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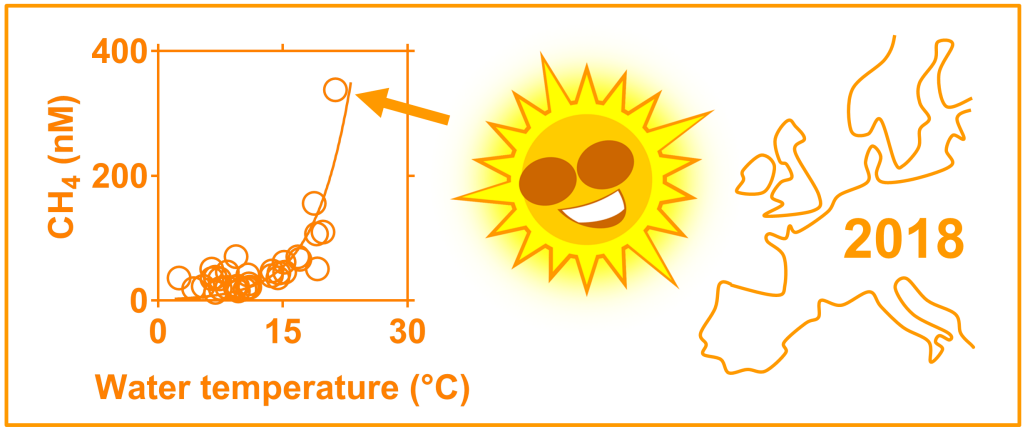

# Response of marine methane dissolved concentrations and emissions in the Southern North Sea to the European 2018 heatwave

Borges A.V.[1,*], C. Royer[1,2], J. Lapeyra Martin[2], W. Champenois[1], N. Gypens[2]

[1] Université de Liège, Unité d'Océanographie Chimique, Institut de Physique (B5), B-4000, Belgium

[2] Université Libre de Bruxelles, Laboratoire d'Ecologie des Systèmes Aquatiques, CP221, Boulevard du Triomphe, B-1050, Belgium

* corresponding author: alberto.borges@uliege.be

## Abstract

During the European heatwave of 2018 that led to record-breaking temperatures in many countries across northern and central Europe, average seawater temperature in July was 2.5°C higher than the mean from 2004 to 2017 for same month in the Belgian coastal zone (BCZ) (Southern Bight of the North Sea). The mean dissolved $CH_4$ concentration in surface waters in July 2018 (338 nmol $L^{-1}$) was three times higher than in July 2016 (110 nmol $L^{-1}$), and an extremely high dissolved $CH_4$ concentration in surface waters (1,607 nmol $L^{-1}$) was observed at one near-shore station. The high dissolved $CH_4$ concentrations in surface waters in the BCZ in July 2018 seemed to be due to a combination of enhancement of methanogenesis and of release of $CH_4$ from gassy sediments, both most likely related to warmer conditions. The emission of $CH_4$ from the BCZ to the atmosphere was higher in 2018 compared to 2016 by 57% in July (599 versus 382 µmol $m^{-2}$ $d^{-1}$) and by 37% at annual scale (221 versus 161 µmol $m^{-2}$ $d^{-1}$). The European heatwave of 2018 seems to have led to a major increase of $CH_4$ concentrations in surface waters and $CH_4$ emissions to the atmosphere in the BCZ.

**Keywords:** European 2018 Heatwave; North Sea; Belgian coastal zone; Methane

## 1. Introduction

Methane ($CH_4$) is the second most important long-lived anthropogenic greenhouse gas (GHG) after $CO_2$ (IPCC 2013), and has numerous anthropogenic and natural sources and sinks in the three major Earth compartments (atmosphere, land and ocean) (Saunois et al. 2016). The open ocean is a very small source of $CH_4$ to the atmosphere (≤2 $TgCH_4$ $yr^{-1}$, Rhee et al. 2009; Weber et al. 2019) compared to other natural $CH_4$ sources (~220 $TgCH_4$ $yr^{-1}$) dominated by wetlands and to anthropogenic $CH_4$ sources (~350 $TgCH_4$ $yr^{-1}$) dominated by agricultural food production (cattle and rice paddies) (Saunois et al. 2016). The origin of $CH_4$ in surface waters of the open ocean is elusive, coined the "ocean $CH_4$ paradox", possibly related to several processes that are probably variable from one system to another such as the transformation of methylated molecules such as dimethylsulfoniopropionate or methylphosphanate (Karl et al. 2008; Florez-Leiva et al. 2013), production of $CH_4$ by phytoplankton itself (Lenhart et al. 2016), or methanogenesis in the guts of some species of copepods (Stawiarski et al. 2019). Coastal waters, and in particular estuaries, are more intense sources of $CH_4$ to the atmosphere (~10 $TgCH_4$ $yr^{-1}$) than open oceanic waters (Bange et al. 1994; Middelburg et al. 2002; Borges and Abril 2011; Upstill-Goddard and Barnes 2016). The $CH_4$ emissions from coastal waters are sustained by methanogenesis in sediments fueled by high organic matter deposition and in some regions by natural gas seeps, mud volcanoes or $CH_4$ hydrates (Dimitrov, 2002; Damm and Budeús 2003; Mau et al. 2007; Malakhova et al. 2010; Shakhova et al. 2010). In coastal waters, the largest source of $CH_4$ seems to be sedimentary and leads to an enrichment of $CH_4$ in bottom waters, the fate of which depends on water column depth. Concentrations of $CH_4$ in surface waters and related $CH_4$ emissions to the atmosphere are higher in shallow regions of the continental shelf (Borges et al. 2016), and, in deeper areas, $CH_4$ in bottom waters is dispersed by lateral transport (advection or turbulent mixing) or removed by microbial oxidation, and is transported very slowly across the thermocline to surface waters (Schneider von Deimling et al. 2011; Mau et al. 2015; Graves et al. 2015). Overall, this leads to a negative relation between $CH_4$ concentration and depth (in absolute values) both locally (Borges et al. 2016; 2018) and globally (Weber et al. 2019).The future evolution of $CH_4$ emissions from coastal waters in response to warming and eutrophication (and related

expansion of hypoxia) remains largely unconstrained and unquantified (Naqvi et al. 2010).

The response to natural oscillations (e.g. El Niño-Southern Oscillation, North Atlantic Oscillation) or extreme weather events (e.g. heatwave, very mild winter) can be used as a natural laboratory to determine how marine ecosystems might respond to climate change (e.g. warming, increased stratification, local change of wind intensity) (e.g. Le Quéré et al. 2002; Champenois and Borges 2012, 2019). Furthermore, heatwave events are predicted to increase in frequency and magnitude as a consequence of global warming (Frölicher et al. 2018). Heatwaves have been shown to affect marine ecosystems worldwide, leading to mortality of some organisms, out-of-range species migrations, or outbreaks of undesirable organisms, in pelagic communities (McCabe et al. 2016; Cavole et al. 2016; Oliver et al. 2017), seagrass beds (Marbà and Duarte 2010; Arias-Ortiz et al. 2018), coral reefs and reef-associated communities (Wernberg et al. 2013; Hughes et al. 2017), kelp forests (Wernberg et al. 2016), and rocky benthic communities (Garrabou et al. 2009). The effect of heatwaves on the marine sources and sinks of greenhouse gases such as $CH_4$ and on its emission to atmosphere have not been documented so far, to our best knowledge.

Borges et al. (2016; 2018) reported the distribution and seasonal variability of dissolved $CH_4$ in the Belgian coastal zone (BCZ) and hypothesized that warming would increase $CH_4$ concentrations in surface waters. This hypothesis builds on the fact that there are large quantities of organic matter from former peatlands (dating from the last glacial period) in near-shore sediments and the presence of gassy sediments, most probably pockets of $CH_4$ (Missiaen et al. 2002). Furthermore, the BCZ is an area of important deposition of sediment and organic matter compared to the rest of the North Sea (de Haas and van Weering 1997), consequently near-shore sediments are muddy and rich in organic matter (Braeckman et al. 2014). Warming could stimulate the release of $CH_4$ from sediments either due to enhanced methanogenesis (mainly limited by temperature, given the high stock of sedimentary organic matter) and/or release of $CH_4$ from gassy sediments (ebullition/flaring). Furthermore, the water column in the BCZ is permanently well-mixed (no summer thermal stratification), so that inputs of $CH_4$ from sediments are efficiently mixed to surface waters; conversely, warming of surface waters propagates to an equivalent warming of surface sediments. The heat should then further propagate deeper into

the sediment by diffusion and advection, depending on sediment porosity (Goto and Matsubayashi 2009) and by pore-water pumping by surface waves (Savidge et al. 2016).

