Fate of methane from sea floor seeps

Lab studies and modelling

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Technology for a better society

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The work is described in two open technical reports, and one peer-reviewed paper:

- https://hdl.handle.net/11250/2730544
- https://hdl.handle.net/11250/3038685
- Nordam, Tor, et al. "Fate of Dissolved Methane from Ocean Floor Seeps." *Environmental Science & Technology* 59.17 (2025): 8516-8526. https://doi.org/10.1021/acs.est.5c03297

The model used is available as open source:

- https://github.com/SINTEF/Fate-of-methane/

I would also like to thank Karsten Bolding for email support on the use of GOTM.

Motivation

"Modeling indicates that less than ~4% of the gas initially released at the seafloor is transported via bubbles into the mixed layer and, ultimately, to the atmosphere.

However, […] almost all of the methane released at the seafloor could be transferred into the atmosphere in the stormy fall and winter time."

Schneider von Deimling et al. (2011)

https://doi.org/10.1016/j.csr.2011.02.012

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Quantification of seep-related methane gas emissions at Tommeliten, North Sea

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ABSTRACT

Tommeliten is a prominent methane seep area in the Central North Sea. Previous surveys revealed shallow gas-bearing sediments and methane gas ebullition into the water column. In this study, the in situ methane flux at Tommeliten is re-assessed and the potential methane transport to the atmosphere is discussed, with regards to the hydrographic setting and gas bubble modeling. We have compiled previous data, acquired new video and acoustic evidence of gas bubble release, and have measured the methane concentration, and its C-isotopic composition in the water column. Parametric subbottom sonar data reveal the three-dimensional extent of shallow gas and morphologic features relevant for gas migration. Five methane ebullition areas are identified and the main seepage area appears to be 21 times larger than previously estimated. Our video, hydroacoustic, subbottom, and chemical data suggest that $\sim 1.5 \times 10^6$ mol CH_4/yr (~26 tons CH_4/yr) of methane gas is being released from the seepage area of Tommeliten. Methane concentration profiles in the vicinity of the gas seeps show values of up to 268 nM (~100 times background) close to the seafloor. A decrease in $\delta^{13}C$-CH_4 values at 40 m water depth indicates an unknown additional biogenic methane source within the well oxygenated thermocline between 30 and 40 m water depth. Numerical modeling of the methane bubbles due to their migration and dissolution was performed to estimate the bubble-derived vertical methane transport, the fate of this methane in the water column, and finally the flux to the atmosphere. Modeling indicates that less than ~4% of the gas initially released at the seafloor is transported via bubbles into the mixed layer and, ultimately, to the atmosphere. However, because of the strong seasonality of mixing in the North Sea, this flux is expected to increase as mixing increases, and almost all of the methane released at the seafloor could be transferred into the atmosphere in the stormy fall and winter time.

1. Introduction

1.1. CH_4 in the atmosphere

Methane is the most abundant hydrocarbon in the atmosphere and is influencing the global climate. Compared to carbon dioxide, the global warming potential (GWP) of methane is about 25–40 times higher on a 100 yr timescale (Shindell et al., 2009). The atmospheric methane concentration growth rate per year has decreased from 1% in the early 1980s (Blake and Rowland, 1988) to close to zero at the turn of the millennium. Following this period of little change, Rigby et al. (2008) present measurements showing renewed growth in early 2007. The reasons for this observation are still under debate, but Arctic wetlands, thawing permafrost, and fires in the high latitudes are among the most favored explanations.

1.2. Marine methane seepage

The net atmospheric methane emission is currently estimated to be 592 Tg CH_4/yr and predominantly consists of anthropogenic contributions (>60%, e.g. agriculture, gas flaring; Reeburgh, 2007). Emissions of submarine methane from mud volcanoes, faults and seepage have been neglected in previous IPCC reports, but were suggested as a potentially important source ranging between 15 and 60 Tg CH_4/yr (Etiope, 2004; Kvenvolden and Rogers, 2005). However, the atmospheric input of this geologic marine methane remains unknown. Methane from marine sediments enters the water column either dissolved in the pore

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Motivation

To address the question of whether almost all the methane could in fact be transferred into the atmosphere during winter,

- We made a model to specifically study the fate of the dissolved methane
- We did some microbial oxidation experiments, to investigate oxidation rates

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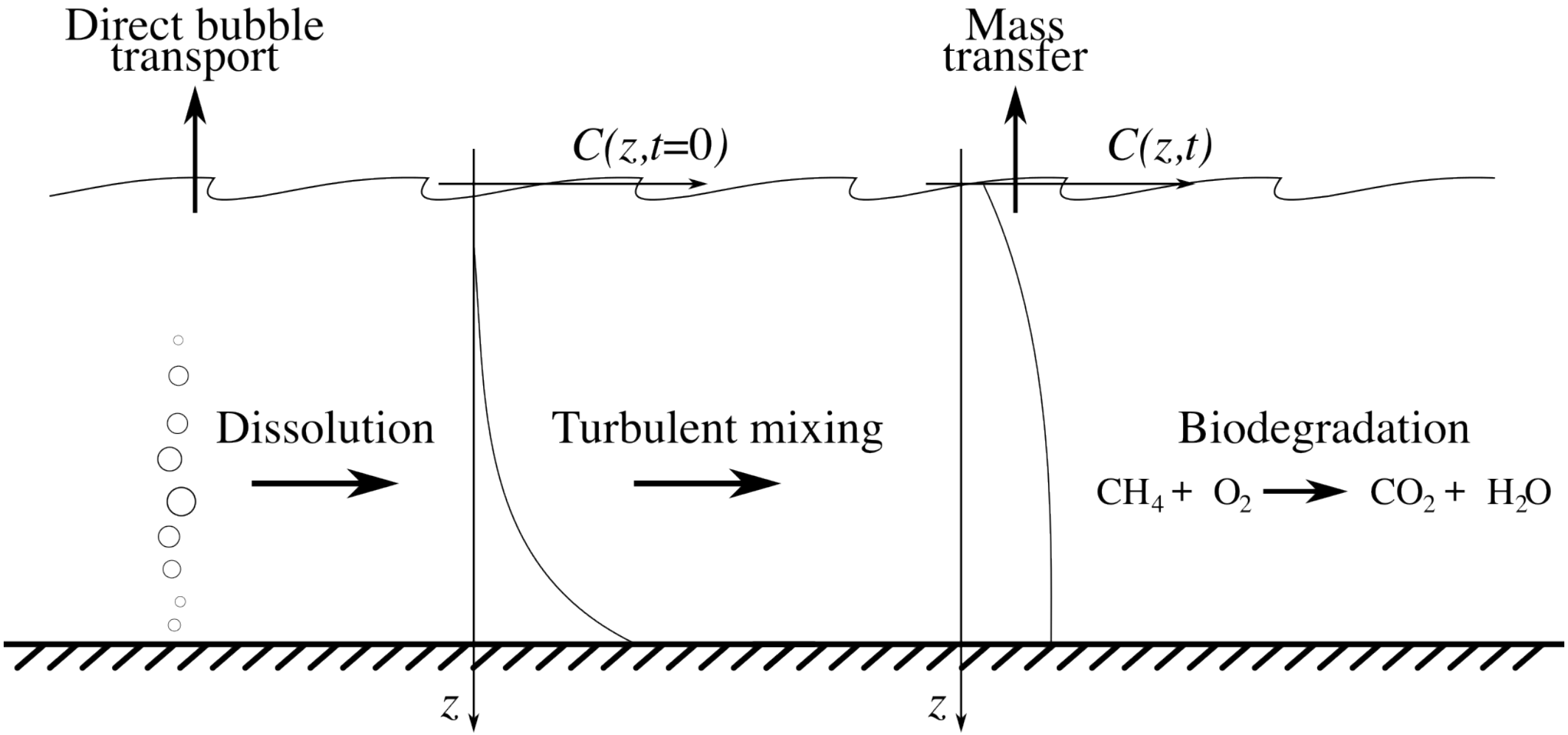
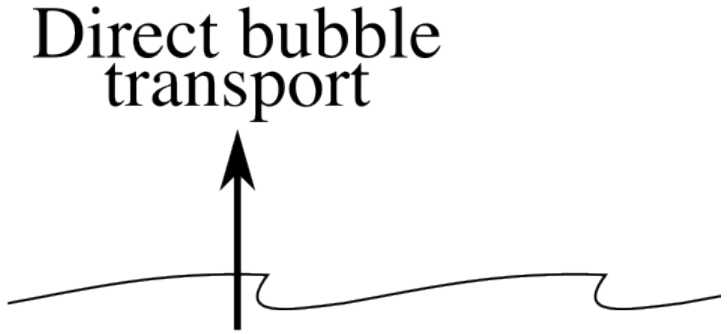
Water column model



Bubble rise and dissolution

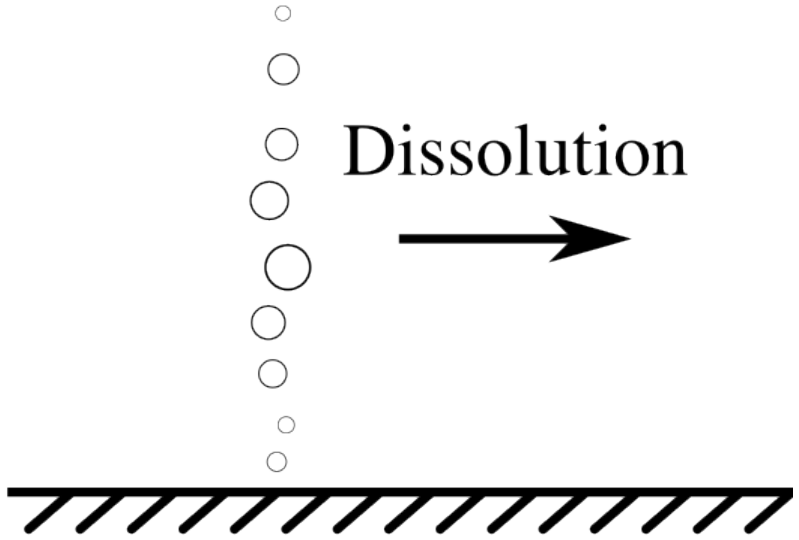
Bubble rise and dissolution modelled with TAMOC[1]

- Rise speed calculated from composition, pressure, etc.
- Methane dissolution via mass transfer into ambient
- Uptake of nitrogen and oxygen from ambient into bubble



