

## ***Interactive comment on “One year of continuous measurements constraining methane emissions from the Baltic Sea to the atmosphere using a ship of opportunity” by W. Gülzow et al.***

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Reviewer 1:

- 1) Large parts of the introduction (see sections 1.1 and 1.2) are very lengthy and therefore should be shortened considerably. It is not necessary to repeat in detail what is known from the literature.

The detailed repetition of central aspects of the literature was chosen to show the state-of-the-art background of the present study and to confine the focus of the manuscript. Furthermore it was attempted to give the reader a collection of important headwords for a better understanding of the following context of controlling factors of the methane

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emissions (water depth, wind, organic matter content ...). Also, we cannot find the introductory part of the manuscript to be of exceptional length.

- One important, recently published, article has been ignored: Bange et al. (2010), *Biogeosciences*, 7, 1279-1284.

We added the reference and a short note on the findings, as well as the recently published paper by Reindl and Bolalek (2012). The paragraph at page 9901, line 16ff reads now:

Bange et al. (1998) describe the seasonal distribution of methane and nitrous oxide in the Bodden Sea, using an equilibrator combined with a gas chromatograph, postulating that methane emissions of this area account for 17 % of the total Baltic Sea methane emissions. Monthly observations of the water column at Bokins Eck (BE) between 2006 and 2008 showed a bimodal seasonality of sedimentary release of methane and elevated methane concentrations in deep water layers after sedimentation of fresh organic material from phytoplankton blooms (Bange et al. 2010). Similar periodic changes of methane fluxes from the sediment into deeper water layers were observed in the coastal zone of Puck Bay along the Hel Peninsula (Reindl and Bolalek 2012). Based on six measuring campaigns (1995, 1996) in the shallow coastal area between the Island of Rügen and Hiddensee (Southern Baltic Sea) using coating measuring chambers with continuously and automatic sampling equipment, Heyer and Berger (2000) found a strong correlation of the amount of organic matter in the sediment and interannual as well as seasonal variations of methane emissions.

- 2) Section 2.2: There is no word about the quality of the  $\chi\text{CH}_4$  data from the NOAA station in Poland. Where is it located? At the coast? Are the data affected by terrestrial/marine  $\text{CH}_4$  sources? What are actual values used? Have the data been altered for pollution events? etc. Please add this information.

We added information on the sampling site. The paragraph at page 9903, line 24ff reads now:

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Therefore, atmospheric methane concentrations  $x\text{CH}_4\text{atm}$  (ppm) of the NOAA station BAL (Baltic Sea, Poland, 55.35 °N 17.22 °E) were used to calculate sea surface equilibrium concentrations. The station is situated 65 m offshore. Atmospheric samples are taken using a surface flask sampler positioned at 3 m above sea level. The air samples are collected weekly and analyzed according to the standard evaluation protocol of NOAA (<http://www.esrl.noaa.gov/gmd/ccgg/>).

- 3) I am wondering why the authors did not make at least an attempt to quantify the overall annual emissions from the Baltic Sea. They have a data set in their hands which opens the door for a first reasonable regional-weighted and temporal resolved emission estimate. . .

The data set shows the sea surface methane concentration of the first year of continuous and autonomous measurements using a VOS-line on the Baltic Sea along three transects. The authors did no area-weighted quantification of an overall annual emission of the Baltic Sea in view of a follow up publication. We now have nearly 3 years of data available, allowing for a better judgment on interannual variations. Also, we can include additional data of several research cruises, covering a significantly larger area of the Baltic Sea surface, including the Bothnian Sea and allowing a closer look at gradients from the coasts to the basins. This comprehensive data set could not be included in the current work, but the result will be more robust and would then have to replace an estimate based only on the data of the year 2010. So we avoided to give this estimate, simply because we have more data to do exactly what is suggested by the reviewer.

- 4) Large parts of the 'Results and Discussion' are very lengthy and therefore should be shortened considerably (see e.g., sections 3.2, 3.3, 3.5). Maybe a table summarizing the most important results would help to improve the clarity and readability of the article.

The comment has been acknowledged. We thoughtfully went through the entire

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manuscript to avoid redundancy and to shorten wherever it seemed possible.

- 5) Fig 3: Usually upwelling is described by plotting CH<sub>4</sub> vs. SST; see e.g. Rehder et al., GBC, 2002. I would like to suggest that such a figure should be added and discussed.

