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Interactive comment on "Seasonal methane accumulation and release from a gas emission site in the central North Sea" *by* S. Mau et al.

S. Mau et al.

smau@marum.de

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Anonymous Referee #2

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the paper aims to constrain the seasonal variation of sea-air methane fluxes originating from shallow gas seepage, which is an important research question. The area of interest is located in a summerly thermally stratified part of the North Sea showing complete mixing in winter. A two layer model is introduced to investigate the seasonal changes of physical methane fluxes and relate those to microbial uptake.

The paper tackles complex tracer oceanographic problems requiring well planned sampling strategies, current measurements, solution of advection-diffusion equations and estimates about seasonal variation of vertical eddy diffusive transport. But the paper only presents an extremely simplified model. The oceanographic understanding appears limited and the model suffers from incompleteness and severe misunderstanding.

Author: The model is only part of the reported work and thought to support our conclusions, which were based on our data. It is not indented to be and nowhere near a detailed oceanographic model, which would need a more comprehensive data base. Our simple model is rather thought to check if our hypothesis to explain our data is correct. Biogeochemical measurements in summer and winter time in a shallow shelf sea area indicate that a seasonal thermocline leads to an enrichment of methane below the thermocline. The winter data, which is currently rare and worthwhile to be published, shows that without a thermocline, there is no enrichment of methane. Furthermore, we discuss that microbial methane oxidation appears to be a small sink of methane. These findings hints to the conclusion, that most of the methane, which piles up below the seasonal thermocline, must be vented to the atmosphere as soon as the thermocline breaks down. We used the model to check if that hypothesis is plausible. Certainly more data collected at different times of the year and revised models are needed to further validate this hypothesis.

(1) The vertical eddy diffusivity kz was estimated constant to 10e-4 from literature (and tested for model uncertainty with 10-3, and 10-5 respectively. A seasonal built-up and destruction of a thermocline gives rise to a non-static kz with variation by orders of magnitude throughout the year. The authors should have derived monthly kz, e.g. by Thorpe Scale analyses, from CTD data.

Author: First of all, we used only one value of kz (Dv in the text) for cross pycnocline mixing in the months from May until August, when there are two different water layers. The rest of the year, the water column is well mixed and there is only one water layer.

Unfortunately, we do not have sufficient CTD data to be able to calculate representative kzs. The data for the months May and June originate form an area within 3 to 6° E and 54 to 56° N as described in the manuscript, but were located closer to the coast than our study area. We lack any CTD-data of August. All CTD-data originate from water sampling and were not taken to investigate the turbulent diffusivity, thus we do not have sufficient replicates for a representative estimation of kz. However, based on the constructive suggestion of the reviewer, we estimated kz by using published dissipation rates of turbulent kinetic energy (Palmer et al., 2008, Thorpe et al., 2008) and calculating the buoyancy frequency from the available CTD-profiles. These results indicate that kz is in the order of 10-4 to 10-6 m2 s-1. Therefore, the value of kz used for the model was appropriately chosen. We will include this estimation in the manuscript.

(2) A 1D model is suggested to describe the flux of methane from the "deep" layer to the upper/mixed layer using Ficks 1st law. The authors derive model parameter dC_CH4/dz from their field data by assuming a 1D case. This would require a distinct dC_CH4/dz gradient with more or less homogeneous horizontal distribution of methane. However, the near field water column methane distribution pattern surrounding individual gas seepage clusters appears highly variable, i.e. with significant variation in three dimensions as shown by the authors themselves (Fig. 4). Surface methane values measured up to 2127nM with UWMS were reported. Obvious reasons are gas bubbles as visualized with acoustics. But the model assumes the only CH4 source is the lowermost layer in their model. In summer the thermocline may reach down by 30m leaving a lowermost layer with 10m thickness. Methane gas bubbles easily bypass a 10-20m bottom layer without losing major fractions of their initial moles as shown in the cited paper McGinnis et al. (2006). No field data is provided about the crucial model parameters initial gas bubble size and methane mole fraction. Overall, bubbles most likely provide a strong source for methane input to the upper layer, but this is totally neglected in the model.

