

Physical and chemical processes affecting release of CO₂ at the seafloor

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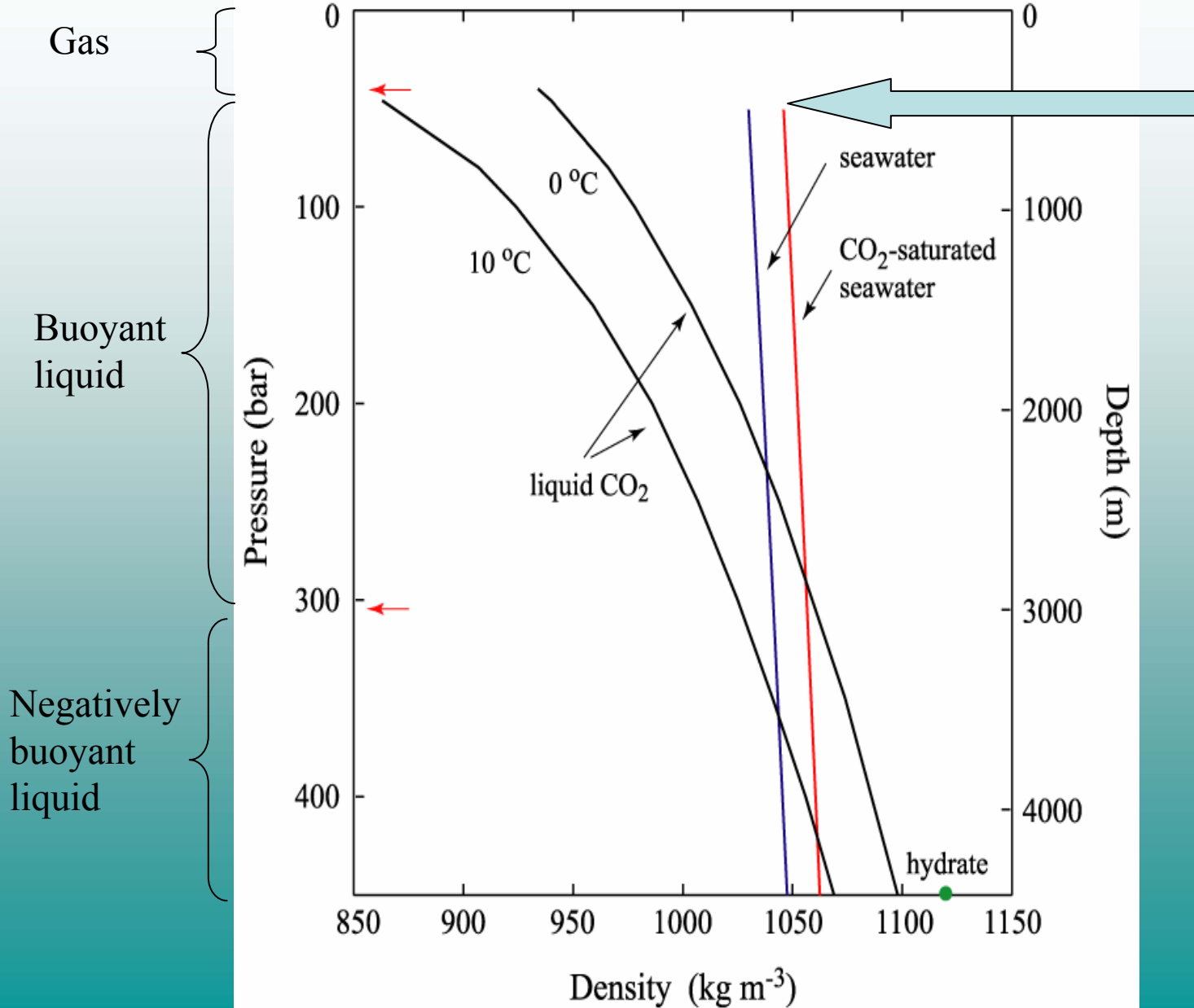
Knowledge base relevant to CO₂ at the seafloor

- General theoretical, modelling and laboratory research on direct ocean storage of CO₂
- 10-15 years ago, the "lake option" was proposed and studied theoretically based on lab properties.
- Lessons from observations of natural CO₂ vent systems, brine pools and gravity currents.
- Recent small scale experimental work with advanced ROV and observational techniques (Brewer et. al.).
- No *in situ* experiments on the bottom boundary layer (BBL) scale.
- Few published studies on deep sea BBL turbulence and hydrodynamics for an active tracer like CO₂.

Outline

- The ways in which CO₂ may be released at the seafloor
 - Gas bubbles
 - Liquid droplets
 - Liquid CO₂ "lake" or "pool"
 - Hydrate
 - Solid (dry ice)
 - Predissolved in water
- Dissolution from a deep sea pool of liquid CO₂
- Mixing in the benthic boundary layer
- Gravity current dynamics
- Challenges and ongoing numerical modelling

Density of seawater, liquid CO₂, hydrate and CO₂-enriched seawater



Dissolving CO₂ in seawater always increases density of seawater.

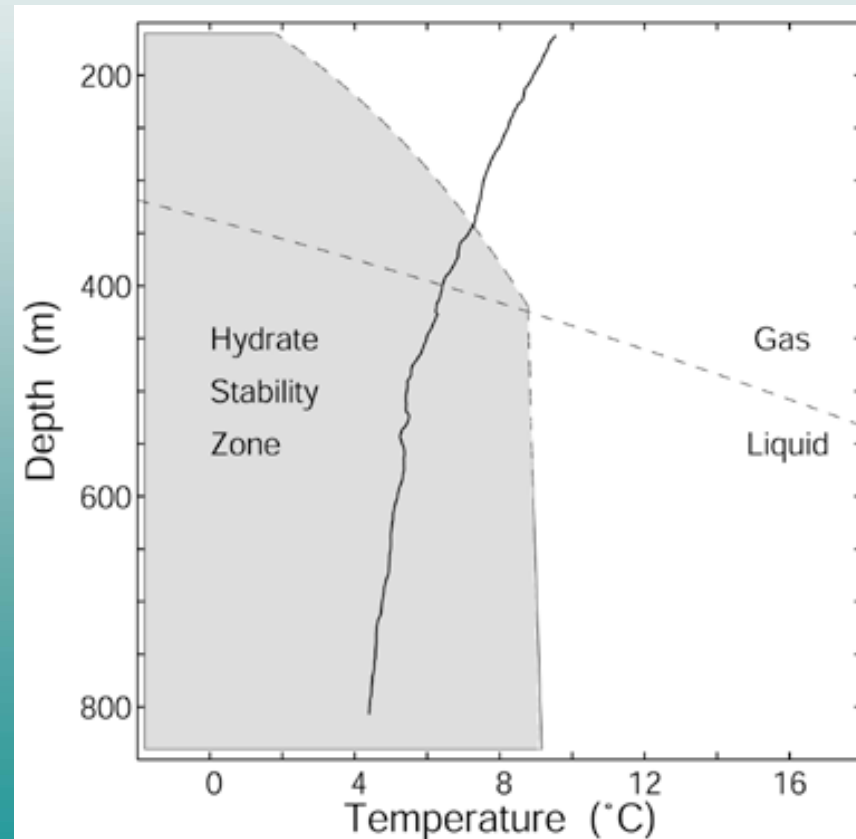
(Fer & Haugan, Limnol. & Oceanogr., 2003)

Different ways of releasing CO₂

- 1: Seepage at less than 400m depth makes CO₂ bubbles. However, CO₂ gas hydrates may form for water depths as shallow as 100m if water is sufficiently cold.