In the present study, we report dissolved $CH_4$ concentrations in surface waters at nine fixed stations in the BCZ during 2018 (Fig. 1) that are compared to an equivalent data-set obtained in 2016. We compare the data-sets obtained with the same analytical methodology in 2016 and 2018 at the same stations and using the same temporal resolution (monthly), to check for inter-annual changes. In particular, we investigate the response of marine $CH_4$ concentration to the European 2018 heatwave (WMO 2018), in order to test the above mentioned hypothesis of enhancement of dissolved $CH_4$ concentrations in response to warming. The European 2018 heatwave led to record-breaking temperatures in many countries across northern and central Europe, and according to the European Center for Medium Weather Forecast, near-surface air temperature anomaly in Europe in the period of April to August, calculated with respect to the 1981-2010 average for those months, was nearly 2°C in 2018, much larger than in any previous year since 1979 (Magnusson et al. 2018).

## 2. Material and methods

Data were collected in the BCZ on board the *RV Simon Stevin* at nine fixed stations (Fig. 1) in 2017, 2018 and 2019 during 23 cruises of one or two days duration (28/29-03-17; 26-04-17; 23/24-05-17; 05-03-18; 21/22-03-18; 04/04-04-18; 24/25-04-18; 02/03-05-18; 29/30-05-18; 25/26-07-18; 27/29-08-18; 25/27-09-18; 24/25-10-18; 21/22-11-18; 18-12-18; 29/30-01-19; 20/21-02-19; 26/27-03-2019; 08/08-04-2019; 23/24-04-2019; 06/08-05-2019; 20/21-05-2019; 04/05-06-2019). Sampling was carried out in surface waters (3 m depth) with a 4L Niskin bottle mounted on a six bottle rosette connected to a conductivity-temperature-depth (CTD) probe (Sea-bird SBE25). Sampling was only made in surface waters because we previously showed that there are no major vertical gradients (between surface and bottom waters) of salinity, temperature and $CH_4$ in the BCZ which is a permanently well-mixed area due to its shallowness and strong tidal currents (Borges et al. 2016). Duplicate water samples for the determination of dissolved $CH_4$ concentration were collected in borosilicate serum bottles (50 mL) with silicone tubing, left to overflow,

poisoned with a saturated solution of $HgCl_2$ (200 µL), sealed with a butyl stopper and crimped with an aluminum cap. The concentration of $CH_4$ was determined with the headspace equilibration technique and a gas chromatograph (GC) equipped with a flame ionization detector (SRI 8610C) calibrated with $CH_4$:$CO_2$:$N_2O$:$N_2$ mixtures (Air Liquide Belgium) of 1, 10 and 30 µatm $CH_4$. The method is described in detail by Borges et al. (2018), and was inter-calibrated with other laboratories in the first large-scale international inter-calibration of marine $CH_4$ and $N_2O$ measurements (Wilson et al. 2018). Precision was about ±3% for $CH_4$ based on analysis of 159 duplicate samples. The air-sea flux of $CH_4$ ($F_{CH4}$) was computed using the gas transfer velocity parameterization as a function of wind speed of Nightingale et al. (2000), and the Schmidt number of $CH_4$ in seawater computed from temperature according to Wanninkhof (1992), using daily wind speed data from the Westhinder platform (2.4378°E 51.3883°N) acquired by the Meetnet Vlaamse Banken (MVB) and retrieved from the Vlaams Instituut voor de Zee (VLIZ) data-center (http://www.vliz.be/en/measurement-network-flemish-banks). We used a constant atmospheric value of the partial pressure of $CH_4$ ($pCH_4$) of 1.9 µatm. The typical variability of atmospheric $pCH_4$ of ±0.2 ppm leads to a small error in the computation of $F_{CH4}$, on average of <±0.5% for our data-set because the observed dissolved $CH_4$ concentrations in the BCZ were always distinctly above the saturation value.

Samples for the determination of chlorophyll-*a* (Chl-*a*) were filtered on Whatman GF/F glass fiber filters (47 mm diameter) and stored frozen (-20°C). The concentration of Chl-*a* was determined on acetone (90%) extracts by fluorimetry (Holm-Hansen et al. 1965) using a Kontron SFM25 fluorimeter. Samples for the determination of total suspended matter (TSM) were filtered on pre-weighted Whatman GF/F glass fiber filters (47 mm diameter) and data retrieved from the VLIZ data-center (http://www.vliz.be/en/lifewatch-0).

Hourly seawater temperature data from 2004 to 2018 were acquired at the two platforms (Westhinder (2.439°E 51.389°N) and Wandelaar (3.047°E 51.395°N)) by the MVB and retrieved from the VLIZ data-center (http://www.vliz.be/en/measurement-network-flemish-banks). Seawater temperature from the platforms compared satisfactorily with the CTD measurements from the cruises (Fig. S1). Daily average air temperatures were acquired at the Oostende airport (2.870°E 51.204°N, < 1km from the seashore) and were retrieved from Weather Underground data-base (https://www.wunderground.com/).

Statistical tests were carried out at 0.05 level, using GraphPad Prism® software. Normality of the distribution was tested with the D'Agostino-Pearson omnibus normality test, and differences were tested with the Wilcoxon matched-pairs signed rank test.

The georeferenced and timestamped data-set from 2017-2019 of dissolved $CH_4$ concentration, salinity, water temperature and Chl-*a* is publically available (Borges and Gypens 2019). The data-set from 2016 is also publically available as a Supplemental File of Borges et al. (2018).

## 3. Results and Discussion

Seawater temperature in the BCZ in July-August 2018 was exceptionally high compared to the last 13 years (Fig. 2). The monthly average of seawater temperature in the BCZ in July 2018 (20.4°C) was 2.5°C higher than the mean from 2004 to 2017 in July (17.9°C), and the monthly average of seawater temperature in August 2018 (21.0°C) was 2.3°C higher than the mean from 2004 to 2017 in August (18.8°C). The monthly average seawater temperature in 2018 was higher by 1.5°C in July and by 1.4°C in August compared to 2016. Monthly air temperature in Oostende in July 2018 (19.0°C) was 2.3°C higher than the mean from 2004 to 2017 in July (16.7°C). The monthly average air temperature in 2018 was higher by 2.2°C in July and by 0.1°C in August compared to 2016. The time-series of air temperature since 2004 shows that two other heatwaves occurred in August 2004 (18.7°C) and July 2006 (20.2°C), but there was not a corresponding increase of seawater temperature, probably reflecting less extensive and more local heatwaves, and/or of shorter duration.

The mean seawater temperature measured at the nine fixed sampling stations (Fig. 1) was 1.5°C higher in July 2018 (21.3°C) than in July 2016 (19.8°C) (Fig.3). Higher seawater temperatures were observed at all nine stations, with differences ranging between 0.7°C at stations 700 and 780 and 2.3°C at station 215 and ZG02 (Fig. S2). There was an onshore-offshore gradient of seawater temperature in July 2018, with lower temperatures offshore (20.8°C on average for stations ZG02, 330 and 780) than onshore (22.0°C on average for stations 120, 130 and 700) (Fig. S2).

