# Reply to reviewer #1

We thank reviewer 1 for the helpful comments that helped to improve the manuscript. Please find our replies to the general and specific comments below.

*For characterising marine ecosystem shifts over time, especially in highly anthropogenically impacted regions, sustained time series data are invaluable, but such records are sparse. Their documentation is essential so papers of this type, in this case presenting decadal records of dissolved methane, dissolved oxygen and chlorophylla from the Boknis Eck time series site in the Baltic, are welcome. The Boknis Eck site is subject to severe eutrophication and is an active site of methane production so this paper has potential to provide important insights into methane temporal variability.*

*As such this paper clearly falls within the scope of Biogeosciences. The authors represent a group that has a long experience of marine methane measurements and of working at the Boknis Eck site. Their methodology is well established and sound, and it is described concisely yet in enough detail to enable their reproduction by others.*

*The observations presented are rather straightforward, and while no novel concepts or ideas are described the data are worth reporting and are adequately set into the wider context, citing relevant sources. Overall the paper is well structured and generally easy to follow, and the figures are clear. I was however, a little unclear as to the authors explanation of the unusually high surface methane observed in December 2014. They mention a major inflow at this time, of high salinity, oxygenated North Sea water but it was not clear to me whether they were implying this water to be high or low in methane (or the same) relative to in situ conditions. I think an additional sentence or two would help clarify this.*

Thank you for your suggestion. A direct comparison of the dissolved $CH_4$ concentrations in the North Sea and Baltic Sea would be necessary to assess the impact of the saline water inflow. According to the published results of Bange et al. (1994) and Rehder et al. (1998), $CH_4$ concentrations in surface North Sea is much lower than in the Eckernförde Bay. Advection of water with high $CH_4$ concentration does seem to be unlikely. We thus hypothesize that the MBI led to lower concentrations in the bottom water, substituting previously high concentration throughout the water column in the lower part below the mixed layer, hence causing the observed anomaly in the $CH_4$ concentration profile.

We will include the above information and the corresponding references in section 4.2.

*They also describe a major outflow period in which sea levels declined prior to this inflow, and extreme weather that could have affected the sediment structures in the Eckernförde Bay. Presumably this could have led to methane release, but I think they stop short of saying this. Instead, they tend to favour hydrostatic pressure release due to the falling sea level as a cause of methane release from the sediments. It is not especially clear to me how this signal is transferred to the surface.*

We suggest that enhanced $CH_4$ concentrations could be attributed to sedimentary release, and high $CH_4$ concentrations could be either homogeneously distributed all over the water column (via gas bubbles) or only detected at the bottom (via porewater exchange) when the hydrostatic pressure decreased at first. The $CH_4$-enriched water was subsequently lifted to the surface by the saline inflow, which is heavier than the low salinity-water in the Eckernförde Bay. This is supported by the negative correlation between $CH_4$ concentrations and salinity in the water column.

The decline of hydrostatic pressure could be one of the potential causes of the enhanced $CH_4$ release from the sediment. There might be other potential causes, for example, sediment resuspension, resulted either from the storm or the flushing of the strong saline inflow, but this is not supported by the variation of Secchi depths. The occurrence of MBI is usually associated with storms and strong winds, but this is beyond the discussion of this study. We do not have any evidence and therefore, did not discuss the potential impact of the extreme weather conditions.

We will add more detail in section 4.2.

*Also, the hydrostatic pressure change, equivalent to the order of 1 metre in a 28-metre water column is rather small relative to the changes that occur in some estuarine and mangrove environments the authors cite. Can they provide evidence that such changes can produce the observations they describe? I wonder how important this mechanism might be relative to other possibilities.*

Lohrberg et al. (2020) reported the detection of a widespread $CH_4$ ebullition event in the Eckernförde Bay in October 2014, shortly before the occurrence of the strong MBI. They demonstrated that storm-associated fluctuations of hydrostatic pressure induced the ebullitions and estimated a sedimentary $CH_4$ flux of ~1900 µmol $m^{-2}$ $d^{-1}$, as a result of the changes in water level (± 0.5 m) and air pressure (± 1500 Pa, equivalent to approximately ± 0.15 m of water level fluctuation). Air pressure is not recorded at the BE time-series station, and we calculated the sea-to-air flux of ~3100 µmol $m^{-2}$ $d^{-1}$, with the changes in water level of ± 1 m. Water level fluctuation, when there was no strong wind or inflow event, was approximately ± 0.2 m in the

Eckernförde Bay. Ignoring the $CH_4$ oxidation in the water column, the sharp increase in sea-to-air $CH_4$ fluxes in December 2014 are generally in good agreement with the sedimentary $CH_4$ release reported by Lohrberg et al. (2020), which provides a strong evidence that the changes in water levels are capable of inducing such strong changes in $CH_4$ release.
We will incorporate this in section 4.2.

*It has been documented for example that current flows across the seabed that could be induced by surface inflows in shallow water, can set up pressure gradients driving pore water flow (e.g. Ahmerkamp et al., The impact of bedform migration on benthic oxygen fluxes. JGR Biogeosciences https://doi.org/10.1002/2015JG003106). I think perhaps a little more in-depth discussion of the various possibilities would be insightful. For example, is it possible to estimate the amount of methane that would be expected to be released from the sediments over the duration of the hydrostatic pressure drop, and is this consistent with the observed effect?*
Thank you for your suggestions. Porewater exchange might be an important benthic $CH_4$ source, and we will add more detail in section 4.2. Sedimentary $CH_4$ release via ebullition from Lohrberg et al. (2020) is generally consistent with our results. Please see the reply above.

*The authors could perhaps also clarify why they chose to use a different equation for calculating flux densities (Nightingale et al., 2000) to that used in their earlier paper (Bange et al. (2010), i.e. Raymond and Cole (2001), which gives a lower gas transfer velocity. The authors point out that the two sets of results agree if the same equation is adopted but I was curious about their reasoning in selecting Nightingale et al (2000) for this study. I am not suggesting they are incorrect in this, rather I just wanted to know their reasoning.*
We choose Nightingale et al. (2000) over Raymond and Cole (2001) because we would like to compare our results with other time-series analysis in section 4.4. As we discussed in section 4.3, there might be a great difference in flux densities originated from the different equations adopted. SI and ALOHA used Nightingale et al. (2000) and Wanninkhof (2014), respectively. Generally fluxes calculated from these 2 equations are close, and we choose the first one because it lies in the middle of many different gas transfer parameterizations, which makes it widely used and well-accepted.

# Reply to reviewer #2

We thank reviewer 2 for the detailed comments. Please find our replies below.

*GENERAL COMMENTS*

*The paper by Ma et al. titled: "A decade of methane measurements at the Boknis Eck Time-series Station in the Eckernförde Bay (Southwestern Baltic Sea)" investigated the CH4 temporal variability (from 2006 to 2017) in the whole water column at the Boknis Eck Time-series Station located in the Eckernförde Bay (SW Baltic Sea). In this system the concentration of CH4 increases with depth due principally to the fluxes from the sediments. Sporadic elevated CH4 concentrations (up to 696 nM) have been observed in the upper layer coinciding with Major Baltic Inflow events. During the period studied the Eckernförde Bay is an intense but highly variable source of atmospheric CH4. The manuscript is very interesting and as the authors state, time-series measurements of CH4 are still sparse, reason why the study can contribute to have a better knowledge of the behaviour of this greenhouse gas in coastal systems, hot spots of CH4 emissions.*

*The data are well presented and the discussion of the dataset is comprehensive and conclusive. However, from my point of view, I have some suggestions to render the work more attractive to readers. Therefore, I suggest its publication after minor revisions.*

*Since part of the behaviour of CH4 is attributed to contributions of more saline water from the North Sea and that it is a seasonal study with significant variations in temperature, it is convenient to include the variations of temperature and salinity in Figures 2 and 3.*

Thank you for your suggestion. Seasonal and inter-annual variations of temperature and salinity will be shown in the figures.

*Throughout the manuscript it have been discussing about good and bad correlations between the different variables studied, however, hardly any statistical data (p values, r2) are provided to indicate the good or bad degree of these correlations. I think it would be convenient to include a table with the annual intervals of variation and mean values and deviation of the studied variables including salinity, temperature and wind speed.*

We will include a table with the variables as suggested.

*SPECIFIC COMMENTS*

*Pg. 1 Ln 27. Missing "l" in oil. Pg2 Ln 52. Include "temperature increment" in : : :.. which is one of the most rapid temperature increment in large marine ecosystems.*

We will revise them as suggested.

*Pg 3 Ln 74. HgCl2 was added to the sample once it was sealed with rubber stopper and aluminium caps? Was the measurement done with a gas-tight syringe? In that case, could a small pore have been left in the rubber stopper to facilitate gas exchange?*

There is a small pore left in the rubber stopper after poisoning, but the gas exchange is negligible for this standard method. We have tested that the vials are gas-tight despite of the pores.

*Pg 3 Ln 83. The concentrations of CH4 standards used should be indicated, because although the average concentration is 51.2 _ 84.2nM, there were some sporadic samples with very high concentrations (more than 600 nM) and those concentrations should be within the calibration line.*

The measurements last for more than a decade and the standard gases we used changed several times. We have adjusted the concentrations of standard gases for every measurement to make sure that the values of the samples fall in the range of the calibration curves. In this case, we think it is not necessary to list all the $CH_4$ standards.

We will include this information in the method section.

*Pg 3Ln 89-90. The accurate in dissolved oxygen and Chla measurements should be indicated. How were temperature and salinity measured? What was the precision of these measurements?*

Temperature and salinity were measured by the sensors equipped on CTD. Different methods might be adopted for individual parameter during the past decades. A more comprehensive overview of temperature, salinity, dissolved oxygen, Chla as well as other parameters at BE can be found in the paper by Lennartz et al. (Lennartz, S. T., Lehmann, A., Herrford, J., Malien, F., Hansen, H. P., Biester, H., and Bange, H. W.: Long-term trends at the Boknis Eck time-series station (Baltic Sea), 1957–2013: does climate change counteract the decline in eutrophication? Biogeosciences, 11, 6323–6339, https://doi.org/10.5194/bg-11-6323-2014, 2014).

We will include this information in the method section.

*Pg 4 Ln 112-113. What H2S concentrations were measured? It would be interesting to include these values*

Unfortunately we did not measure the $H_2S$ concentrations. The presence of $H_2S$ was recognized by the strong smells of the bottom water.
We will add it to the text.

*Pg 4 Ln 115. Indicate the value of the DO concentration that was obtained in the surface waters. This upwelling has also been appreciated in other variables such as nutrients?*
We have shown the approximate $O_2$ saturation in the surface water, which is a better proof than the actual DO concentration. The occurrence of the upwelling can be identified by nutrients, too, but not as clearly as indicated by temperature and $O_2$. In this case, we think it is not necessary to include the variation of nutrients in the water column.
We will add more detail here.

*Pg 4 Ln 115-116. Since the authors write about behaviour of temperature and salinity, it would be convenient to include graphs of these variables in Figures 2 and 3.*
The variations of temperature and salinity will be shown in figure 2 and 3.

*Pg 5 Ln 124. What is the reason that in BE the Chla has elevated concentrations only in the upper layers in March and not occupied the whole water column as other works realised in this system? Are Chla and Secchi depth well related to the entire study?*
*Perhaps it could be included in the figure of the Chla the Secchi depth graph. If we look at figure 2 in 2006 and 2012 the Chla occupies the entire water column. What could have happened in these years for the Chla behaviour to be different?*
It remains unclear why elevated Chla concentrations were only detected in the upper layers in March. The overall correlation between Chla and Secchi depth is poor ($r^2$=0.17, p<0.0001, n=111). High Chla concentrations all over the water column in November/December 2006 and March 2012 were coinciding with slightly enhanced nutrients and high temperatures. Nutrients and temperature might be potential environmental controls on Chla distribution.
We will incorporate this additional information in section 4.1.

*Pg 5 Ln 130. To show seasonal and inter-annual variations, a table could also be presented showing the variation interval and annual mean value of each variable. Figures 2 and 3, although very illustrative, have been made with interpolations and do not show the specific data that it is interesting to know.*
A table will be shown in the main text as suggested.

*Pg 5 Ln 154. Is there any work in the area where CH4 benthic fluxes have been measured? If so, it would be interesting to include the value.*

There are several papers reporting benthic $CH_4$ fluxes in the Eckernförde Bay. Sedimentary $CH_4$ release, via pockmarks or ebullition, was discussed in section 4.3.

*Pg 6 Ln 166-167. Was the water more turbid? Did Secchi's disc reach less depth?*

We did not see a strong decline in Secchi depth.

We will add this information in the text.