An interim hydrate stage with subsequent dissolution will contribute to keeping the CO₂ in place rather than escaping upwards in the gas phase.

One may speculate that a dense, CO₂-enriched seawater layer could be created near the seafloor if seepage is slow.



Different ways of releasing CO₂

2: Slow seepage of small droplets into quiescent water in isolated depression or non-tidal basin may also create high concentration of dissolved CO₂.

3: Seepage of pore water with predissolved CO₂.

All of 1-3 may be relevant to geological subseabed storage. Impact research for direct ocean storage could also be appropriate for seepage from geological storage (Haugan, Waste Mgmt., 1997).

4: Direct deep ocean storage of seawater with predissolved CO₂ (e.g. Saito et al. (2000) Gas Lift Advanced Dissolution concept).

Different ways of releasing CO₂

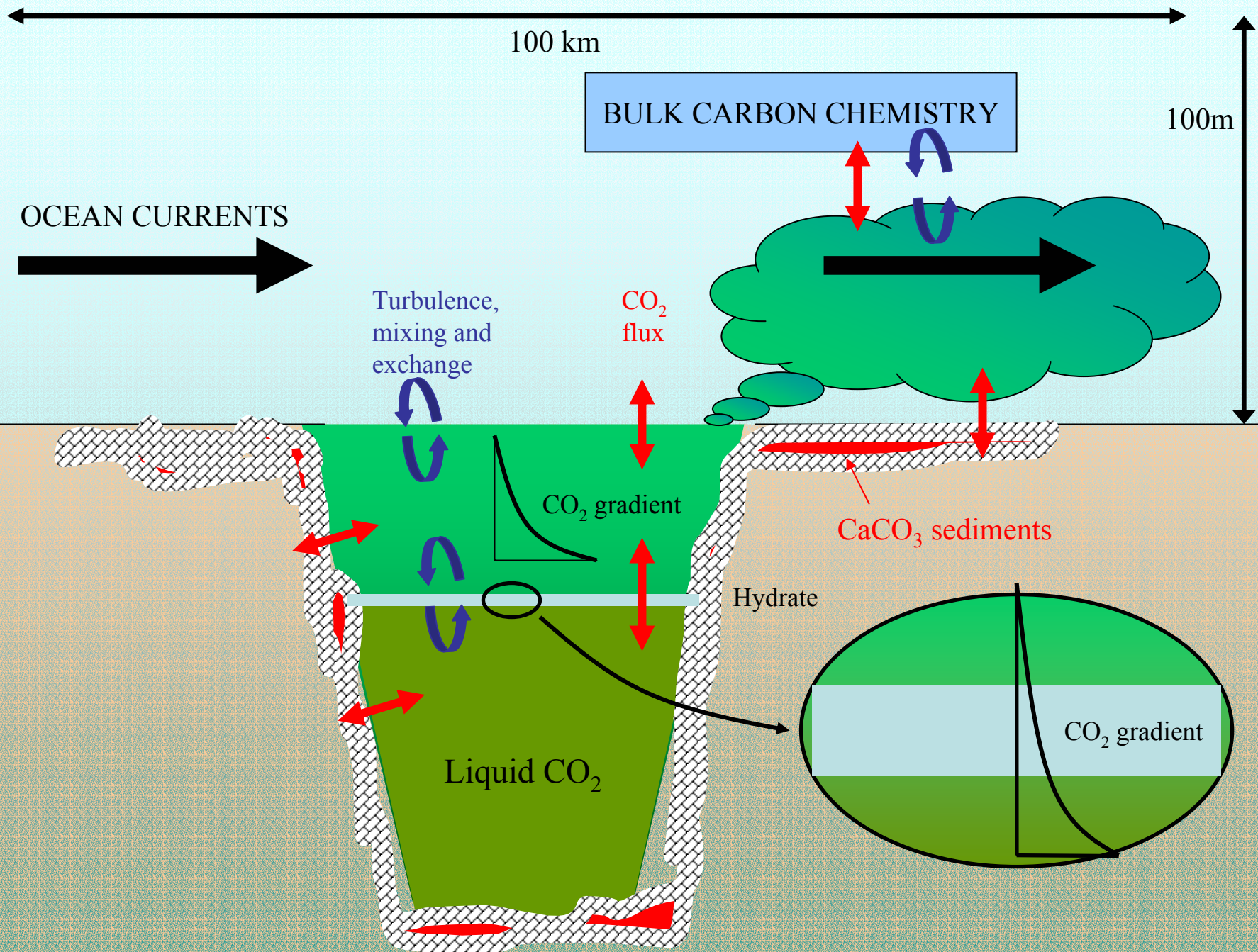
All of 1 - 4 will create a layer of dense, low pH water spreading along the seafloor as a gravity current unless obstructed by topography.

5: Release at less than 400m in warm water implies rapid escape to the atmosphere.

6: Release of dry ice is impractical and costly.

7: Release of buoyant droplets and dynamics of droplet plumes are covered in talk by Chen.

8: Main focus hereafter on deep sea pool dissolution, vertical mixing, gravity current.