Figure 3 was extended by the requested plot (Fig.3E) and according amendments of the discussion of figure 3 were made. The paragraph page 9911, line 11ff reads now:

In Fig. 3, methane (A), methane saturation (B), partial pressure of carbon dioxide (C) and sea surface temperature (D) are shown for the area north-west of Gotland for the year 2010. Sharp drops of the sea surface temperature (Fig. 3D) on 24 July (light green line) and 3 September (dark green line) can be pointed out with values reaching well down below 5°C. We highlight these two lines in the following, though various weaker events were observed over the course of the year. For both dates, a property-property plot of methane versus sea surface temperature (Fig. 3E) shows a significant inverse correlation demonstrating the strong coherence of rising cold water masses and increased methane values at the surface (Rehder et al. 2002). Whereas methane concentrations in July (Fig. 3A, transparent light green lines) vary in general between 3.5 nM and 4.5 nM, methane values reach up to 6.8 nM in the surface water on 24 July (Fig. 3A light green line). . . .

The paragraph page 9912, line 26 reads now:

Though both properties generally increase underneath the mixed layer (Thomas and Schneider, 1999; Schmale et al., 2010), the pCO<sub>2</sub> increase, mostly resulting from organic matter degradation, will start immediately underneath the mixed layer, while stronger enhancement of methane is generally observed in greater depth. The oval shaped characteristic of the plot of methane concentration versus sea surface temperature and methane (Fig. 3E), can be explained by the faster response time of the temperature signal compared to the slower MCA-signal in respect to changing water masses. The signal reflects the "in and out"- passage through the inhomogeneous

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upwelling water mass.

- Minor comments Page 9899, line 7: Bates et al. (1996) 'A reevaluation of the open ocean source of methane to the atmosphere' (JGR, 101, 6953-6961) has been ignored.

The reference was added. The paragraph page 9899, line 6 reads now:

Emission estimates of the world's ocean range from 2 % (Bange et al., 1994) to 10 % (Grunwald et al., 2009) of the total global methane source strength to the atmosphere. Significantly smaller emission estimates were calculated for open ocean areas like the Pacific Ocean by Bates et al. (1996) with less than 0.1% of the atmospheric source strength, and the authors point out the underestimation of coastal zones. Bange et al. (1994, 2006) also comment the underestimation of shallow marine areas due to the lack of data and the uncertainty of the role of estuaries, shelf and coastal areas, which may contribute 75 % of the total marine methane emissions to the atmosphere.

- Page 9926, lines 22-23: I could not find a 'model-based estimation of sea-air exchange of methane' in Bange (2006). Please correct.

Unfortunately a typing error of the reference year led to the wrong citation. The error was corrected and the paragraph page 9926, line 23 reads now:

A model-based estimation of sea-air exchange of methane as described in Bange (2004) was not attempted at this point due to the lack of available time series of water column methane concentration profiles of the Baltic Sea in the period of August to November 2010.

Reviewer 2:

- I suggest authors to consider to add some additional information in lines 100-106 of the manuscript about methane flux from sediment into water column in coastal areas of Baltic Sea.

We incorporated some recent findings from Reindl and Bolalek (2012) (citation added);

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see new form of the paragraph at page 9901, line 16ff above (first large text block in reply to reviewer 1).

Figure caption:

Fig. 3: (A) Methane, (B) methane saturation, (C) carbon dioxide, and (D) sea surface temperature plotted versus longitude for the year 2010 of the area West of Gotland for the year 2010. All data in transparent thin lines color-coded for each month. Two significant upwelling events in July and September are highlighted with outstanding light green (24 July) and dark green (3 September) lines. For these dates, methane versus sea surface temperature is shown using the same color coding (E).

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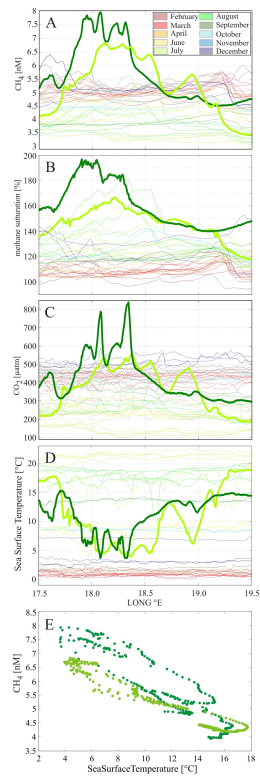


Fig. 1.

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