*Pg 6 Ln 178. What is the r2 of the relation between salinity and CH4?*

$r^2$=0.84, and this value together with p and n are now included in the text.

*Pg6 Ln 184. Include variation of dissolved oxygen values to change from hypoxic to oxic condition in the bottom layer.*

The values are now included as suggested.

*Pg 6 Ln 189. Include the correlation coefficient (r2) of the relation between salinity and CH4 in November 2013. Pg 6 Ln 199. Include the correlation coefficient (r2) of the relation between salinity and CH4 in March 2014.*

$r^2$, p and n are now described in the text.

*Pg 7 Ln 213-214. Since CH4 saturation has been obtained from the surface methane concentration and equilibrium concentrations of CH4 in seawater, it is obvious that surface CH4 saturations are directly proportional to its concentrations in the surface water, I would omit this from the manuscript.*

The calculation of $CH_4$ saturation involves concentration, as well as temperature and salinity. The authors would like to point out that surface $CH_4$ saturations are directly proportional to its concentrations, despite of the significant seasonality in temperature and salinity. In another paper about dissolved $N_2O$ at BE, the influence of temperature is stronger. We think this comparison might be interesting.

*Figures:*

*In the figures the letters and numbers are in Arial and not in Times New Roman like the rest of the manuscript.*

BG does not require a consistency for the fonts in the figures and in the text. As long as the fonts in all of the figures are consistent, it would not be a problem.

*Figure 1. The quality of the figure must be improved. Figures 2 and 3. The axis titles should appear with capital letters as: Depth, not depth and Dissolved oxygen not only oxygen. It should be convenient to include isolines in these figures for a better appreciation of the concentration variations. Figures with temperature and salinity variations should be included.*

The figures will be revised as suggested, except for the isoclines. We tried to include isoclines in figure 2 and 3, but they seem crowded and messy because of the strong gradients.

According to the comments from the reviewers, the following changes are made in the manuscript:

1. Include a table of inter-annual variations of temperature, salinity, wind speed and dissolved $CH_4$ concentrations.
2. Improve the quality of Fig. 1.
3. Show inter-annual and seasonal variations of temperature and salinity in Fig. 2 and 3, respectively.
4. Add more details about standard gases (in lines 85–86) and the overview of other parameters (in lines 94–95).
5. Include the information about $H_2S$ recognition (in line 117).
6. Add more details about upwelling signal from nutrients in lines 121–122.
7. Analyze the potential correlations between Chlorophyll *a* and nutrients or temperature in lines 130–133.
8. Compare $CH_4$ concentrations from the North Sea and the Eckernförde Bay in lines 171–175.
9. Briefly discuss the potential $CH_4$ contribution from porewater exchange and sediment resuspension in lines 180–183.
10. Reanalyze the impact of ebullition as a result of water level change and compare the values with our results in lines 188–196.
11. Add the specific numbers about correlations and $O_2$ concentrations in lines 200, 206, 212 and 219.
12. Few typos in the manuscript were corrected.

# A decade of methane measurements at the Boknis Eck Time-series Station in the Eckernförde Bay (Southwestern Baltic Sea)

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**Abstract.** Coastal areas contribute significantly to the emissions of methane ($CH_4$) from the ocean. In order to decipher its temporal variability in the whole water column, dissolved $CH_4$ was measured on a monthly basis at the Boknis Eck Time-series Station (BE) located in the Eckernförde Bay (SW Baltic Sea) from 2006 to 2017. BE has a water depth of about 28 m and dissolved $CH_4$ was measured at six water depths ranging from 0 to 25 m. In general, $CH_4$ concentrations increased with depth, indicating a sedimentary release of $CH_4$. Pronounced enhancement of the $CH_4$ concentrations in the bottom layer (15–25 m) was found during February, May–June and October. $CH_4$ was not correlated with Chlorophyll *a* or $O_2$ over the measurement period. Unusually high $CH_4$ concentrations (of up to 696 nM) were sporadically observed in the upper layer (0–10 m) (e.g. in November 2013 and December 2014) and were coinciding with Major Baltic Inflow (MBI) events. Surface $CH_4$ concentrations were always supersaturated throughout the monitoring period, indicating that the Eckernförde Bay is an intense but highly variable source of atmospheric $CH_4$. We did not detect significant temporal trends in $CH_4$ concentrations or emissions, despite of ongoing environmental changes such as warming and deoxygenation in the Eckernförde Bay. Overall, the $CH_4$ variability at BE is driven by a complex interplay of various biological and physical processes.

## 1. Introduction

Methane ($CH_4$) is an atmospheric trace gas which contributes significantly to global warming (IPCC, 2013) and the evolution of stratospheric ozone (WMO, 2018). Atmospheric $CH_4$ mole fractions have been increasing by about 150 % since the industrial revolution (IPCC, 2013).

The oceanic release of $CH_4$ to the atmosphere plays a minor role for the global atmospheric $CH_4$ budget (Saunois et al. 2016). However, coastal areas have been identified as hot spots of $CH_4$ emissions (see e.g. Bange et al., 1994; Upstill-Goddard et al., 2000; Borges et al., 2016). Dissolved $CH_4$ in coastal waters is mainly resulting from the interplay of (i) sedimentary sources such as anaerobic methanogenesis during the decomposition of organic matter (Xiao et al., 2018; Dale et al., 2019) or seepage from oil ~~l~~and natural gas reservoirs (Bernard et al., 1976; Hovland et al., 1993; Judd et al., 2002) and (ii) microbial $CH_4$ consumption which occurs under oxic conditions in the water column and under anoxic conditions in the sediments (Pimenov et al., 2013; Steinle et al., 2017; Egger et al., 2018). Only recently, Weber et al. (2019) estimated the global oceanic $CH_4$ emissions to range from 6 to 12 Tg $yr^{-1}$, of which about 0.8–3.8 Tg $yr^{-1}$ were attributed to coastal waters. Occasional studies of the $CH_4$ production and consumption pathways in coastal waters and the associated $CH_4$ emissions to the atmosphere have received increasing attention during the last decades (Bange et al., 1994; Reeburg 2007; Naqvi et al., 2010). However, time-series measurements of $CH_4$ which would allow identifying short- and long-term trends in view of the ongoing environmental changes in coastal regions (such as eutrophication, warming and deoxygenation) are still sparse. In this paper we present the monthly measurements of $CH_4$ from a time-series station in the Eckernförde Bay (Baltic Sea) during 2006–2017.

Due to severe eutrophication, sediments in the Eckernförde Bay receive large amount of organic matter (Smetacek et al., 1987; Oris et al., 1996; Nittrouer et al., 1998) and thus are active sites of $CH_4$ formation (Schmaljohann, 1996; Whiticar, 2002; Treude et al., 2005; Maltby et al., 2018). Seasonal and inter-annual $CH_4$ variations in concentration, saturation and air-sea flux density were investigated for more than a decade. The aim of this study was to assess the seasonal dynamics of and the environmental controls on $CH_4$ variability in the Eckernförde Bay which is affected by high nutrient concentrations, increasing water temperatures and ongoing loss of dissolved oxygen (Lennartz et al., 2014).

## 2. Study site

The Boknis Eck (BE) time-series station is one of the oldest continuously conducted marine time-series stations in the world. The first sampling took place in 1957, and has been conducted on a monthly base with only minor interruptions since then (Lennartz et al., 2014). It is situated in the Eckernförde Bay in the southwestern (SW) Baltic Sea, with a depth of approximately 28 m (Fig. 1). The sediments in the Bay are characterized by high organic matter load and sedimentation rate (Orsi et al., 1996; Whiticar, 2002), which is closely associated with the spring and autumn algae blooms (Smetacek, 1985).

The Baltic Sea has only a limited water exchange with the North Sea through the Kattegat, which makes this area very sensitive to climate change and anthropogenic impacts. As a result of global warming, the increasing trend for the global sea surface (< 75 m) temperatures (SST) was about 0.11 °C per decade (IPCC, 2013), while a net SST increase of 1.35 °C was observed in the Baltic Sea during 1982–2006, which is one of the most rapid temperature increment in large marine ecosystems (Belkin, 2009). Lennartz et al. (2014) reported a warming trend of up to 0.2 °C per decade at the BE time-series station for the period of 1957–2013. Nutrients in the Baltic Sea have been increasing until 1980s as a result of the intensive agricultural and industrial activities, and then started to decline due to effective wastewater control (HELCOM, 2018). However, hypoxia and anoxia have been increasing in the Baltic Sea during the past several decades (Conley et al., 2011; Carstensen et al., 2014). Similar trends in nutrients and $O_2$ were also detected at the BE time-series station (Lennartz et al., 2014), indicating that the Eckernförde Bay is representative for the biogeochemical setting of the SW Baltic Sea. In concert with the declining nutrient concentrations, Chlorophyll *a* concentrations at the BE time-series station were declining as well (Lennartz et al., 2014).

Located close to the bottleneck of the water exchange between the North Sea and the Baltic Sea, the BE time-series station is also sensitive to hydrographic fluctuations such as inflows of saline North Sea Water. There is no riverine input to the Eckernförde Bay, and thus, the saline water inflow from the North Sea plays a dominant role in the hydrographic setting at BE. Because the inflowing North Sea water has a higher salinity compared to Baltic Sea water, a pronounced summer stratification occurs which leads to the development of a pycnocline at about 15 m water depth. The seasonal stratification occurs usually from mid-March until mid-September. During this period, vertical mixing is restricted and bacterial decomposition of organic material in the deep layer causes pronounced hypoxia and sporadically occurring anoxia during late summer (Lennartz et al., 2014). Pronounced phytoplankton blooms occur regularly in autumn (September–November) and spring (February–March) and to a lesser extent during summer (July–August) (Smetacek et al. 1985).

## 3. Methods

### 3.1 Sample collection and measurement

Monthly sampling of $CH_4$ from the BE time-series station started in June 2006. Seawater was collected from 6 depths (1, 5, 10, 15, 20 and 25 m) with 5 L Niskin bottles mounted on a CTD rosette. 20 mL brown glass vials were filled in triplicates without any bubbles. The vials were sealed immediately with rubber stoppers and aluminum caps. These samples were poisoned with 50 µL saturated aqueous mercury chloride ($HgCl_2$) solution as soon as possible, and then stored in a cool, dark place until measurement. The storage time of the samples before the measurements was less than 3 months.

A static headspace-equilibrium method was adopted for the $CH_4$ measurements. A 10 mL Helium (99.9999 %, AirLiquide, Düsseldorf, Germany) headspace was created inside the vial with a gas-tight syringe (VICI Precision Sampling, Baton Rouge, LA). The sample was vibrated with Vortex (G-560E, Scientific Industries Inc., New York, USA) for approximately 20 s and then left for at least 2 h to reach the $CH_4$ equilibrium between air and water phases. A 9.5 mL subsample of headspace was injected into a gas chromatograph equipped with a flame ionization detector (GC-FID, Hewlett-Packard 5890 Series II, Agilent Technologies, Santa Clara, CA, USA). Separation took place on a packed column (SS, 1.8 m length, packed with molsieve 5A, Grace, Columbia, Maryland, USA). Standard gas mixtures with varying mole fractions of $CH_4$ in synthetic air (Deuste-Steininger GmbH, Mühlhausen, Germany and Westfalen AG, Münster, Germany) were used daily to calibrate the response of FID before measurements. The concentrations of standard gases were adjusted for every measurement to make sure that the values of the samples fall in the range of the calibration curves. The standard gas mixtures were calibrated against NOAA primary gas standard mixtures in the laboratory of the Max-Planck-Institute for Biogeochemistry in Jena, Germany. Further details about the measurements and calculations of the dissolved $CH_4$ concentration can be found in Bange et al. (2010). The mean precision of the $CH_4$ measurements, calculated as the median of the estimated standard errors (see David, 1951) from all triplicate measurements, was ± 1.3 nM. Samples with an estimated standard error of >10 % were omitted. Dissolved $O_2$ concentrations were measured with Winkler titrations, and Chlorophyll *a* concentrations were measured with a Fluorometer (Grasshoff et al., 1999). Secchi depth was measured with a white disk

(~30 cm in diameter). Sea levels were measured at Kiel-Holtenau, which is about 15 km away from the BE time-series station (http://www.boos.org/). A more comprehensive overview of temperature, salinity, dissolved $O_2$, Chlorophyll *a* as well as other parameters at the BE time-series station can be found in Lennartz et al. (2014).