Summer-time $CH_4$ concentrations (July-August) were significantly higher in 2018 than 2016 (Wilcoxon matched-pairs signed rank test $p$=0.0432<0.05). The

mean $CH_4$ concentration in surface waters in July 2018 (338 nmol $L^{-1}$) was three times higher than in July 2016 (110 nmol $L^{-1}$) (Fig. 3). Higher $CH_4$ concentrations in July 2018 than July 2016 were observed at seven of the nine individual stations (Fig. 4 and S3). The differences of $CH_4$ concentrations in July 2018 compared to July 2016 were particularly marked at stations 130 and 700, with extreme $CH_4$ concentrations of 1,074 and 1,607 nmol $L^{-1}$, respectively. Such dissolved $CH_4$ concentration values are much higher than those typically reported in estuarine polyhaline regions, as maximum $CH_4$ concentrations are observed mostly in the oligohaline estuarine regions with values typically < 500 nmol $L^{-1}$ (Borges and Abril 2011). These dissolved $CH_4$ concentration values were higher than any other previous report in natural surface waters of the North Sea, and equivalent to the maximum concentration reported above an abandoned borehole in the Northern North Sea ($CH_4$ concentration of 1,453 nmol $L^{-1}$, Rehder et al. 1998). Such high dissolved $CH_4$ concentrations are extremely uncommon in continental shelves in general (typically $CH_4 \leq 10$ nmol $L^{-1}$) (e.g. Bange et al. 1994; Bange 2006; Weber et al. 2019), and are only observed in areas of shallow and intense gas seeps such as near Coal Oil Point in Santa Barbara Channel (Mau et al. 2007) or the bays in the Black Sea around Sevastopol (Malakhova et al. 2010). The dissolved $CH_4$ concentrations in the BCZ in late winter and spring (January-May) were similar in 2016, 2017, 2018 and 2019 (Fig. 3), even if the late winter (January-March) seawater temperatures were particularly low in 2018 (Figs. 2 and 3). The dissolved $CH_4$ concentrations in the BCZ in early winter (October-December) were also similar as those in 2016 and 2017 for the monthly averages of all stations (Fig. 3). At station 130, the shallowest among the nine sampled stations, high values of dissolved $CH_4$ concentrations were observed in September 2016 and November 2018 (Fig. 4) possibly to transient increases in relation sediment resuspension due to autumn storms. Indeed, in November 2018, TSM values were higher in the most coastal stations (700, 130, 120) (50.0±19.0 mg $L^{-1}$) than the most off-shore stations (ZG02, 330, 780) (13.0±10.4 mg $L^{-1}$), indicative of vigorous sediment resuspension, and the highest TSM value among the nine sampled stations was at stations 130 (69.0 mg $L^{-1}$). In conclusion, the major difference among years of dissolved $CH_4$ concentrations in the BCZ was observed in July 2018 compared to 2016, except for small-scale variations in autumn due sediment resuspension at the shallowest station (130).

The concentration of $CH_4$ in estuarine environments depends on the balance of source terms (riverine and lateral inputs and sedimentary fluxes) and loss terms (emission to the atmosphere and microbial oxidation). Salinity values in July 2018 were lower in most stations than in July 2016, but values were within the range typically observed at each station (Fig. S4). However, at station 700 where the highest $CH_4$ concentration was observed, salinity was higher in July 2018 than in July 2016. Furthermore, Borges et al. (2018) showed using a simple model that $CH_4$ brought from the Scheldt estuary is rapidly lost during transport in the BCZ mainly due to emission to the atmosphere, and that inputs of $CH_4$ from the Scheldt do not contribute significantly to the observed high $CH_4$ values in the BCZ. We conclude that the difference in dissolved $CH_4$ concentration in the BCZ between July 2018 and 2016 was unrelated to differences in estuarine inputs of $CH_4$ from the Scheldt.

Borges et al. (2018) showed that $CH_4$ at the different stations of the BCZ behaved differently to inputs of phytoplankton organic matter and to seasonal temperature change, depending on the organic matter content of sediments (sandy and organic matter poor *versus* muddy and organic matter rich). At stations with organic poor and sandy sediments (stations 215, ZG02 and 330), $CH_4$ production in sediments seemed to rapidly increase in response to the sedimentation of organic matter from the spring phytoplankton bloom, and then declined by the start of summer. This is in line with the observation of a peak of $CH_4$ production in the sediments that follows the spring phytoplankton bloom with a time lag of 1 month in Eckernförde Bay in the Baltic Sea (Bange et al. 2010; Steinle et al. 2017). In the stations of the BCZ with sandy sediments, the time lag between the $CH_4$ peak and the phytoplankton bloom seemed to be shorter (14 to 21 d) because more shallow (Borges et al. 2018). Consequently, it seems unlikely that changes in the intensity of the spring phytoplankton bloom might explain changes in $CH_4$ production in the BCZ several months later, either in summer or fall. Indeed, the most off-shore western stations (ZG02, 330, 215 and 230) showed in 2019 distinctly higher peaks of $CH_4$ in mid-May compared to the other years (Figs. 4 and S3). This was probably in response to the higher delivery of freshly produced organic matter from the phytoplankton bloom. Indeed, average Chl-*a* at stations ZG02, 330, 215 and 230 in April was higher in 2019 (11.7±3.6 µg $L^{-1}$) than 2016 (4.1±2.1 µg $L^{-1}$) and 2018 (10.3±7.1 µg $L^{-1}$) (the higher mean value in 2018 compared to 2016 was driven by the values at station 230, the mean value in 2018 excluding this station was 6.9±3.0

µg $L^{-1}$). At these stations, $CH_4$ peaked in Mid-May 2019 in response to higher spring-time Chl-*a* but decreased again by early June 2019, confirming that the response of $CH_4$ to the spring phytoplankton bloom is fast (< 1 month) and short-lived.

Average Chl-*a* was higher in March and early April 2018 than 2016 (Fig. 3). This seemed to be a generalised feature at all nine stations (Fig. S5) suggesting better light conditions in early 2018 than 2016, as winter-time dissolved inorganic nutrients were similar in 2018 and 2016 (not shown). However, average Chl-*a* concentrations in the BCZ were similar from late April to July in 2016 and 2018 (Fig. 3). Furthermore, the highest $CH_4$ concentrations in July 2018 were observed at stations 130 and 700 where sedimentary $CH_4$ production does not respond to inputs from the spring phytoplankton bloom, but, instead is a function of seasonal temperature variations because they have muddy sediments rich in organic matter (Borges et al. 2018). We conclude that the difference in $CH_4$ concentration in the BCZ between July 2018 and 2016 was unrelated to marginal differences in the early spring phytoplankton bloom in particular in the near-shore muddy stations (130 and 700) where the highest $CH_4$ concentrations were observed, although it might have played a small role in the off-shore sandier stations (ZG02 and 330).

The average dissolved $CH_4$ concentration for each cruise was positively correlated to water temperature (Fig. 5), as previously reported in the area (Borges et al. 2016; 2018), although extending markedly the upper bounds of the range of variations of both dissolved $CH_4$ concentration and seawater temperature values. This might possibly be explained by the optimum temperature for methanogenic archaea around 35-40°C for mesophiles and 60-65°C for thermophiles (Zeikus and Winfrey 1976; Schulz et al. 1997; Yvon-Durocher et al. 2014). The most common lineages of methanogenic archaea in marine sediments are *Methanoculleus* and *Methanosaeta* followed by *Methanolinea,* and organisms from all three groups are either mesophiles or thermophiles (Wen et al. 2017). Psychrophile methanogenic archaea (optimum growth at 15-20°C) have only been reported in cold deep lake sediments but not in marine sediments (Blake et al. 2017), although psychrotolerant methanogenic archaea (optimum growth temperature similar to mesophiles but capable of survival at temperatures of 0-5°C) have been reported in Arctic marine sediments (Kendall et al. 2007). Below the optimum temperature and in an environment rich in organic matter, a positive relationship between methanogenesis and temperature is expected (Yvon-Durocher *et al.* 2014). Indeed, warming has been

shown experimentally to increase $CH_4$ production in freshwater sediments (Yvon-Durocher et al. 2017; Comer-Warner et al. 2018).

Although station 120 is characterized by muddy sediments rich in organic matter (Braeckman et al. 2014), the increase of $CH_4$ in July 2018 compared to 2016 was not as spectacular as in stations 130 and 700 (Fig. 4, S3). Stations 130 and 700 are located above a zone of gassy sediments (Missiaen et al. 2002). Release of $CH_4$ from gassy sediments is enhanced in warmer conditions due the decrease of the solubility of $CH_4$ and thermal expansion of gas pockets (Martens et al. 1998; Wever et al. 1998). We conclude that the increase of temperature related to the heatwave of 2018 most likely led to a general increase of $CH_4$ in surface waters (at nearly all stations) due to enhancement of methanogenesis (stimulation of microbial metabolism), and that, in addition, the higher temperature also most likely led to enhanced $CH_4$ release from gassy sediments (in particular at stations 130 and 700 situated in the area of acoustic turbidity indicative of gassy sediments).

