentrations" (Rows 6-7 p 1526). Row 9 p 1526 "... our approach was to reutilize 2005 values", state where these concentrations come from also in the abstract (i.e. measured or modelled?). Try to avoid diffuse statements like "Dry gas deposition and volatilisation of γ -HCH increase in the future relative to the present" (Rows 16-17 p 1526), give a number on how much, for example average % of present or similar, and also mention what aspect of climate change that caused this effect. "PCB 153 in sediment decreases exponentially in all three runs, but even faster in the future, both of which are the result of climate change" (Rows 19-20 p 1526) unclear what is meant by "even faster in the future". Was the conclusion really that this decrease was mainly due to climate change? Degradation in sediment is mentioned in the results section as an important process, and this is not clearly dependent on climate change and would occur even if the climate was not changing. It is not clear from Fig 4-6 that the decrease in mass in sediments of PCB153 is faster in the 2090-2099 scenario compared to the

other scenarios as stated in the abstract.

Answer (3):

Perhaps we were a little sloppy here.

We will complete the abstract.

We will mention the FANTOM by name.

We will clear the language up.

We will avoid diffuse statements and give precise numbers, and we will mention which aspect of climate change caused the increase of deposition.

We will clear up the issue of PCB153 in sediment degrading "even faster in the future".

Yes, the conclusion was that degradation increases in the future due to climate change.

We will clear up Fig 4-6 and the issue of decreasing mass of PCB153 in sediment in the future.

Anonymous Referee #1 comment (4):

Introduction

The introduction lacks several crucial components. First, it is not adequately motivated why this study is necessary. The first paragraph is not logical. The fact that (many, but not all!) POPs bioaccumulate in many organisms is not a good motivation to why the impact of climate change on POP levels is necessary to assess. Instead, the introduction should discuss what processes that determine the fate of organic chemicals that may be influenced by future climate change and to what extent.

Answer (4):

In the introduction to the resubmitted manuscript, we will motivate why the study is necessary by discussing processes determining the fate of POPs and the extent to which they may be influenced by future climate change.

Anonymous Referee #1 comment (5):

Second, a number of previous modeling studies of POPs in the North Sea are listed, which is good, but is not stated what were the conclusions or key-results of these studies that can be related to the current study. For example, what knowledge is still lacking and how will this study fill these knowledge gaps? Do the previous studies address climate change for example? Some key publications are also not referred to in the introduction. Although not focusing specifically on coastal and shelf areas and not using high resolution models, there are publications addressing POPs and climate change which should be mentioned here. For example, the review by Gouin et al 2013 (Env TOx Chem 32:20) (this publication state that the direct impact of climate change on POP levels in the environment are in general fairly small, merely a factor 2 in most cases) and the modelling study by Lamon et al 2009 (ES&T 43:5818) addressing PCBs in future climates could be mentioned.

Answer (5):

We will state the conclusions and key results of studies listed in the introduction and how they can be related to the present study, by stating, for example, what knowledge is still lacking and how the present study helps fill the knowledge gap.

We will also mention other publications addressing POPs and climate change and describe their conclusions. We thank the reviewer for the suggested list of papers.

Anonymous Referee #1 comment (6):

The methodology is adequate, although using the same forcing and initial concentrations in all scenarios is not realistic. It is understandable and makes it possible to look at climate change impact only, but the actual years chosen become meaningless.

Answer (6):

Yes, we agree with the reviewer that the actual years chosen are meaningless. Since, to date, we have no idea about input changes of POPS through the atmosphere or rivers in the 21st century (North Atlantic input into the North Sea is negligible), we cannot perform a projection for the POPs as we do for the climate itself. So we just use the 10-year slices from the A1B climate scenario run.

Anonymous Referee #1 comment (7):

Model description

The authors refer to their previous publication with a more detailed model description. However, they repeat the description of air-sea gas exchange. Units are not given, and it is not stated what "D" is. It is also not logical to repeat the description only for this process, since other processes turn out to be important for POP fate later in the manuscript, for example sinking of POC, resuspension, degradation in sediment etc.

Answer (7):

We repeated the details of air-sea gas exchange because most of the changes occurred at this interface due to changing atmospheric forcing (air-temp, wind, precipitation) and water temperature. We will include a more detailed description of the processes in the revised manuscript.

Anonymous Referee #1 comment (8):

It is not completely clear, but I assume that constant concentrations were used in air and rivers throughout all simulations?

Answer (8):

Yes. This is how we argue that changes in POPs are due to climate effects only. We will clarify this.

Anonymous Referee #1 comment (9):

The statement that only one climate scenario is used (and not several) because the authors are interested in climate change impact only is not logical. This can be done, and would perhaps be done better, by using several climate scenarios and run simulations for a longer time period. "Again, we stress that we run only one climate scenario, because we are interested in the impact of the climate on the fate of the POPs in the North Sea. For investigating the future development of the POPs, different projections and even ensemble runs would be necessary" (Rows 12-15 p 1531).

Answer (9):

Yes, the statement is incorrect and will be removed. We chose the A1B scenario because it is a moderate choice. Since a more extreme scenario introduces more variance, the conclusions become more biased. Additionally, there is a huge amount of work involved in setting up another scenario, which we did not have the manpower or time to do.

Anonymous Referee #1 comment (10):

Results

The results are clearly discussed and the mechanisms behind observed changes are explained in a good way. However, in general the significance of these differences is not discussed, and the results are not related to findings by other researchers. This should be improved.