Release from a pool of CO₂

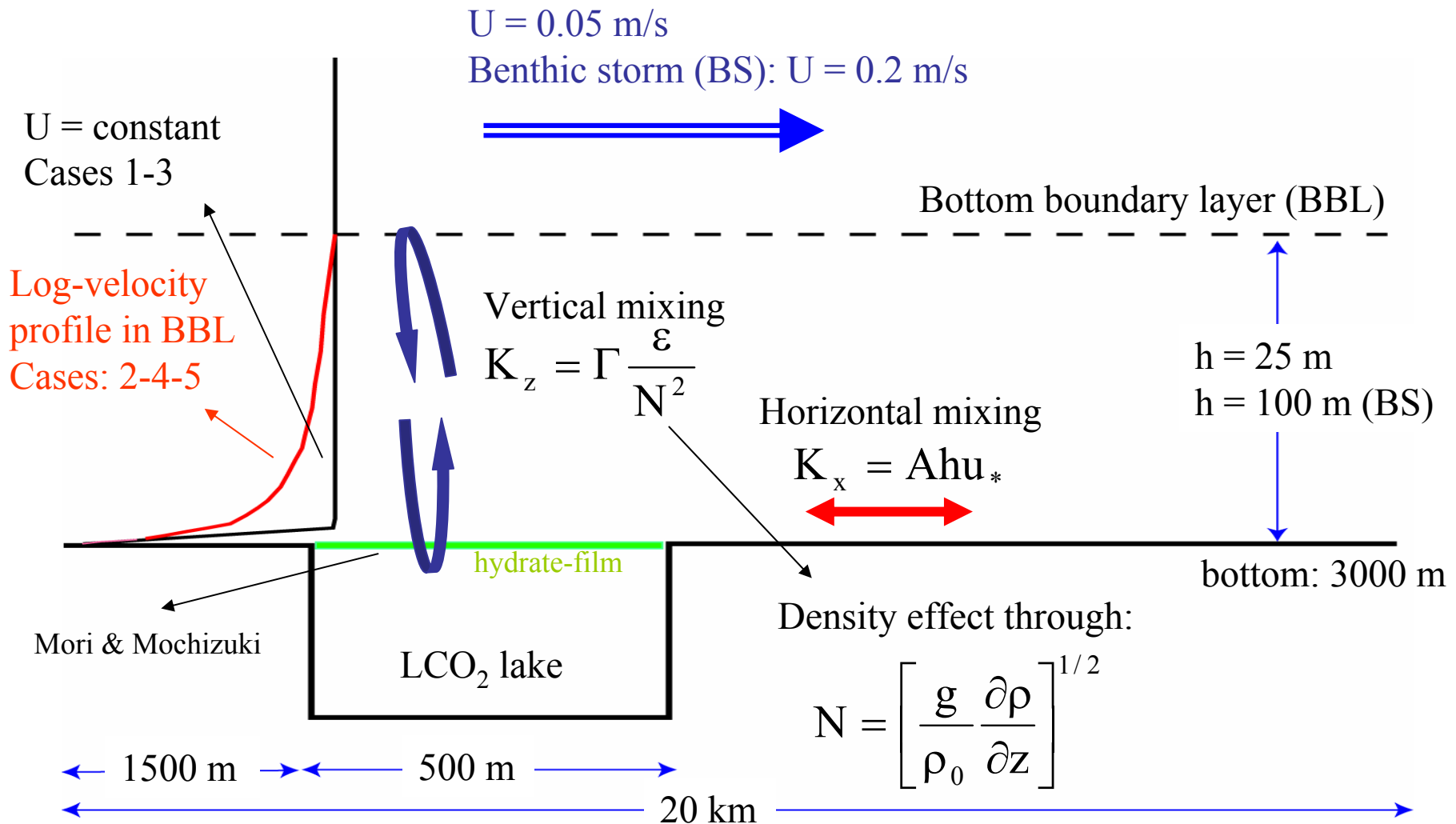
- One experiment with CO₂ exposed to natural currents at 684m depth (Brewer et al., J.O., 2004, in press)
- One experiment with controlled dynamic perturbation (thruster and wavemaker) ~ 50 liter, 3940m:



Preliminary results from controlled experiment

- Interfacial wave dispersion relation from analysis of video imagery for various wavemaker wavelengths
 - Interfacial deep "water" gravity waves
- Stability – instability transition from sequence of thruster experiments
 - Critical velocity identified (~ 18 cm/s at 3940m depth)
- Energy content by wavelet analysis of interface
 - Peak energy at about 1cm, close to theoretical wavelength of maximum growth rate for Kelvin-Helmholtz instability between CO₂ and water (depending on interfacial tension)

A model with dissolution-turbulence coupling



Unsteady, 2D advection-diffusion equation:

$$\frac{\partial C}{\partial t} + \frac{\partial (uC)}{\partial x} + \frac{\partial (wC)}{\partial z} = \frac{\partial}{\partial x} \left(K_x \frac{\partial C}{\partial x} \right) + \frac{\partial}{\partial z} \left(K_z \frac{\partial C}{\partial z} \right)$$

Domain: 20 km x 200 m at flat ocean bottom at 3000 m depth.

Source: A liquid CO₂ lake of 500 m span at 1500-2000 m from origin.

Grid resolution: 250 m in horizontal, 1.25 - 5 m in vertical.

Simple BBL theory:

$$\varepsilon(z) = \frac{\tau_0}{\rho} \frac{\partial U}{\partial z} = \frac{u_*^3}{\kappa z}$$

where ε is turbulent kinetic energy dissipation rate,

τ_0 is shear stress at the bottom,

u_* is friction velocity, and

κ is von Karmans constant.

Mass transfer proportional to u^*

Capillary permeation model (Mori & Mochizuki, 1997)

Molar flux of liquid CO₂ into the water phase:

$$J_{\text{CO}_2} = \frac{1}{1 - \tilde{C}_s} \left[K_m \frac{\rho_{\text{mix}}}{M_{\text{mix}}} (\tilde{C}_s - \tilde{C}_{\text{amb}}) - \frac{\tilde{C}_s}{1 - \tilde{C}_s} J_w \right]$$

Molar flux of water through the hydrate:

$$J_w = f(\delta, M_{\text{mix}}, v_{\text{mix}})$$

Hydrate thickness:

$$\delta = f(\gamma, \phi, r_c, p, \tau, \eta_{\text{mix}}, C_s, n, K_m)$$

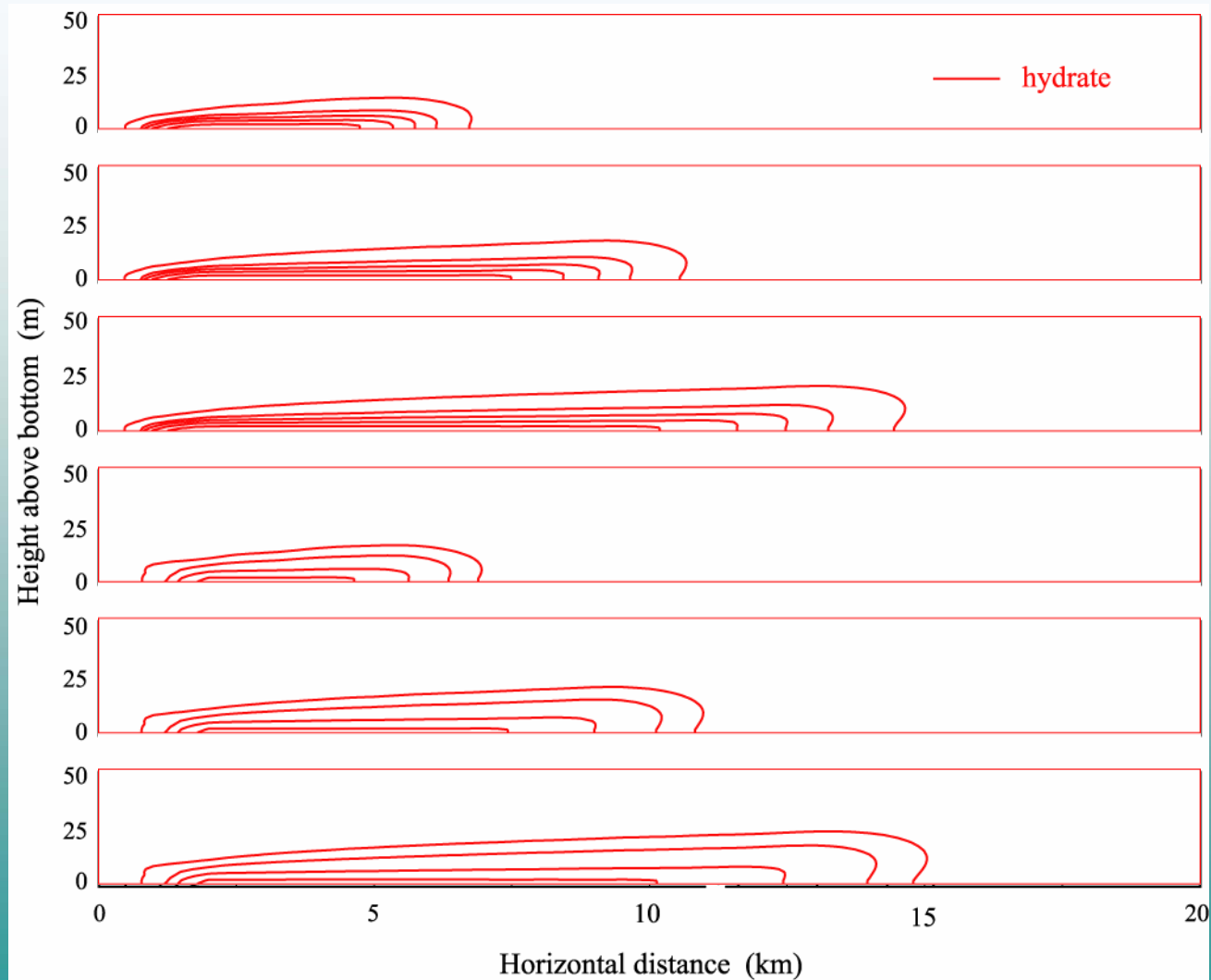
Tortuosity, $\tau = 2$; Porosity, $p = 10^{-3}$, Capillary radius, $r_c = 10^{-8}$ m

Interfacial tension, $\sigma = 19.4 \times 10^{-3}$ N m⁻¹, Contact angle, $\phi = 0^\circ$