Output from TAMOC, used for further modelling

- Fraction of methane that reaches surface directly
- Depth-distribution of dissolved methane

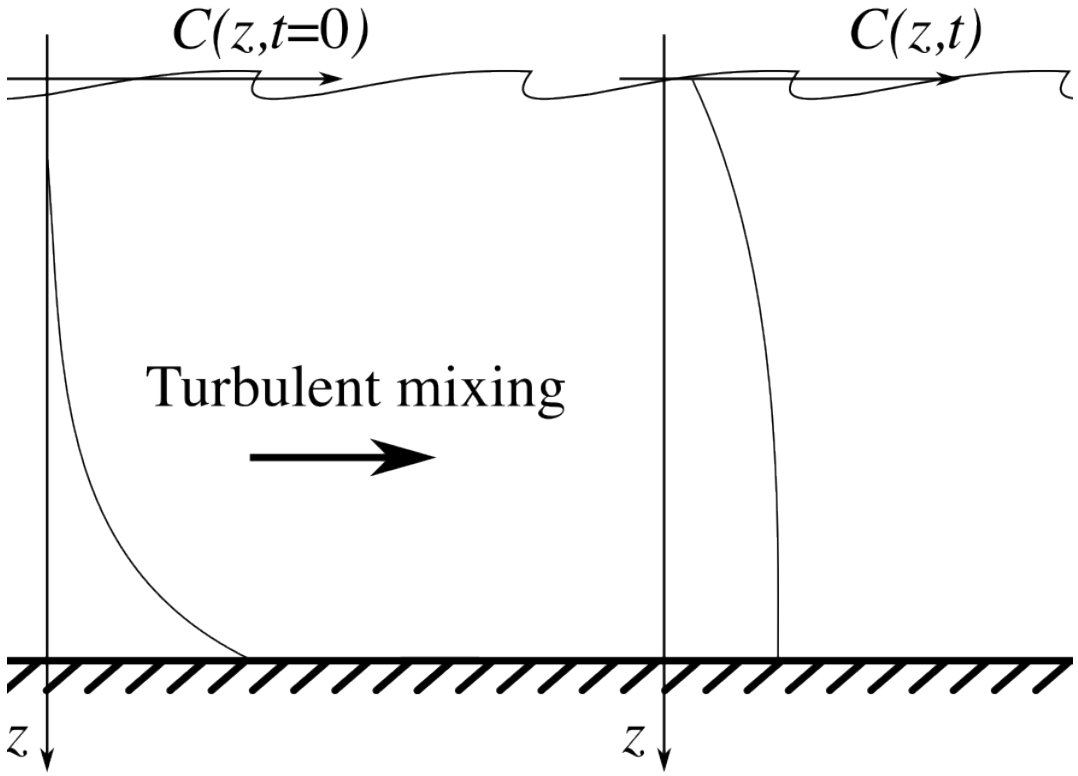


[1] Socolofsky et al. (2016) "Texas A&M Oilspill Calculator (TAMOC): Modeling suite for subsea spills". In: *Proceedings of the 38th AMOP technical seminar*.

Vertical mixing of dissolved methane

Modelled with the diffusion equation

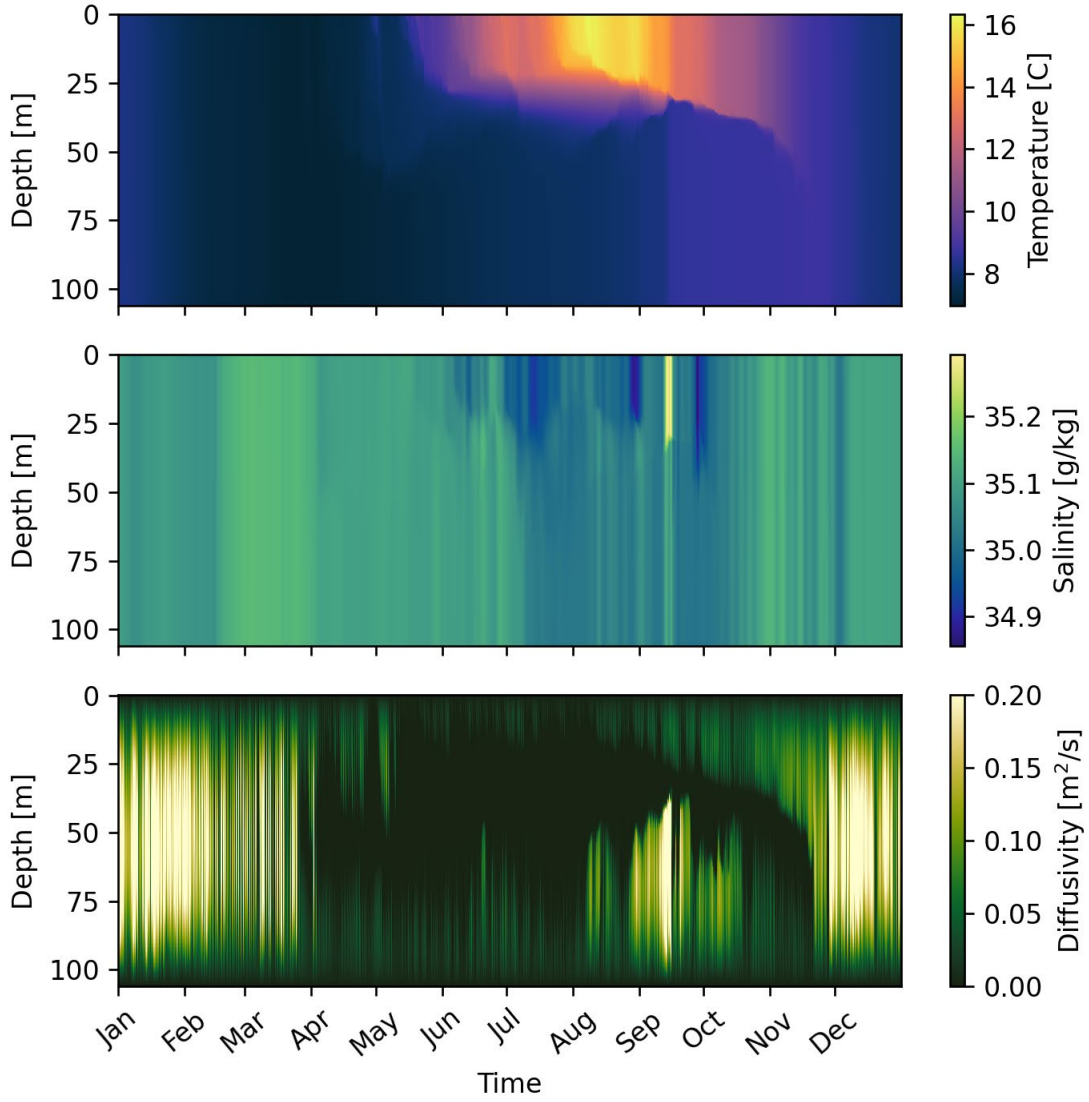
- Initial concentration profile comes from bubble model
- Initial concentration is highest near the sea floor
- Vertical mixing will bring methane to the surface, where it can escape
- Intensity of vertical mixing changes throughout the year
- Stable stratification will delay exchange between deeper layers and surface mixed layer



Vertical mixing of dissolved methane

Vertical eddy diffusivity is input to the model

- Turbulence modelled with GOTM[2]
- Based on temperature, salinity, currents, wind, air temperature, etc.
- We use GOTM to generate one year of vertical diffusivity
- Then we run the diffusion model for one year to capture seasonal variations

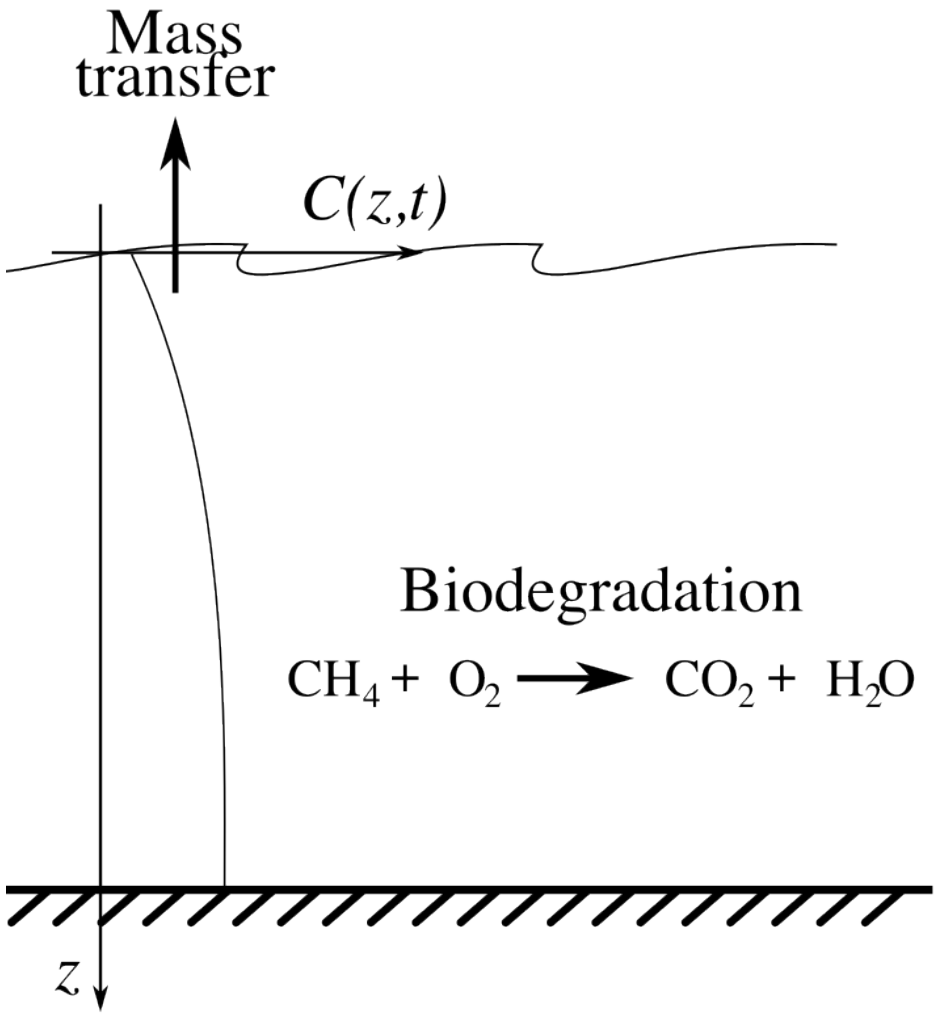


[2] https://gotm.net/

Mass transfer and biodegradation

- Dissolved methane can escape to atmosphere via mass transfer at surface
- Transfer rate depends on near-surface concentration and mass-transfer coefficient
- Mass-transfer coefficient parameterized from wind speed
- Mass transfer is described via boundary condition

- Dissolved methane in water column is also oxidized to CO_2 and water by microbes
- Microbial oxidation is modelled as first-order decay, with a given half-life



What do we mean by “fate of methane”?