Author: 1D models are also used for modeling transport and reactions in marine sed-

iments where methane and other chemical compounds also considerably vary over space. Similarly, bubble dissolution models do not take horizontal advection into account. These models (Leifer and Patro, 2002, McGinnis et al., 2006) suggest highest dissolution close to the sediment surface with an exponential decline towards shallower depth, thus, the main methane contribution occurs in the bottom layer, which is coherent with our data. Indeed part of the bubble dissolution takes place also in the upper layer, but if we assume that bubble dissolution is not affected by stratification as shown by Schneider von Deimling et al. (2011) at the Tommeliten site in the North Sea, then the source term for the upper layer would always be the same. Therefore, the model values that are presented are conservative estimates and the model results are qualitatively correct. Gas bubble size and methane mole fraction were not measured during the two field campaigns as our intention was to investigate the seasonal variable methane distribution.

(3) monthly mean wind speed was taken for sea-air flux modeling. But the sea-air gas transfer is highly non-linear with wind speed and a monthly mean approach needs discussion. The sea-air flux potential is also governed by an interplay between strength and continuation of wind in relation to the remaining dissolved methane pool in the "wind-exhausted" layer. I.e. strong wind will not necessarily drive enhanced sea-air flux once the upper CH4 layer was exhausted already.

Author: The formation and erosion of the seasonal thermocline forming a kind of barrier for dissolved methane was the objective of the model, thus, we used months as appropriate time period. Furthermore, monthly values of wind speed were readily accessible. However, based on this comment, we conducted a sensitivity check varying the parameters by +/- 10%. The sensitivity analysis showed that wind speed is the most sensitive parameter inducing a change of $\sim$25% whereas surface water temperatures only yielded a difference by 1.5%. We will add the sensitivity check to the discussion.

(4) The box model approach is only feasible in a closed system, but most likely the sampled area is an open system with significant advection and methane loss in various

directions, and gas bubble methane dissolution up to the sea surface. The paper refers to using the disputable approach from Mau et al. (2012).

Author: We used a 1D model that indeed ignores any horizontal movement of methane. This very simple approach was used to test our understanding of the system. If horizontal effects would be taken into account, then the source term would need to be increased. The added quantity of methane would be horizontally advected and dispersed, but the vertical exchange processes would remain the same, thus, the outcome of the model is qualitatively correct. If we move horizontally away from our modelled source, then all concentrations would be lower due to horizontal eddy diffusion. Therefore, the limitation due to the thermocline would diminish with distance to the source and the sea-air flux would decrease. As we did not extrapolate over an area, but focused on the emission site, we argue that the model is sufficient for identifying a hypothetical seasonal cycle. More importantly, as the results fit to our observations, we think that the model is a valid approach.

(5) No current data are presented for the study site. But the North Sea is highly affected by the tides and the dominating M2 tide will likely cause significant current changes in amplitude and phase on an hourly timescale. Therefore the tempo-spatial methane distribution and the respective sampling are highly controlled by the actual current around the seep sites. The methane distribution pattern in winter shown in Fig. 4b is interpreted as a result from enhanced mixing. However, it could also be caused from current amplitude and direction “flushing” the seepage area with background water during the time of measurement (e.g. frontal jets have been discussed for the Dogger Bank with currents exceeding 15 cm s -1, but are not mentioned in the paper).

Author: We agree that currents affect methane distribution, however, currents transport the methane, but do not decrease the concentration as no concentration gradient is included in the equation of the advective flux. We drew a sketch to show the influence of advection/currents (Fig. 1) showing that independent of the current velocity, the vertical concentration profile would always look similar: the concentration would

exponentially decrease with distance to the seafloor. In addition, the seasonal thermocline will always limit vertical transport, thus, the model is qualitatively correct, but not necessarily quantitatively. However, we included a description and added a plot to the supplementary material showing the east-west and north-south velocities of the currents during the sampling campaigns. The current data are modelled data by the BSH using wind and air temperature forecasts, thus, might be deficient. These data indicate that current direction was for most CTD stations to the north-west. Also the velocities were similar ranging between 0.1 and 0.2 m s-1 during sampling campaigns. The lower concentrations in winter (i.e., western part of transect) were measured when the currents were less intense than the currents during sampling the eastern part of the transect, where higher concentrations were measured. Therefore, there was most likely no flushing.