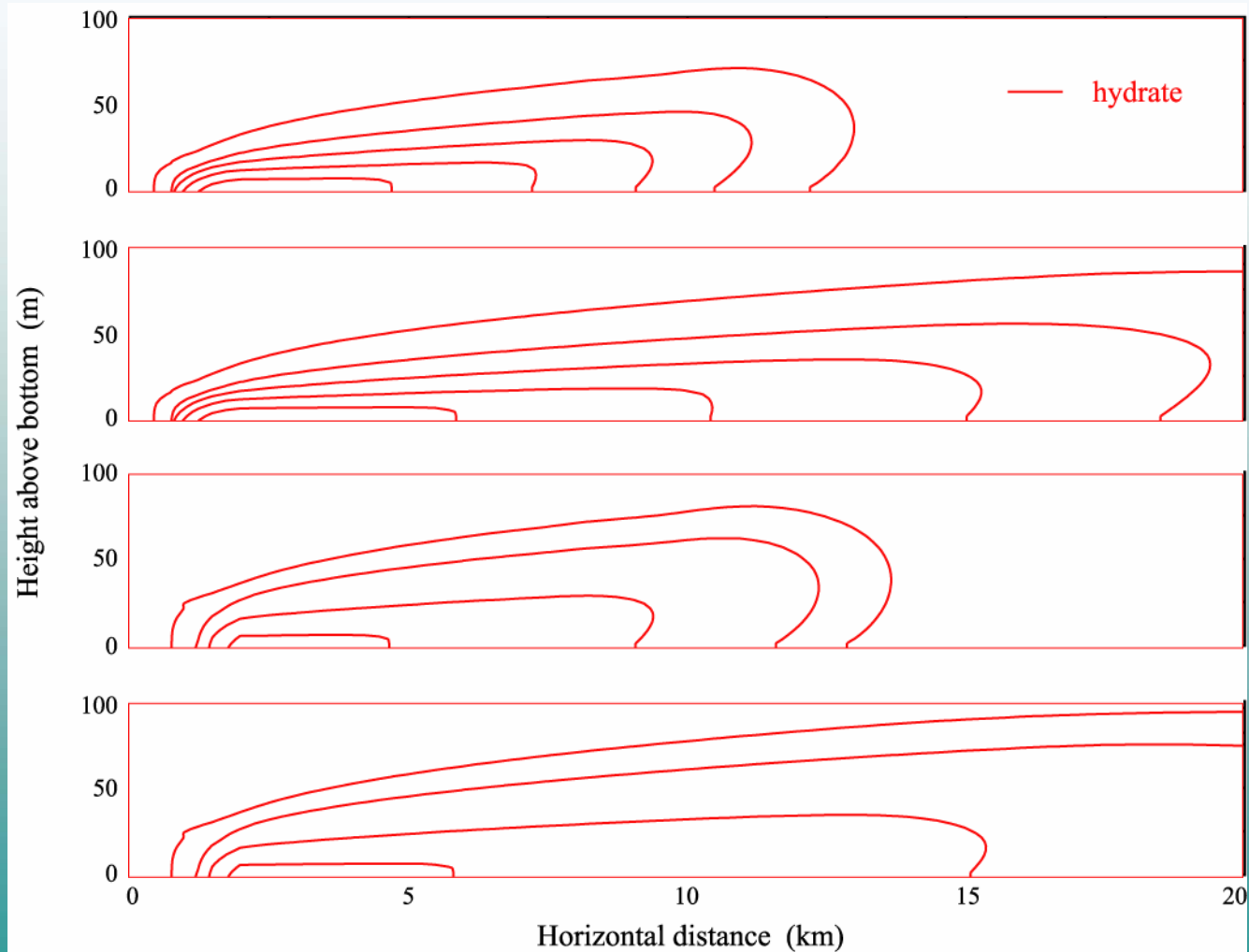
Hydration number, $n = 5.75$

Mass transfer coefficient: $K_m = 0.1 u_* \text{Sc}^{-0.67}$

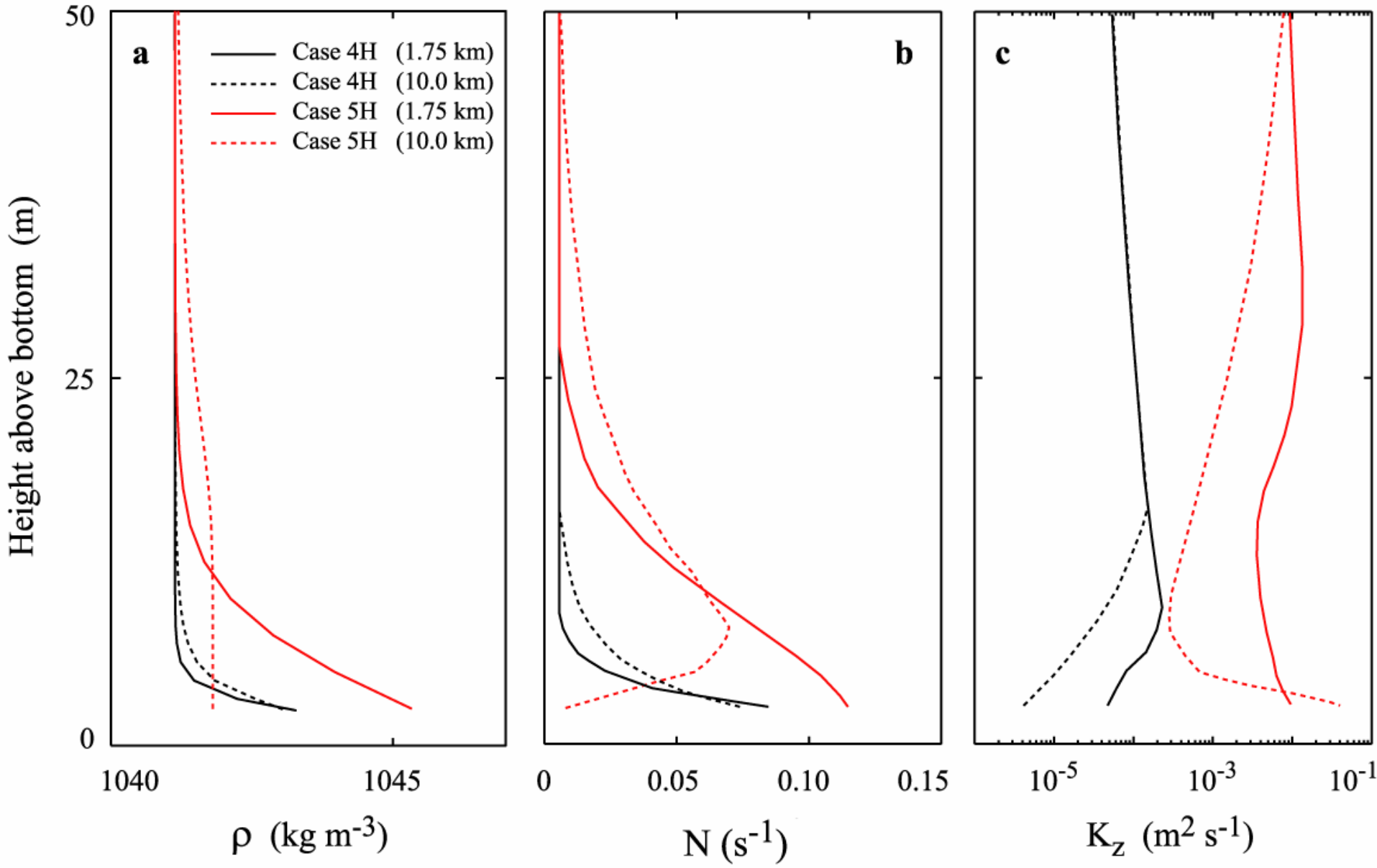
Standard case with BBL profiles (case 4 and 4H)



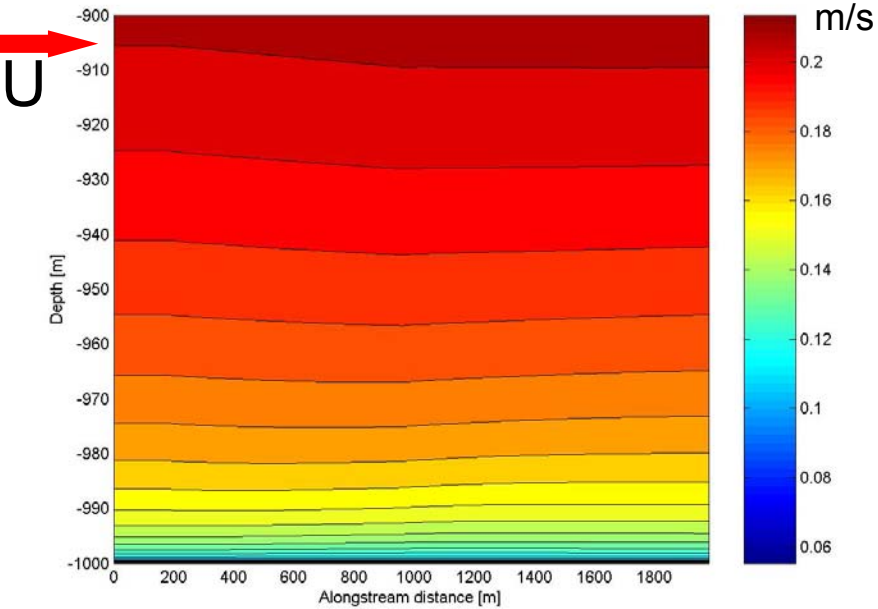
Benthic storm (case 5 and 5H)