We consider three pathways for methane from sea floor seeps

- It can reach the atmosphere directly with the bubble
- It can dissolve, and then reach the atmosphere via mass transfer at surface
- It can dissolve, and then be oxidized to CO_2 by bacteria
- (We ignore e.g. hydrate formation)

The point is to calculate the fraction that is oxidized, and the fraction that reaches the atmosphere

What do we mean by “fate of methane”?

A note on fractions:

- The diffusion-reaction equation (with a first-order reaction term) is linear in concentration:

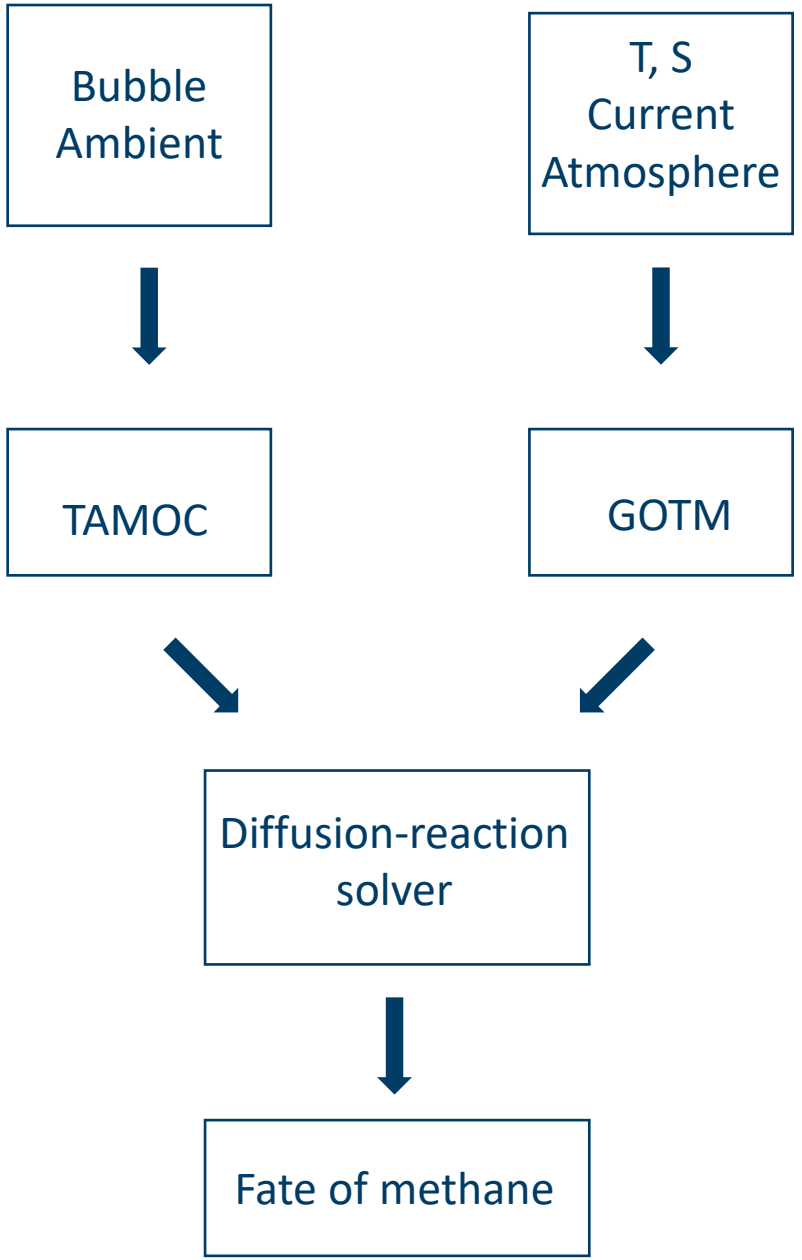
$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial z}\left(D\frac{\partial C}{\partial z}\right) - k_1 C$$

- We do not need to know the actual release rate at the seep.
- We do not need to calculate actual concentration.
- The ratio between the two fates stays the same if the concentration is scaled.
- Assumption: Concentration of methane in atmosphere is small enough not to retard the mass transfer from the surface water

Modelling chain

- TAMOC models bubble rise and dissolution
- GOTM models turbulence

- Diffusion-reaction[3] solver models:
  - Vertical mixing of dissolved methane
  - Mass transfer to atmosphere
  - Microbial oxidation
- Diffusion-reaction solver:
  - Solves PDE with finite-volume method
  - Crank-Nicolson method
  - Implicit, second order in space and time



[3] https://github.com/SINTEF/Fate-of-methane/

Model inputs

Input to TAMOC

- Initial bubble size and composition
- Temperature and salinity profiles
  - Taken from ocean models
- Ambient concentrations of dissolved gasses (oxygen and nitrogen)
  - Some data available on dissolved oxygen, less on nitrogen
  - Can be calculated based on T, S profiles and assumption of equilibrium with atmosphere

Input to GOTM

- Temperature and salinity profiles
  - Taken from ocean models
- Currents
  - Taken from ocean models
- Wind, air pressure, air temperature, humidity, solar heating
  - Available from models, e.g., ERA5

Model inputs

Input to diffusion-reaction model

- Initial concentration of dissolved methane deposited by bubble
  - Output from TAMOC
- Mass transfer coefficient at surface
  - Parameterised from wind speed
- **Vertical diffusivity**
  - Output from GOTM
  - Also available from some ocean models
- **Microbial oxidation rate (half-life)**
  - Available from literature sources

The last two, and in particular the microbial oxidation rate, has considerable uncertainty

Intermezzo – Microbial oxidation experiments

- Microbial oxidation rates are very important for the modelling
- Uncertainty in oxidation rate drives uncertainty in fate
  - We will return to this point

- A brief look at oxidation rates reported in the literature
- A summary of experimental work we did

Microbial oxidation rates

- A literature review found very large variations in reported oxidation rates
- Converted to half-lives, results range from 1-2 days to several years
  - Assumption: Decay rate is first-order in methane concentration

- Possible reasons for variations include:
  - Local differences (bacterial communities, available oxygen, nutrients, etc.)
  - First-order assumption may be unjustified -> different methane concentrations give different results
  - Uncertainties in experimental results

Tritium method

- Add known activity of tritium-labelled methane (^3H-CH_4) to water sample
- Incubate for some time
- Oxidation produces tritium-labelled water:

$$^3H\text{-}CH_4 + O_2 \rightarrow {}^3H\text{-}H_20 + CO_2$$

- Sparge the sample with nitrogen, to remove un-reacted ^3H-CH_4
- Measure radioactivity in the water sample
- Calculate what fraction of the initially added activity was converted to water

- Advantage: Can in principle work with very low concentrations
- Disadvantage: Working with radioactive material
  - May carry risk to health
  - May require inconvenient HSE routines

Tritium method - Experimental approach

- Stock solution of tritium-labelled methane was created
- Normal methane was added to water samples, and pre-incubated
- Stock solution of labelled methane was added to water samples
  - Activity in some samples were measured by scintillation counter
- Samples were incubated for 0, 4, 13, 17 and 27 days, at 5 °C
- At each sampling:
  - Seal was broken, and an amount of water extracted
  - Water was sparged with nitrogen for two hours
  - Remaining activity in water measured by scintillation counter
  - Note: Bottles were sacrificed for sampling, each datapoint comes from a separate bottle

Tritium method – Analysis of results

Assumptions:

- As time goes to infinity, all the labelled methane will be converted to labelled water
- Reaction is first-order in methane concentration, giving a constant half-life
- After sparging, the 0-day incubation samples had some remaining activity
  - We called this a “background”, and assumed that all sparged samples would have this

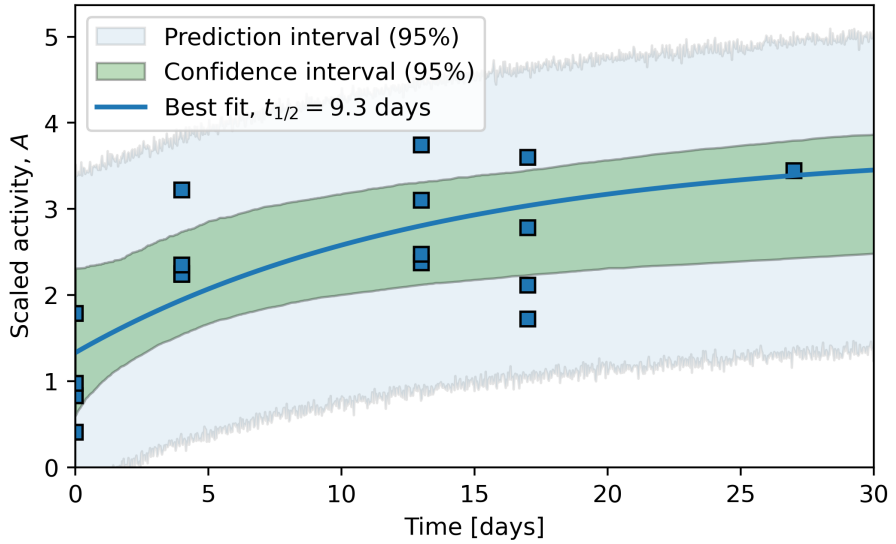
This leads to a generating model for the activity, $A(t)$, in the samples:

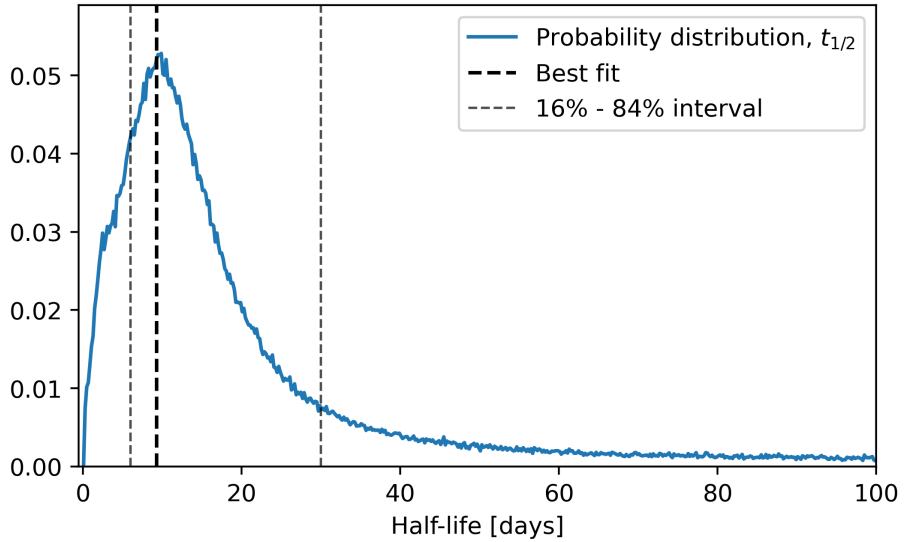
$$A(t) = (A_{tot} - A_b)\left(1 - \exp\left(-\ln 2 \frac{t}{t_{1/2}}\right)\right) + A_b$$

Activity starts at A_b at $t = 0$, and goes towards A_{tot}, with half-life $t_{1/2}$.