However, the above interpretation of the $CH_4$-temperature relation given in Figure 5 requires that the extra heat in the water column during the heatwave of 2018 partly propagated into the sediments, and presumably also led to higher sediment temperatures. The BCZ is shallow (<30 m deep) and experiences strong tidal currents, consequently, the water column is permanently well mixed so that the surface of sediments should have experienced warmer conditions in 2018 compared to the other years, and we assume that this extra heat propagated deeper in the sediment. The extent of additional warming of sediments should depend on the sediment thermal diffusivity that increases with porosity and is highest in sandy sediments (Goto and Matsubayashi 2009). Furthermore, in sandy and permeable sediments such as those present in the BCZ, surface gravity waves may drive advective pore-water exchange that can increase 50-fold the fluid exchange between sandy sediment and overlying water relative to the exchange by molecular diffusion (Precht and Huettel 2003), and should also increase heat propagation in sediments (Savidge et al. 2016). In shallow and sandy sediments of the Gulf of Mexico, Jackson and Richardson (2001) showed that temperature within the sediment down to 1 m below surface tracked closely seasonal changes of the overlying water at time-scales compatible with the long-lasting heatwave experienced in Europe in 2018.

The $FCH_4$ in July 2018 was 202% higher at station 130 (1,935 versus 639 µmol $m^{-2}$ $d^{-1}$) and 165% higher at station 700 (2,845 versus 1,070 µmol $m^{-2}$ $d^{-1}$) than

in July 2016. The overall average of $FCH_4$ for the nine stations in July 2018 was 57% higher than in July 2016 (599 versus 382 µmol $m^{-2}$ $d^{-1}$) (Table 1). The yearly average of $FCH_4$ for the nine stations was 37% higher in 2018 than 2016 (221 versus 161 µmol $m^{-2}$ $d^{-1}$), showing that the European heatwave of 2018 most likely had a major impact on the $CH_4$ emissions from the BCZ.

## 4. Conclusion

The European heatwave of 2018 most likely led to a major increase of the emissions to the atmosphere of $CH_4$ from the BCZ both in summer and at annual scale. This indicates that emissions of $CH_4$ to the atmosphere in coastal environments similar to the BCZ (shallow and with organic rich sediments) should most likely increase in future because heatwave events are predicted to increase in frequency and magnitude as a consequence of global warming (Frölicher et al. 2018). Also, the response of the BCZ to the European of 2018 heatwave seems to have provided a natural in-situ experiment of the response to future warming of $CH_4$ emissions from shallow marine areas.

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597

598 **Table 1:** Average at nine stations in the Belgian coastal zone (Fig. 1) of dissolved
599 $CH_4$ concentration in surface waters (nmol $L^{-1}$), wind speed (m $s^{-1}$), gas transfer
600 velocity ($k_{600}$ in cm $h^{-1}$) and air-water $CH_4$ flux ($FCH_4$ in µmol $m^{-2}$ $d^{-1}$). The date
601 corresponds to the last day of the cruise (duration 1-2 days). Wind speed was
602 averaged over the 15 days prior to the last day of the cruise to provide seasonally
603 representative values and smooth out transient weather events (windy or calm
604 spells). Data of $CH_4$ were not acquired in January and February 2018, so to provide
605 annual averaged $FCH_4$, the yearly cycle was completed with data acquired in
606 January and February 2019. Only the cruises of 2016 concurrent in the yearly cycle
607 with those of 2018/9 were used.

| Dates | | $CH_4$ nmol $L^{-1}$ | | Wind speed m $s^{-1}$ | | $k_{600}$ cm $h^{-1}$ | | $FCH_4$ µmol $m^{-2}$ $d^{-1}$ | |
|---|---|---|---|---|---|---|---|---|---|
| | | 2016 | 2018/9 | 2016 | 2018/9 | 2016 | 2018/9 | 2016 | 2018/9 |
| 28-01-16 | 29-01-19 | 20 | 23 | 10.9 | 9.5 | 33.8 | 25.9 | 90 | 79 |
| 16-02-16 | 21-02-19 | 38 | 50 | 11.2 | 8.8 | 34.7 | 22.4 | 192 | 167 |
| 04-03-16 | 05-03-18 | 13 | 36 | 9.9 | 10.2 | 28.5 | 28.3 | 47 | 131 |
| 31-03-16 | 22-03-18 | 21 | 19 | 8.9 | 9.6 | 22.7 | 24.5 | 70 | 58 |
| 11-04-16 | 04-04-18 | 23 | 35 | 8.4 | 7.3 | 21.3 | 14.6 | 76 | 75 |
| 27-04-16 | 24-04-18 | 15 | 27 | 8.6 | 6.4 | 20.4 | 12.6 | 45 | 56 |
| 04-05-16 | 03-05-18 | 18 | 20 | 8.6 | 8.9 | 21.0 | 22.7 | 58 | 73 |
| 25-05-16 | 30-05-18 | 48 | 61 | 7.0 | 6.8 | 14.3 | 13.5 | 126 | 163 |
| 26-07-16 | 26-07-18 | 110 | 338 | 7.3 | 5.1 | 15.4 | 7.5 | 382 | 599 |
| 24-08-16 | 27-08-18 | 51 | 96 | 8.3 | 8.2 | 19.3 | 18.8 | 209 | 391 |
| 27-09-16 | 26-09-18 | 156 | 87 | 7.3 | 8.0 | 15.7 | 19.0 | 530 | 543 |
| 26-10-16 | 24-10-18 | 40 | 45 | 8.4 | 7.2 | 19.8 | 14.8 | 142 | 207 |
| 23-11-16 | 22-11-18 | 41 | 71 | 10.5 | 10.1 | 30.6 | 27.5 | 208 | 313 |
| 19-12-16 | 18-12-18 | 35 | 46 | 7.2 | 11.2 | 14.3 | 34.8 | 74 | 245 |
| **Average** | | **45** | **68** | **8.8** | **8.4** | **22.3** | **20.5** | **161** | **221** |

608

Figure captions

**Figure 1:** Map of the nine sampling stations (circles), the West Hinder platform (wind speed measurements), bathymetry, and sediment acoustic turbidity in the Belgian coastal zone. Acoustic turbidity corresponds to gassy sediments (from Missiaen et al. 2019).

**Figure 2:** Time series of monthly seawater temperature (°C) in the Belgian coastal zone and monthly air temperature (°C) in the city of Oostende from 2004 to 2018. Horizontal dotted line indicates the value of July 2018.

**Figure 3:** Average of the nine sampled stations in the Belgian coastal zone (Fig. 1) of seawater temperature (°C), chlorophyll-*a* concentration (Chl-*a* in µg $L^{-1}$), and dissolved $CH_4$ concentration ($CH_4$ in nmol $L^{-1}$) in 2016, 2017, 2018 and 2019. Error bars represent the standard error (that in some cases are smaller than the symbol and do not appear on the plot).

**Figure 4:** Seasonal variations of dissolved $CH_4$ concentration ($CH_4$ in nmol $L^{-1}$) at nine stations in the Belgian coastal zone in 2016, 2017, 2018, 2019. The plots are arranged to correspond to the spatial distribution of the stations (Fig. 1), left to right corresponding to West to East, and top to bottom corresponding from off-shore to near-shore. The same data are presented in Figure S2 with Y-axis individually scaled for each station.

**Figure 5:** Dissolved $CH_4$ concentration (nmol $L^{-1}$) as function of seawater temperature (°C) averaged for the nine stations (Fig. 1) in the Belgian coastal zone in 2016, 2017, 2018 and 2019. The dotted line shows the exponential fit for data with seawater temperature (T) > 10°C: $CH_4 = 1.7527 \exp(0.2283*T)$ ($r^2=0.93$), where $CH_4$ is in nmol $L^{-1}$ and T in °C.