### 3.2 Calculation of saturation and air-sea flux density

The $CH_4$ saturation ($S_{CH4}$, %) was calculated as:

$$S_{CH4} = 100 \times CH_{4obs} / CH_{4eq} \quad (1)$$

where $CH_{4obs}$ and $CH_{4eq}$ are the observed and equilibrium concentrations of $CH_4$ in seawater, respectively. $CH_{4eq}$ was calculated with the in-situ temperature and salinity of seawater (Wiesenburg and Guinasso, 1979), and the dry mole fraction of atmospheric $CH_4$ at the time of sampling, which was derived from the monthly atmospheric $CH_4$ data measured at Mace Head, Ireland (AGAGE, http://agage.mit.edu/).

The air-sea $CH_4$ flux density ($F_{CH4}$, in μmol $m^{-2}$ $d^{-1}$) was calculated as:

$$F_{CH4} = k \times (CH_{4obs} - CH_{4eq}) \quad (2)$$

where k (in cm $h^{-1}$) is the gas transfer velocity calculated with the equation given by Nightingale et al. (2000), as a function of the wind speed and the Schmidt number (Sc). Sc was computed with the empirical equations for the kinematic viscosity of seawater (Siedler and Peters, 1986) and the diffusion coefficients of $CH_4$ in water (Jähne et al., 1987). Wind speed data were recorded at the Kiel Lighthouse (www.geomar.de/service/wetter/), which is approximately 20 km away from the BE time-series station. The wind speeds were normalized to the height of 10 m ($u_{10}$) with the method given by Hsu et al. (1994).

## 4. Results and discussion

### 4.1 Seasonal variations of environmental parameters and dissolved $CH_4$

Seasonal hypoxia were observed every year at the BE time-series station during 2006–2017 (Fig. 2). $O_2$ depletion was detected in the bottom layer (~15–25m) during July–October with minimum $O_2$ concentrations usually occurring in September (Fig. 3). Lennartz et al. (2014) found a significant decrease in dissolved $O_2$ concentrations in the bottom water at the BE time-series station over the past several decades and suggested that temperature-enhanced $O_2$ consumption and a prolonged stratification period might be the causes of deoxygenation. Anoxia with the presence of hydrogen sulfide ($H_2S$, identified from the strong smell) in the period of concurrent $CH_4$ measurements were found in the autumn of 2007, 2014 and 2016, respectively. The anoxic event in 2016 lasted from September until November and was the longest ever recorded at the BE time-series station. In September 2017, a pronounced undersaturation of $O_2$ (~50%) was observed in surface water (Fig. 2). The low temperature together with enhanced salinity in the surface water in September 2017 suggests the occurrence of an upwelling event, which transported $O_2$-depleted and colder bottom waters to the surface. An upwelling signal was also present in the nutrient concentrations (not shown), but less pronounced than in temperature and dissolved $O_2$ concentrations. Similar events were also detected in September 2011 and 2012.

Enhanced Chlorophyll *a* concentrations, which can be used to indicate phytoplankton blooms, were usually observed in spring or autumn, but not in every year (Fig. 2). Seasonal variations of Chlorophyll *a* concentrations were generally consistent with the annual plankton succession reported by Smetacek (1985). During 2006–2017, high Chlorophyll *a* concentrations were usually found in the upper layers in March (Fig. 3), which is different from the seasonality during 1960–2013 where on average, high concentrations occupied the whole water column (Lennartz et al., 2014). Another difference is that no prevailing 'winter dormancy' of biological activity was observed: Chlorophyll *a* concentrations usually remained high throughout the autumn–spring period. In November and December 2006 and March 2012 when high Chlorophyll *a* concentrations were observed all over the water column, nutrients and temperature were generally higher. Although the overall correlation between Chlorophyll *a* and nutrients ($NO_3^-$, $r^2=0.01$, $p<0.01$, $n=674$) or temperature ($r^2=0.02$, $p<0.0001$, $n=671$) is poor, nutrients or temperature might be potential environmental controls on Chlorophyll *a* distribution. As a proxy of water transparency, Secchi depth was lowest in March indicating a high turbidity, coincident with the Chlorophyll *a* maximum. Chlorophyll *a* concentrations and Secchi depths have been decreasing over the past decades in the Baltic Sea (Sandén and Håkansson, 1996; Fleming-Lehtinen and Laamanen, 2012; Lennartz et al., 2014), but this trend cannot be identified from the median slope at the BE time-series station during 2006–2017.

$CH_4$ concentrations at the BE time-series station showed strong seasonal and inter-annual variability (Fig. 2, Table. 1). During 2006–2017, dissolved $CH_4$ concentrations ranged between 2.9 to 695.6 nM, with an average of 51.2 ± 84.2 nM. High concentrations were generally observed in the bottom layer (~15–25 m). Enhanced $CH_4$ concentrations were mainly observed during February, May–June and October (Fig. 3). Steinle et al. (2017) measured aerobic $CH_4$ oxidation at the BE time-series station and found that lowest rates occurred in winter, which might be one of the reasons for the enhanced $CH_4$ concentrations in February.

The $CH_4$ accumulation in May and June can be linked to enhanced methanogenesis fueled by organic matter from the spring algae bloom. Capelle et al. (2019) found a positive correlation between mean monthly $CH_4$ concentrations and Chlorophyll *a* concentrations in the upper layers of time-series measurements from Saanich Inlet. Bange et al. (2010) also reported correlations between seasonal $CH_4$ variation and Chlorophyll *a* or Secchi depth, albeit with a time lag of one month, at the BE time-series station during 2006–2008. Although we did not detect such relationships for the extended measurements during 2006–2017, in 2009 and 2016, when no spring algae blooms were detected, $CH_4$ concentrations in following summer months were lower than average (Fig. 2).

Maximum $CH_4$ concentrations were usually observed in October, at the end of the seasonal hypoxia (Fig. 3). Due to the long-lasting anoxic event, strong $CH_4$ accumulations were observed in autumn 2016 (~600 nM), which are the highest in the bottom layer during 2006–2017. Prevailing for several months, depletion of bottom $O_2$ concentrations exerts a strong influence on the underlying sediment. Maltby et al. (2018) detected a shoaling of the sulfate reduction zone in autumn and enhanced methanogenesis in the sediments at the BE time-series station. Reindl and Bolalek (2012) found similar variations in sedimentary $CH_4$ release in the coastal Baltic Sea. In-situ production in the anoxic bottom water might be a potential $CH_4$ source as well (Scranton and Farrington, 1977; Levipan et al., 2007). We, therefore, suggest that the accumulation of $CH_4$ in the bottom water in October is caused by its release from the sediments and in-situ production in the overlying water column in combination with the pronounced water column stratification during autumn which prevents ventilation of $CH_4$ to the surface layer.

### 4.2 Enhanced $CH_4$ concentrations in the upper water layer

In agreement with Schmale et al. (2010) and Bange et al. (2010), we found that $CH_4$ concentrations generally increase with water depth, indicating a prevailing release of $CH_4$ from the sediments into the water column in the Baltic Sea (see Sect. 4.1). Nonetheless, unusual high $CH_4$ concentrations in the upper layers were detected sporadically at the BE time-series station during 2006–2017 (Fig. 2). In November 2013 and March 2014, average $CH_4$ concentrations in the upper waters were 187.2 ± 13.9 nM (1–10 m) and 217.8 ± 1.4 nM (5–10 m), which are about 16 and 5 times higher than those found in the bottom layers, respectively (Fig. 4). The most striking event occurred in December 2014, when $CH_4$ concentrations in the upper layer (1–15 m) were as high as 692.6 ± 3.4 nM (19,890 ± 115 %), whereas dissolved $CH_4$ in the bottom layer (20–25 m) was ~50 nM. The surface $CH_4$ concentration in December 2014 was the highest observed during 2006–2017. In December 2014, a major Baltic inflow (MBI) event occurred, carrying large amounts of saline and oxygenated water from the North Sea into the Baltic Sea (Mohrholz et al., 2015). ~~It~~Dissolved $CH_4$ concentrations in the surface North Sea were much lower than in the Eckernförde Bay (Bange et al., 1994; Rehder et al., 1998), and therefore a direct $CH_4$ contribution from the North Sea by oxygenated waters seems unlikely. We hypothesize that this inflow substituted the lower part of the water column which had high $CH_4$ concentration throughout the water depth before, opposite to, e.g., an in-situ production of $CH_4$ at the surface being responsible for the observed concentration profile anomaly. The MBI is the third strongest event ever recorded, and an unusual outflow period was detected in the Eckernförde Bay: Sea levels declined since mid-November and reached minimum on 10 December, and then began to increase with the inflow (Fig. 5). The sampling at the BE time-series station took place on 16 December, during the main inflow period. Extreme weather conditions (wind speed >15 m s$^{-1}$) were observed several days before the sampling date, and storm-generated waves and currents could have affected the sediment structures in the Eckernförde Bay (Oris et al., 1996). Currents across the seabed can result in pressure gradients that drive porewater flow within the permeable sediments (Ahmerkamp et al., 2015), which might be a potential $CH_4$ source. Sediment resuspension might also contribute to enhanced $CH_4$ release, but we did not observe a significant decline in Secchi depths in December 2014 (Fig. 2).

The significant decrease in sea level alleviated the static pressure on the sediments. Enhanced $CH_4$ release from the sediments, via gas bubbles or exchange from porewater, may have led~~leads~~ to the accumulation of $CH_4$ in the water column. Similar hydrostatic pressure effects were also reported in tidal systems such as mangrove creeks and estuaries (see e.g. Barnes et al. 2006; Maher et al., 2015; Sturm et al., 2017). Atmospheric pressure also contributes to the overall pressure on the sediments, but it is not recorded at the BE time-series station and thus was omitted. Although the water level fluctuation of ± 1 m (Fig. 5) seems rather small compared to the water depth (28m), it might exert a strong influence on the sediments. Water level fluctuation, when there was no strong wind or inflow event, was approximately ± 0.2 m in the Eckernförde Bay. Lohrberg et al. (2020) detected a change in water level (± 0.5 m) and air pressure (± 1500 Pa, equivalent to approximately ± 0.15 m of water level fluctuation) during a weak storm in the fall of 2014. The fluctuation in hydrostatic pressure induced a pronounced $CH_4$ ebullition event in the Eckernförde Bay, and a sedimentary $CH_4$ flux of 1916 µmol $m^{-2}$ $d^{-1}$ was estimated (Lohrberg et al., 2020). ~~Lohrberg et al. (2020) identified a pronounced $CH_4$ ebullition event in the Eckernförde Bay in the fall of 2014 as a result of the decline in hydrostatic pressures during a weak storm.~~ This value is generally in good agreement with the sharp increase in the sea-to-air $CH_4$ fluxes in December 2014 (see section 4.3). The outflow period of the MBI in 2014 lasted for almost a month, and bulk ebullitions and supersaturated water with $CH_4$ could be anticipated. During the inflow period, large amounts of North Sea water flooded into the Eckernförde Bay and presumably pushed the $CH_4$-enriched water to the surface. A negative correlation was found between salinity and $CH_4$ concentration in the water column (Fig. 4a, $r^2$=0.84, p=0.01, n=6), indicating that vertical $CH_4$ distributions were linked to the mixing of saline water in the bottom and less-saline water in the upper layers. We suggest that $CH_4$ release driven by hydrostatic pressure fluctuations and the MBI-associated mixing are responsible for the abnormal $CH_4$ profile in December 2014.

The $CH_4$ anomaly in November 2013 can be linked to saline water inflow as well. Nausch et al. (2014) reported the occurrence of an inflow event from 27 October to 7 November in 2013. The sampling at the BE time-series station took place on 5 November, and an increase in salinity was detected in the bottom water (Fig. 4b). The rapid transition from hypoxic (9.8 µM/L, 25 m in October) to oxic condition (239.2 µM/L, 25 m in November) in the bottom layer also supports the occurrence of the inflow (Fig. 2). Steinle et al. (2017) found a change in the temperature optimum of aerobic $CH_4$-oxidizing bacteria (MOB) in November 2013 at the BE time-series station and linked it to a displacement of the local MOB community as a result of saltwater injection. Although enhanced $CH_4$ concentrations and high net methanogenesis rates were detected in the sediments in November 2013 (Maltby et al., 2018), the saline inflow with less dissolved $CH_4$ was sandwiched between the sediments and the upper layer waters. As a result, we also found a negative salinity-$CH_4$ correlation in the water column (Fig. 4b, $r^2$=0.86, p<0.01, n=6). This inflow event was much weaker than the MBI in December 2014, and no obvious outflow or inflow period can be identified from sea level variations. There was no strong fluctuation in hydrostatic pressure and thus sedimentary $CH_4$ release and $CH_4$ supersaturations in the water column were lower than in December 2014. Another difference is that the decrease in salinity and increase of $CH_4$ concentrations were observed between 10–20 m, which is at shallower depths compared to the MBI in December 2014, indicating that the saline water volume in the bottom layer was larger at the time of the sampling in November 2013.