Answer (10):

We will discuss the significance of these results, while relating them to the findings of other researchers.

Anonymous Referee #1 comment (11):

Some specific comments:

Results for hydrodynamics are in general relevant, but the salinity reduction is unnecessary to mention unless the salinity is related to e.g. POP solubility in water in the model?

Answer (11):

Water density is calculated using temperature and salinity. So it is important for hydrodynamic processes such as advection and mixing.

We will remove mention of salinity reduction.

Anonymous Referee #1 comment (12):

Avoid using words like “quite” as in “which are quite important for our POP dynamics” (Row 12 p 1532).

Answer (12):

We will consider this in the revised manuscript.

Anonymous Referee #1 comment (13):

The scale in Fig 3 c and d is difficult to read.

Answer (13):

We will change this.

Anonymous Referee #1 comment (14):

Row 25-28 p 1532: It is not explained why there are seasonal differences in dry gas deposition. Is this the result of emission seasonality or other mechanisms, e.g. temperature dependence of atmospheric concentrations? Authors should mention why dry gas deposition is increased (i.e. changes in temperature and wind speed, and hence the mass transfer coefficients?) and that it is the increased mass in the water column that leads to increased volatilization for HCH.

Answer (14):

Yes, atmospheric concentrations are based on monthly concentrations for 2005. Deposition is then a function of atmospheric concentrations, but also of the different atmospheric and ocean conditions, e.g., air and water temperature and wind speed.

We will clarify this in the revised manuscript.

Anonymous Referee #1 comment (15):

How can the decrease in total mass in water in the first year of all runs be consistent with the 1996-2005 trend, which is due to decreasing atmospheric concentrations and river input, if these concentrations are held constant in the future simulations? Or are these concentrations not constant? Maybe clarify this.

Answer (15):

Since river and atmospheric concentrations were reduced up to 2005, the result is that concentrations in the water column decrease up to the end of 2005. Although 2005 values are used repeatedly in the three 10-year time slices (2006-2015, 2046-2055, 2090-2099), values are less in 2005 because the response is not instantaneous. There is a recovery time for concentrations to become steady. Since 2015, 2055 and 2099 are at the end of 10 years of steady input, the concentrations have become steady, to 1st order, at this time. We will clarify this point in the manuscript.

Anonymous Referee #1 comment (16):

Row 29 p 1534 "These events are also found in the sediment and γ -HCH records." Please clarify, are the authors referring to measured data? Please give a reference in that case.

Answer (16):

No. These are seen in the model results. We will clarify this.

Anonymous Referee #1 comment (17):

Row 11 p 1535: "During storm events, erosion causes mobilisation and resuspension of PCB 153 into the water column, increasing its concentration there, thus resulting in a consequent increase in volatilisation." At what depths can storms resuspend bottom sediments? How big fraction of the North Sea bottoms will be resuspended in this way? A brief discussion of this issue would be helpful here.

Answer (17):

This mostly occurs in shallow water, close to coasts. Stronger storms will mean that these strong currents will get to deeper waters. We will clarify this.

Anonymous Referee #1 comment (18):

Row 16 p 1535: "This is in contrast to γ -HCH, which is generally net depositional from atmosphere to ocean, has total mass in water more than two orders of magnitude greater than that in sediment". Looking at Fig 4-6, this is only true for the 2006-2015 scenario, and only the last 10 years or so.

Answer (18):

Third panels in Fig.4-6 show that PCB 153 is generally net volatilizational. Fourth panels Fig.4-6 show that total mass of PCB 153 is two orders of magnitude greater than that in water. Note the different scales on left and right hand side. We will make this note in the revised manuscript.

Anonymous Referee #1 comment (19):

Section 3.4: In general, very small changes in annual mass fluxes are observed due to the climate change. But this is not commented!

Answer (19):

We will state this in the revised manuscript.

Anonymous Referee #1 comment (20):

Section 3.5: Row 3 p 1538: "First, concentration in gas in the atmosphere is greatest along the British coast", re-word this sentence. Again, it is not clear what scenario was used for the atmospheric and riverine concentrations. Are these concentrations decreasing during the simulations?

Answer (20):

*No, concentrations are fixed, with an annual cycle (2005 values).
We will clarify this in the revised manuscript.*

Anonymous Referee #1 comment (21):

Figure 9-11: Using the absolute difference makes it difficult to judge the significance of the differences. An increase of at maximum 0.05 ng/L seems negligible. It would be better to use the ratio between 2005 and the other years.

Answer (21):

We will do this in the revised manuscript.

Anonymous Referee #1 comment (22):

Last sentence in this paragraph (Row 17 p 1532): "It is the effect of these changes on the studied POPs that are analysed." It is not clear which changes that the authors refer to.

Answer (22):

We refer to the changes in physical forcing and variables described in the previous paragraph of the manuscript: 'The effect of the change in wind conditions during the 21st century on bottom erosion and deposition of suspended particulate matter (SPM), which are quite important for our POP dynamics, is shown in Fig. 3c and d, respectively. Plots show trends of relative frequency of erosion and deposition periods in percentage points per 100 yr, based on annual mean percentages of erosion and deposition for the 21st century. A slight increase/decrease in annual periods of erosion/deposition are found.'

We will clarify this in the revised manuscript.