FINAL REPORT PROGRAM LEFE

Program LEFE/ CYBER	**MEMESTRA**: **ME**thane dynamics in **M**arine **E**nvironments based on in **S**i**T**u obse**R**v**A**tions.	**Years 2019-2021**

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Context. Methane (CH_4) emissions constitute the second largest contribution to historical warming after carbon dioxide (CO_2). The role of the oceans is often neglected due to large uncertainties on the distribution of the marine sources of CH_4, their dynamics, and the nature and kinetics of the biogeochemical processes involved.

Objectives / scientific questions. Evaluating the CH_4 dynamics and the biogeochemical processes that control its distribution across the water column in various marine systems combining *in situ* observations and discrete sampling and assessing the main processes driving the fluxes taking into account their spatial and temporal variability.

Main results.

1. CH_4 dynamics in coastal megatidal environments of the south Western English Channel (sWEC)

Coastal ecosystems and estuaries are considered as hotspots of marine methane (CH_4) emissions and present large variability in terms of air-sea carbon dioxide (CO_2) fluxes with poorly constrained driving mechanisms. From January 2018 to December 2020, monthly surveys were carried out in two contrasted small estuaries of the Bay of Morlaix (South-Western English Channel (sWEC), NE Atlantic) influenced by both agricultural and urbanized watersheds to collect dissolved CH_4, carbonate system and ancillary biogeochemical parameters. These parameters were also monitored at 2 proximate stations of the French coastal monitoring SOMLIT network representative of coastal waters of the sWEC. Dissolved Inorganic Carbon (DIC), Total Alkalinity (TA) and $\delta^{13}C_{DIC}$ showed a rather conservative behaviour typical of North Atlantic waters mixing with freshwater from non-limestone watersheds throughout a gradient from the inner estuary to the coastal stations. Mixing models showed a well-constrained linear correlation between ΔTA and ΔDIC in the inner estuaries, which indicated a potential source of DIC/TA controlled by the combination of organic matter (OM) degradation via denitrification and sulphate reduction at the water-sediment interface during spring/summer. A difference in conservativity was observed between pCO_2 and CH_4 between the inner (conservative) and outer (perturbations linked to the stratification) estuary. The mean CH_4 and CO_2 fluxes were 120.3 µmol C m^{-2} yr^{-1} (6500% of saturation) and 18.1 mol m^{-2} yr^{-1} in Penzé estuary and 309.7 µmol C m^{-2} yr^{-1} (14600% of saturation) and 13.0 mol C m^{-2} yr^{-1} in the urbanized Morlaix estuary, respectively, showing a different order of magnitude compared to the SWEC (Fig. 1). This study revealed high GHG emissions in small, poorly documented estuaries, that could change the GHG balance of the European estuarine waters, considering the numerous similar systems in the Brittany region.

2. CH_4 distribution in the upper water column of the subtropical North Atlantic Ocean

The distribution of dissolved CH_4 concentrations and associated air–sea fluxes during winter 2020 were investigated as part of the JC191 cruise in the subtropical North Atlantic Ocean (26° N, 80′ W and 26° N, 18′ W). Water samples from 64 stations were collected from the upper 0-400 m water column. The upper oxic mixed layer was oversaturated in dissolved CH_4 (120 – 280 %), with the highest concentrations of 7–10 nmol L^{-1} found to the east of the transect, consistent with observations from other subtropical regions of the world's oceans. These high anomalies were found to be associated with phosphate-depleted waters and regions where the abundance of the ubiquitous picocyanobacterial *Synechococcus* and *Prochlorococcus* were elevated (Fig. 2). Although other phytoplanktonic phyla cannot be excluded, this suggests that cyanobacteria contribute to the release of CH_4 in this region. This study confirms the subtropical North Atlantic Ocean as a source of CH_4 and provides evidence to corroborate the key role that picocyanobacteria play in helping to explain the oversaturation of CH_4 found in surface mixed layer of the open ocean, otherwise known as the “ocean methane paradox”.

3. CH4 distribution in the upper water column of the Western Tropical South Pacific

As part of the TONGA cruise in the Western Tropical South Pacific Ocean (WTSP), we sampled the 0-400 m water column along a 1,500 km W-E transect from Noumea (New Caledonia) to the Central Pacific Gyre (165°W) to

determine the CH_4 concentrations as well as the genetic and functional diversity of marine picocyanobacteria. Results confirm that CH_4 oversaturation is a common feature of the oxic mixed layer over the whole transect, and is strongly correlated to phosphate depletion, the abundance of *Prochlorococcus* and *Synechococcus* cells as well as the relative abundance of specific picocyanobacterial taxa. These results are not only in agreement with previous observations in other regions of the world, but also with the recent findings from lab-based experiments showing the ability of cyanobacteria to produce CH_4 under both light and dark conditions.