The situation in March 2014 is different. We did not find any evidence for saline water inflow or hydrostatic pressure fluctuation, and the correlation between $CH_4$ concentration and salinity is poor (Fig. 4c, $r^2$=0.43, p=0.16, n=6). The occurrences of the unusual $CH_4$ profiles were accompanied by the enhanced Chlorophyll *a* concentrations in the upper waters. $CH_4$ productions by widespread marine phytoplankton have been reported and might be potential sources of surface $CH_4$ supersaturations (Lenhart et al, 2016; Klintzsch et al., 2019). However, spring or autumn algae blooms at the BE time-series station were often observed without $CH_4$ accumulation and surface $CH_4$ contribution from phytoplankton remains to be proven. Potential sources for the enhanced $CH_4$ in March 2014 are still unclear.

In summary, we suggest that saline water inflow and the subsequent upwelling of water are the most potential causes for the $CH_4$ surface accumulation in November 2013 and December 2014. Nonetheless, the occurrence of inflow does not necessarily lead to enhanced $CH_4$ concentrations in the upper waters. Inflow events are relatively common, for example, in 2013, besides the inflow in November, three other events with similar estimated inflow volumes were detected in January, February and April (Nausch et al., 2014), but no $CH_4$ anomaly was found during that period. The magnitude of the $CH_4$ anomalies might depend on the strength of the inflow events and other factors, such as storms and sediment resuspension. Besides, there is a high chance that the monthly sampling at the BE time-series station only captured few $CH_4$ pulses. Inflow events usually last days to weeks, but the accumulated $CH_4$ in the upper layers might last even shorter because of effective aerobic $CH_4$ oxidation (Steinle et al., 2017) and strong vertical mixing in winter. The occurrences of surface $CH_4$ accumulations at the BE time-series station might be more frequent than been observed.

### 4.3 Surface saturation and flux density

Surface $CH_4$ saturations are directly proportional to its concentrations in the surface water ($S_{CH4}=31.40 \times [CH_4] + 10.29$, $R^2=0.9794$, $n=77$, $p<0.0001$; Fig. 6a, b), despite of the pronounced seasonal variations in temperature (Fig. 3). This indicates that the net $CH_4$ production at BE is overriding the temperature-driven variability of the $CH_4$ concentrations. Excluding the extreme value from December 2014, surface $CH_4$ saturations at the BE time-series station varied between 129–5563 %, with an average of 615 ± 688 %. The surface layer was supersaturated with $CH_4$ and thus emitting $CH_4$ to the atmosphere throughout the sampling period.

The coastal Baltic Sea, especially the southwestern part, is a hot spot for $CH_4$ emissions. Area-weighted mean $CH_4$ saturations for the entire Baltic Sea (113 % and 395 % in winter and summer 1992, respectively; Bange et al., 1994) were lower than at the BE time-series station. Schmale et al. (2010) extensively investigated dissolved $CH_4$ distributions in the Baltic Sea, and found that surface $CH_4$ supersaturations were stronger in the shallow western areas.

Sea-to-air $CH_4$ flux densities fluctuated between 0.3–746.3 $\mu$mol $m^{-2}$ $d^{-1}$, with an average of 43.8 ± 88.7 $\mu$mol $m^{-2}$ $d^{-1}$ (excluding the extreme value in December 2014, Fig. 6c). Comparable results in saturation and flux density were observed at the pockmark sites in the Eckernförde Bay (Bussmann and Suess, 1998). Although surface $CH_4$ saturations in this study are consistent with the previously published results by Bange et al. (2010) (554 ± 317 %), calculated $CH_4$ flux densities in this study are much higher than in Bange et al. (2010) (6.3–14.7 $\mu$mol $m^{-2}$ $d^{-1}$). The discrepancy derives from different flux calculation methods. Bange et al. (2010) adopted the equations by Raymond and Cole (2001) with a lower gas transfer velocity, and they used the median of surface $CH_4$ concentrations for computation, which eliminated the extreme values. Our results are in good agreement with the ones reported by Bange et al. (2010) if we adopt the same method.

$CH_4$ emissions from coastal waters could be roughly considered as the difference between formation and oxidation of $CH_4$ in the water column and sediments. Although sediments are substantial $CH_4$ sources, most $CH_4$ is consumed before evading to the atmosphere (Martens et al., 1999; Treude et al., 2005; Steinle et al., 2017). Treude et al. (2005) compared the potential and field rates of anaerobic oxidation of methane (AOM) in the sediments of the Eckernförde Bay and suggested that the AOM-mediating organisms are capable of fast response to changes in $CH_4$ supply. Steinle et al. (2017) reported that 70–95 % of dissolved $CH_4$ were effectively removed in the water column during summer stratification. Apart from MBI-driven uplift of $CH_4$-enriched bottom water to the surface (see below), wind-driven upwelling events can lead to a ventilation of the accumulated $CH_4$ to the atmosphere. For example, Gülzow et al. (2013) observed elevated $CH_4$ concentrations in the Gotland Basin as a result of wind-induced upwelling. The influence of upwelling at the BE time-series station, however, is more prominent due to the shallow water depth. In September 2012 and 2017, when upwelling occurred (see Sect. 4.1), sea-to-air $CH_4$ flux densities were 65.9 $\mu$mol $m^{-2}$ $d^{-1}$ and 132.3 $\mu$mol $m^{-2}$ $d^{-1}$, respectively, which were about 50 % and 200 % higher than the mean value.

Enhanced $CH_4$ saturations and associated emissions at the BE time-series station were also strongly promoted by saline inflows (see Sect. 4.2). We found very high surface $CH_4$ saturation and flux density in November 2013 and December 2014 (Fig. 6). In December 2014, surface $CH_4$ saturations were as high as 19,770 % and the calculated flux density reached 3104.5 $\mu$mol $m^{-2}$ $d^{-1}$. Inflows of saline waters usually occur in winter, when the well-ventilated water column, relatively low $CH_4$ oxidation rates and high wind speeds are all favorable for high $CH_4$ emissions (Wanninkhof, 2014; Steinle et al., 2017). Assuming that there was no continuous mixing or supply of $CH_4$ to the surface layer, it took about 3.3 days for the accumulated $CH_4$ to come back to equilibrium values under the calculated flux density, during which the annual $CH_4$ emissions from the Eckernförde Bay would increase by approximately 66 % in 2014. This is also in line with our speculation in Sect. 4.2 that the monthly sampling at the BE time-series station might have missed some of the short-lived $CH_4$ pulses. Moreover, methanogenesis in the sediments of the Eckernförde Bay is sufficient for $CH_4$ bubble formation (Whiticar, 2002). Hydrostatic pressure fluctuations associated with saline water inflow could have triggered $CH_4$ seepage and gas bubble plumes from the seafloor to the atmosphere (Wever et al., 2006; Lohrberg et al., 2020). Gas ebullition sites were usually found accompanied by pockmark structures (Schneider von Deimling et al., 2011) and Jackson et al. (1998) provided sonar evidences for $CH_4$ ebullition in the Eckernförde Bay. However, recently Lohrberg et al. (2020) reported a widespread $CH_4$ ebullition event in the Eckernförde Bay and found no direct linkage between pockmarks and ebullitions. They estimated the bubble-driven $CH_4$ flux during a weak storm in the fall of 2014 was 1916 $\mu$mol $m^{-2}$ $d^{-1}$. These findings point to the fact that ebullition might be an important, but highly variable, additional $CH_4$ efflux to the atmosphere. However, our measurements did not capture gas bubbles and, thus, the estimate of the overall $CH_4$ emissions resulting from the MBI might be too low. In this case, a time-series monitoring of saline inflows and sea level variations, combined with a continuous observation of $CH_4$ variability, especially in winter, are essential in quantifying $CH_4$ emissions from the Eckernförde Bay.

### 4.4 Comparison with other time-series measurements

Besides this study, time-series measurements of $CH_4$ have also been reported from Saanich Inlet (SI), British Columbia, Canada (Capelle et al., 2019) and ALOHA station in the North Pacific Subtropical Gyre (Wilson et al., 2017).

Located in a seasonally anoxic fjord, the time-series station in SI has a similar hydrographic setting compared to BE, but a deeper water depth (230 m, Capelle et al., 2019). Surface $CH_4$ saturations at SI fell in the lower end of the range observed here for BE (Fig. 7). Despite the fact that the mean surface saturation in SI was higher, $CH_4$ flux densities were much lower than at BE. Since the air-sea exchange approach of Nightingale et al. (2000) was used in both studies, the discrepancy is resulting from the higher wind speeds at BE. $CH_4$ saturations from ALOHA were only slightly supersaturated (close to the equilibrium saturation) and the flux densities were consequently low as well, which is resulting from the fact that ALOHA is a deep water (~4800 m) station located in the oligotrophic open ocean where potential strong $CH_4$ sources such as sedimentary release or methanogenesis under low $O_2$ in the water column are negligible (Wilson et al., 2017).

Wilson et al. (2017) analyzed the time-series $CH_4$ data from ALOHA during 2008–2016 and observed a decline in the surface $CH_4$ concentrations since 2013. They attributed the potential decrease in $CH_4$ production to fluctuations in phosphate concentrations. Capelle et al. (2019) also detected a significant decline of $CH_4$ concentrations in the upper water column over time at SI and proposed a link with the shoaling of the boundary of the hypoxic layer. However, no significant trend was detected in $CH_4$ concentrations or flux densities at the BE time-series station (Fig. 6), despite of the relatively long observation period. The different situations can be explained by the shallow water depth in the Eckernförde Bay, which makes the $CH_4$ distribution sensitive to the variability of its sedimentary release and events such as MBI and wind-driven upwelling.

## 5. Conclusions

The $CH_4$ measurements at the BE time-series station showed a strong temporal variability and variations with depths. A pronounced enhancement of the $CH_4$ concentrations was usually found in the bottom layer (15–25 m) during February, May–June and October which indicates that the release from the sediments is the major source of $CH_4$. Organic matter and dissolved $O_2$ are usually considered as the main controlling factors for $CH_4$ production and consumption pathways, but we did not detect correlations of $CH_4$ with Chlorophyll *a* or $O_2$ during 2006–2017.

Obviously non-biological processes such as local wind-driven-upwelling and the inflow of saline North Sea waters play a significant role for the observed variability of $CH_4$ at BE. However, these phenomena, which occur on relatively short time scales of day or weeks, were not frequently detected; most probably due to the monthly sampling frequency. The surface layer at BE was always supersaturated with $CH_4$ and therefore, BE was a persistent and strong, but highly variable, source of $CH_4$ to the atmosphere. We did not detect significant temporal trends in $CH_4$ concentrations or emissions, despite of ongoing environmental changes (warming, deoxygenation) in the Eckernförde Bay. Overall, the $CH_4$ variability at BE is driven by a complex interplay of various biological (i.e. methanogenesis, oxidation) and physical (i.e. upwelling, inflow events) processes. Continuous observations at the BE time-series station, with an emphasis on the period when upwelling and saline inflow usually occur is therefore, of great importance in quantifying $CH_4$ variability and the associated emissions as well as for predicting future $CH_4$ variability in the SW Baltic Sea.

***Data availability.*** Data are available from the Boknis Eck Database: https://www.bokniseck.de (Bange and Malien, 2020) and MEMENTO (the MarinE MethanE and NiTrous Oxide database, https://memento.geomar.de (Kock and Bange, 2015).

***Author contributions***. XM, MS, STL, and HWB designed the study and participated in the fieldwork. $CH_4$ measurements and data processing were done by XM, MS and STL. XM wrote the article with contributions from MS, STL and HWB.

***Competing interests***. The authors declare that they have no conflict of interest.

***Acknowledgements.*** The authors thank the captain and crew of the RV *Littorina* and *Polarfuchs* as well as many colleagues and numerous students who helped with the sampling and measurements of the BE time-series through various projects. Special thanks to A. Kock for her help with sampling, measurements and data analysis. The time-series at BE was supported by DWK Meeresforschung (1957–1975), HELCOM (1979–1995), BMBF (1995–1999), the Institut für Meereskunde (1999–2003), IfM-GEOMAR (2004–2011) and GEOMAR (2012–present). The current $CH_4$ measurements at BE are supported by the EU BONUS INTEGRAL project which receives funding from BONUS (Art 185), funded jointly by the EU, the German Federal Ministry of Education and Research, the Swedish Research Council Formas, the Academy of Finland, the Polish National Centre for Research and Development, and the Estonian Research Council. The Boknis Eck Time-Series Station (www.bokniseck.de) is run by the Chemical Oceanography Research Unit of GEOMAR, Helmholtz Centre for Ocean Research Kiel. The sea level data used in this study was made available by the EMODnet Physics project (www.emodnet-physics.eu/map), funded by the European Commission Directorate General for Maritime Affairs and Fisheries.