Density, buoyancy frequency and vertical diffusivity profiles



Lower 100m

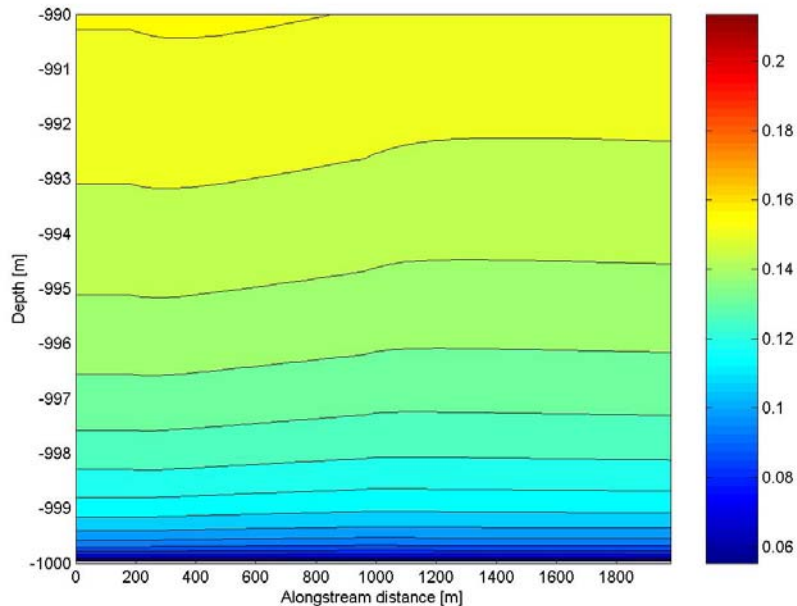


New model study in progress

Near equilibrium velocity (after 20 hours)

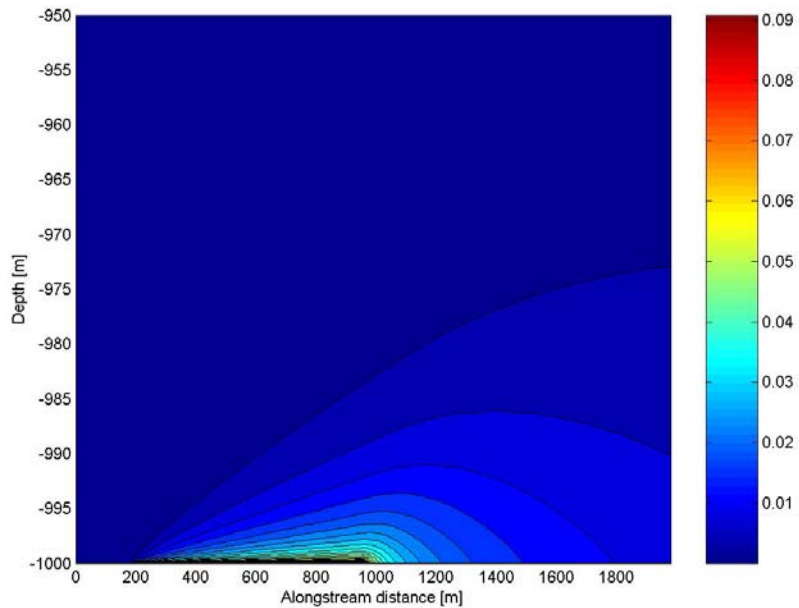
- 2000m by 1000m domain
- Specified CO_2 flux at seafloor between $x=200\text{m}$ and $x=1000\text{m}$
- $dx = 20\text{m}$
- dz varying from 7cm to several m in 400 points over 1000m.
- $k-\varepsilon$ turbulence model
- 20 cm/s background current from left to right in this case

Lower 10m

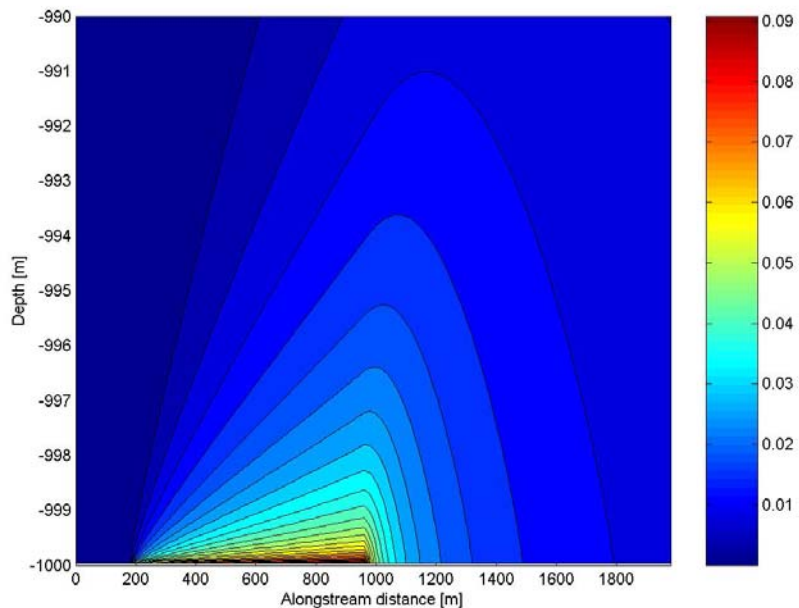


← CO_2 source →

Lower 50m



Lower 10m



CO₂ source

Carbon concentration (20 hours, 20 cm/s current)

Unpublished work addressing:

- Concentrations, pH values, horizontal and vertical scales
- Turbulence closure models
- Effects of ambient stratification
- Importance of density effect for different conditions
- Characterization of outflowing plume which may progress by stable gravity current dynamics

Conclusions on dissolution and mixing

- Dissolution and vertical mixing processes are coupled.
- In models, CO₂-induced stable stratification above the hydrate layer suppresses mixing considerably. This reduces the importance of hydrate to retard dissolution.
- Modelled dissolution from less than 1 cm y⁻¹ in purely diffusive regime with no velocity (Ohsumi, 1997) to about 10 cm y⁻¹ in low velocity regime and more than 1 m y⁻¹ in benthic storms (Fer & Haugan, 2003).
- Kelvin-Helmholtz instability has recently been experimentally confirmed in deep sea CO₂ experiment, but no build-up of dissolved CO₂ concentration.

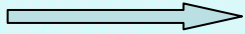
Conclusions on dissolution and mixing

- Topography may shield against strong currents and reduce dissolution (Kobayashi, 2003).
- On the other hand, recirculation could lead to build up of high concentration, and overspill associated with internal seiching could occur in sill basin.
- Long term large rate experiment would be required to check concentration profile in water above CO₂ pool.
- Brine pools are natural analogues, but often have larger density contrast.
- Outflowing plume from pool area is expected to form gravity current.