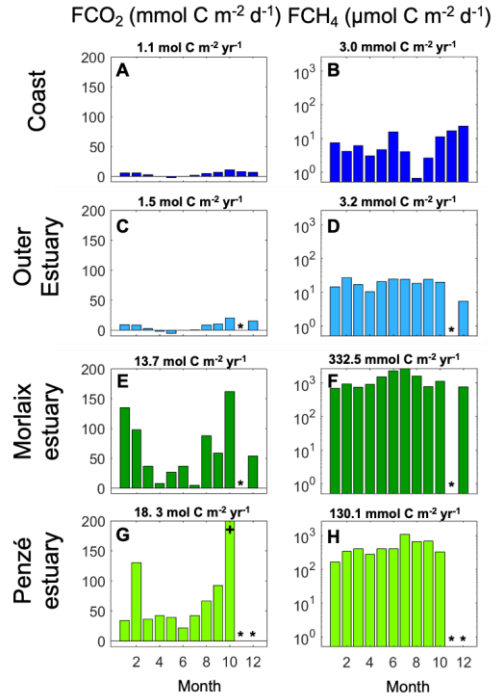

Figure 1

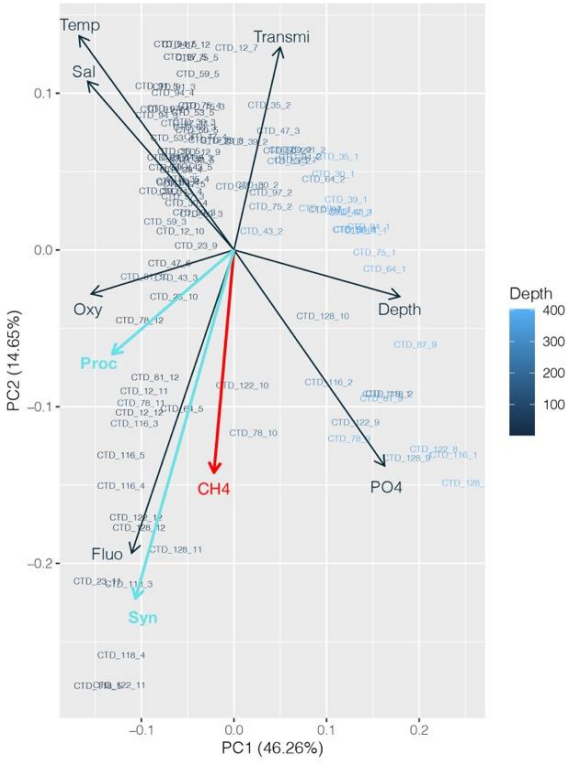

Figure 2

Figure 1. Monthly sea-to-air fluxes of CO_2 and CH_4 in the Western English Channel (from Gac et al, in prep)
Figure 2. CH_4 production in the upper euphotic layer of the North Atlantic Ocean appears to be associated to primary production and the presence of picocynaobacteria (Kolomijeca et al., Ocean Science, 2022)

Future of the project. The main results obtained over the course of the MEMESTRA project clearly underline the lack of knowledge on the contribution of the oceans to the sea-to-air fluxes of CH_4. From these preliminary data, the project will evolve towards 1) a better comprehension of the contribution of the small estuaries to the CH_4 budget, including a better assessment of the CO_2-CH_4 relationships and 2) the identification of the metabolic pathways involved in the production of CH_4 by phytoplankton in open oceans and macroalgae in coastal systems. Indeed, these processes may counterbalance the C intake due to the photosynthesis, and hence on the carbon budget that is taken into account for the climate models. The future of the project will require new observing tools for the measurement of CH_4 in marine systems. This will be proposed as part of calls for projects (ANR, ERC).

Number of publications, communications and theses

Articles:

Boulart, C., Garczarek, L., Le Moal, P., Partensky F., Bigeard E., Ferrieux M., Ratin M., Le Gall F., Romac S., Baudoux A-C., Jeanthon, C., Bonnet, S., Guieu, C. Marine picocyanobacteria, an important player in the distribution of dissolved methane in the Western Tropical South Pacific Ocean? *in prep for Frontiers in Marine Science (TONGA Special Issue)*

Gac J.-P., Boulart C., Cariou T., Crec'hriou R., Macé E., Bureau S., Vernet M. Marrec P., and Bozec Y. Dynamics of the CO_2 system and CH_4 in two estuaries of a megatidal embayment with contrasting watersheds (South-Western English Channel). In prep for Frontiers in Marine Science

Kolomijeca A, Marx L, Reynolds S, Cariou T, Mawji E, Boulart C. 2022. An update on dissolved methane distribution in the subtropical North Atlantic Ocean. *Ocean Sci.* 18(5):1377–88

Thèse.

Gac J-P. 2021. Etude multi-échelles des échanges air-mer de CO2 et de l'acidification océanique en Manche Occidentale. Sorbonne Université, Paris.

Data availablility

SOMLIT Network, TONGA (DOI 10.17882/88169)