*Financial support*. Xiao Ma is grateful to the financial support provided by the China Scholarship Council (grant no. 201306330056) and the BONUS INTEGRAL project (grant no. 03F0773B).

## References

Ahmerkamp, S., Winter, C., Janssen, F., Kuypers, M. M., and Holtappels, M.: The impact of bedform migration on benthic oxygen fluxes, J. Geophys. Res.-Biogeo, 120, 2229–2242, https://doi.org/10.1002/2015JG003106, 2015.

Barnes, J., Ramesh, R., Purvaja, R., Nirmal Rajkumar, A., Senthil Kumar, B., Krithika, K., Ravichandran, K., Uher, G., and Upstill-Goddard, R.: Tidal dynamics and rainfall control $N_2O$ and $CH_4$ emissions from a pristine mangrove creek, Geophys. Res. Lett., 33, L15405, doi:15410.11029/12006GL026829, 2006.

Bange, H. W., Bartell, U. H., Rapsomanikis, S., and Andreae, M. O.: Methane in the Baltic and North Seas and a reassessment of the marine emissions of methane, Global Biogeochem. Cy., 8, 465–480, https://doi.org/10.1029/94GB02181, 1994.

Bange, H. W., Bergmann, K., Hansen, H. P., Kock, A., Koppe, R., Malien, F., and Ostrau, C.: Dissolved methane during hypoxic events at the Boknis Eck Time Series Station (Eckernförde Bay, SW Baltic Sea), Biogeosciences, 7, 1279–1284, https://doi.org/10.5194/bg-7-1279-2010, 2010.

Bange, H. W. and Malien, F.: Boknis Eck Time-series Database, Kiel Datamanagement Team, http://www.bokniseck.de/, last accessed: 23 March 2020.

Belkin, I. M.: Rapid warming of large marine ecosystems, Prog. Oceanogr., 81, 207–213, doi:10.1016/j.pocean.2009.04.011, 2009.

Bernard, B. B., Brooks, J. M., and Sackett, W. M.: Natural gas seepage in the Gulf of Mexico, Earth Planet Sc. Lett., 31, 48–54, 1976.

Borges, A. V., Champenois, W., Gypens, N., Delille, B., and Harlay, J.: Massive marine methane emissions from near-shore shallow coastal areas, Sci. Rep.-UK, 6, 27908, DOI: 10.1038/srep27908, 2016.

Bussmann, I., and Suess, E.: Groundwater seepage in Eckernförde Bay (Western Baltic Sea): Effect on methane and salinity distribution of the water column, Cont. Shelf Res., 18, 1795–1806, https://doi.org/10.1016/S0278-4343(98)00058-2, 1998.

Capelle, D. W., Hallam, S. J., and Tortell, P. D.: Time-series $CH_4$ measurements from Saanich Inlet, BC, a seasonally anoxic fjord, Mar. Chem., 215, 103664, https://doi.org/10.1016/j.marchem.2019.103664, 2019.

Carstensen, J., Andersen, J. H., Gustafsson, B. G., and Conley, D. J.: Deoxygenation of the Baltic Sea during the last century, P. Natl. Acad. Sci. USA, 111, 5628–5633, https://doi.org/10.1073/pnas.1323156111, 2014.

Conley, D. J., Carstensen, J., Aigars, J., Axe, P., Bonsdorff, E., Eremina, T., and Lannegren, C.: Hypoxia is increasing in the coastal zone of the Baltic Sea, Environ. Sci. Technol., 45, 6777–6783, doi: 10.1021/es201212r, 2011.

Dale, A. W., Flury, S., Fossing, H., Regnier, P., Røy, H., Scholze, C., and Jørgensen, B. B.: Kinetics of organic carbon mineralization and methane formation in marine sediments (Aarhus Bay, Denmark), Geochim. Cosmochim. Ac., 252, 159–178, https://doi.org/10.1016/j.gca.2019.02.033, 2019.

David, H. A.: Further applications of range to analysis of variance, Biometrika, 38, 393–409, 1951.

Egger, M., Riedinger, N., Mogollon, J. M., and Jørgensen, B. B.: Global diffusive fluxes of methane in marine sediments, Nat. Geosci., 11, 421–425, https://doi.org/10.1038/s41561-018-0122-8, 2018.

Fleming-Lehtinen, V., and Laamanen, M.: Long-term changes in Secchi depth and the role of phytoplankton in explaining light attenuation in the Baltic Sea, Estuar. Coast Shelf S., 102, 1–10, https://doi.org/10.1016/j.ecss.2012.02.015, 2012.

Grasshoff, K., Kremling, K., and Ehrhardt, M.: Methods of seawater analysis, 3rd edition, WILEY-VCH, Weihheim, Germany, 1999.

Gülzow, W., Rehder, G., Schneider von Deimling, J., Seifert, S., and Tóth, Z.: One year of continuous measurements constraining methane emissions from the Baltic Sea to the atmosphere using a ship of opportunity, Biogeosciences, 10, 81–99, doi:10.5194/bg-10-81-2013, 2013.

Hansen, H. P., Giesenhagen, H. C., and Behrends, G.: Seasonal and long-term control of bottom water oxygen deficiency in a stratified shallow-coastal system, ICES J. Mar. Sci., 56, 65–71, doi: 10.1006/jmsc.1999.0629, 1999.

HELCOM: State of the Baltic Sea - Second HELCOM holistic assessment 2011–2016, Baltic Sea Environ. Proc., 155, http://stateofthebalticsea.helcom.fi/, 2018.

Hovland, M., Judd, A. G., and Burke Jr., R. A.: The global flux of methane from shallow submarine sediments, Chemosphere, 26, 559–578, https://doi.org/10.1016/0045-6535(93)90442-8, 1993.

Hsu, S. A., Meindl, E. A., and Gilhousen, D. B.: Determining the power-law wind-profile exponent under near-neutral stability conditions at sea, J. Appl. Meteorol., 33, 757–765, https://doi.org/10.1175/1520-0450(1994)033<0757:DTPLWP>2.0.CO;2, 1994.

IPCC: Climate Change 2013: The physical science basis. Contribution of Working Group I to the fifth assessment report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, UK and New York, NY, 2013.

Jackson, D. R., Williams, K. L., Wever, T. F., Friedrichs, C. T., and Wright, L. D.: Sonar evidence for methane ebullition in Eckernförde Bay, Cont. Shelf Res., 18, 1893–1915, https://doi.org/10.1016/S0278-4343(98)00062-4, 1998.

Jähne, B., Heinz, G., and Dietrich, W.: Measurements of the diffusion coefficients of sparingly soluble gases in water, J. Geophys. Res.-Oceans, 92, 10767–10776, https://doi.org/10.1029/JC092iC10p10767, 1987.

Judd, A. G., Hovland, M., Dimitrov, L. I., Garcia Gil, S., and Jukes, V.: The geological methane budget at continental margins and its influence on climate change, Geofluids, 2, 109–126, https://doi.org/10.1046/j.1468-8123.2002.00027.x, 2002.

Klintzsch, T., Langer, G., Nehrke, G., Wieland, A., Lenhart, K., and Keppler, F.: Methane production by three widespread marine phytoplankton species: release rates, precursor compounds, and potential relevance for the environment, Biogeosciences, 16, 4129–4144, https://doi.org/10.5194/bg-16-4129-2019, 2019.

Kock, A. and Bange, H. W.: Counting the ocean's greenhouse gas emissions, Eos (Washington DC), 96, 10–13, https://doi.org/10.1029/2015EO023665, 2015.

Lenhart, K., Klintzsch, T., Langer, G., Nehrke, G., Bunge, M., Schnell, S., and Keppler, F.: Evidence for methane production by the marine algae Emiliania huxleyi, Biogeosciences, 13, 3163–3174, doi:10.5194/bg-13-3163-2016, 2016.

Lennartz, S. T., Lehmann, A., Herrford, J., Malien, F., Hansen, H. P., Biester, H., and Bange, H. W.: Long-term trends at the Boknis Eck time-series station (Baltic Sea), 1957–2013: does climate change counteract the decline in eutrophication? Biogeosciences, 11, 6323–6339, https://doi.org/10.5194/bg-11-6323-2014, 2014.

Levipan, H. A., Quiñones, R. A., Johansson, H. E., and Urrutia, H.: Methylotrophic methanogens in the water column of an upwelling zone with a strong oxygen gradient off central Chile, Microbes Environ., 22, 268–278, 2007.

Lohrberg, A., Schmale, O., Ostrovsky, I., Niemann, H., Held, P., and Schneider von Deimling, J.: Discovery and quantification of a widespread methane ebullition event in a coastal inlet (Baltic Sea) using a novel sonar strategy, Sci. Rep.-UK, 10, 4393, https://doi.org/10.1038/s41598-020-60283-0, 2020.

Maher, D. T., Cowley, K., Santos, I. R., Macklin, P., and Eyre, B. D.: Methane and carbon dioxide dynamics in a subtropical estuary over a diel cycle: Insights from automated in situ radioactive and stable isotope measurements, Mar. Chem., 168, 69–79, https://doi.org/10.1016/j.marchem.2014.10.017, 2015.

Maltby, J., Steinle, L., Löscher, C. R., Bange, H. W., Fischer, M. A., Schmidt, M., and Treude, T.: Microbial methanogenesis in the sulfate-reducing zone of sediments in the Eckernförde Bay, SW Baltic Sea, Biogeosciences, 15, 137–157, https://doi.org/10.5194/bg-15-137-2018, 2018.

Martens, C. S., Albert, D. B., and Alperin, M. J.: Stable isotope tracing of anaerobic methane oxidation in the gassy sediments of Eckernförde Bay, German Baltic Sea, Am. J. Sci., 299, 589–610, 1999.

Mohrholz, V., Naumann, M., Nausch, G., Krüger, S., and Gräwe, U.: Fresh oxygen for the Baltic Sea-An exceptional saline inflow after a decade of stagnation, J. Marine Syst., 148, 152–166, https://doi.org/10.1016/j.jmarsys.2015.03.005, 2015.

Naqvi, S.W.A., Bange, H.W., Farías, L., Monteiro, P.M.S., Scranton, M.I., and Zhang, J.: Marine hypoxia/anoxia as a source of $CH_4$ and $N_2O$, Biogeosciences, 7, 2159–2190, https://doi.org/10.5194/bg-7-2159-2010, 2010.

Nausch, G., Naumann, M., Umlauf, L., Mohrholz, V., and Siegel, H.: Hydrographisch-hydrochemische Zustandseinschätzung der Ostsee 2013, Leibniz-Institut für Ostseeforschung Warnemünde, Germany, 2014.

Nightingale, P. D., Malin, G., Law, C. S., Watson, A. J., Liss, P. S., Liddicoat, M. I., Boutin, J., and Upstill-Goddard, R. C.: In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers, Global Biogeochem. Cy., 14, 373–387, https://doi.org/10.1029/1999GB900091, 2000.

Nittrouer, C. A., Lopez, G. R., Wright, L. D., Bentley, S. J., D'Andrea, A. F., Friedrichs, C. T., Craig, N. I., and Sommerfield, C. K.: Oceanographic processes and the preservation of sedimentary structure in Eckernförde Bay, Baltic Sea. Cont. Shelf Res., 18, 1689–1714, https://doi.org/10.1016/S0278-4343(98)00054-5, 1998.

Orsi, T. H., Werner, F., Milkert, D., Anderson, A. L., and Bryant, W. R.: Environmental overview of Eckernförde bay, northern Germany, Geo-Mar. Lett., 16, 140–147, 1996.

Pimenov, N. V., Ul'yanova, M. O., Kanapatskii, T. A., Mitskevich, I. N., Rusanov, I. I., Sigalevich, P. A., Nemirovskaya, I. A., and Sivkov, V. V.: Sulfate reduction, methanogenesis, and methane oxidation in the upper sediments of the Vistula and Curonian Lagoons, Baltic Sea, Microbiology+, 82, 224–233, DOI: 10.1134/S0026261713020136, 2013.

Raymond, P. A. and Cole, J. J.: Gas exchange in rivers and estuaries: Choosing a gas transfer velocity, Estuaries, 24, 312–317, https://doi.org/10.2307/1352954, 2001.

Reeburgh, W. S.: Oceanic methane biogeochemistry, Chem. Rev., 107, 486–513, https://doi.org/10.1021/cr050362v, 2007.

