# **Reply to Anonymous Referee #1**

## General comments:

This article is an interesting study demonstrating the importance of including short-term (hourly) variability in atmospheric CO2 concentrations in calculations of air-sea CO2 flux for the Baltic Sea and Danish inner waters. Currently, large-scale air-sea CO2 exchange estimates for coastal margins and seas are poorly constrained due to large spatial and temporal variability in these environments and a lack of measurements. Past studies presenting estimates of air-sea CO2 flux both in this study region and in coastal regions generally use low temporal resolution atmospheric CO2 (i.e., averaged weekly, monthly, or yearly) in addition to low temporal resolution surface ocean pCO2.

Recent studies have shown that atmospheric CO2 concentrations can vary significantly over marine areas that border populated regions and/or regions with large fluxes between the atmosphere and terrestrial biosphere. This study uses a model with nests over Europe and northern Europe capable of reproducing variability in the atmospheric CO2 concentrations on a 16.7 x 16.7 km grid over the study region at hourly intervals. The authors acknowledge that even at this resolution the model underestimates the diurnal variability in atmospheric CO2 during the Northern Hemisphere summer. The study also provides an estimate for the contribution of air-sea CO2 exchange for the territorial waters of Denmark to its national carbon budget.

The study region is found to be an annual sink for CO2 with strong summer uptake partially offset by winter outgassing. The magnitude of the sink from a simulation including short-term atmospheric CO2 variability is significantly smaller than the magnitude of the sink in a simulation where constant (i.e., monthly averaged) atmospheric CO2 values are used. The authors indicate that this bias is due in part to diurnal boundary layer dynamics where constant atmospheric CO2 values are too high relative to observations during the day when wind speeds are typically strongest. The authors emphasize that since diurnal variability in the model is smaller than observed, the magnitude of this bias may be larger than indicated in the current study. The choice of gas exchange parameterization has an even larger impact on the magnitude of the regional CO2 sink – the parameterization of Weiss et al. (2007) yields a CO2 sink more than three times greater than a simulation using the Wanninkhof (1992) parameterization. This study represents a contribution to the growing literature on air-sea CO2 exchange in coastal regions and inland seas.

Reply: We thank the reviewer for pointing out that this is an interesting study. We also thank the reviewer for constructive review. In the following we answer the reviewer point by point and describe the related changes we have made to the manuscript.

p. 4, line 3: What about the impact of short-term variability in atmospheric CO2 concentrations on large-scale estimates of air-sea CO2 flux? Are there studies that demonstrate the potential bias of omitting short-term atmospheric CO2 variability on estimates of air-sea CO2 flux for the coastal seas as a whole?

Reply: To our knowledge no such studies have previously been conducted.

p. 10, lines 24-26: Wanninkhof (1992) is now out of date (use Sweeney et al. 2007 or another more recent parameterization) – the Sweeney et al. (2007) parameterization was used by Wanninkhof himself in a recent study of global air-sea CO2 flux (Wanninkhof et al., 2013). See discussion of the gas transfer coefficient (k) in section 3.1 p. 1988-1989 of this paper. The Nightingale et al. (2000) parameterization yields k values for wind speeds from 5 to 10 m s-1 that are close to Sweeney et al. (2007) and could also be appropriate since it is based on experiments conducted close to the study region. The Weiss et al. (2007) parameterization yields k values for open ocean studies; this should be mentioned in the text.

Reply: The choice of using the parameterisation by Wanninkhof 1992 was made, as to be able to compare with results from previous studies within the study area, as many of these have also used this parameterisation (*Löffler et al., 2012; Rutgersson et al., 2008; Wesslander et al., 2010*). Thus, the uncertainty bound to the choice of transfer velocity parameterisation is for this comparison eliminated. However, we have then tested the sensitivity of the choice of parameterisation by executing simulations, where Nightingale et al. 2000 and Weiss et al. 2007 have been used. N00 and Weiss07 were chosen amongst the many existing parameterisations, as these experiments have been made close to and within the study region. With this said, other parameterisations could also have been applied such as Sweeney et al. 2007, especially since the study by Sweeney et al. 2007 is based on an updated and improved version ocean bomb DI14C inventory. We will in any future studies also include this parameterisation.