Tritium method – Analysis of results

- Python library lmfit[4] was used to fit model parameters to all datapoints simultaneously
- Python library emcee[5] was used to do Markov-Chain Monte Carlo sampling, to estimate uncertainty
- Showing results from one of three sets of experiments (see report and paper for rest).
- Results from these samples:
  - Most likely half-life: 9.3 days
  - 16% – 84% interval: 6 – 30 days





[4] https://lmfit.github.io/lmfit-py/

[5] https://emcee.readthedocs.io/en/stable/

Other methods we have used

GC-FID:

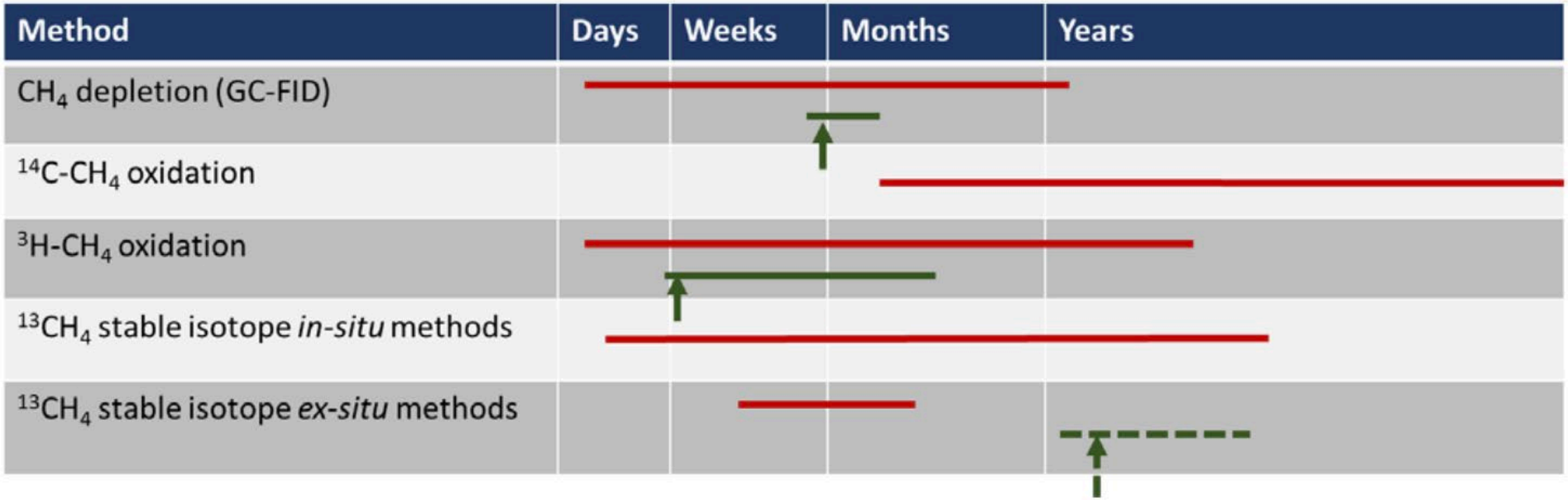
- Advantages:
  - Easy to use, familiar instrument, no HSE issues
  - Does not require fancy isotopically labelled methane
  - We get much better repeatability across parallel samples than with the tritium method
- Disadvantages:
  - Cannot work with as low concentrations as isotope methods
  - Must measure in gas phase, and calculate back to water concentration via calibration curve

^{13}C-labelled methane:

- Oxidation produces labelled CO^2, which must be transferred to gas phase
- Reaction rate can be determined from ratio between ^{13}C and ^{12}C in gas phase
- Advantage: Stable isotope, no HSE issues
- Disadvantage: Was not found to be as straightforward and reliable as other methods
  - Caveat: We do not have Isotope Ratio Mass Spectrometer in-house, and had to send samples away

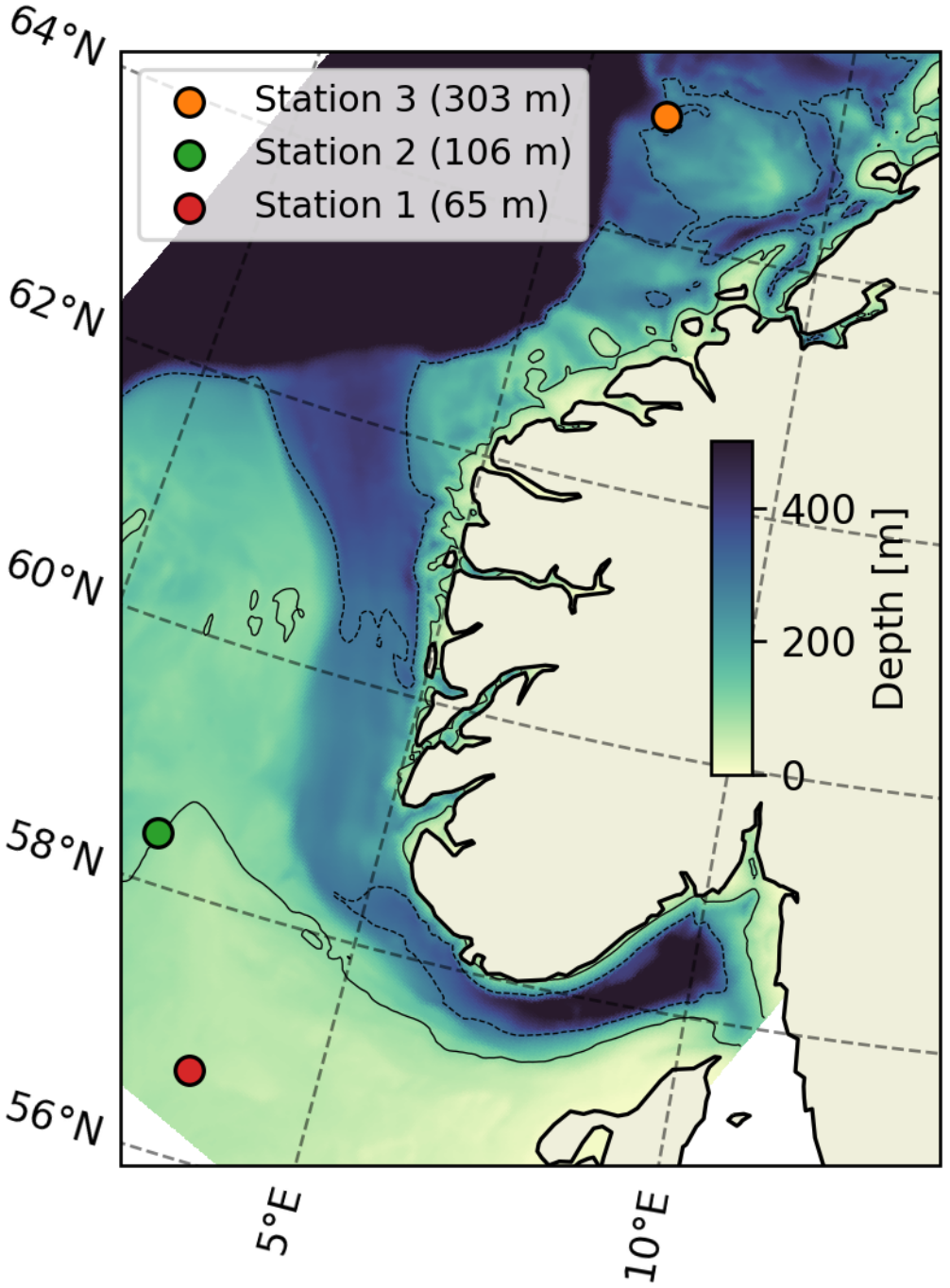
Other methods we have used

- Green lines show results from our studies (green arrows are harmonic average)
- ^{13}C data (dashed greed lines) have uncertain quality



Back to modelling

- Will go through some example results
  - Single bubble model
  - Diffusion-reaction model for dissolved methane
  - Annual averages
- Three locations, picked for different depths
- Two in North Sea, one in Norwegian Sea
- Water column data from NorShelf[6], a 2.5 km ocean model by MET Norway
- Atmosphere data from ERA5[7]