Rehder, G., Keir, R. S., Suess, E., and Pohlmann, T.: The multiple sources and patterns of methane in North Sea waters. Aquat. Geochem., 4, 403–427, https://doi.org/10.1023/A:1009644600833, 1998.

Reindl, A. R., and Bolałek, J.: Methane flux from sediment into near-bottom water in the coastal area of the Puck Bay (Southern Baltic), Oceanol. Hydrobiol. St., 41, 40–47, https://doi.org/10.2478/s13545-012-0026-y, 2012.

Sandén, P., and Håkansson, B.: Long-term trends in Secchi depth in the Baltic Sea, Limnol. Oceanogr., 41, 346–351, https://doi.org/10.4319/lo.1996.41.2.0346, 1996.

Saunois, M., Bousquet, P., Poulter, B., Peregon, A., Ciais, P., Canadell, J. G., Dlugokencky, E. J., Etiope, G., Bastviken, D., Houweling, S., Janssens-Maenhout, G., Tubiello, F. N., Castaldi, S., Jackson, R. B., Alexe, M., Arora, V. K., Beerling, D. J., Bergamaschi, P., Blake, D. R., Brailsford, G., Brovkin, V., Bruhwiler, L., Crevoisier, C., Crill, P., Covey, K., Curry, C., Frankenberg, C., Gedney, N., Hoglund-Isaksson, L., Ishizawa, M., Ito, A., Joos, F., Kim, H. S., Kleinen, T., Krummel, P., Lamarque, J. F., Langenfelds, R., Locatelli, R., Machida, T., Maksyutov, S., McDonald, K. C., Marshall, J., Melton, J. R., Morino, I., Naik, V., O'Doherty, S., Parmentier, F. J. W., Patra, P. K., Peng, C. H., Peng, S. S., Peters, G. P., Pison, I., Prigent, C., Prinn, R., Ramonet, M., Riley, W. J., Saito, M., Santini, M., Schroeder, R., Simpson, I. J., Spahni, R., Steele, P., Takizawa, A., Thornton, B. F., Tian, H. Q., Tohjima, Y., Viovy, N., Voulgarakis, A., van Weele, M., van der Werf, G. R., Weiss, R., Wiedinmyer, C., Wilton, D. J., Wiltshire, A., Worthy, D., Wunch, D., Xu, X. Y., Yoshida, Y., Zhang, B. W., Zhang, Z., and Zhu, Q.: The global methane budget 2000-2012, Earth Syst. Sci. Data, 8, 697–751, doi:10.5194/essd-8-697-2016, 2016.

Schmale, O., Schneider von Deimling, J., Gülzow, W., Nausch, G., Waniek, J. J., and Rehder, G.: Distribution of methane in the water column of the Baltic Sea, Geophys. Res. Lett., 37, https://doi.org/10.1029/2010GL043115, 2010.

Schmaljohann, R.: Methane dynamics in the sediment and water column of Kiel Harbour (Baltic Sea), Mar. Ecol. Prog. Ser., 131, 263–273, doi:10.3354/meps131263, 1996.

Schneider von Deimling, J., Gülzow, W., Ulyanova, M., Klusek, Z., and Rehder, G.: Detection and monitoring of methane ebullition, BONUS-Baltic GAS Final report WP 3.3, 2011.

Scranton, M. I., and Farrington, J. W.: Methane production in the waters off Walvis Bay, J. Geophys. Res., 82, 4947–4953, https://doi.org/10.1029/JC082i031p04947, 1977.

Siedler, G., and Peters, H.: Properties of sea water: physical properties, in: Oceanography, edited by: Sündermann, J., Springer, Berlin, Germany, 233–264, 1986.

Smetacek, V.: The annual cycle of Kiel Bight plankton: A long-term analysis, Estuaries, 8, 145–157, 1985.

Smetacek, V., von Bodungen, B., von Bröckel, K., Knoppers, B., Martens, P., Peinert, R., Pollehne, F., Stegmann, P., and Zeitzschel, B.: Seasonality of plankton growth and sedimentation, in: Seawater-Sediment Interactions in Coastal Waters, edited by: Rumohr, J., Walger, E., and Zeitzschel, B., Springer, Berlin, Germany, 34–56, 1987.

Steinle, L., Maltby, J., Treude, T., Kock, A., Bange, H. W., Engbersen, N., Zopfi, J., Lehmann, M. F., Niemann, H.: Effects of low oxygen concentrations on aerobic methane oxidation in seasonally hypoxic coastal waters, Biogeosciences, 14, 1631–1645, https://doi.org/10.5194/bg-14-1631-2017, 2017.

Sturm, K., Werner, U., Grinham, A., and Yuan, Z.: Tidal variability in methane and nitrous oxide emissions along a subtropical estuarine gradient, Estuar. Coast Shelf S., 192, 159–169, https://doi.org/10.1016/j.ecss.2017.04.027, 2017.

Treude, T., Krüger, M., Boetius, A., and Jørgensen, B. B.: Environmental control on anaerobic oxidation of methane in the gassy sediments of Eckernförde Bay (German Baltic), Limnol. Oceanogr., 50, 1771–1786, https://doi.org/10.4319/lo.2005.50.6.1771, 2005.

Upstill-Goddard, R. C., Barnes, J., Frost, T., Punshon, S., and Owens, N. J.: Methane in the southern North Sea: Low-salinity inputs, estuarine removal, and atmospheric flux, Global Biogeochem. Cy., 14, 1205–1217, https://doi.org/10.1029/1999GB001236, 2000.

Wanninkhof, R.: Relationship between wind speed and gas exchange over the ocean revisited, Limnol. Oceanogr. : Method., 12, 351–362, https://doi.org/10.4319/lom.2014.12.351, 2014.

Weber, T., Wiseman, N. A., and Kock, A.: Global ocean methane emissions dominated by shallow coastal waters, Nat. Commun., 10, 4584, https://doi.org/10.1038/s41467-019-12541-7, 2019.

Wever, T. F., Lühder, R., Voβ, H., and Knispel, U.: Potential environmental control of free shallow gas in the seafloor of Eckernförde Bay, Germany, Mar. Geol., 225, 1-4, https://doi.org/10.1016/j.margeo.2005.08.005, 2006.

Whiticar, M. J.: Diagenetic relationships of methanogenesis, nutrients, acoustic turbidity, pockmarks and freshwater seepages in Eckernförde Bay, Mar. Geol., 182, 29–53, https://doi.org/10.1016/S0025-3227(01)00227-4, 2002.

Wiesenburg, D. A., and Guinasso Jr, N. L.: Equilibrium solubilities of methane, carbon monoxide, and hydrogen in water and sea water, J. Chem. Eng. Data, 24, 356-360, 1979.

Wilson, S. T., Ferrón, S., and Karl, D. M.: Interannual variability of methane and nitrous oxide in the North Pacific Subtropical Gyre, Geophys. Res. Lett., 44, 9885–9892, https://doi.org/10.1002/2017GL074458, 2017.

WMO: Scientific assessment of ozone depletion: 2018, World Meteorological Organization, Geneva, Switzerland, Global Ozone Research and Monitoring Project–Report No. 58, 588 pp., 2018.

Xiao, K. Q., Beulig, F., Roy, H., Jørgensen, B. B., and Risgaard-Petersen, N.: Methylotrophic methanogenesis fuels cryptic methane cycling in marine surface sediment, Limnol. Oceanogr., 63, 1519-1527, https://doi.org/10.1002/lno.10788, 2018.

Table 1. Annual mean (arithmetic average ±standard deviation) of water temperature, salinity, wind speed and dissolved $CH_4$ concentrations at the BE time-series station during 2006–2017. Water temperatures, salinity and $CH_4$ concentrations were averaged over the water column (0–25 m). Wind speeds were recorded at Kiel lighthouse.

| year | Temperature (°C) | Salinity | Wind speed ($u_{10}$, m $s^{-1}$) | $CH_4$ (nM) |
|---|---|---|---|---|
| 2006 | 9.19±5.75 | 20.14±3.11 | 7.5±2.6 | 39.3±38.1 |
| 2007 | 9.68±4.55 | 17.78±2.14 | 7.5±2.5 | 44.9±45.5 |
| 2008 | 10.11±4.20 | 19.14±3.43 | 6.2±2.1 | 36.9±41.9 |
| 2009 | 9.20±4.81 | 18.36±2.22 | 7.3±2.3 | 27.8±26.2 |
| 2010 | 8.47±5.20 | 17.80±3.22 | 5.5±2.7 | 34.8±39.3 |
| 2011 | 8.74±5.16 | 19.14±2.78 | 6.8±3.1 | 36.9±29.1 |
| 2012 | 9.47±3.89 | 18.67±2.63 | 8.7±2.1 | 46.4±44.3 |
| 2013 | 9.04±5.45 | 17.89±3.74 | 5.9±2.8 | 67.7±83.1 |
| 2014 | 10.38±4.93 | 19.17±2.79 | 7.0±3.3 | 101.4±183.3 |
| 2015 | 9.19±4.28 | 19.71±3.30 | 6.1±2.8 | 35.7±36.3 |
| 2016 | 10.09±4.71 | 18.80±3.19 | 5.9±1.7 | 52.6±111.4 |
| 2017 | 10.21±4.86 | 19.50±2.11 | 6.8±2.4 | 30.5±22.9 |

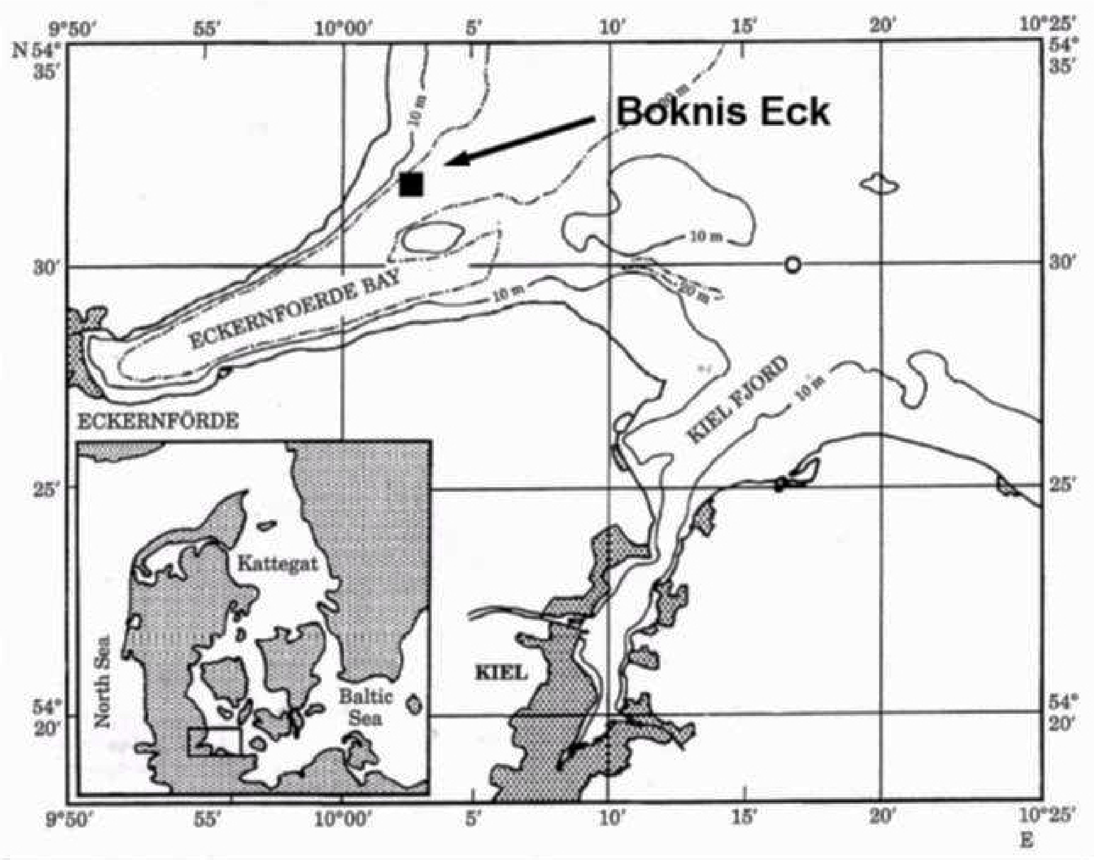
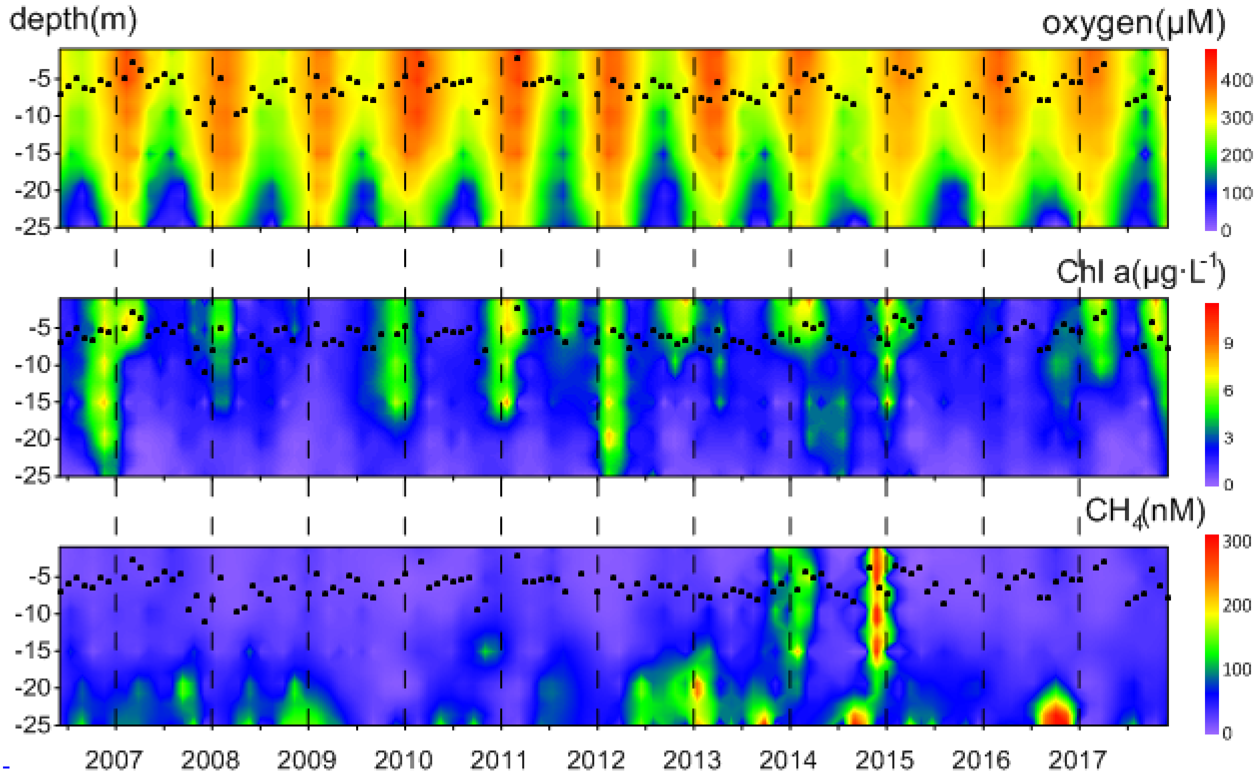