DEVELOPMENT OF GRAVITY CURRENT

100m

BACKGROUND CURRENT



Turbulence,
mixing and
exchange



CO₂
flux



BACKGROUND + GRAVITY CURRENT



CO₂ gradient



CaCO₃ sediments

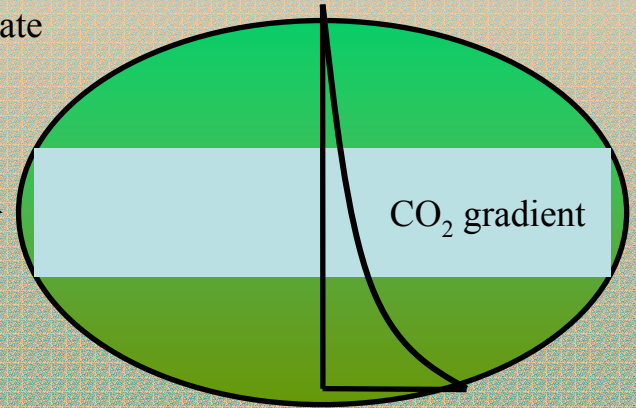


Hydrate



Liquid CO₂

CO₂ gradient



Gravity current dynamics

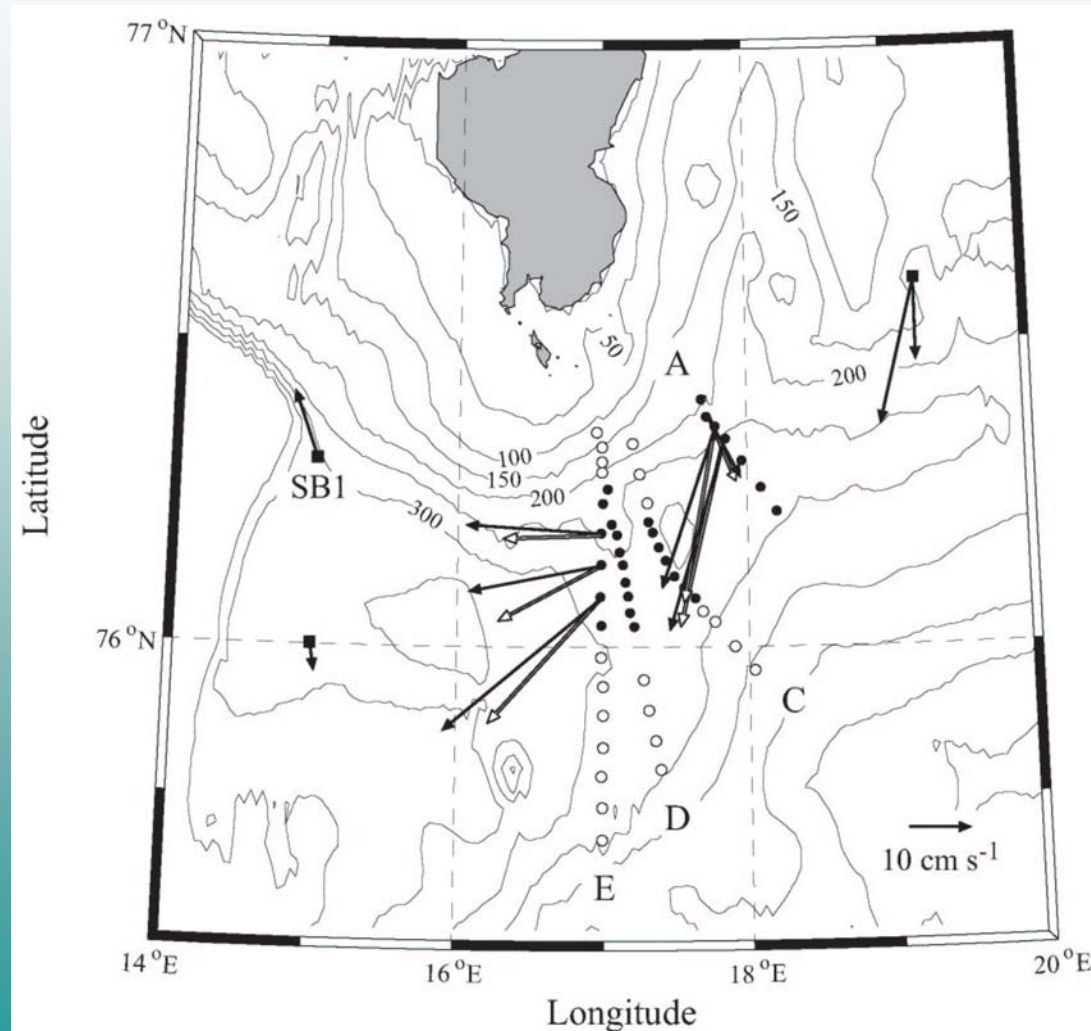
- A slope of only about 4×10^{-3} (2 times the drag coefficient) may lead to a steady gravity current.
- Classical gravity current models are formulated for homogeneous layers. A rich literature is based on laboratory studies.
- Gravity currents with CO₂-enriched water have been modelled and studied as a vertical transport mechanism for CO₂ (Drange & Haugan, 1993, Adams et al., 1995, Alendal, 1996).
- Gravity currents entrain ambient water. Mixing and entrainment is particularly strong near the transient head.
- Natural gravity currents in the ocean occur where dense water, typically at high latitudes, descend to ventilate deep water.
- Recent *in situ* observations have begun to explore internal structure previously only studied in models.

Natural analogue gravity current: Observed outflow from basin with cooling and freezing/brine rejection

Storfjorden, Svalbard -
Identified as two layers:

- 1) a lower layer (~15 m) with relatively uniform vertical structure
- 2) an upper, thick mixing layer (~30 m) with larger vertical density gradient

The widening of the lower layer is comparable to Ekman veering (friction from the bottom)



Cross-section outflow

Vertical diffusivity (bars)