A few changes will be made in the text:

#### Change on page 17005 line 9 from:

In the present study we use the parameterisation of Wanninkhof (1992) (hereafter referred to as W92), which is the parameterisation most frequently applied. This transfer velocity is a function of the wind speed at 10 m above the surface  $(u_{10})$  and when normalised to Sc at 20°C in salt water it has the form

То

In the present study we use the parameterisation of Wanninkhof (1992) (hereafter referred to as W92). This parameterisation has been used in many previous studies within the study area (Löffler et al., 2012; Rutgersson et al., 2008; Wesslander et al., 2010), and by using W92 this allows for a direct comparison of the estimated fluxes. W92 is a function of the wind speed at 10 m above the surface ( $u_{10}$ ) and when normalised to Sc at 20°C in salt water it has the form

#### Text added to page 17005 line 21:

The parameterisation by Weiss et al. (2007) often yields greater values than other transfer velocity parameterisations; however, it will be applied here, as the experiment was conducted within the study area.

### Change on page 17019 line 6-12 from:

The largest uncertainty connected to the estimated air–sea flux is related to the choice of transfer velocity. The results from the VAT simulation presented here were calculated using the parameterisation by Wanninkhof (1992), but model simulations using parameterisation of Nightingale et al. (2000) and Weiss et al. (2007) have also been conducted. With these parameterisations the annual flux for the study area is changed to –667 and –858 GgCyr–1, respectively

То

As to assess the uncertainty connected to the choice of transfer velocity on the estimated air-sea flux model, simulations using parameterisations of Nightingale et al. (2000) and Weiss et al. (2007) have also been conducted. Throughout the seasons the parameterisation by Weiss et al. (2007) gives more extreme values than Nightingale et al. (2000), but the annual sum for the study area results in -667 and -858 Gg C yr<sup>-1</sup> for Nightingale et al. (2000) and Weiss et al. (2007), respectively. Other transfer velocity parameterisations could also have been interesting to use in the presents study. An example is the parameterisation by Sweeney et al. (2007), which is based on an updated and improved version of the radiocarbon method used in W92. Here, the two different parameterisations by Weiss et al. (2007) and Nightingale et al. (2000) were chosen, as these experiments were conducted within and close to the study area, respectively.

# p. 15, line 23: Could indicate the interannual variability in the air-sea CO2 flux for the study region here and mention whether this is believed to represent method uncertainty or the actual interannual variability.

Reply: Due to the limitation of pCO2 data for the study area, we have made and used a climatology for the pCO2. Thus, the interannual variability arising will only be a result of the variation in the atmospheric CO2. Therefore, it is not reasonable to examine the inter-annual variability in the air-sea flux. When more pCO2 data exists for the study area, the inter-annual variability could be investigated.

# Text added to page 17011 line 10

Here, the annual average of 2616 Gg C yr<sup>-1</sup> is reported. The inter-annual variability of the estimated flux will solely be a result of the inter-annual variations in the atmospheric  $CO_2$ , as a climatology is used for the surface water pCO2, due to the limited amount of data.

p. 20, lines 9-10: Isn't there a reduction in winter outgassing in the CAT simulation relative to the VAT simulation (i.e., seen from comparison of Fig. 3b top left panel and Fig. 8)?

Reply: yes, the outgassing is reduced in the CAT simulation relative to the VAT simulation. And yes, there has been made a mistake in text here.

The text on p. 20 line 9-10 will be change from

The deviation between the two simulations in the study region is mainly caused by a reduction in the winter uptake in the CAT simulation To

The deviation between the two simulations in the study region is mainly caused by the reduced winter outgassing in the CAT simulation relative to the VAT simulation.

p. 22, line 16: Since the Nightingale et al. (2000) gas exchange parameterization typically yields smaller k values (except at wind speeds lower than 4 m s-1) in comparison to Wanninkhof (1992) I would have thought that the magnitude of the uptake using N00 would be smaller than the simulation using W92. I also would have thought that the magnitude of uptake calculated using the Weiss et al. (2007) parameterization would have been nearly twice that calculated with N00.