**Fig. 1 Location (black square) of the Boknis Eck time-series station in the Eckernförde Bay, southwestern Baltic Sea. (from Hansen et al., 1999)**


depth(m)
oxygen(μM)
Chl a(μg·L-1)
CH4(nM)
-5
-10
-15
-20
-25
400
300
200
100
0
9
6
3
0
300
200
100
0
2007
2008
2009
2010
2011
2012
2013
2014
2015
2016
2017

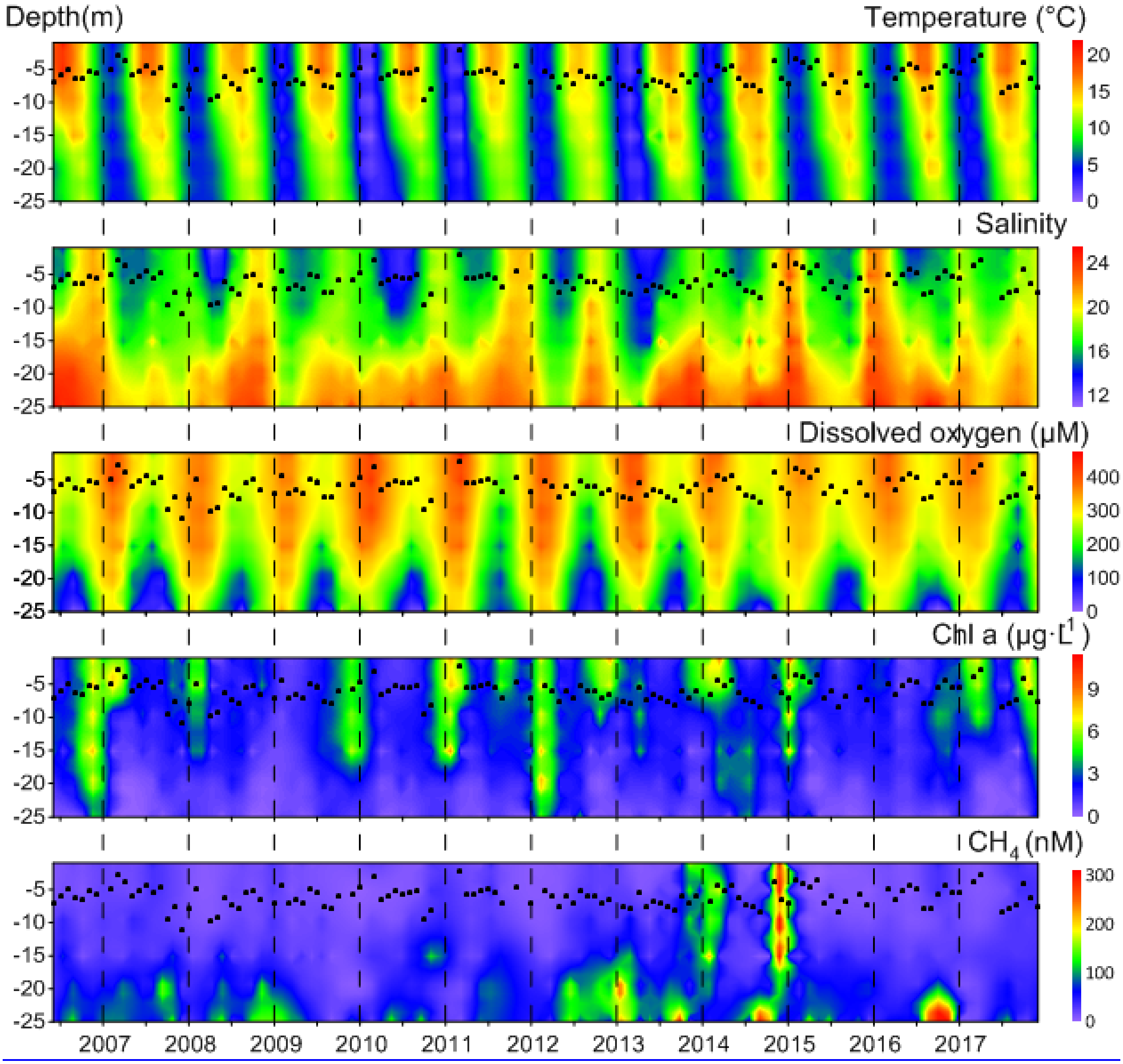
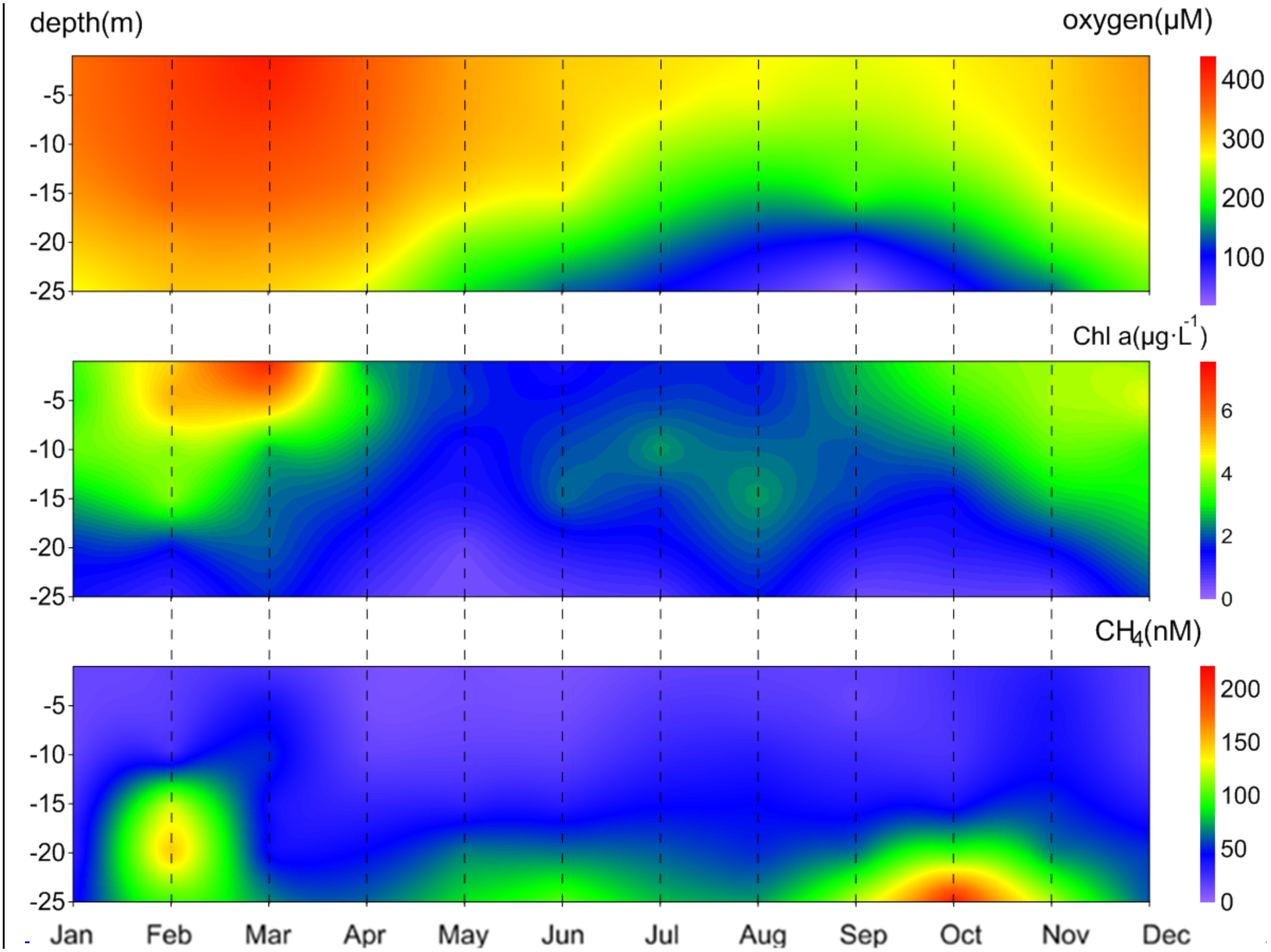


**Fig 2. Distributions of temperature, salinity, dissolved $O_2$, Chlorophyll *a* and $CH_4$ at the BE time-series station during 2006–2017. Black dots indicate the monthly measurements of Secchi depth. To get a better visualization, the maximum color bar for $CH_4$ concentration is 300 nM, but some of the actual concentrations are higher (for example, in December 2014 and in autumn 2016).**


depth(m)
oxygen(μM)
400
300
200
100
Chl a(μg·L$^{-1}$)
6
4
2
0
CH$_4$(nM)
200
150
100
50
0
-5
-10
-15
-20
-25
Jan
Feb
Mar
Apr
May
Jun
Jul
Aug
Sep
Oct
Nov
Dec