No background CTD is available, and the amount of lateral input of methane into the layers remains unknown.

Author: Methane concentrations measured at a reference station were mentioned and we will provide the data of the background CTD sampled 32 km away from the central station in summer. Methane concentrations at the reference station ranged between 17-25 nM and agree well with the ones published by Grundwald et al. (2009) for the German Bight.

The observation of enhanced MOx activity at depth is a valid observation. Also the high resolution in situ mass spectrometer CH4 data in the near-field of gas seepage is valuable, because such data are very rare (but the respective 3D methane distribution it is not presented in the paper). The authors could think about a complete new story using such data. With the severe shortcomings of the model and missing current information the content of the paper can not support the conclusions. Therefore this paper can not be suggested for publication.

Author: We think that our conclusions are justified. Our main conclusion is that dis-

solved methane accumulates below the seasonal thermocline in summer and does not accumulate in winter when there is no stratification. Furthermore, detailed analysis of the methane oxidation measurements illustrate that methane oxidation is insufficient to significantly reduce the methane load. In combination, this suggests that the accumulated methane is at some time transferred to the atmosphere. Therefore, the conclusions are drawn from our measured data and, in addition, are supported by the model, which we argue to be qualitatively valid. Our conclusions and measurements fit to and extend the current knowledge of the sinks of methane emitted from shallow seep sites. We provide analysis in winter time and methane oxidation measurements, which are both rare data and reinforce the hypothesis of a higher methane release from the sea to the atmosphere in fall as reported by Gülzow et al., 2013 for the Baltic Sea.

Technical Comments - equations are missing to allow for reconstructing the individual model steps

Author: On page 18014 line 12 we describe that eq. 3 and 4 are used in the model.

- Fig. 1: the wind recording station can hardly be detected. The flow pattern of the North Sea currents are provided, but the ones prevailing in the study area remain unclear, also in the text!

Author: We changed the color of the wind recording station for better visibility and will include a description of the general oceanography and available modelled currents during the samplings campaigns.

- Fig. 2: poor quality and unclear. The three figures/inserts have three different color codes for the depth, confusing: : :.The UWMS sampling path could be better presented in 3D together with the methane concentration distribution in the results chapter.

Author: We will change the color code of the UWMS sampling path matching it to the flare imaging color scale, the other depth scale refers to the smooth topography. If we would have a scale for all, then the topography cannot be illustrated (would be one

color). We think the discrete measured water samples and the UWMS-data are best compared as we illustrated it.

- Fig. 3: The CH4 concentration profiles should be included here.

Author: The different methane concentration profiles are not as similar as e.g. the different temperature or salinity profiles, thus, we presented the data as contour plots to show more details.

- Sea-air flux calculations: rather provide classical and comprehensive work introducing the generic sea-air flux equation equ. 4 than self-citation.

Author: The citations will be deleted.

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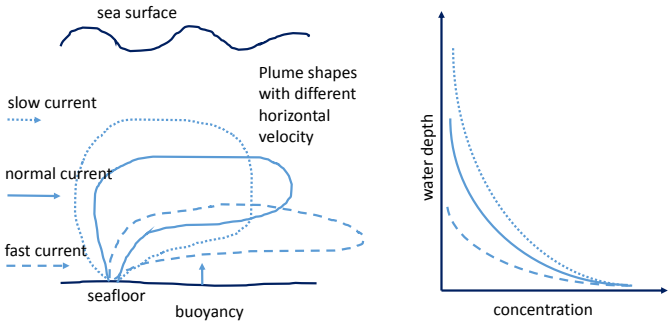

Fig. 1. Sketch of a methane plume with a constant source and buoyancy, but different water velocities. The shape of the vertical methane concentration profile would not change.