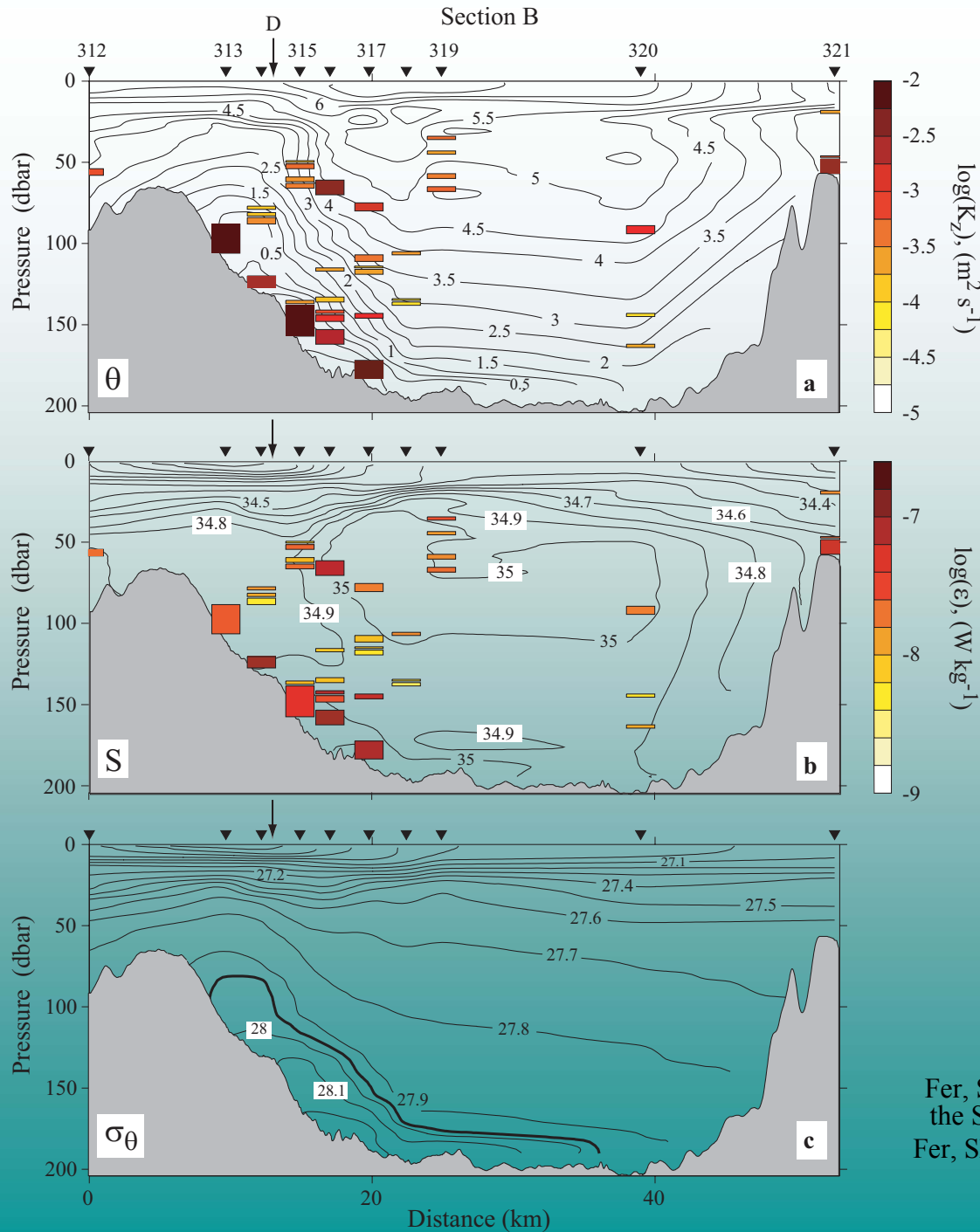
Temperature (contoured)

TKE dissipation (bars)

Salinity (contoured)

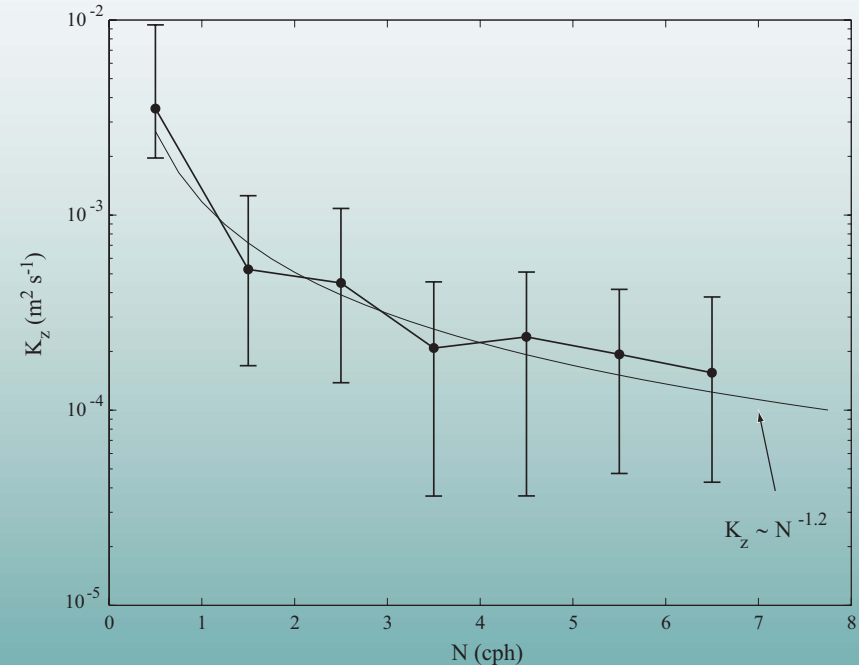
Potential density

Fer, Skogseth, Haugan and Jaccard: Observations of the Storfjorden overflow, *Deep-Sea Research* 2003.
Fer, Skogseth and Haugan: Mixing of the Storfjorden overflow (Svalbard Archipelago) inferred from density overturns, *J. geoph. res.* 2004.



Dynamics of a dense outflow

- Lower part of dense plume is homogenized by turbulence from bottom irregularities.
- Upper part of dense plume mixes with ambient water due to shear induced mixing (vertical diffusivity is decreasing with increasing buoyancy frequency as $N^{-1.2}$).
- This may be relevant to deep sea CO_2 -enriched seawater plume depending on slope and carbon concentration.



Turbulence Profiling

- microstructure, turbulence, and standard CTD Sensors
- allows direct estimate of mixing and elucidation of mixing process
- presently free fall from surface
- could be run free rising from seafloor in the deep sea



MSS



Conclusions and challenges

Many technical options for direct storage would ultimately deliver the CO₂ in dissolved form in a dense, low pH, near bottom layer.

The carbon concentration in such a layer and its mixing and spreading would be crucial to sediment interaction, benthic life, dilution in the pelagic domain, and efficiency.

Preliminary model studies based on very limited *in situ* data give dissolution estimates for deep sea liquid CO₂ pools depending on the current regime.

Models furthermore suggest the development of a dense internal boundary layer within the BBL.

Challenges

What is the lifetime and space scale of elevated carbon concentrations? When does the BBL homogenize at sufficiently low carbon concentrations that further diffusion is reduced to a passive tracer problem?

Obtain general understanding of mixing between the BBL and ambient water.

The response of CaCO_3 sediments to elevated carbon concentration.

On the microscale: The fascinating complexities of hydrate formation and dissolution.

Make "the world" understand that these topics need to be addressed now.