Fig. 1

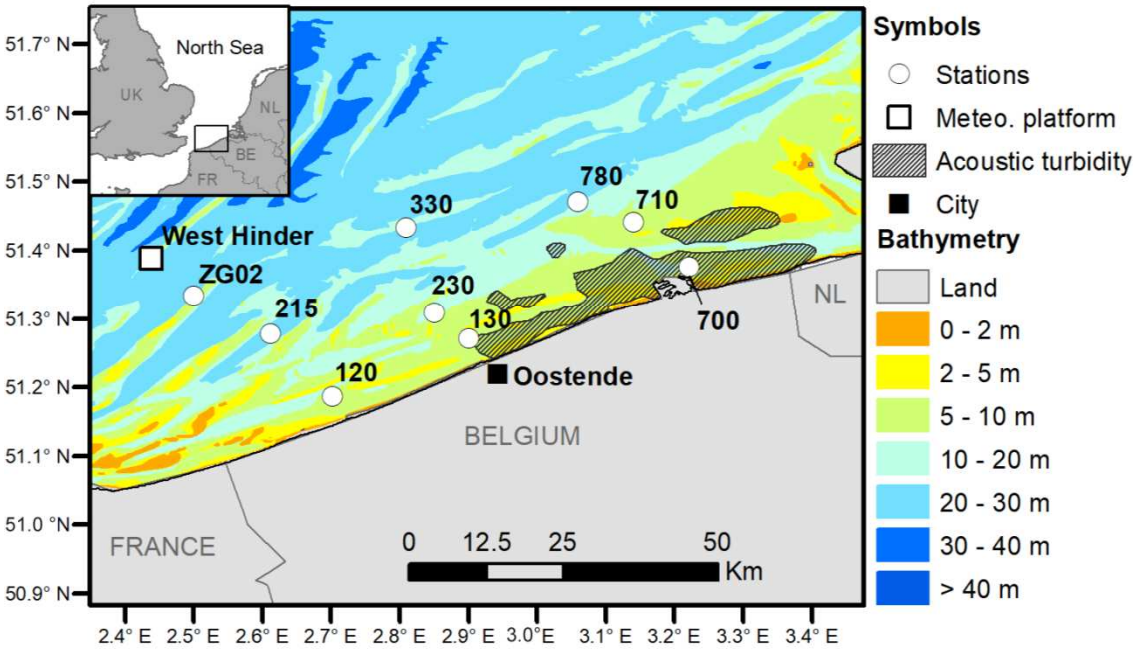


Fig. 2

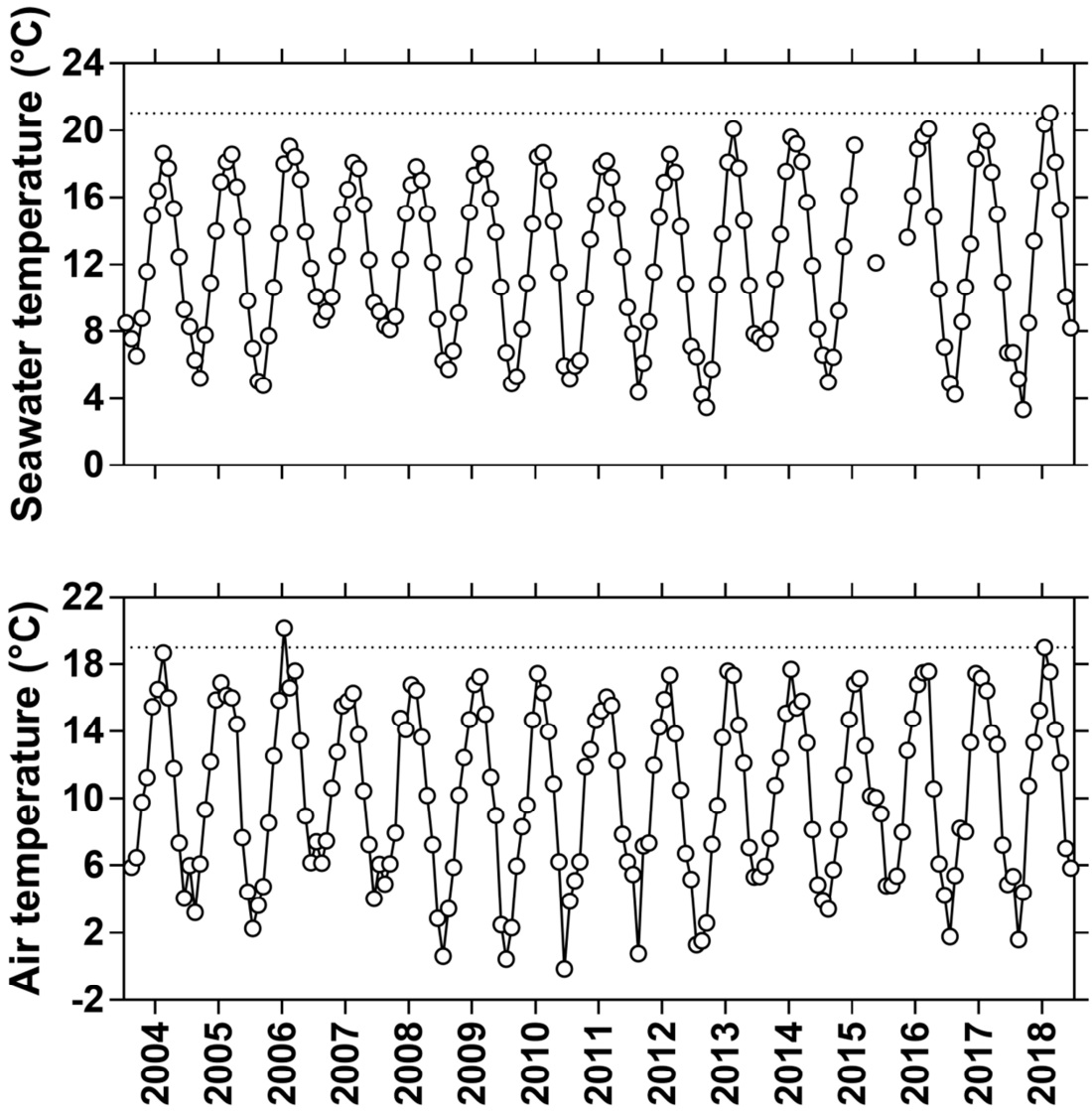


Fig. 3