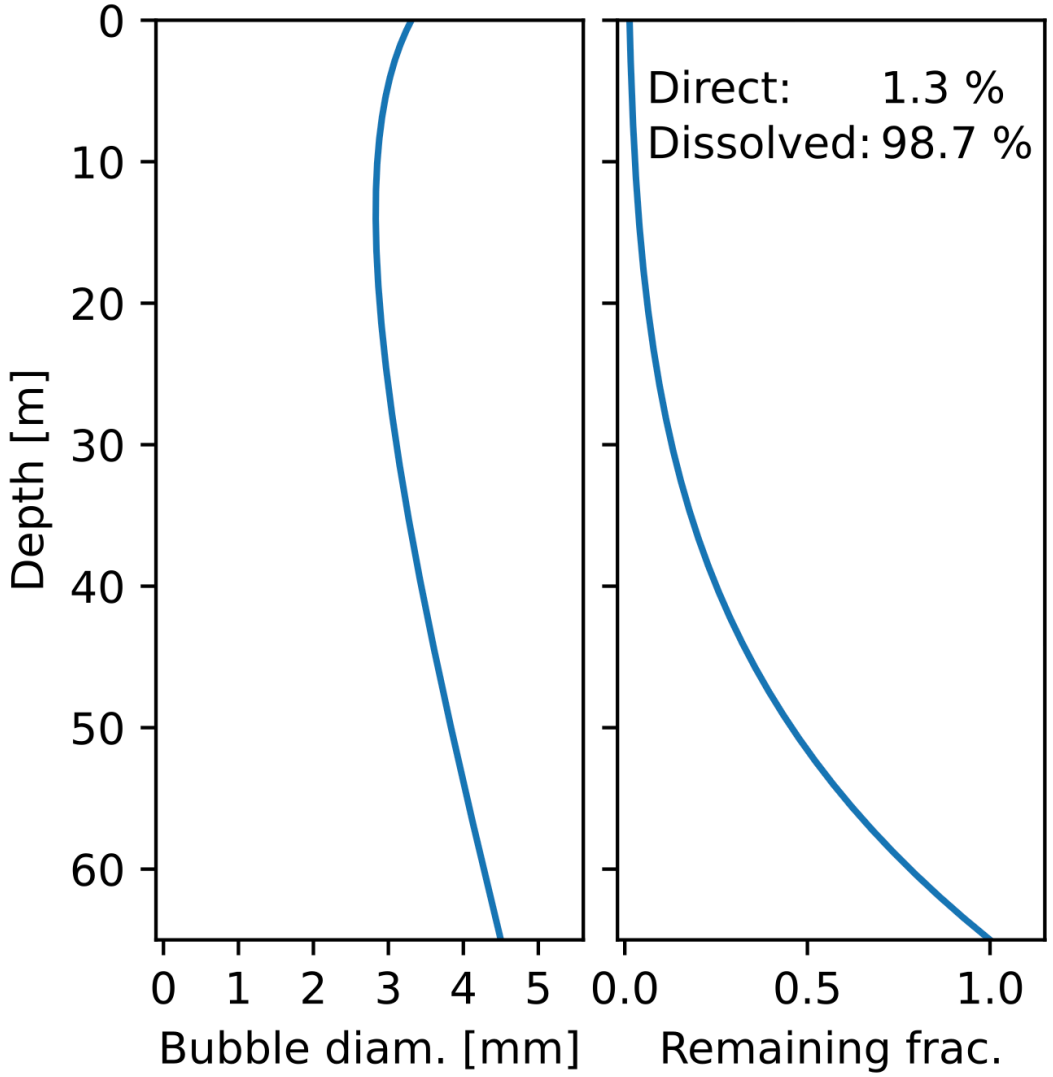
[6] Röhrs, J.; Sperrevik, A. K.; Christensen, K. H. NorShelf: A reanalysis and data-assimilative forecast model for the Norwegian Shelf Sea; Report 04/2018; Norwegian Meteorological Institute, 2018.

[7] Hersbach, H.; Bell, B.; Berrisford, P.; Hirahara, S.; Horányi, A.; Muñoz-Sabater, J.; Nicolas, J.; Peubey, C.; Radu, R.; Schepers, D.; et al. The ERA5 global reanalysis. Quarterly Journal of the Royal Meteorological Society 2020, 146, 1999–2049.

Example results – Bubble model

Station 1: 65 m depth

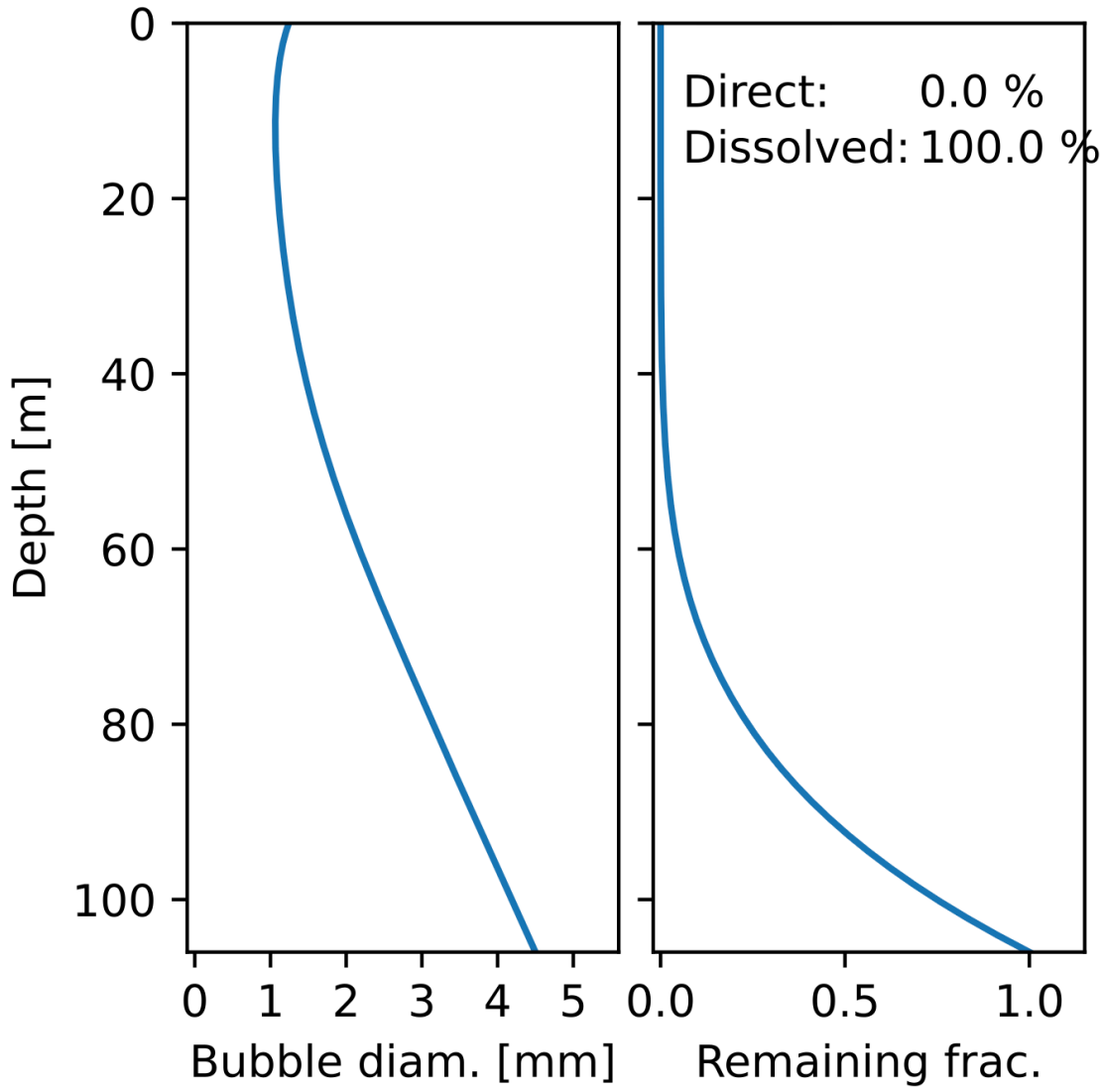
- Bubble reaches the surface
- 98.7% of original methane dissolved
- 1.3% of methane reaches atmosphere directly with bubble
- Assumptions:
  - Initial bubble size 4.5 mm
  - Initially pure methane
  - Clean bubble mass transfer (no surfactants)



Example results – Bubble model

Station 2: 106 m depth

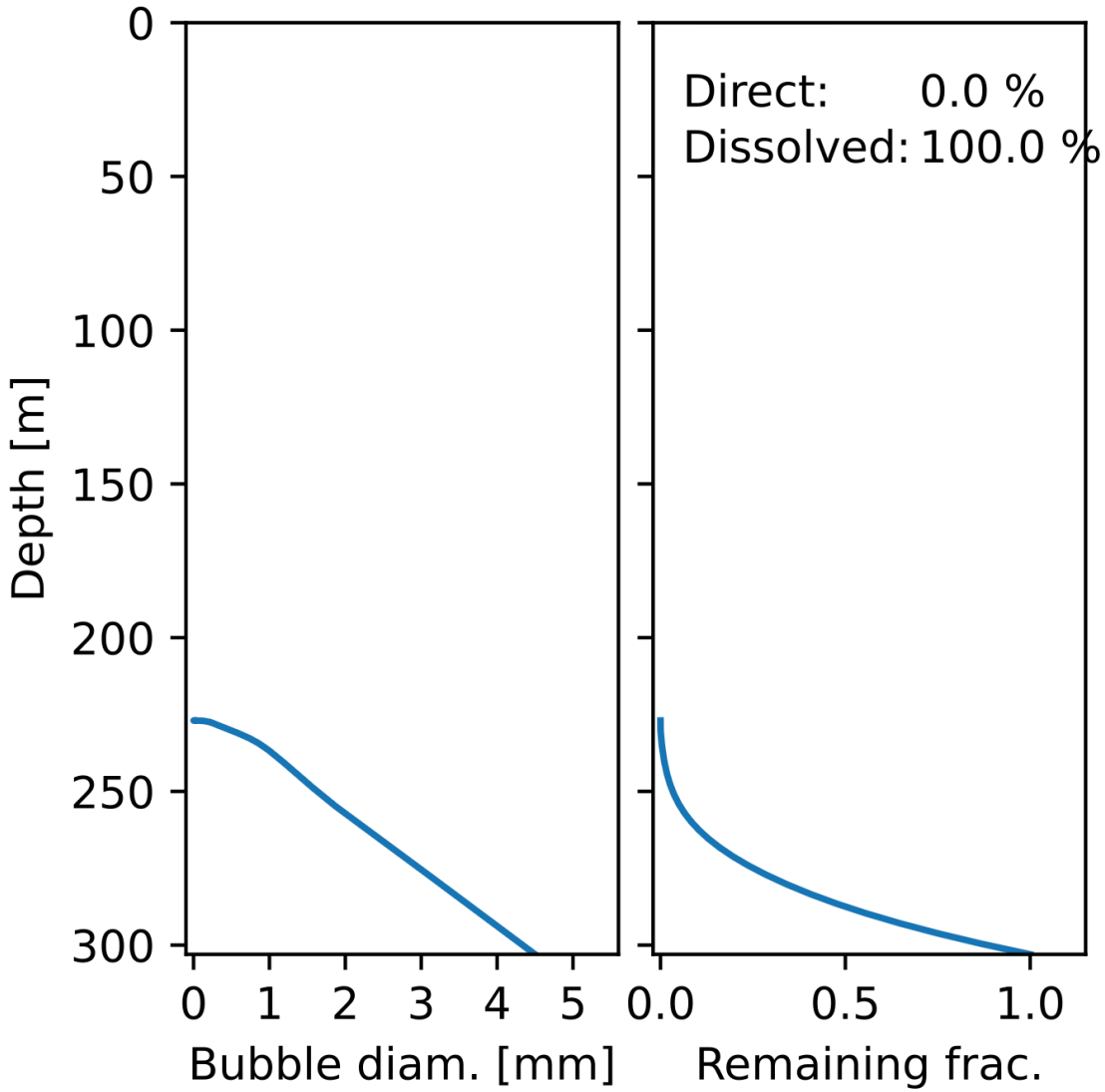
- Bubble reaches the surface
- 100% of original methane dissolved
- 0% of methane reaches atmosphere directly with bubble
- Assumptions:
  - Initial bubble size 4.5 mm
  - Initially pure methane
  - Clean bubble mass transfer (no surfactants)