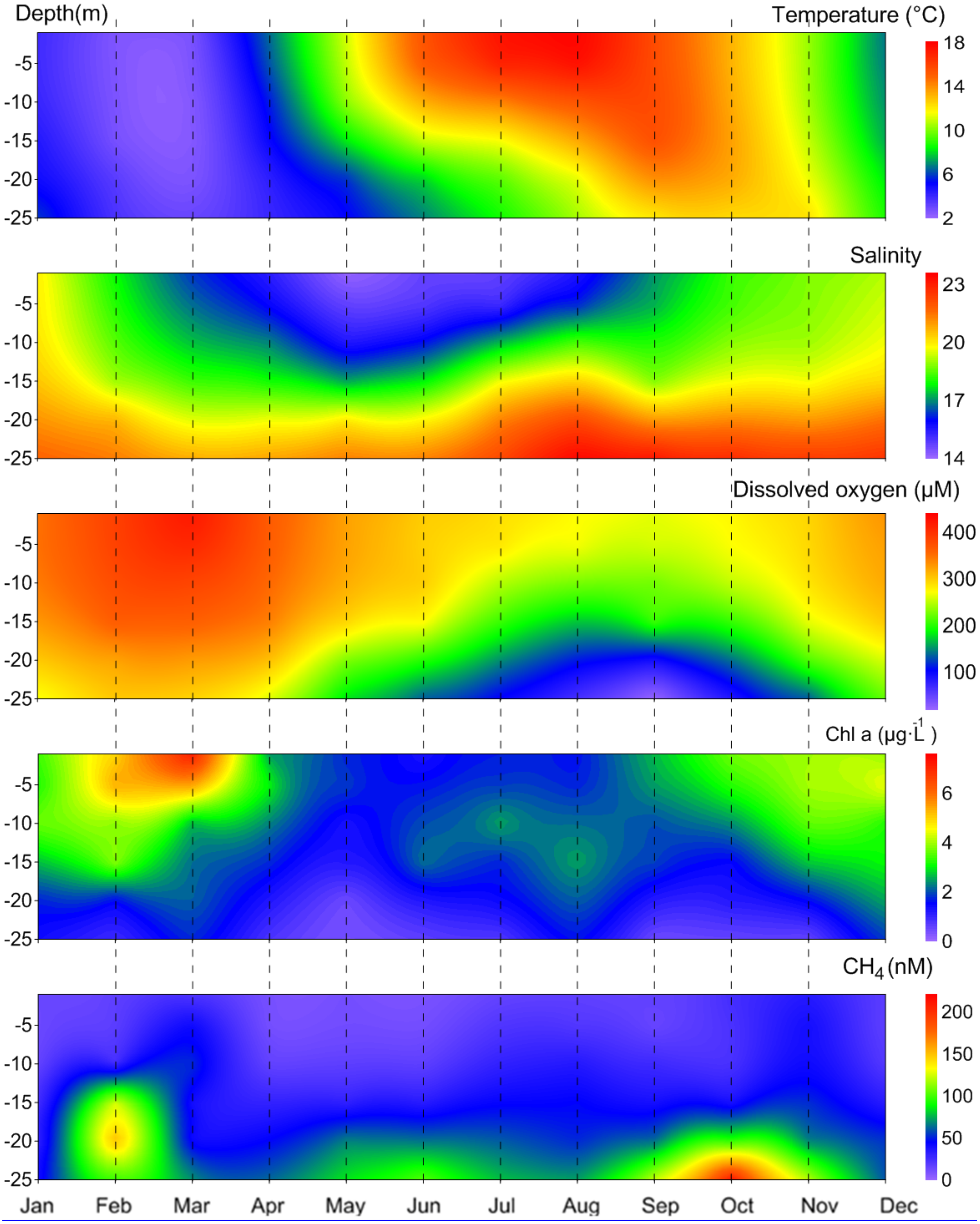
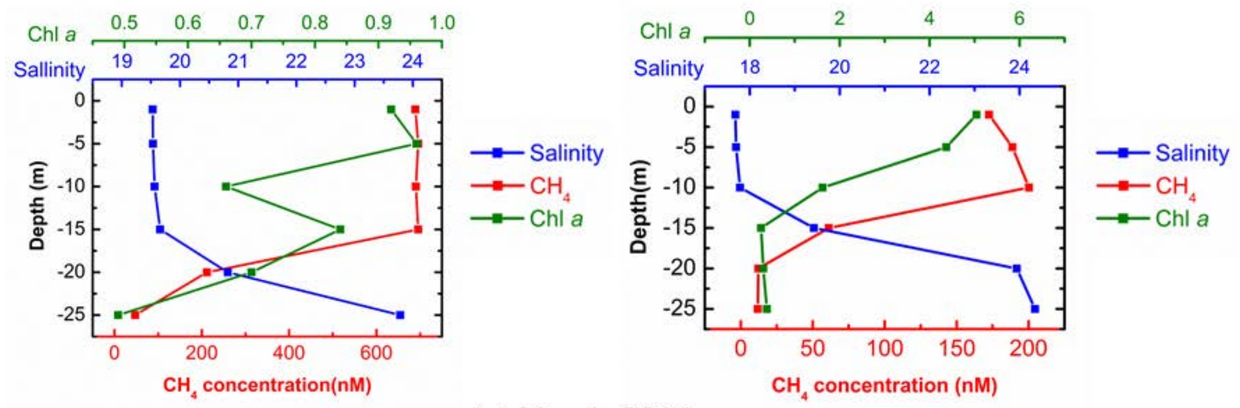

Depth(m)
Temperature (°C)
18
14
10
6
2
Salinity
23
20
17
14
Dissolved oxygen (μM)
400
300
200
100
Chl a (μg·L-1 )
6
4
2
0
CH4 (nM)
200
150
100
50
0
-5
-10
-15
-20
-25
Jan
Feb
Mar
Apr
May
Jun
Jul
Aug
Sep
Oct
Nov
Dec

**Fig 3. Mean seasonal variations of temperature, salinity, dissolved $O_2$, Chlorophyll *a* and $CH_4$ at the BE time-series station during 2006–2017. $CH_4$ concentrations in December 2014 were excluded in plotting.**

(a) December 2014 (b) November 2013



(c) March 2014

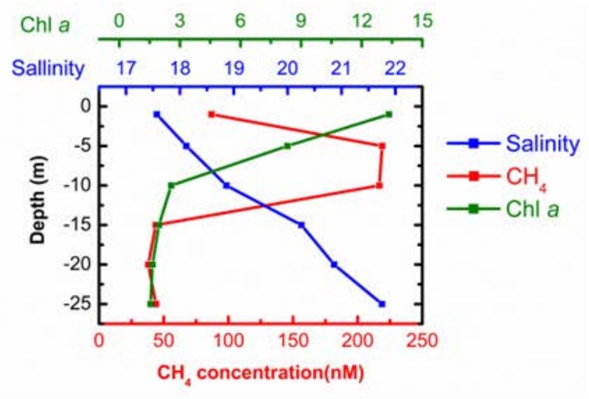
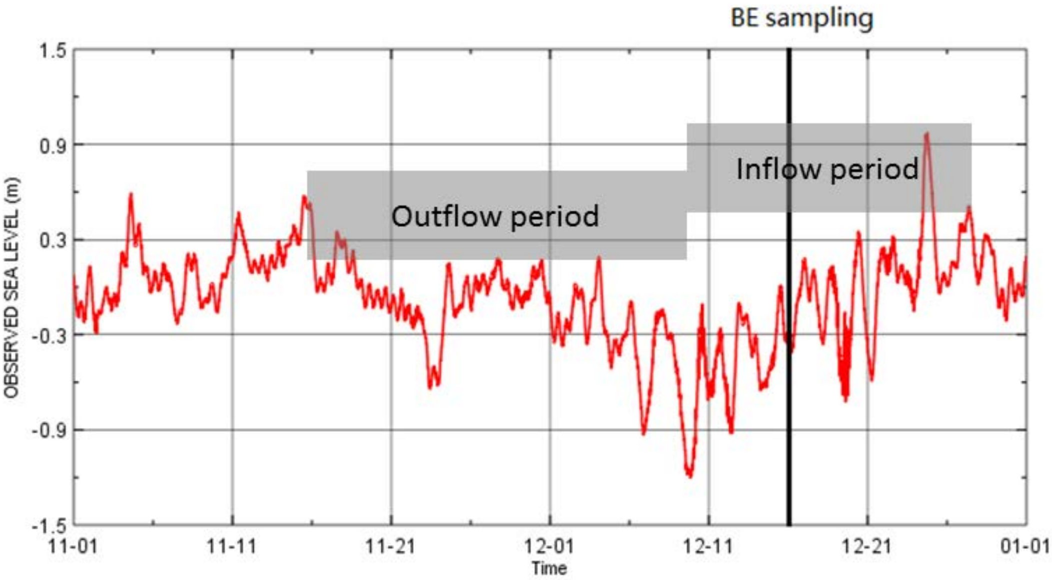


**Fig 4. Vertical distribution of Chlorophyll *a*, salinity and $CH_4$ concentrations in the water column in December 2014 (a), November 2013 (b) and March 2014 (c).**



525

**Fig 5. Sea level variations in November and December, 2014. The black line indicates the occurrence of BE sampling in December 2014.**

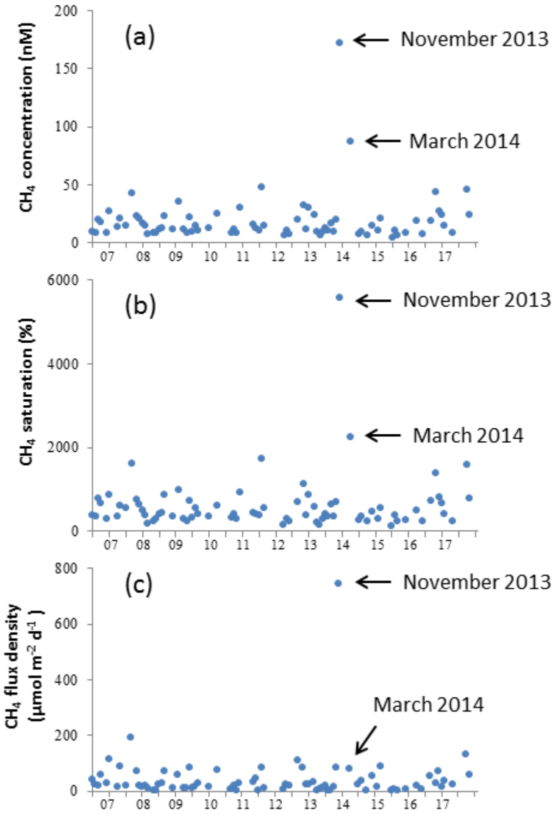


**Fig 6. Inter-annual variations of dissolved CH$_4$ concentration (a), saturation (b) and flux density (c) at the BE time-series station during 2006–2017. Data collected from December 2014 were not shown.** 530

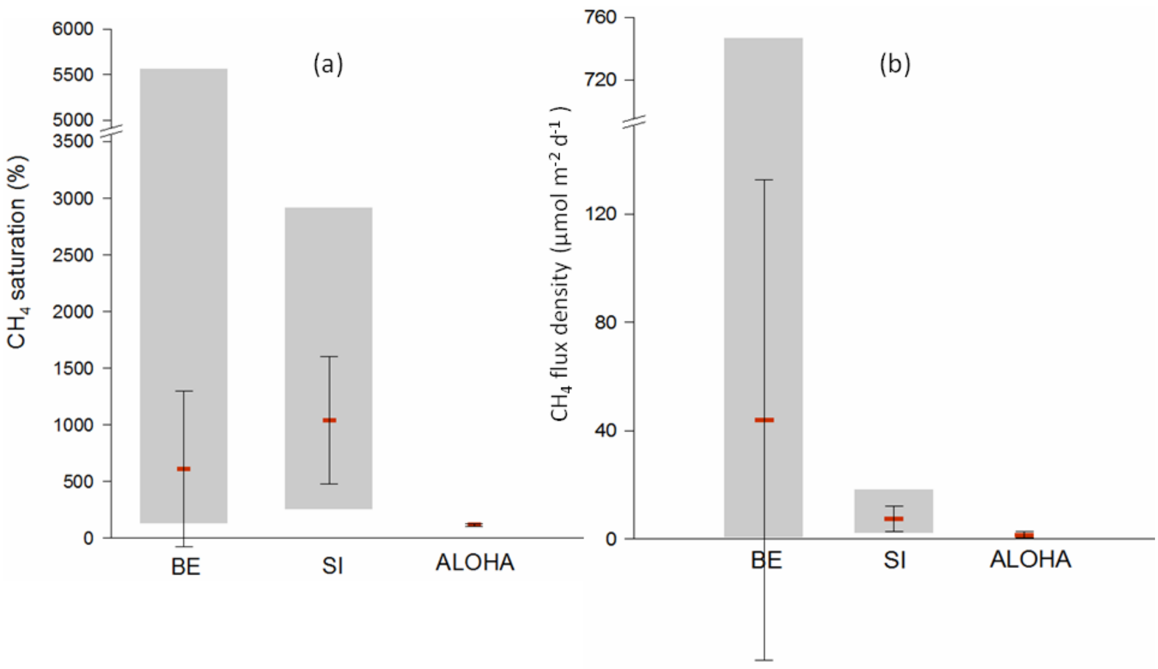


**Fig 7. Comparison of surface $CH_4$ saturations (a) and flux densities (b) from time-series stations of BE, Saanich Inlet (SI) and ALOHA. For the computation of flux density, the equations of Nightingale et al. (2000) and Wanninkhof (2014) were used for SI and ALOHA, respectively. Data in December 2014 at the BE time-series station were not included. Please note the break on the y axis for both charts.**