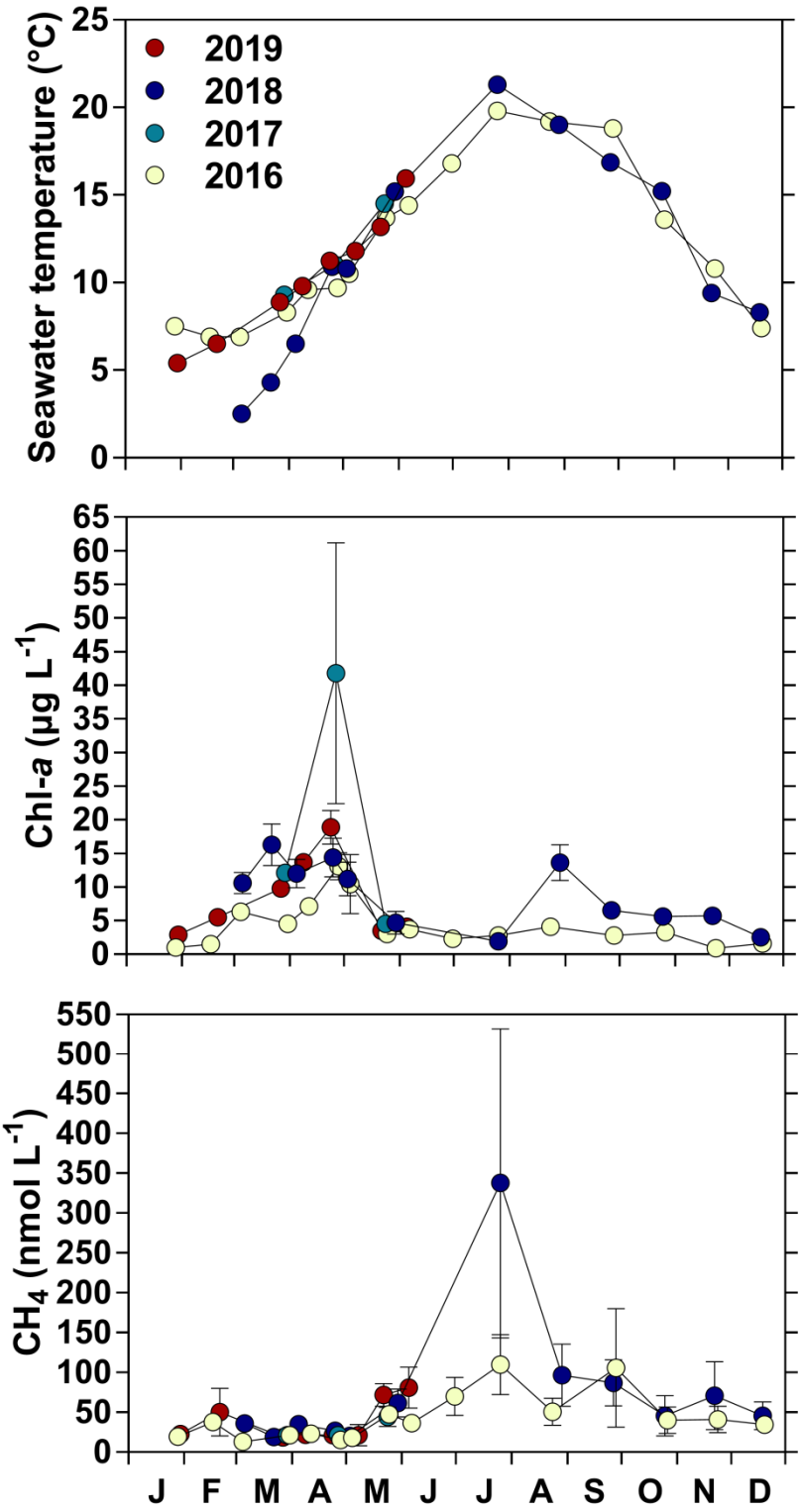


Fig. 4

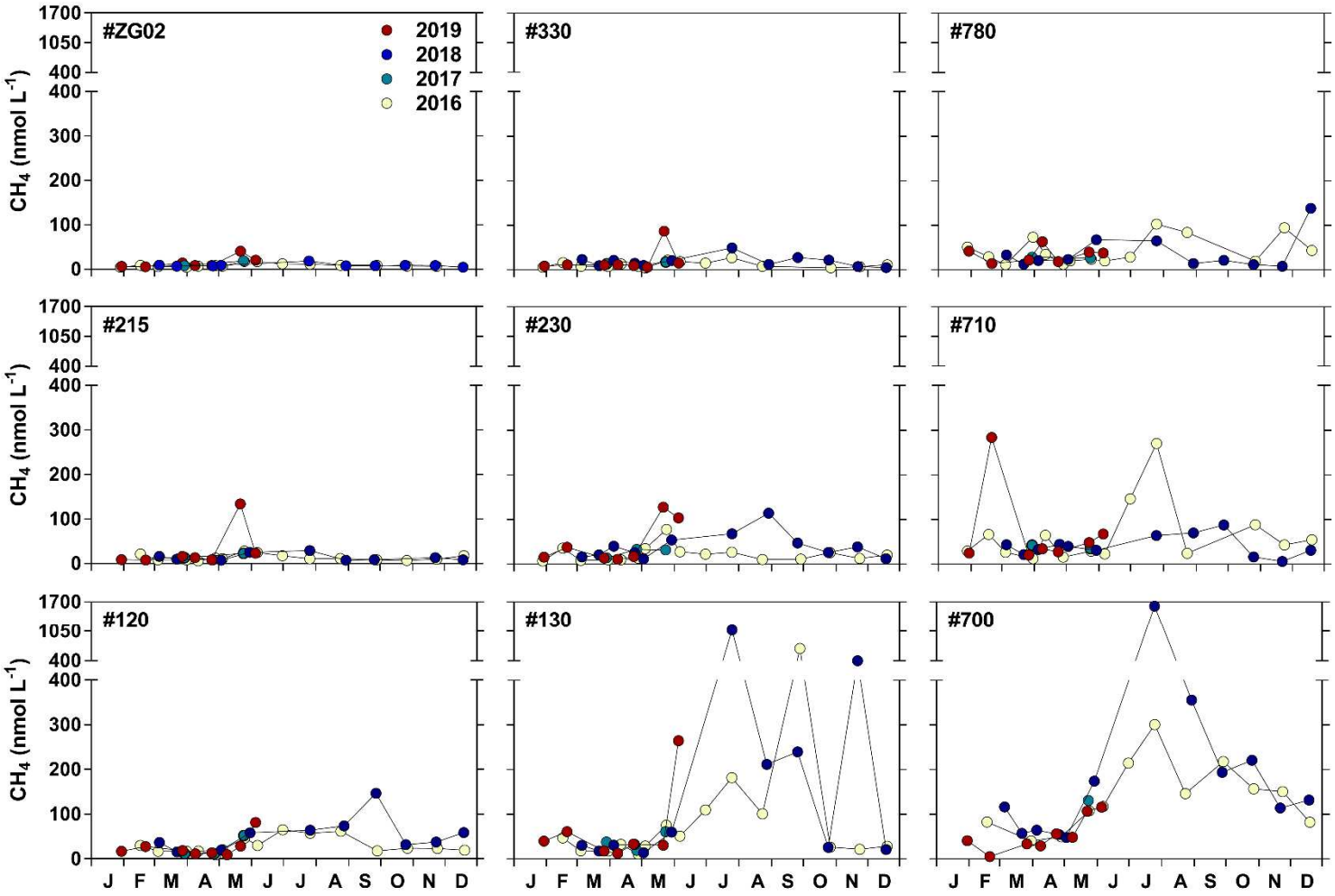
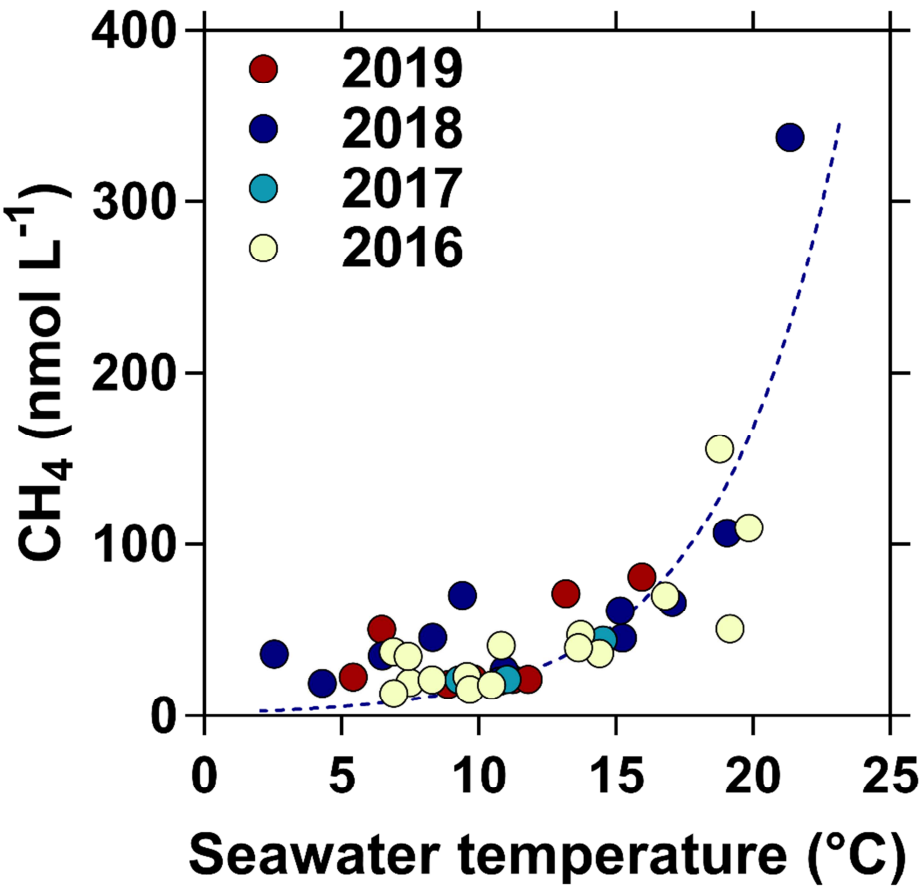