Reply: The seasonality in the surface water pCO2 allows for uptake of atmospheric CO2 during summer and release of CO2 to the atmosphere during winter. Using N00 gives smaller release of CO2 during winter than W92, and a smaller uptake during summer. However, on an annual net basis the total flux sums up to give a greater uptake when using N00 than W92. The same is the case for N00 vs. Weiss, where using Weiss results in greater uptake during summer than when using N00, and greater release of CO2 during winter. On a monthly basis the deviation between the two parameterisations is on average 35%, but because of the sign difference in the flux during the course of the year, the difference of net annual flux between Weiss and N00 is only 22%.

To make this more clear in the text we have changed the text on page 17019 line 6-12 from:

The largest uncertainty connected to the estimated air–sea flux is related to the choice of transfer velocity. The results from the VAT simulation presented here were calculated using the parameterisation by Wanninkhof (1992), but model simulations using parameterisation of Nightingale et al. (2000) and Weiss et al. (2007) have also been conducted. With these parameterisations the annual flux for the study area is changed to –667 and –858 GgCyr–1, respectively

То

As to assess the uncertainty connected to the choice of transfer velocity on the estimated air-sea flux model, simulations using parameterisations of Nightingale et al. (2000) and Weiss et al. (2007) have also been conducted. Throughout the seasons the parameterisation by Weiss et al. (2007) gives more extreme values than Nightingale et al. (2000), but the annual sum for the study area results in -667 and -858 Gg C yr<sup>-1</sup> for Nightingale et al. (2000) and Weiss et al. (2000) and Weiss et al. (2007), respectively. Other transfer velocity parameterisations could also have been interesting to use in the presents study. An example is the parameterisation by Sweeney et al. (2007), which is based on an updated and improved version of the radiocarbon method used in W92. Here, the two different parameterisations by Weiss et al. (2007) and Nightingale et al. (2000) were chosen, as these experiments were conducted within and close to the study area, respectively.

#### Technical/grammatical corrections/suggestions:

Reply: all grammatical corrections have been implemented. When suggestions of re-writing and additions to the text have been made, the changes can be seen accordingly below.

Throughout paper: Could use "pCO2surf" to represent surface ocean pCO2 Reply:  $pCO_2^w$  has been used throughout the paper for the surface ocean pCO2.

p. 2, line 19: "Biogeochemical" should be "biogeochemically"

p. 3, line 4: "though" should be "through"

p. 3, line 28: Could change "not always can be ignored" to "can't always be ignored"

p. 3, line 29: For clarity, indicate that uncertainty in the transfer velocity is much greater than the uncertainty related to temporal variability in atmospheric CO2.

Reply: a sentence has been added here with, thus

*Further, Rutgersson et al. (2008) note that the uncertainties connected with the transfer velocity are much greater.* has been changed to

Further, Rutgersson et al. (2008) note that the uncertainties connected with the transfer velocity are much greater than uncertainties related to temporal variations in atmospheric  $CO_2$ 

p. 4, lines 5-8: Could combine these two sentences. Example: "The present study aims to determine the importance of variability in atmospheric CO2 concentrations on the net air-sea CO2 flux of the Baltic Sea and Danish inner waters (which consist of Kattegat, the Danish Straits, Oresund and the Belt Seas)."
Reply: The above suggestion has been implemented.

p. 4, line 12: "resolutions" should be "resolution"

- p. 4, line 13: "did" should be "do"
- p. 4, line 16: "details" should be "detail"
- p. 4, line 19: "in" should be "for"

p. 4, line 24-26: This description of the organization of the paper could be omitted.

p. 5, line 3-4: It's already been indicated that the Danish inner waters consist of Kattegat and the Danish Straits. Could simply state: "The marine areas investigated in this study are shown in Fig. 1." Reply: The above suggestion has been implemented.

p. 5, line 9: "relative" should be "relatively"

- p. 5, line 10: "low-saline" should be "low-salinity"
- p. 5, line 11: "high-saline" should be "high-salinity"
- p. 5, line 23: No need to capitalize "Northern" (This is the case when direction refers to a general area)
- p. 6, line 23: "Kortzinget" should be "Kortzinger"
- p. 8, line 3: "month" should be "months"
- p. 8, line 5: "value increase," should be "increases" with no comma
- p. 8, line 7: "particularly" should be "particular"

p. 8, lines 10-11: Could rewrite sentence for clarity.