Example results – Bubble model

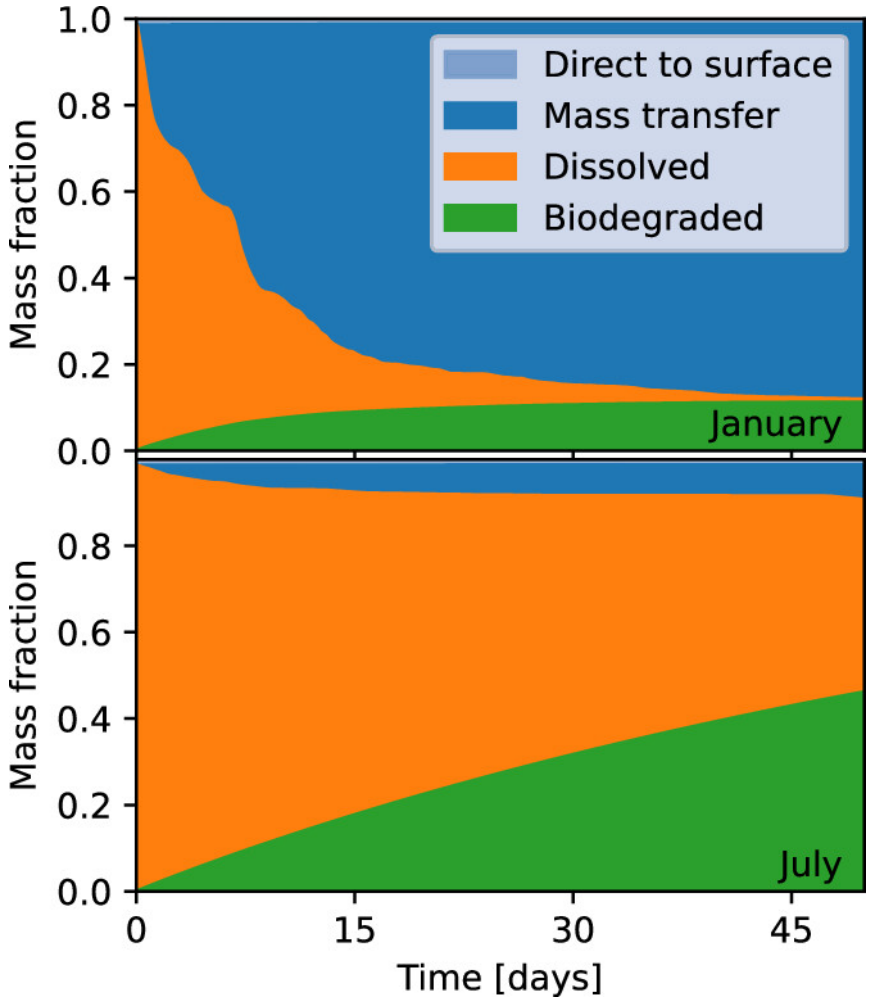
Station 3: 303 m depth

- Bubble does not reach the surface
- 100% of original methane dissolved, mainly in the first 50 m of rise
- 0% of methane reaches atmosphere directly with bubble
- Assumptions:
  - Initial bubble size 4.5 mm
  - Initially pure methane
  - Clean bubble mass transfer (no surfactants)

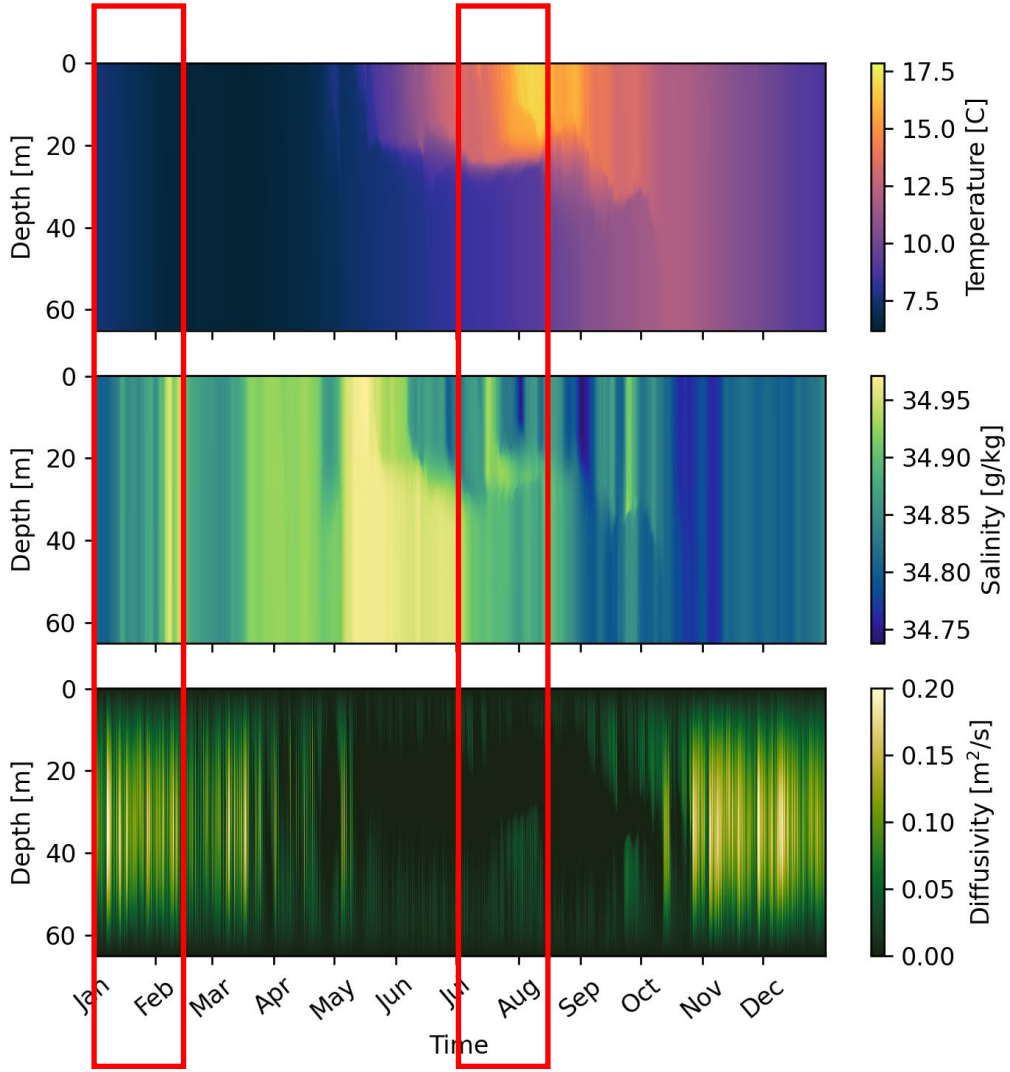


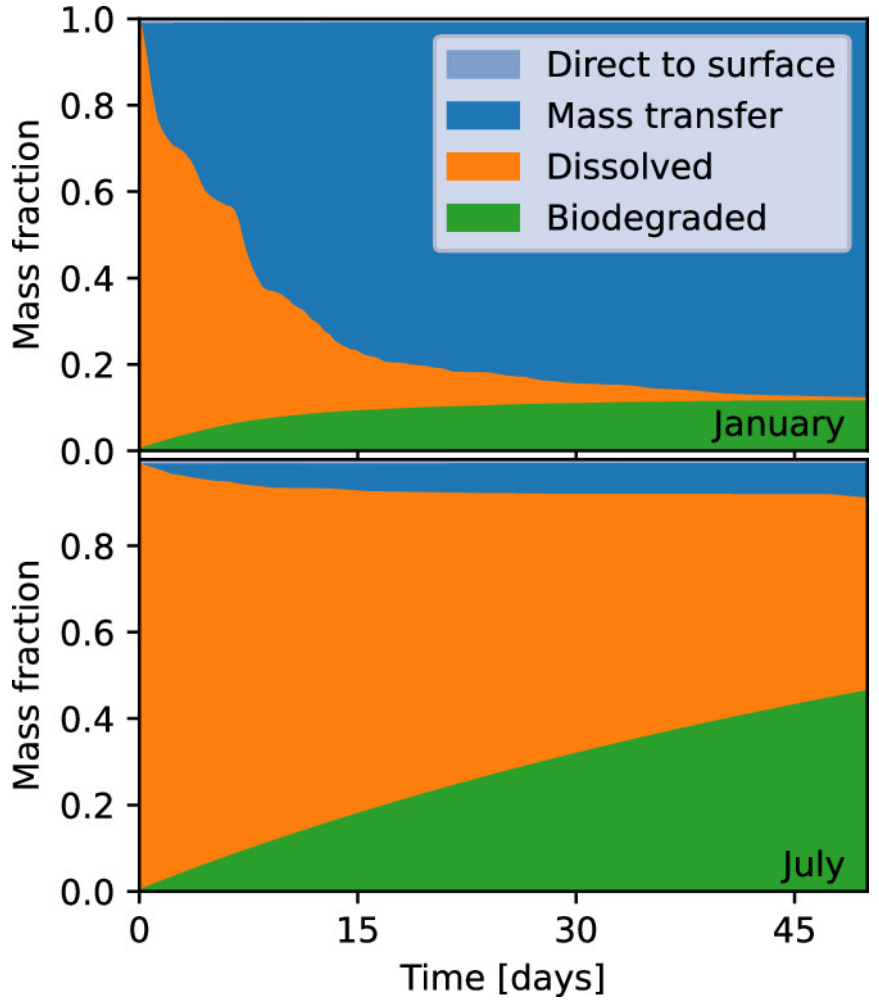
Example results – Diffusion-reaction model