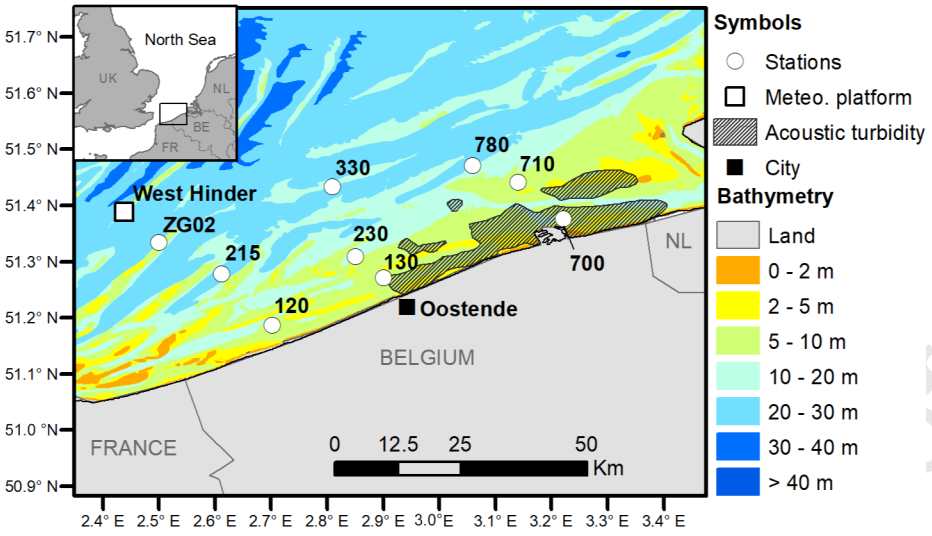
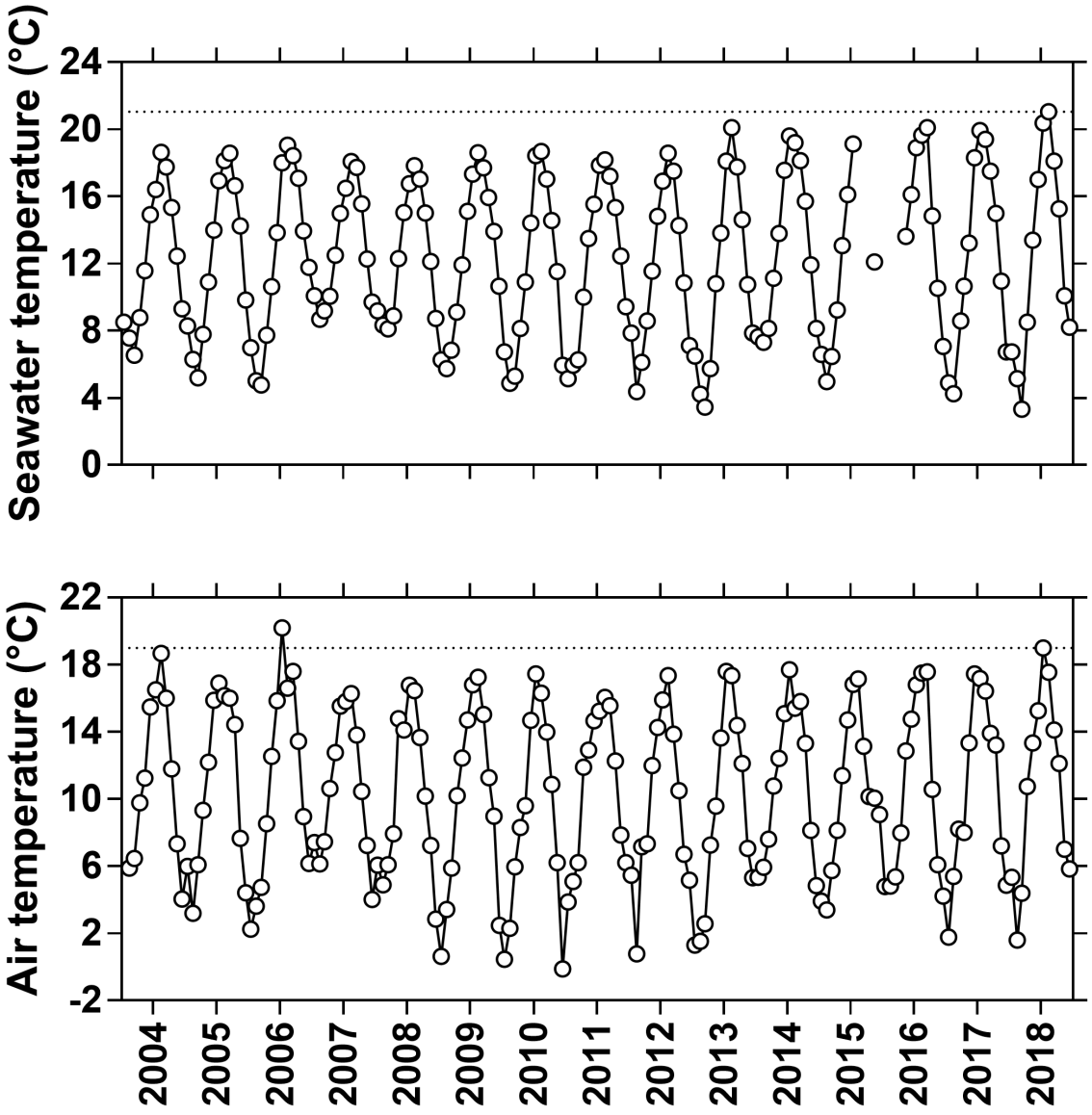
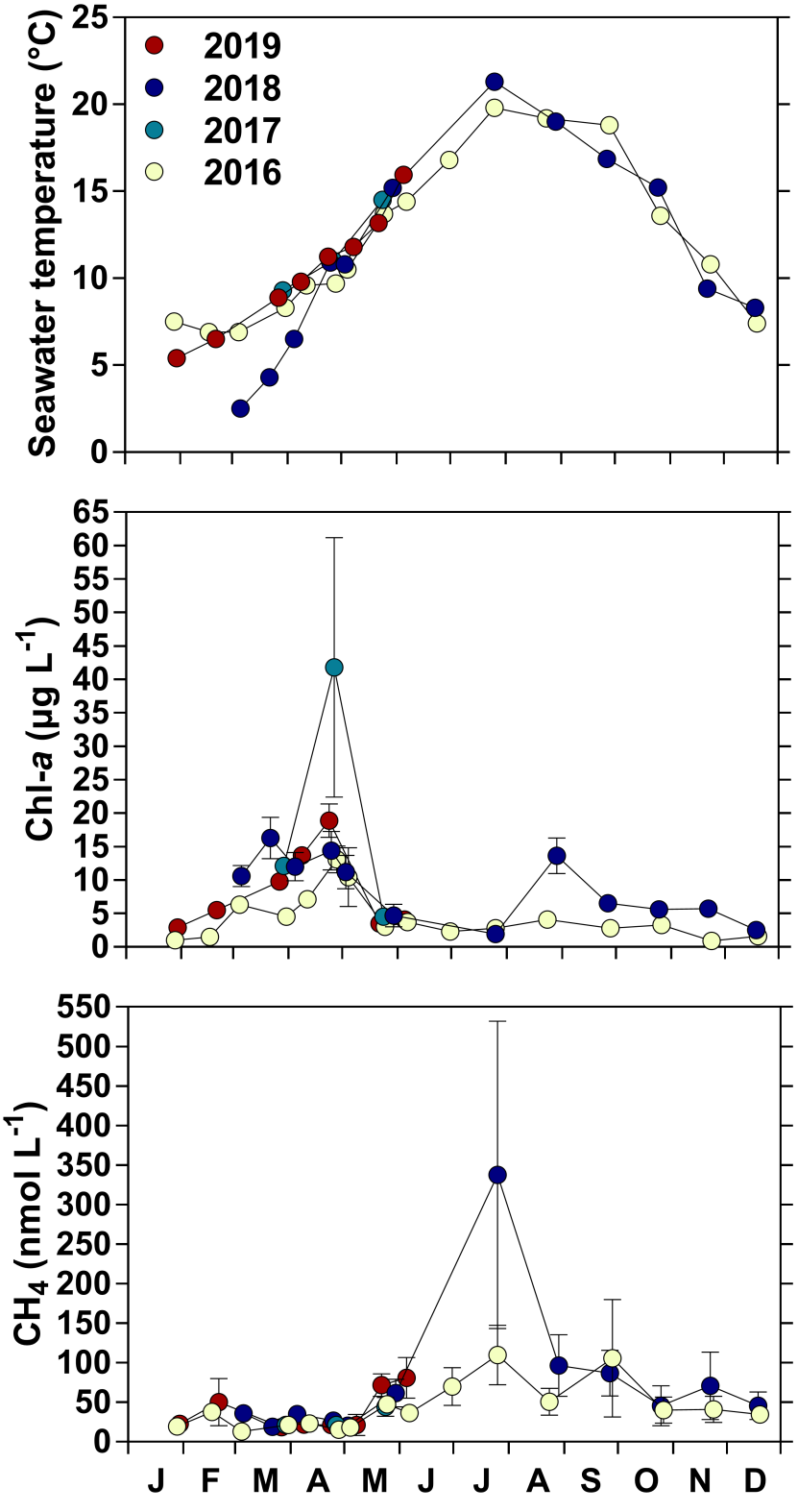