Reply: Page 17002 line 1-2 is changed from:

In general a good accordance between the  $pCO_2$  values calculated from the monitoring stations and underway  $pCO_2$  data from the sub-domains exists.

То

The calculated  $pCO_2$  values at the monitoring stations agree well the underway  $pCO_2$  data

p. 8, line 22: "use" should be "uses"

p. 9, line 3: delete "in the present study"

p. 9, lines 5-6: No need for a new paragraph for this statement.

p. 9, lines 23-24: Could rewrite for clarity. Example: "For the European area, CT values are replaced by a fossil fuel emission inventory ..."

Reply: Page 17003 line 19 – 21 is changed from

CT is for the European area substituted by a fossil fuel emission inventory with a higher spatiotemporal resolution (hourly, 10 km x 10 km) developed by the Institute of Energy Economics and the Rational Use of Energy (Pregger et al., 2007). To

For the European area, the CT values are replaced by a fossil fuel emission inventory with a higher spatiotemporal resolution (hourly, 10 km x 10 km) developed by the Institute of Energy Economics and the Rational Use of Energy (Pregger et al., 2007).

p. 13, line 22: "is contributing" should be "contributes"

p. 13, line 23: Could insert "atmospheric" before "CO2" for clarity.

p. 13, lines 27-29: This sentence should be rewritten for clarity. Example: "In order to investigate the effect of short-term variability in atmospheric CO2 concentration on the air-sea CO2 flux, two different model simulations were conducted." Reply: The above suggestion has been implemented.

p. 14, line 17: Insert "the" before "east"

p. 15, line 10: "has" should be "have"

p. 15, line 12: Could also indicate the interannual variability in this section Reply: see reply to the similar comment above.

p. 15, line 27: "EZZ" should be "EEZ"

p. 16, line 15: "represent" should be "represents"

p. 16, line 17: Replace "Further" with "In addition" or "Furthermore"

p. 16, line 20: "determines" should be "determine"

p. 17, lines 3-5: These two sentences could be combined for clarity

p. 17, line 25: Conclude sentence with a period

p. 18, line 16: Remove "Although,"

p. 18, line 22: Could replace "across the marine areas in focus here." with " across the study area. Reply: The above suggestion has been implemented.

p. 19, lines 8-10: Could indicate the range of annual estimates either here or in section 3 above. Reply: The above suggestion has been implemented.

p. 19, line 11: Delete "as"

p. 19, line 12: Reference Table 3

p. 19, line 14: Insert "the" before "eddy"

p. 19, line 25: "while" should be "and"

p. 19, line 26: delete "in"

p. 19, line 27: "Additional" should be "Additionally"

p. 20, line 5: insert "in this region" after "annual air-sea CO2 flux"

p. 20, lines 7-8: This sentence could be clarified since wind speeds are the same in both simulations.

Page 17016 line 19-20 is changed from:

But large differences are also found over open water areas in spite of a less variable atmospheric  $CO_2$  concentration here. The generally higher wind speeds over open water might lead to the large flux difference here. To

But large differences are also found over open water areas in spite of a less variable atmospheric  $CO_2$  concentration here, i.e. a smaller difference in the atmospheric  $CO_2$  concentration between the two simulations. Despite the small concentration difference, the tendency to higher wind speeds over open oceans leads to the large flux difference here. The same wind fields are applied in both simulations.

p. 21, line 11: "arears," should be "areas" with no comma

Fig. 3b: Indicate in the caption for this figure that fluxes are from the VAT simulation.

Fig. 7: Indicate in the caption and/or title which simulation (i.e., VAT or CAT) these fluxes represent.

Table 3: Column heading to the far right should be "Present study" or "This study"