- Bubble is released at 65 m depth, in the North Sea
- Within minutes, bubble carries about 1% of methane directly to the surface
- Over a period of days and weeks:
  - Dissolved methane is mixed vertically
  - Some methane escapes to atmosphere via mass transfer
  - Some methane is oxidised by microbes
- Half-life of methane in this example is set (somewhat arbitrarily) to 50 days
- Top panel: January, homogenous water column
  - Methane escapes relatively quickly to atmosphere
- Bottom panel: July, stratified water column
  - Escape to atmosphere slower, biodegradation dominates



Example results – Diffusion-reaction model





Annual average results

For each of the three chosen locations:

- Obtain one year (2019) of water column and atmosphere data
- Generate one year of diffusivity data with GOTM
- Run the TAMOC bubble model and the diffusion-reaction solver
  - 36 start times (bubble release), evenly spaced throughout the year
  - Run diffusion-reaction model until no methane is left in water column
  - When stepping past 31st of December, 2019, loop back to 1st of January, 2019
- Repeat for different microbial oxidation half-lives, ranging from 1 to 500 days

Annual average results

Fate of the methane depends on:

- Depth
- Typical conditions at location (stratification)
- Microbial oxidation rate (half-life)

Discussion of results – Biot number

Mass-transfer Biot number

$$Bi = \frac{k_w L}{D}$$

- $Bi \ll 1$: Rate of escape to atmosphere limited by mass transfer coefficient, k_w
- $Bi \gg 1$: Rate of escape to atmosphere limited by diffusivity, D

- Station 1 (65 m depth)
  - Mostly limited by mass transfer
- Station 2 (106 m depth)
  - Mostly limited by mass transfer, except in summer (May-Aug)
- Station 3 (303 m depth)
  - Always limited by diffusion

Discussion of results – Ventilation rate

- We define the ventilation rate as

$$v = \dot{M}/M$$

- $\dot{M}$ is rate of escape to atmosphere, M is remaining dissolved methane.
- v has units of inverse time, just like the microbial oxidation rate, k_1.

- $v \gg k_1$: Methane escapes faster than it is oxidised
- $v \ll k_1$: Methane is oxidised faster than it escapes
- Ventilation rate and oxidation rate gives branching ratio between the two fates (for the dissolved part)

Discussion of results – Ventilation rate

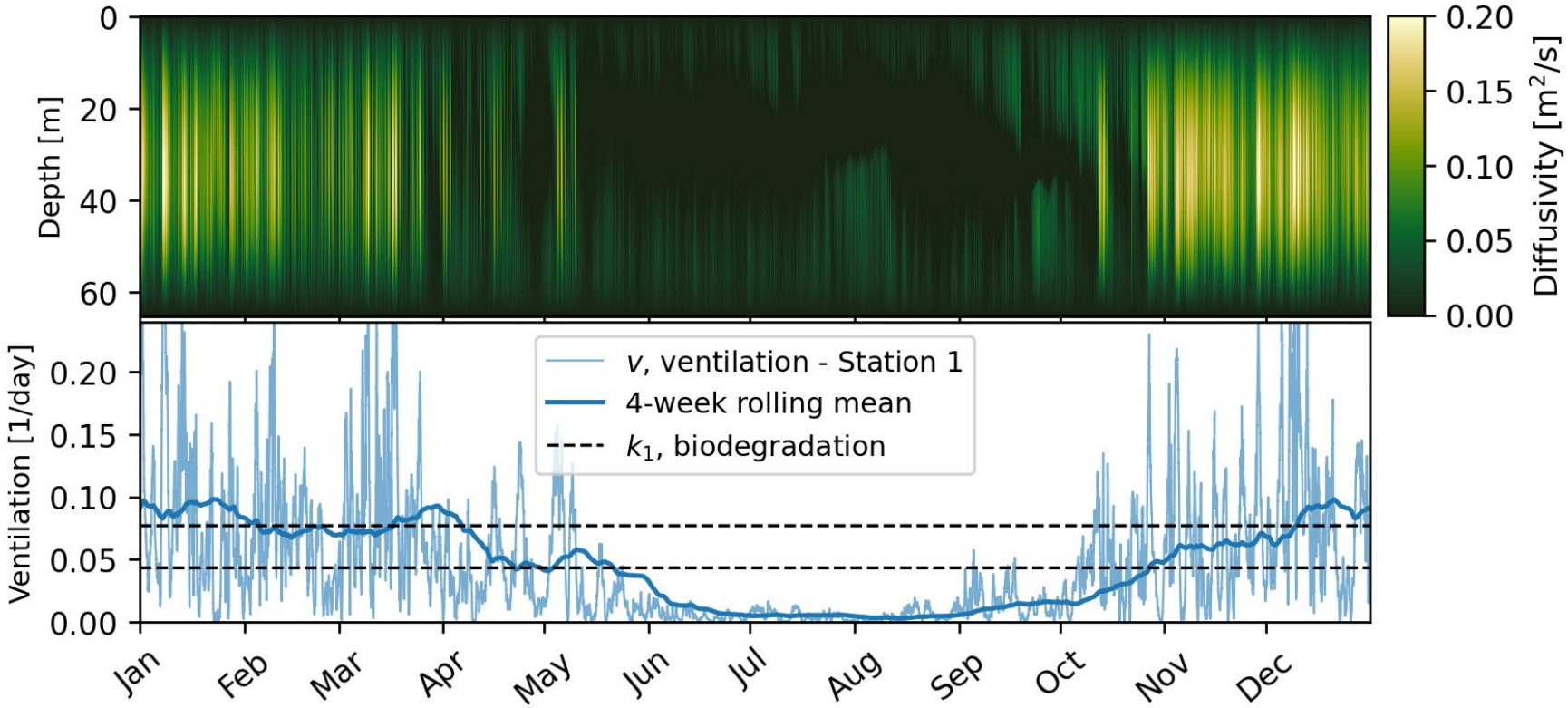
In the following I will compare:

- Ventilation rates found from modelling, for the three stations
  - Will show how this depends on diffusivity
- Microbial oxidation half-lives in the range 9 – 16 days (constant throughout the year)

Discussion of results – Ventilation rate

Station 1 (65 m depth)

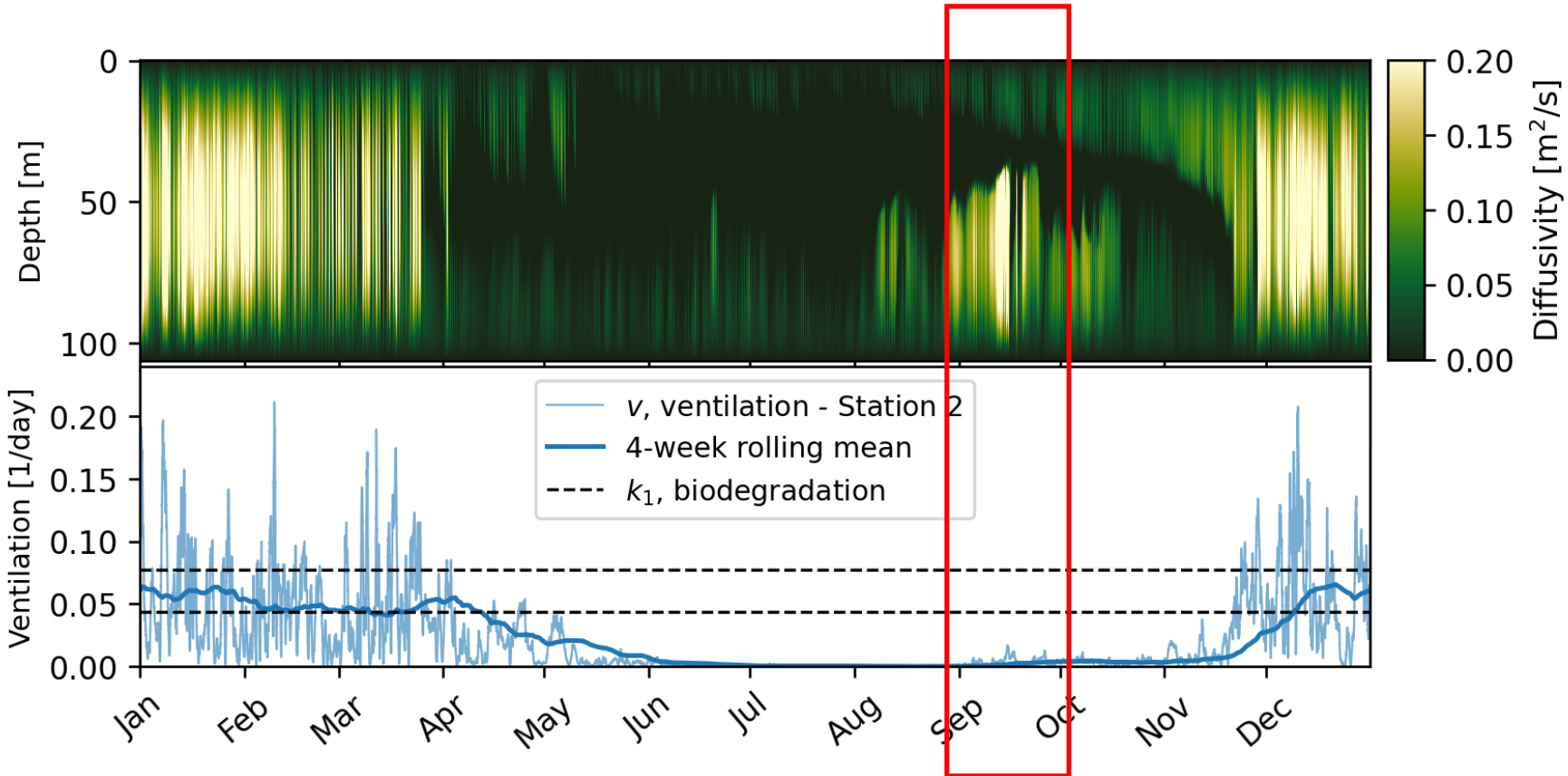
- Ventilation rate and biodegradation rate comparable for most of the year
- Biodegradation dominating during summer, when exchange to the surface is low



Discussion of results – Ventilation rate

Station 2 (106 m depth)

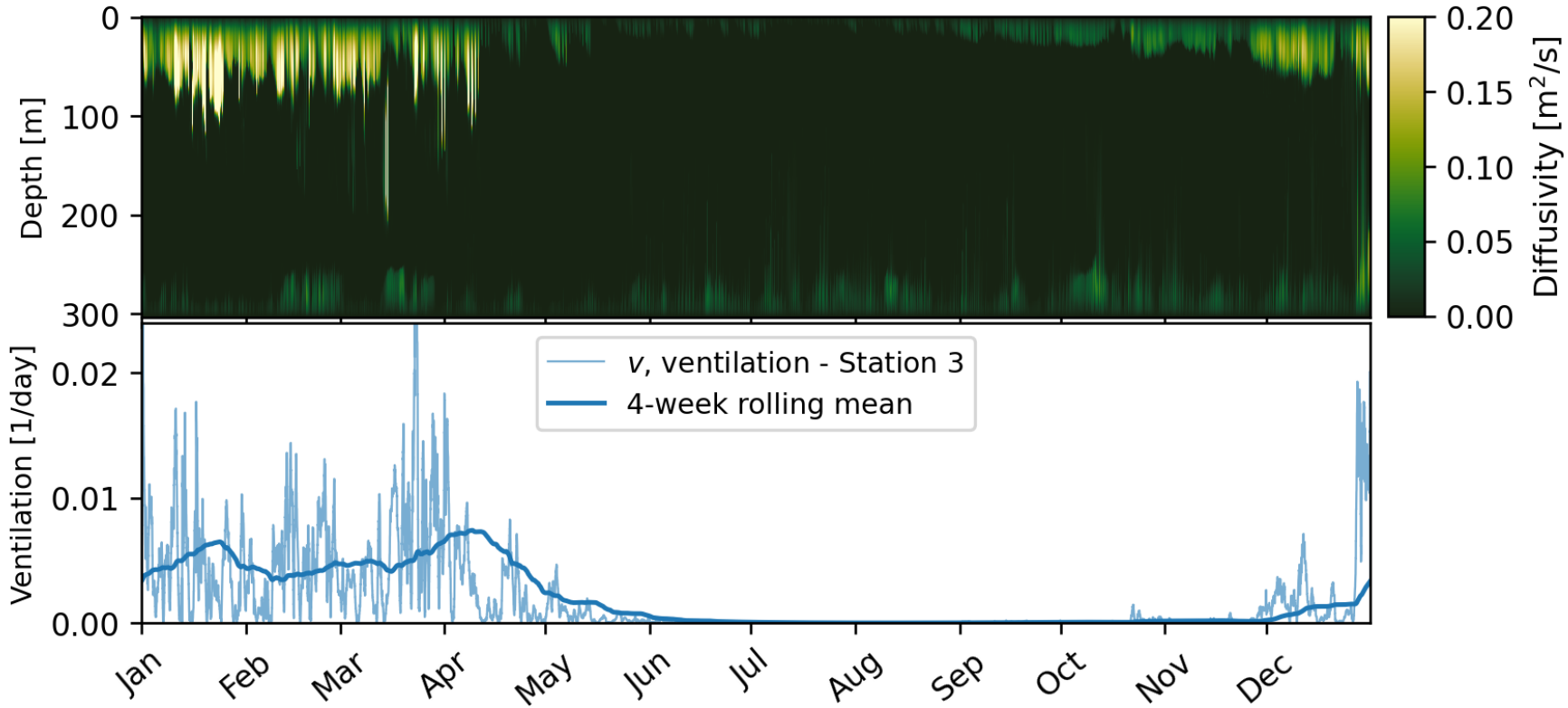
- Ventilation rate and biodegradation rate comparable during winter
- Biodegradation dominating during (May – Nov), when exchange to the surface is low



Discussion of results – Ventilation rate

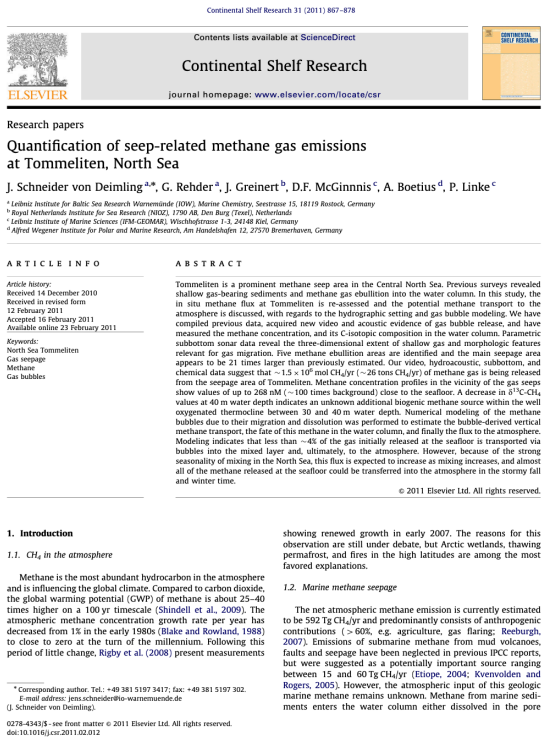
Station 3 (303 m depth)

- Biodegradation dominates throughout the year
- (biodegradation rates not visible on this scale, as ventilation rate is so slow)



Returning to the motivation

- How much of the methane does actually reach the atmosphere during winter?
- Answer: It depends
  - On the depth
  - On the mass-transfer coefficient
  - On the biodegradation rate
  - On the vertical diffusivity
- Sensitivity to different parameters depend on situation
  - Contribution of each parameter to uncertainty depends also on uncertainty in the parameter



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Research papers

Quantification of seep-related methane gas emissions at Tommeliten, North Sea

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ABSTRACT

Tommeliten is a prominent methane seep area in the Central North Sea. Previous surveys revealed shallow gas-bearing sediments and methane gas ebullition into the water column. In this study, the in situ methane flux at Tommeliten is re-assessed and the potential methane transport to the atmosphere is discussed, with regards to the hydrographic setting and gas bubble modeling. We have compiled previous data, acquired new video and acoustic evidence of gas bubble release, and have measured the methane concentration, and its C-isotopic composition in the water column. Parametric subbottom sonar data reveal the three-dimensional extent of shallow gas and morphologic features relevant for gas migration. Five methane ebullition areas are identified and the main seepage area appears to be 21 times larger than previously estimated. Our video, hydroacoustic, subbottom, and chemical data suggest that ~1.5 × 10^9 mol CH_4/yr (~26 tons CH_4/yr) of methane gas is being released from the seepage area of Tommeliten. Methane concentration profiles in the vicinity of the gas seeps show values of up to 268 nM (~100 times background) close to the seafloor. A decrease in $\delta^{13}C$-CH_4 values at 40 m water depth indicates an unknown additional biogenic methane source within the well oxygenated thermocline between 30 and 40 m water depth. Numerical modeling of the methane bubbles due to their migration and dissolution was performed to estimate the bubble-derived vertical methane transport, the fate of this methane in the water column, and finally the flux to the atmosphere. Modeling indicates that less than ~4% of the gas initially released at the seafloor is transported via bubbles into the mixed layer and, ultimately, to the atmosphere. However, because of the strong seasonality of mixing in the North Sea, this flux is expected to increase as mixing increases, and almost all of the methane released at the seafloor could be transferred into the atmosphere in the stormy fall and winter time.

1. Introduction

1.1. CH_4 in the atmosphere

Methane is the most abundant hydrocarbon in the atmosphere and is influencing the global climate. Compared to carbon dioxide, the global warming potential (GWP) of methane is about 25–40 times higher on a 100 yr timescale (Shindell et al., 2009). The atmospheric methane concentration growth rate per year has decreased from 1% in the early 1980s (Blake and Rowland, 1988) to close to zero at the turn of the millennium. Following this period of little change, Rigby et al. (2008) present measurements showing renewed growth in early 2007. The reasons for this observation are still under debate, but Arctic wetlands, thawing permafrost, and fires in the high latitudes are among the most favored explanations.

1.2. Marine methane seepage

The net atmospheric methane emission is currently estimated to be 592 Tg CH_4/yr and predominantly consists of anthropogenic contributions (>60%, e.g. agriculture, gas flaring; Reeburgh, 2007). Emissions of submarine methane from mud volcanoes, faults and seepage have been neglected in previous IPCC reports, but were suggested as a potentially important source ranging between 15 and 60 Tg CH_4/yr (Etiope, 2004; Kvenvolden and Rogers, 2005). However, the atmospheric input of this geologic marine methane remains unknown. Methane from marine sediments enters the water column either dissolved in the pore

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"Modeling indicates that less than ~4% of the gas initially released at the seafloor is transported via bubbles into the mixed layer and, ultimately, to the atmosphere.

However, […] almost all of the methane released at the seafloor could be transferred into the atmosphere in the stormy fall and winter time."

Schneider von Deimling et al. (2011)

https://doi.org/10.1016/j.csr.2011.02.012

What drives the uncertainty in the outcome?

Probably not so important for uncertainty:

- Depth
  - Certain, if talking about a specific case.
  - Less certain, if talking about overall distribution of seeps.
- Mass-transfer coefficient
  - Calibrated against large datasets.
  - Seems to be on reasonably robust footing.
- Bubble model
  - Different models broadly agree about how fast the bubbles dissolve.
  - Results also seem consistent with data from echo sounders.

What drives the uncertainty in the outcome?

Probably more important for uncertainty:

- Vertical diffusivity
  - Obtained from models.
  - Hard to compare against measurements, “eddy diffusivity” is not a thing you measure.
  - Can be compared against observed temperature and salinity profiles, and checked for consistency with time development.
- Biodegradation rate
  - Rates found in literature vary by several orders of magnitude (days to years).
  - Not clear if the variations are due to actual variations in conditions, or experimental factors.
  - Not clear what rate to use for a given case.

Conclusions

If we want to estimate fluxes of methane from seeps to the atmosphere we should:

- Use (simple) models to understand what drives uncertainty, and focus research
- Investigate how well modelled vertical diffusivity represents tracer transport
  - How fast is transport during winter, with homogeneous water column
  - How effective a barrier is the pycnocline during summer, with stable stratification
- Do large, systematic studies of microbial oxidation rates
  - Water from relevant locations
  - Range of relevant concentrations
  - Many replicates
  - Long(er) incubation times
  - Time series showing decay
  - Ideally also genetic studies of microbial communities

Questions?

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