#ZG02
2019
2018
2017
2016
#330
#780
#215
#230
#710
#120
#130
#700
CH4 (nmol L-1)
1700
1050
400
300
200
100
0
J F M A M J J A S O N D

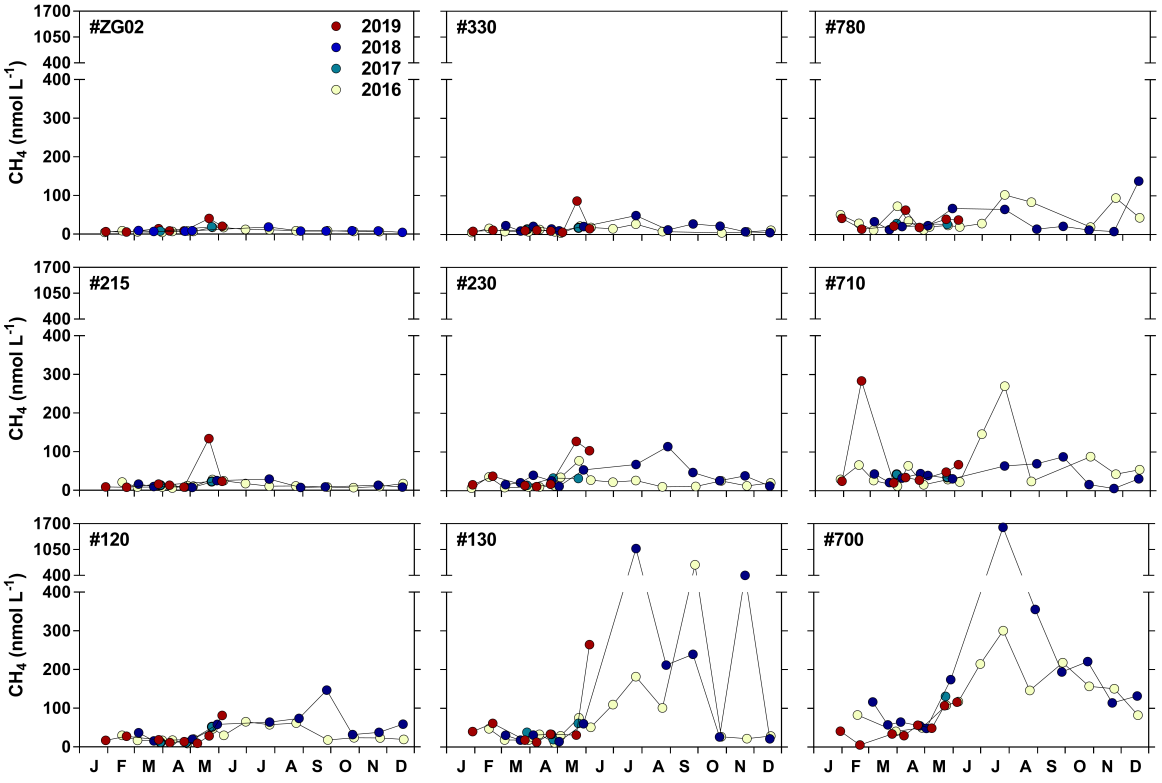
Fig. 5

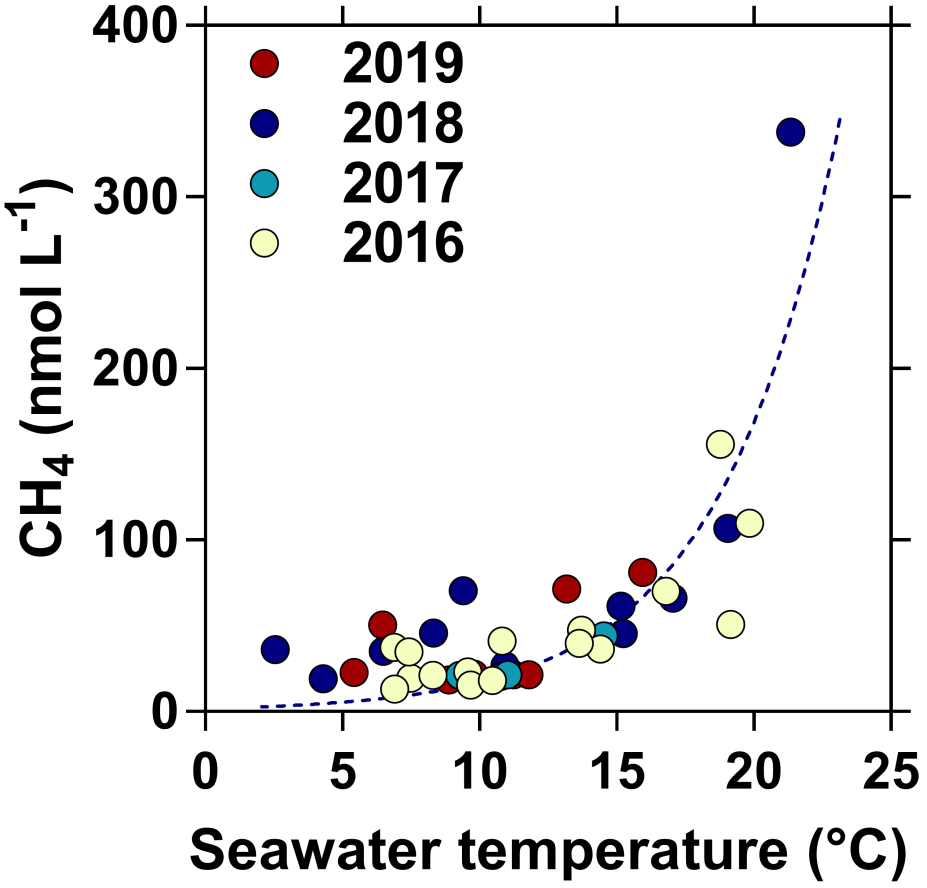





Seawater temperature (°C)
24
20
16
12
8
4
0
Air temperature (°C)
22
18
14
10
6
2
-2
2004
2005
2006
2007
2008
2009
2010
2011
2012
2013
2014
2015
2016
2017
2018


Seawater temperature (°C)
2019
2018
2017
2016
Chl-a (µg L-1)
CH4 (nmol L-1)
J F M A M J J A S O N D


#ZG02
2019
2018
2017
2016
#330
#780
#215
#230
#710
#120
#130
#700
CH4 (nmol L-1)
1700
1050
400
300
200
100
0
J F M A M J J A S O N D


2019
2018
2017
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400
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100
0
$CH_4$ (nmol $L^{-1}$)
0
5
10
15
20
25
Seawater temperature (°C)

- In July 2018 water temperature was 2.5°C higher than other years in the Belgian coast
- In July $CH_4$ concentration was 3 x higher in 2018 than 2016 in the Belgian coast
- Extremely high $CH_4$ concentrations at some stations
- Major impact of European heatwave of 2018 on marine $CH_4$ in the Belgian coast

**Declaration of interests**

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: