Gas solubility and exchange

Learning objectives:
- Familiarity with gas/pressure units
- Convert / calculate between concentration in water, mole fraction in air, partial pressure, etc.
- Understand how to adjust for water vapor content in the atmosphere
- Basic appreciation for the complexities of air-sea gas fluxes
- Ability to use a simple gas flux parameterization equation

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Outline:
- Gas composition, units, laws
  - Dalton's law, ideal gas law, partial pressure vs. fugacity, water vapor, Henry's law
- Gas solubility and example calculations
- Examples of mechanisms that cause anomalies in saturation
- Basic theory of gas exchange
- Empirical gas exchange experiments
- Current (and simplified) flux parameterizations

Why are gases useful?

- Tracers for physical processes:
  - mixing, deep water formation, air-sea exchange, circulation
- Biological processes: biological production, respiration, oxygen deficient zones
- Atmospheric chemistry/global climate change
- Tracers of large-scale ocean changes

What controls gas concentrations?

- Air-sea gas exchange + solubility
- Mixing in the ocean
- Production and removal within the ocean

Air $$P_T = pN_2 + pO_2 + pAr + pH_2O + \cdots$$
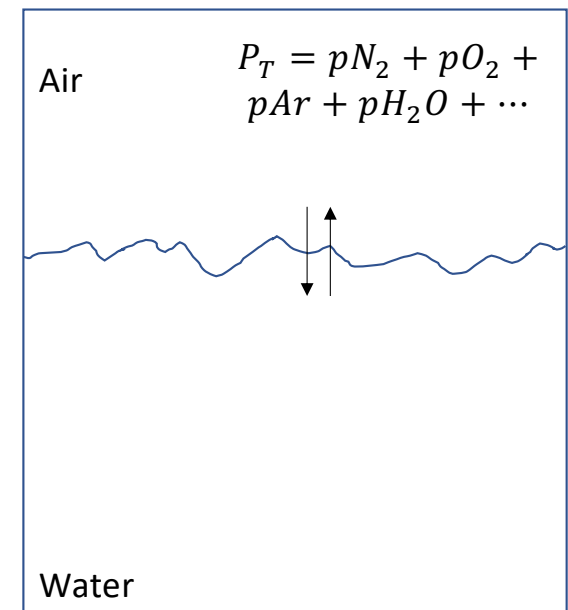
Water

Composition of the atmosphere

Table 1.5. *The major gases of the atmosphere excluding water vapor, which has a concentration of a few percent at saturation in the atmosphere*

Seawater equilibrium concentrations were calculated from the Henry's Law coefficients at 20 °C and $S = 35$.

Gas	Atmospheric mole fraction (atm)	Seawater equilibrium concentration ($\mu mol\ kg^{-1}$)
N_2	7.808×10^{-1}	4.18×10^2
O_2	2.095×10^{-1}	2.25×10^2
Ar	9.34×10^{-3}	1.10×10^1
CO_2	3.65×10^{-4}	1.16×10^1
Ne	18.2×10^{-6}	7.0×10^{-3}
He	5.24×10^{-6}	2.0×10^{-3}
Kr	1.14×10^{-6}	2.0×10^{-3}
Xe	0.87×10^{-7}	3.0×10^{-4}

~ 99%

Mole fraction is the amount (atm or moles) of a gas relative to atmosphere as a whole

Often denoted using X_i (e.g. X_{O2})

Emerson and Hedges

Pressure units

In the atmosphere:

(Std) atmospheric pressure: 1 atm or 1013.25 mb

(varies by ~15%)

1 pascal = 1 N m^{-2}

1 atm   = 760 torr = 760 mmHg (29.92 in Hg)

= 1.013 x 10^5 N m^{-2} = 1.013 x 10^5 Pa

1 bar = 100,00 Pa

=1.013 bar = 1013.25 mb (millibar)

= 14.7 lbs in^{-2}

In the ocean:

0.1 bar = 1 decibar = ~1 m in seawater

An increase in depth of 1000 m in seawater increases
the pressure by ~100 bar (or 1000 db)

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Gas units

Mole fraction, X_c: ppm
- Gas in a gas mixture or a liquid
- $X_c = \frac{n_c}{n_t} = \frac{p_c}{P_T}$, where n_c is the number of moles of gas C (i.e. the mole fraction gives the fraction of the total pressure that each gas contributes to dry air)

Partial pressure, p_c: μatm (micro atmospheres)

Fugacity, f_c: μatm
- like partial pressure, but accounts for non-ideality

Molar: mol kg^{-1}
- moles of gas per kg seawater

Volumetric: L kg^{-1}
- liter of gas @ STP per kg seawater

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Dalton's law of partial pressures

- Partial pressure (p_i) = Mole Fraction (X_i)*P_T
- Total pressure in a fixed volume is equal to the sum of the partial pressures of the components of the mixture

$$P_T = \sum p_G = p_{N_2} + pO_2 + pAr + pH_2O + \cdots$$

- Dalton's Law implies ideal behavior, i.e. all gases behave independently of one another.
- Gases are dilute enough that this is a pretty good assumption (~1% errors in the surface ocean)

Fugacity (f)

- Fugacity of a gas (i) corrects the partial pressure for non-ideal behavior

- f_i and p_i have units of "atm" (atmospheres)

- The fugacity of a gas in analogous to the activity of a solute
  - Unlike solutes: common elemental gases (e.g., O_2, N_2, Ar) deviate from ideal behavior by <0.1%, so we can use fugacity (f) and partial pressure (p) interchangeably
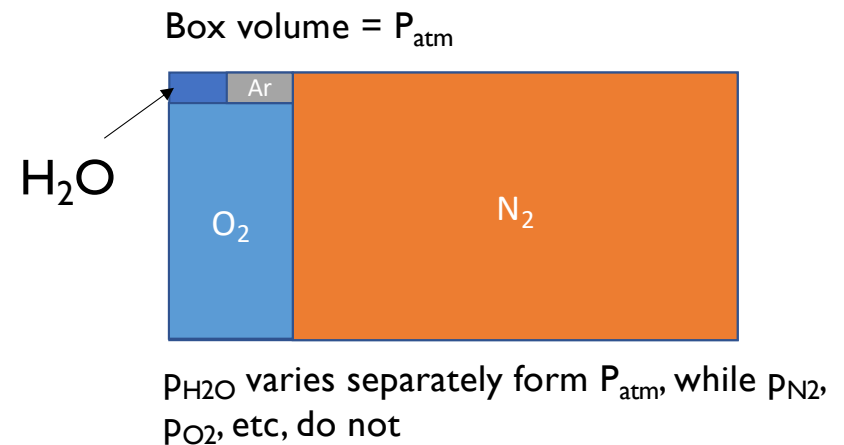 - Some molecular gases (e.g. N_2O, CO_2) have a larger non-ideal behavior (<1%) and it is usually important to correct for this

Ideal gas law

$$P_c V = n_c R T$$

- P_c = pressure exerted by gas c
- V = volume
- T = temperature in K
- n_c = # mol of gas I
- R = ideal gas constant

- At STP (i.e. 273.15 K and 1013.25 hPa), volume of a gas = 22.414 L mol^{-1}
- STP for gases is 0C, not 25C!

Water vapor: Clausius-Clapeyron relation
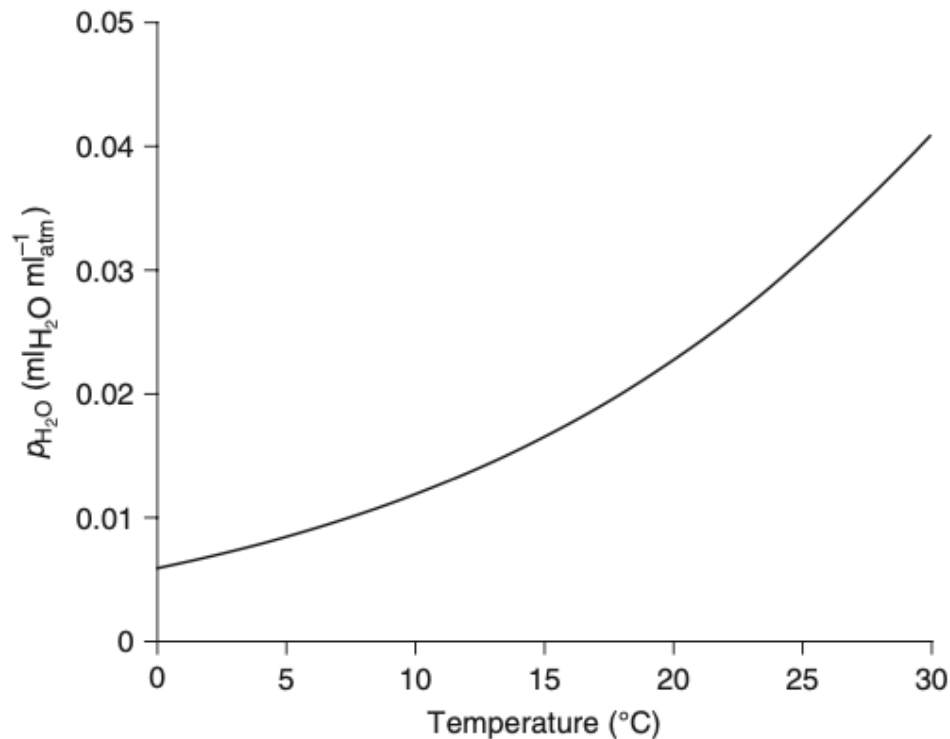
- Water vapor is different than other gases in the atmosphere
- Water vapor is between ~0.6 and 4 % of total atmospheric pressure
- Must be accounted for when determining saturation concentrations from the mole fraction of atmospheric gases

Box volume = P_{atm}

H_2O

Ar

O_2

N_2

p_{H2O} varies separately form P_{atm}, while p_{N2}, p_{O2}, etc, do not

Emerson and Hedges

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Water vapor correction

where p_G = partial pressure of gas G
x_G = mole fraction of gas in dry air
P_T = Total atmospheric pressure
h = % relative humidity
p_0 = saturation vapor pressure of water = $f(T)$

- Mole fractions in the atmosphere are given in dry air
- Total atmospheric pressure includes the partial pressure of water vapor (pH_2O)
- To calculate the partial pressure that a gas exerts on the surface of the water, we must remove the water vapor pressure
- $p_c = X_c(P_T - pH_2O)$
  - p_c = partial pressure of gas C
  - X_c = mole fraction of gas in dry air
  - P_T = Total atmosphere pressure
  - $pH_2O = (h/100) \times p_0$, where h is % relative humidity and p_0 is the saturation vapor pressure of water

Composition of the atmosphere is generally homogenous for major constituents and trace gases, except for H_2O, CO, N_2O, CO_2, and CH_4.

Gas solubility

- The solubility of a gas determines its partitioning between the atmosphere and water
  - The amount of a gas that will be present in water at a given T and P_T (total atmospheric/headspace pressure)
- Solubility coefficients are a type of chemical equilibrium constant that defines the solubility of a gas in water

- Solubility coefficients can be expressed for water in equilibrium with Earth's atmosphere, or with a theoretical atmosphere of a pure gas of interest – need to be careful of what coefficients you use
  - Converting between them can be confusing
  - In this class I will give you solubility information, but if you are performing your own calculations be careful that you are consistent and use published values to check your work

Henry's Law

- The amount of gas that will dissolve in water at thermodynamic equilibrium is described by Henry's Law:

$$[C] = K_{H,C} f_c$$

[C] = the gas concentration mol kg^{-1} atm^{-1}

$K_{H,C}$ = Henry's law solubility coefficient (mol kg^{-1} atm^{-1})

fc = pc = fugacity or partial pressure of a gas

Henry's law coefficients are empirically determined and a function of temperature and salinity.

Saturation: Chemical equilibrium

Air

$$P_T = pN_2 + pO_2 + pAr + pH_2O + \cdots$$
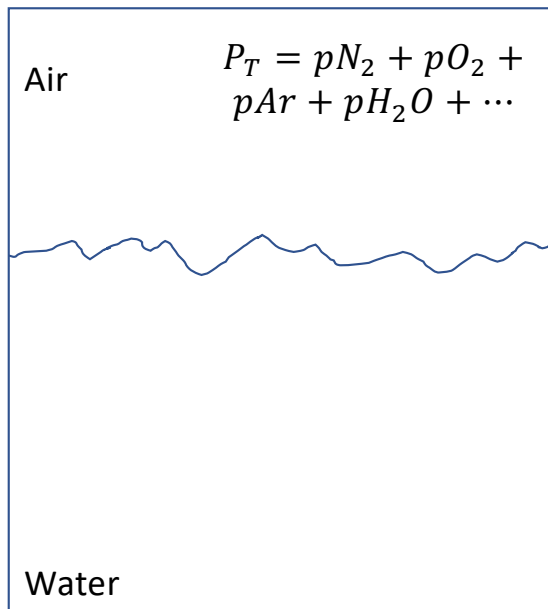
Water

- At equilibrium, the $[C]_{obs.} = [C]_{sat}$, which is determined by pC

$$pC = P_T X_c (1 - p_{H2O})$$

$$[C]_{sat} = K_{H,C} p_c$$

P_T=atmospheric pressure
X_c=atmos. mole fract.
p_c= partial pressure of gas C

- Supersaturated water: $[C]_{obs.} > [C]_{sat}$
  - Gas will tend to move from the water to the atmosphere
- undersaturated water: $[C]_{obs.} < [C]_{sat}$
  - Gas will tend to move from the atmosphere to the water

Gas solubility as a function of temperature
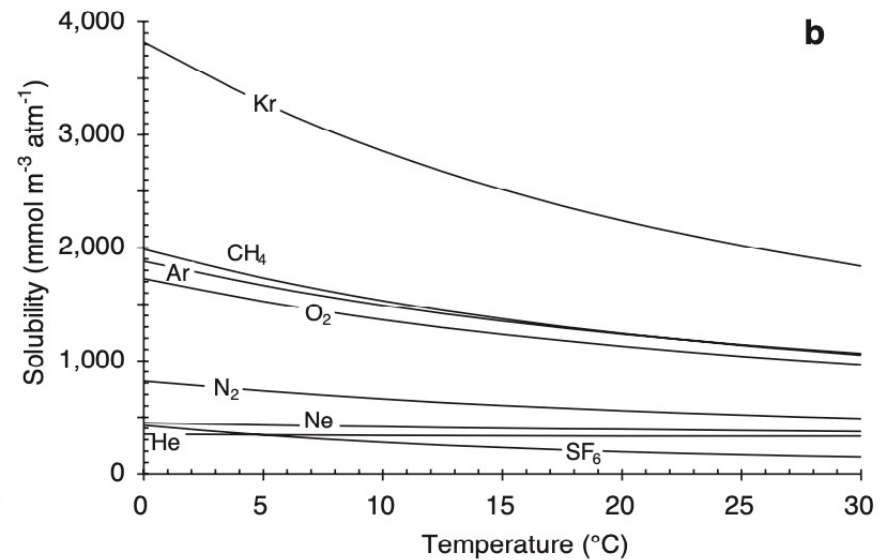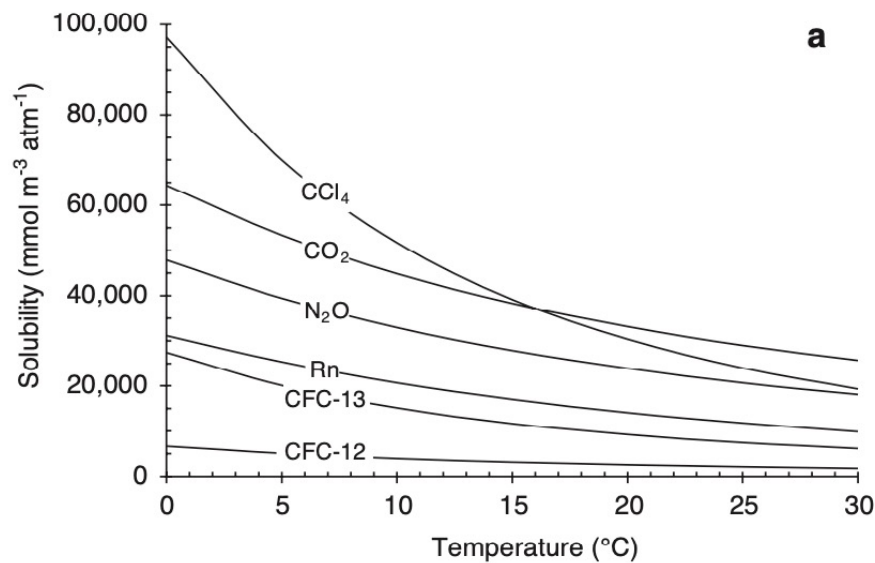
Note the change in scale

Example calculation

What is the saturation concentration of oxygen at 20°C? How about 5°C?

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Example calculation

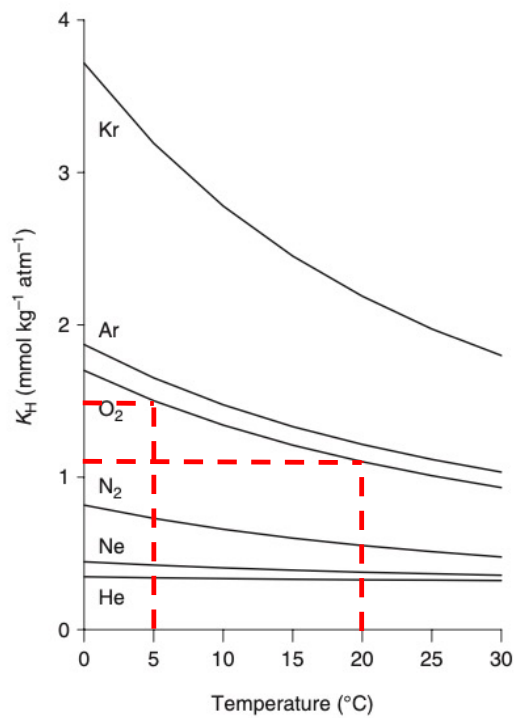What is the saturation concentration of oxygen at 20°C? How about 5°C?

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Emerson and Hedges, at 1atm and S=35

Example calculation

K_H (mmol kg^{-1} atm^{-1}) vs Temperature (°C)

Kr, Ar, O$_2$, N$_2$, Ne, He

Emerson and Hedges, at 1atm and S=35

What is the saturation concentration of oxygen at 20°C? How about 5°C?

$$pO_2 = P_T X_{O2}(1 - p_{H2O})$$

X_{o2} = 0.20946 – that is the mole fraction in dry air

p_{H2O}(20°C, S=35) = 0.0226 (i.e. ~2 % of the total atmospheric pressure is water vapor)

So at 1atm, 20°C the pO_{2atm} = 1 * 0.20946*(1-0.0226) = 0.2030 atm

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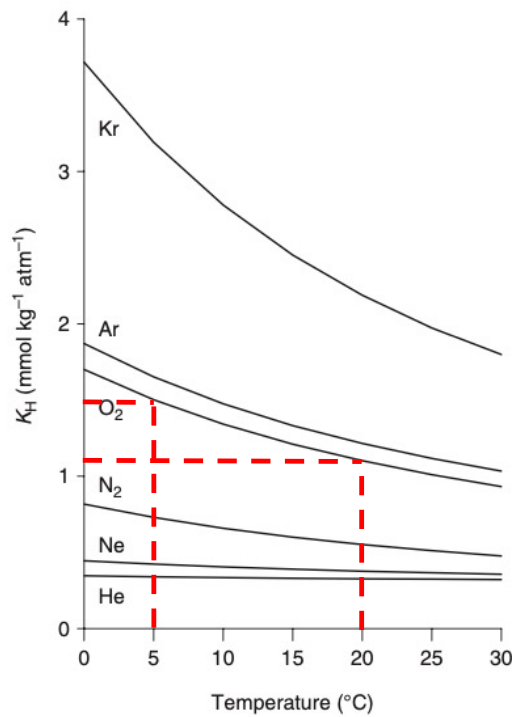p_{H2O}(20°C, S=35) = 0.0226 (i.e. ~2 % of the total atmospheric pressure is water vapor)

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$$[O_2]_{sat} = K_{H,O2} p_{o2}$$
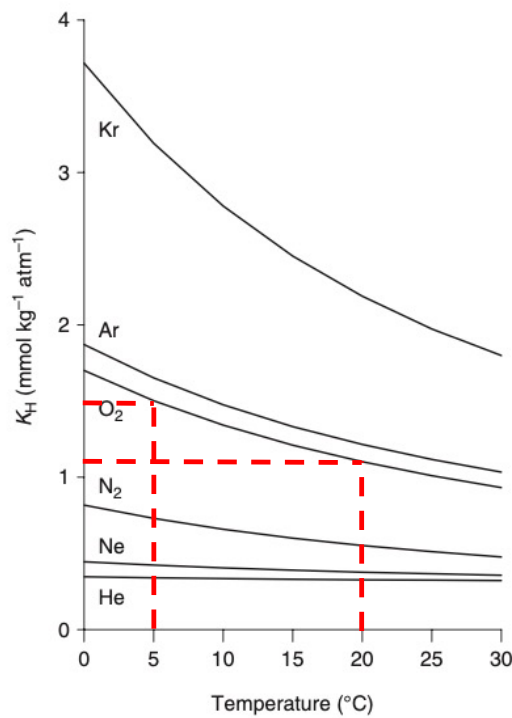
$$[O_2]_{sat,25C} = 1.1 \frac{mmol}{kg\,atm} 0.2030 atm = 0.2233 \frac{mmol}{kg} = 223.3 \frac{\mu mol}{kg}$$

Emerson and Hedges, at 1atm and S=35

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Note the lower pH$_2$O

pO_{2atm} = 1 * 0.20946*(1-0.0084) = 0.2077 atm

$$[O_2]_{sat,5C} = 1.5 \frac{mmol}{kg\,atm} 0.2077 atm = 0.3115 \frac{mmol}{kg} = 311.5 \frac{\mu mol}{kg}$$

Solubility vs. partial pressure: O_2 and N_2

- Oxygen: ~20 % of the atmosphere
  - Partial pressure (pO_2=fO_2) = 0.209
- Nitrogen: ~78 % of the atmosphere
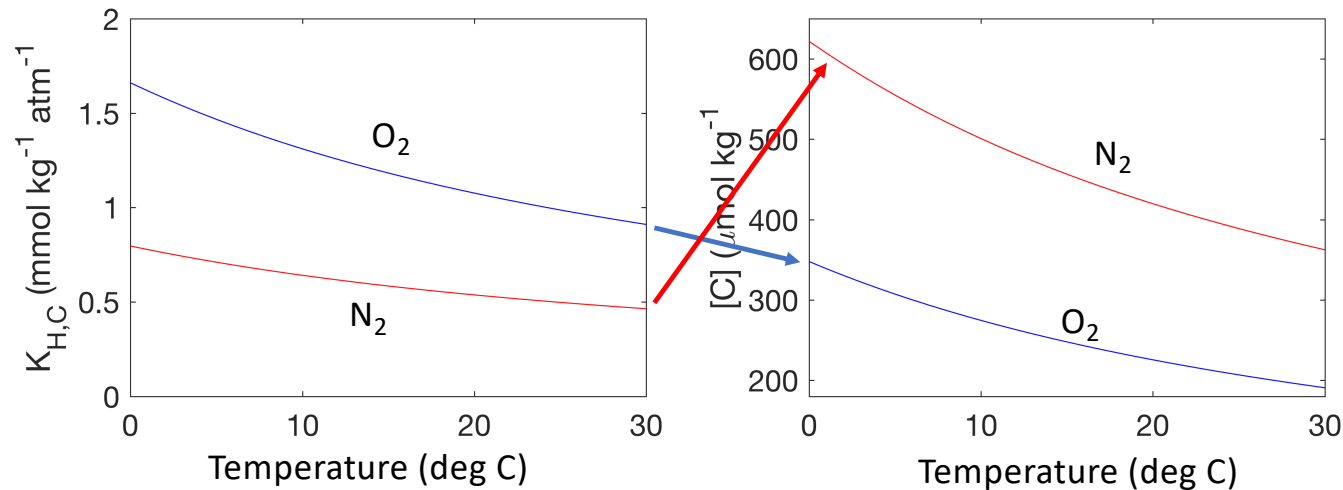 - Partial pressure = 0.781

Solubility vs. Temperature
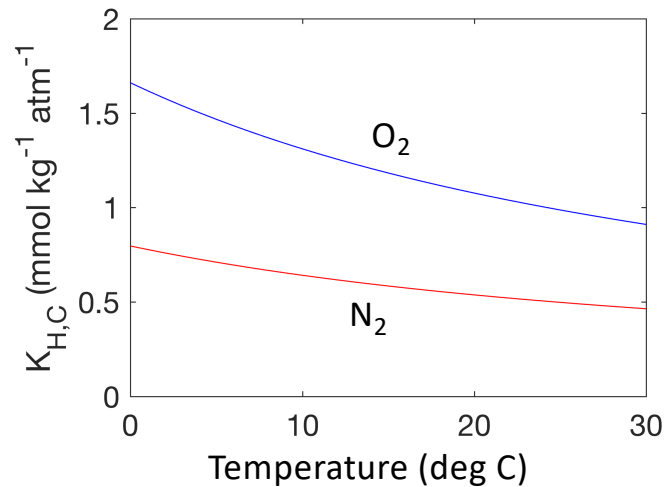
Solubility vs. partial pressure: O_2 and N_2

- Oxygen: ~20 % of the atmosphere
  Partial pressure ($pO_2 = fO_2$) = 0.209
- Nitrogen: ~78 % of the atmosphere
  Partial pressure = 0.781

N_2 is 4x more plentiful in the atmosphere, but only half as soluble as oxygen, so saturation concentration is ~2x oxygen

Solubility vs. Temperature

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Saturation anomalies: Mechanisms

Causes of deviation from equilibrium:

1. Warming or cooling of the sea surface (gas exchange is not instantaneous)
2. Bubble injection from winds/breaking waves
3. Mixing of different water masses
4. Biological activity (e.g. denitrification, photosynthesis/respiration)
5. Change in atmospheric pressure (again, gas exchange is not instantaneous)

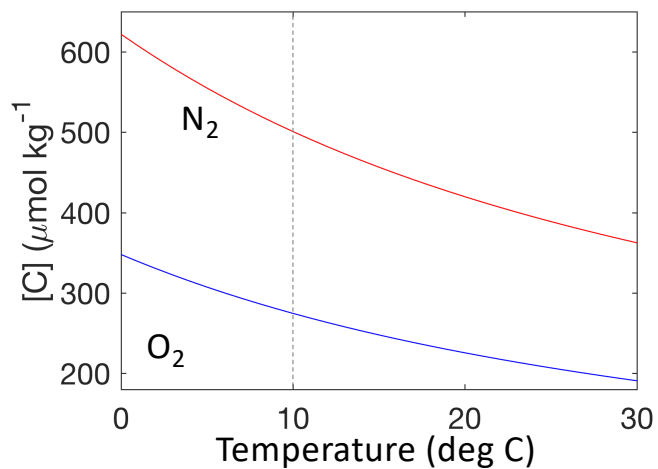# Saturation anomalies: Mechanisms

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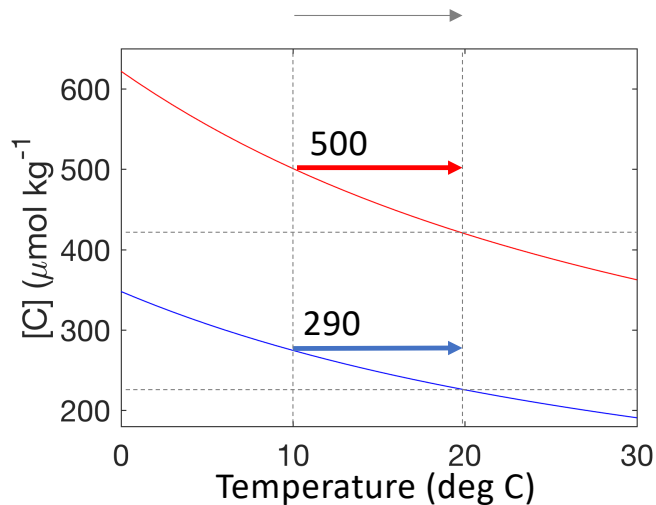5. Change in atmospheric pressure (again, gas exchange is not instantaneous)

Saturation anomalies: Temperature change

Saturation anomalies can either be expressed in concentration or percent

$$\Delta[C] = [C] - [C]_{sat}$$

$$\% \, sat = \frac{[C]}{[C_{sat}]} \times 100$$

$$\% \, supersaturation = \left(\frac{[C]}{[C_{sat}]} - 1 \right) \times 100$$

Saturation anomalies: Temperature change

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Water is warmed by 10 deg, no gas exchange allowed

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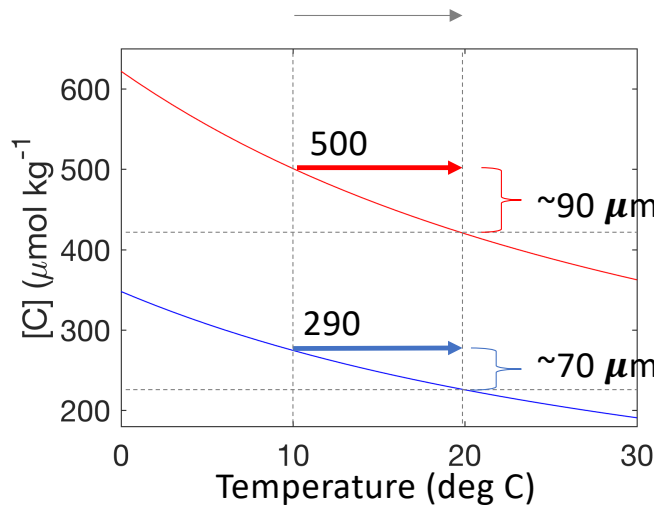
~90 μmol; $\quad \Delta N_2 = 500 - 410 = 90 \frac{\mu mol}{kg} \ or \ \frac{500}{410} \times 100 \approx 122\%$

~70 μmol/kg; $\quad \Delta O_2 = 290 - 200 = 70 \frac{\mu mol}{kg} \ or \ \frac{290}{220} \times 100 \approx 132\%$
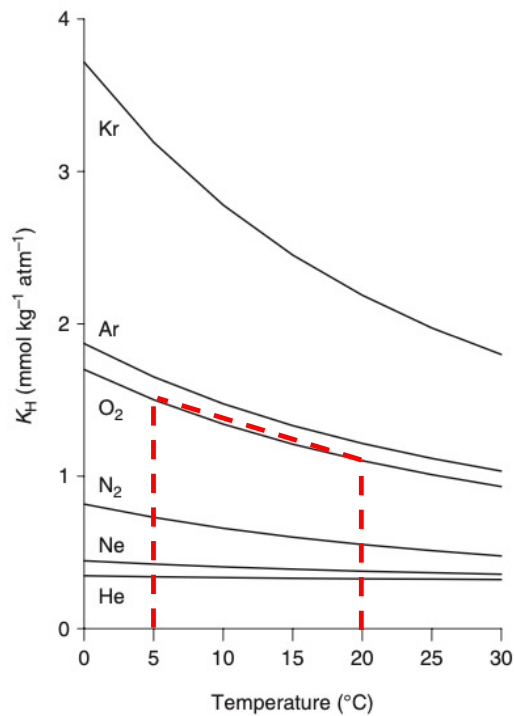
Saturation anomalies: Mixing

What happens when you mix two equal size water masses with different concentrations?

Temperature?
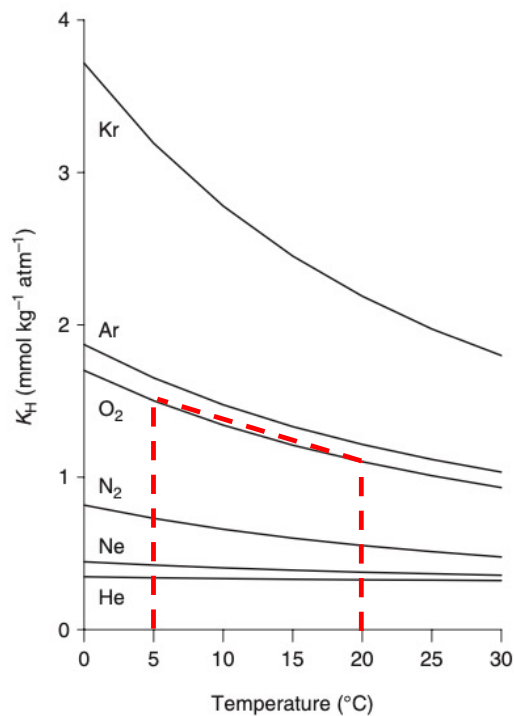Concentration?
Saturation?
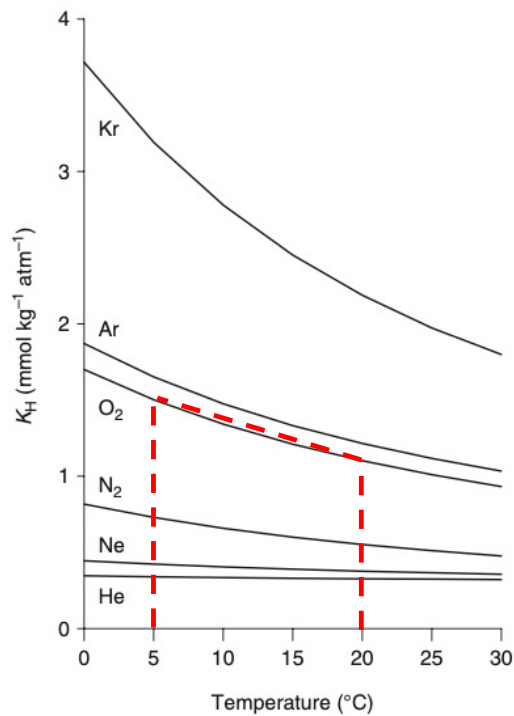
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Temperature? – Conservative (i.e. T_{new} = 12.5 C)
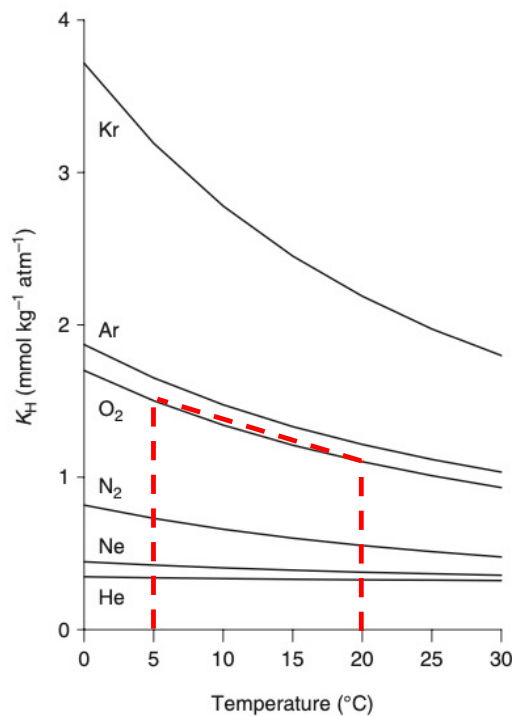Concentration?
Saturation?

Saturation anomalies: Mixing

What happens when you mix two equal size water masses that are both equilibrated with the atmosphere?

Temperature? – Conservative (i.e. T_{new} = 12.5 C, halfway between 20 and 5 degC)
Concentration? – Conservative (i.e. C_{new}= 268 µmol/kg)

Saturation?

Saturation anomalies: Mixing

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Temperature? – Conservative (i.e. T_{new} = 12.5 C, halfway between 20 and 5 degC)
Concentration? – Conservative (i.e. C_{new}= 268 µmol/kg)

% Saturation? – non-conservative

$C_{sat,12.5}$ = 260 µmol/kg

The water is now supersaturated by 8 µmol/kg or ~3%

Why do we care about air-sea gas exchange?

- Globally –
  - understand cycling of important trace cases: CO_2, N_2O, DMS, CH_4
  - Predict / monitor oxygen loss

- Smaller scales:
  - Capturing rates of biological activity
  - Predict evasion of volatile pollutants

What affects gas exchange?

- ΔC (water concentration disequilibrium from saturation) determines driving force for air-sea exchange

- But how does gas actually move between the air and water?
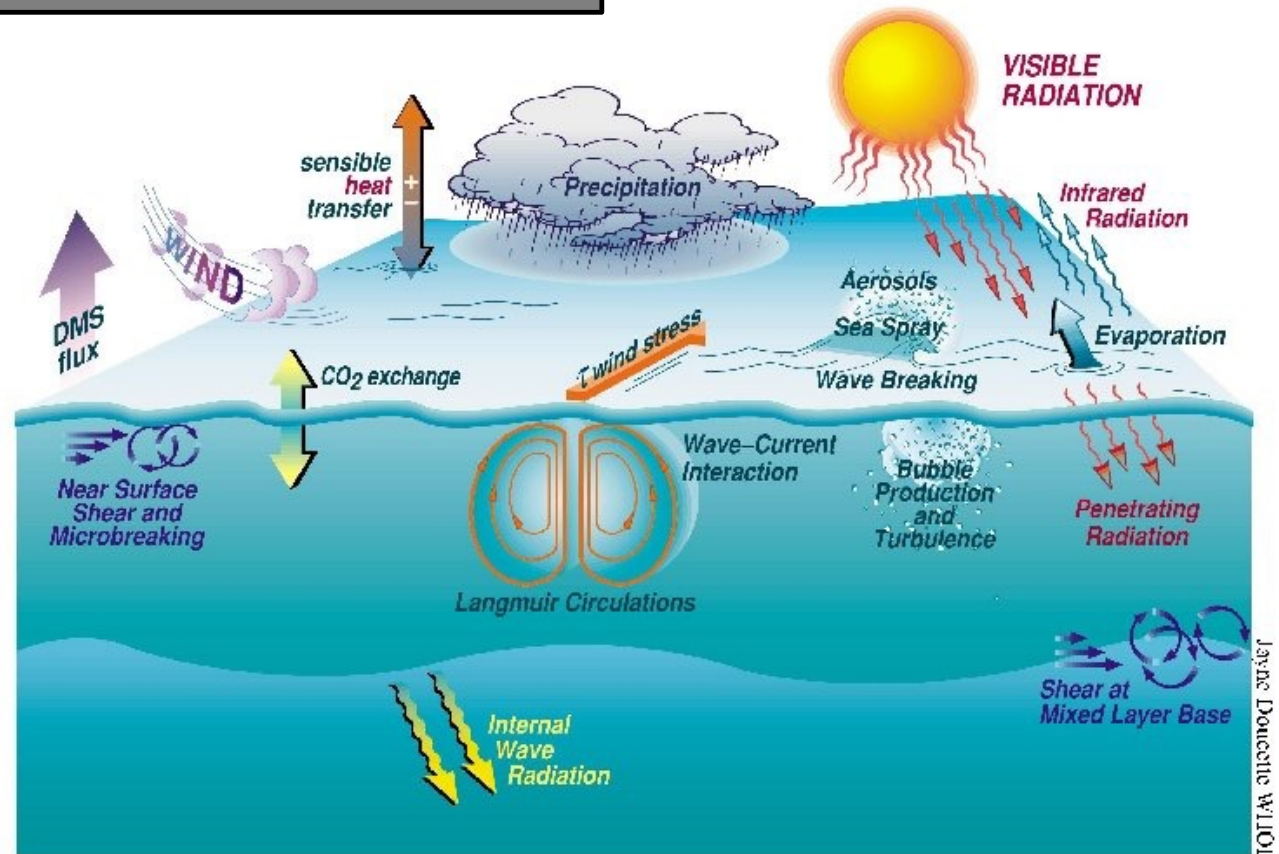
Basic flux equation

- Solubility
  - How much gas water can hold at equilibrium
- Gas transfer coefficient
  - How quickly will a gas cross the air-sea boundary

$F_c = Flux (mol m^{-2} time^{-1})$ — rendered below

F_c = Flux (mol m^{-2} time^{-1})
G_c = Gas transfer coefficient (m/time)*

$$F_C = G_C([C] - [C_{sat}])$$

$$\frac{mol}{m^2\,d} = \frac{m}{d}\left(\frac{\mu mol}{kg}\right)\left(\frac{kg}{m^3}\right)\left(\frac{mol}{\mu mol}\right)$$

[C] = Surface concentration (µmol/kg)
[C]$_{sat}$ = Saturation concentration

$$F_C = G_C K_{H,c}(f_c^w - f_c^a)$$

f_c^w = fugacity of the water
f_c^a = fugacity of the air

*Note that k is often used instead of G
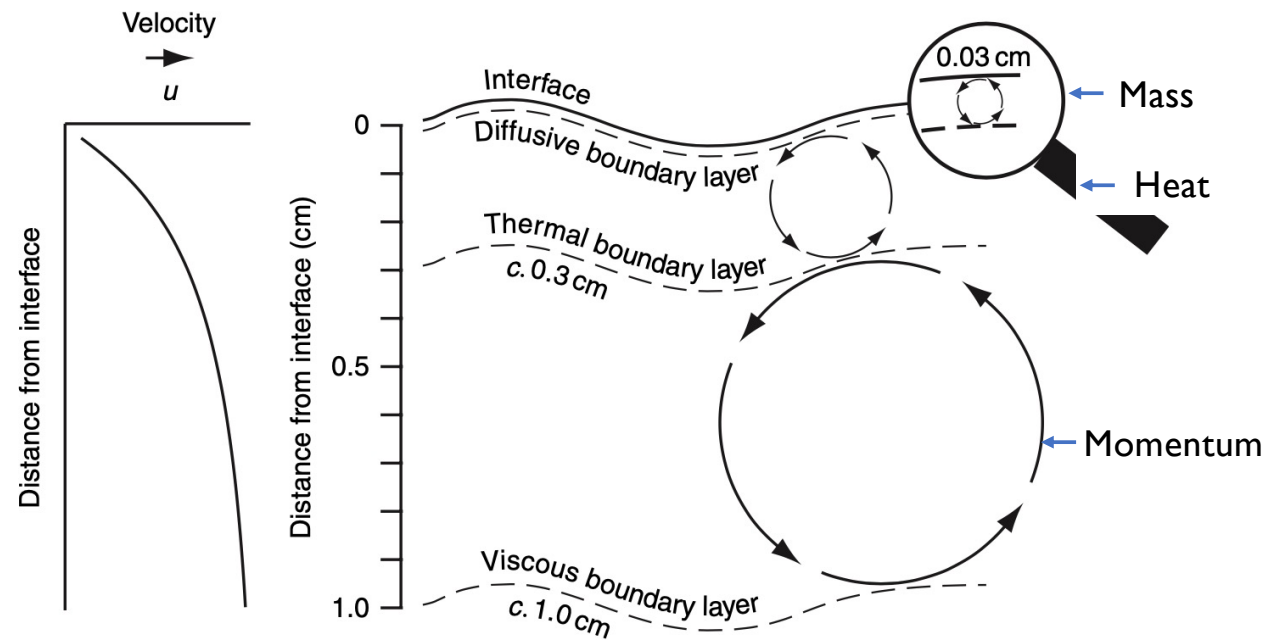
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Boundary layers: Schmidt number

Velocity
u

Distance from interface

Water moves slower
close to the surface

Distance from interface (cm)

0

0.5

1.0

Interface

Diffusive boundary layer

Thermal boundary layer
c. 0.3 cm

Viscous boundary layer
c. 1.0 cm

0.03 cm

← Mass

← Heat

← Momentum

Emerson and Hedges 2008

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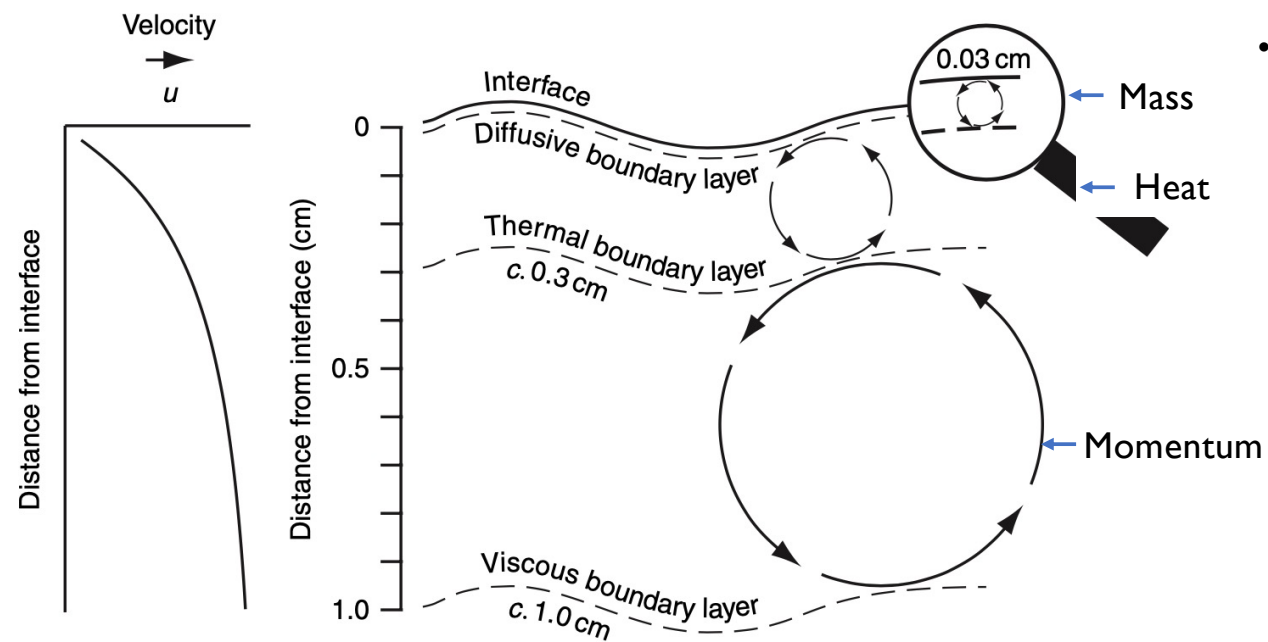
Emerson and Hedges 2008

- Gas exchange is partially a molecular diffusion process
- The layer over which molecular processes become important is dependent on the ratio of the molecular diffusion of a gas relative to the kinematic viscosity

$$G_C = G^* \times \left(\frac{D_C}{v}\right)^n$$

G^* = empirical constant (cm s^{-1})
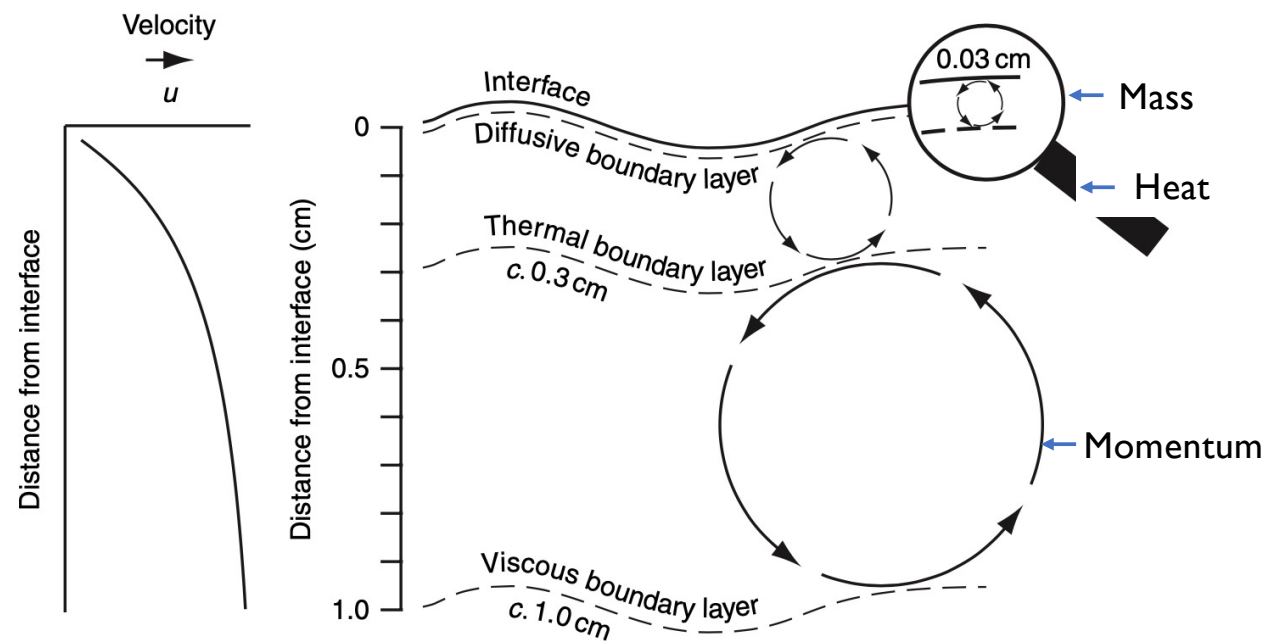D_C = molecular diffusivity of a gas c (cm^2 s^{-1})
v = kinematic viscosity (cm^2 s^{-1})

$$Sc_C = \frac{v}{D_c}$$

Sc_C= Schmidt number for gas C

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Boundary layers: Schmidt number

Velocity
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1.0

Distance from interface (cm)

0.03 cm

← Mass
← Heat
← Momentum

Emerson and Hedges 2008

Sc_C = Schmidt number for gas C

$$Sc_C = \left(\frac{v}{D_c}\right)^n$$

- v and D_C are strongly temperature dependent
- S_c can vary by a factor of 5-6 over oceanic temperature ranges, which makes gas transfer velocity a strong function of temperature
- n is somewhere between ½ and 1, depends on empirical model
- Sc lets us relate gas transfer coefficients for different gases

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Gas exchange models: Stagnant Film

Fugacity

f^a

δ_g

f^a_{sat}

Air

Water

δ_w

$[C^{Sat}]$

$[C]$

Concentration
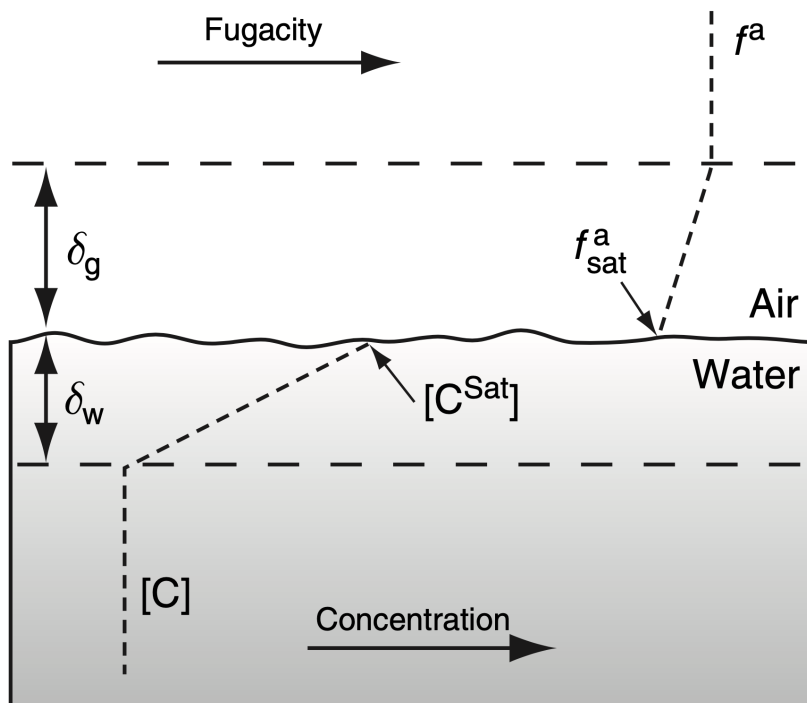
- Simple – gas exchange is purely due to supply of gas to the surface by molecular diffusion – both from the air and water sides

$$G_C = \frac{D_c}{\delta}$$

- Ignores wind speed, assumes there is always some stagnant layer of invariant thickness

- Gas exchange is dependent on molecular diffusion coefficient and therefor Schmidt number to the -1 (n=1)

Emerson and Hedges, Whitman, 1923

Gas exchange models: Surface renewal
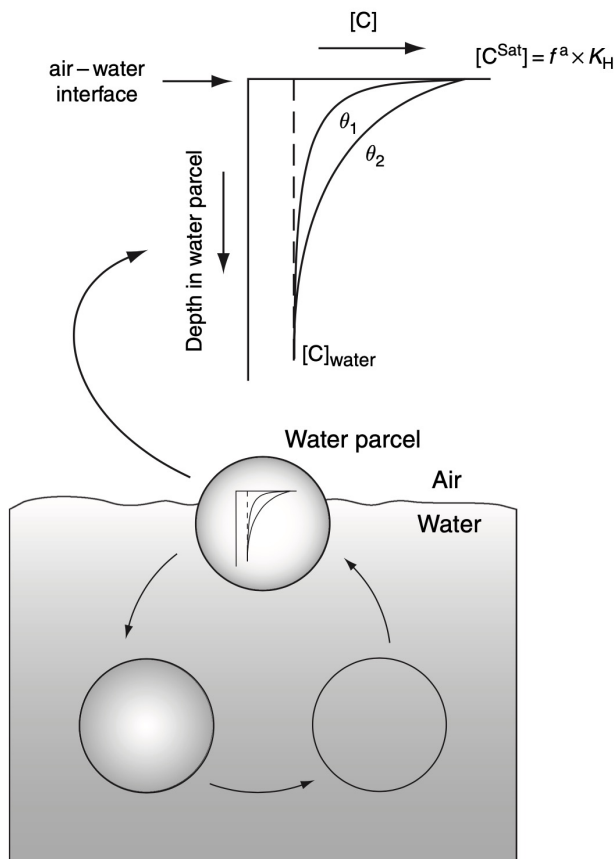
- Assumes gas exchange is primarily limited by supply of water parcels to the surface
- Yields an n=1/2 dependence on Schmidt number
- These are theories – in practice models have been tested empirically with n (and other coefficients/terms) fit to data
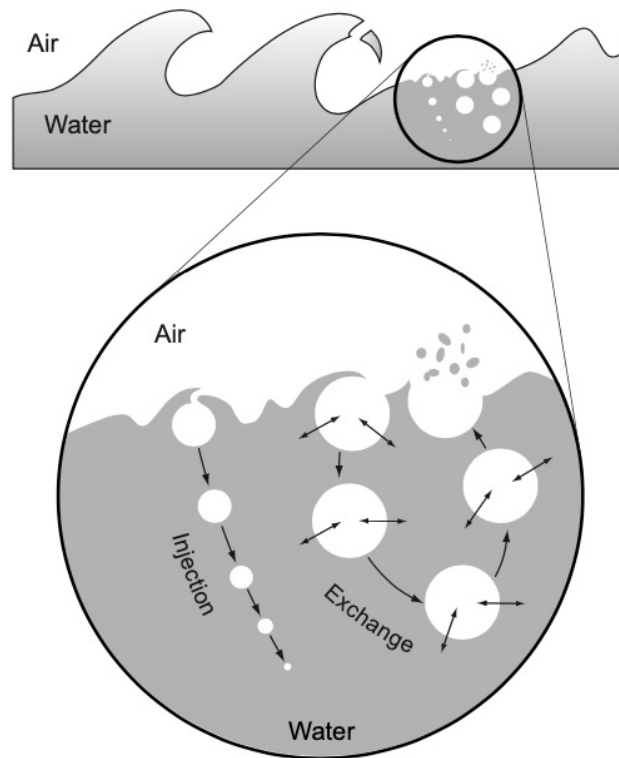
What actually happens in the ocean?

Mechanisms?
- wind speed
- wave height
- wave shape
- breaking vs. non-breaking (bubbles)
- spray
- relative direction of wind and waves

Bubble injection

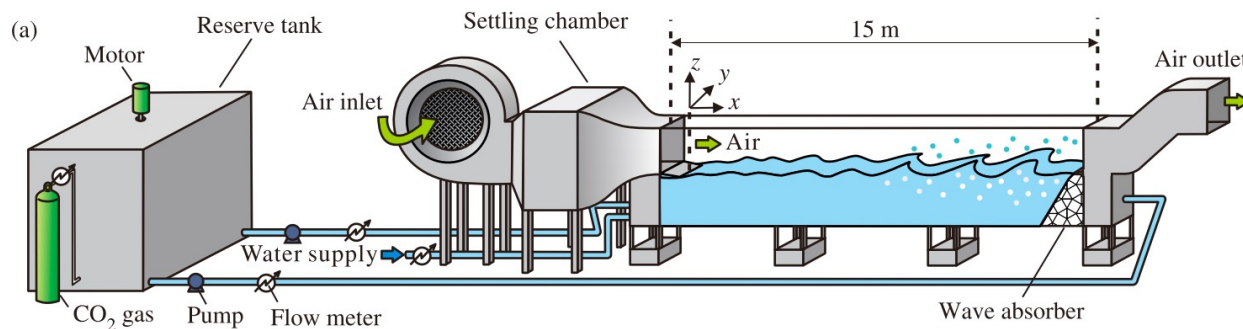Bubbles injected into the ocean can suffer two fates:
1. Forced into dissolution by increased pressure (completely dissolved/collapsed)
2. Sink down, exchanging gas with the surrounding water under higher pressure before returning to the surface (partially dissolved/collapsed)

Bubbles are a significant fraction of gas exchange at high winds for low solubility gases.

Emerson and Hamme, 2021

Empirical determinations (and uses) of G (k)

- Lab experiments
  - Wave tanks

- Large-scale
  - Ocean inventory vs. atmospheric production

- Small-scale
  - Ocean inventory changes
  - Flux co-variance
  - Purposeful tracer release experiments
  - Upper ocean mass balances

- Modeling
  - Empirical or not?

# Lab-based parameterizations: Wave tanks

(a) Motor, Reserve tank, Air inlet, Settling chamber, 15 m, Air outlet, Air, Water supply, CO$_2$ gas, Pump, Flow meter, Wave absorber
(b) (c) (d)

Wave tanks
- Controlled environment
- Repeatable
- Limited fetch, no gusts, no wind/wave mismatches, etc.
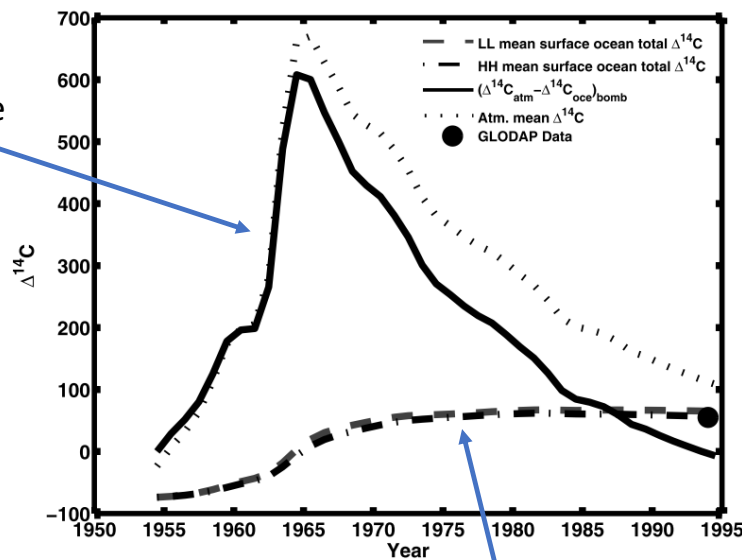- Good for process understanding, but not for magnitude of mass transfer coefficient

Iwano et al. 2013

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Experimental methods: Radon
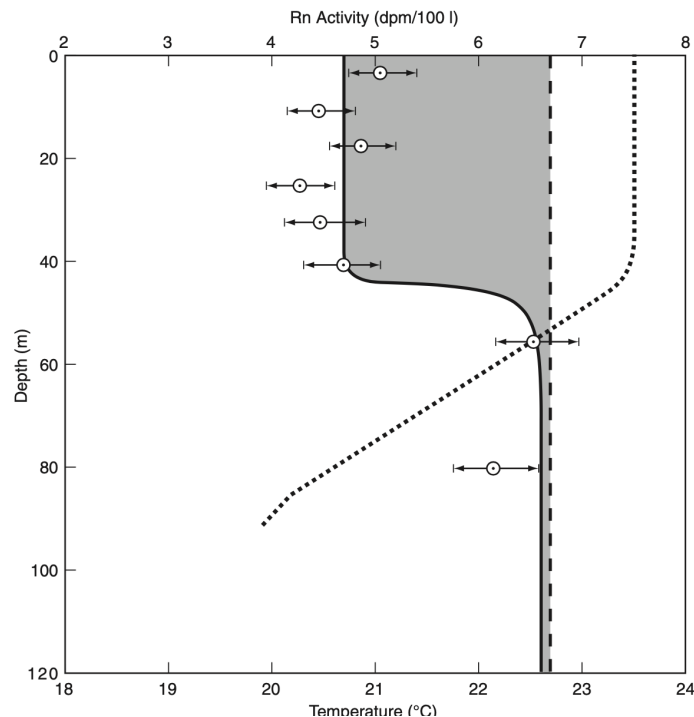
- Radon 226 is present in the ocean and readily measured (half-life of 1620 years)
- Decays to Radon 222 (half-life of 3.85 days)
- Radon 222 escapes to the atmosphere
- Deficit relative to expected can be used in a mass balance to determine the air-sea exchange term over a few days/weeks

Sarmiento and Gruber, 2006

Experimental methods: Radiocarbon

Nuclear bomb tests
released ^{14}C into the
atmosphere

LL mean surface ocean total Δ^{14}C
HH mean surface ocean total Δ^{14}C
$(\Delta^{14}C_{atm} - \Delta^{14}C_{oce})_{bomb}$
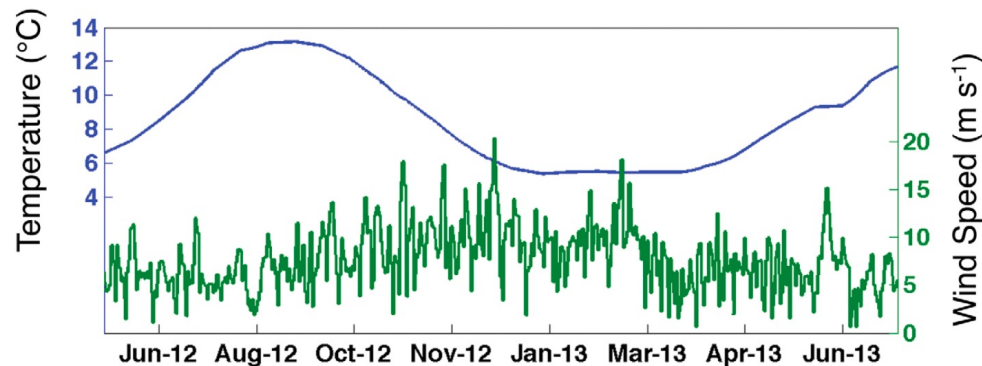Atm. mean Δ^{14}C
GLODAP Data

Sweeney et al. (2007)

Which then accumulated
in the ocean.

- We can calculate air-sea exchange by measuring ^{14}C in the ocean and calculating what the flux must have been to match ocean measurements to atmospheric measurements.
- Provides a long-term estimate
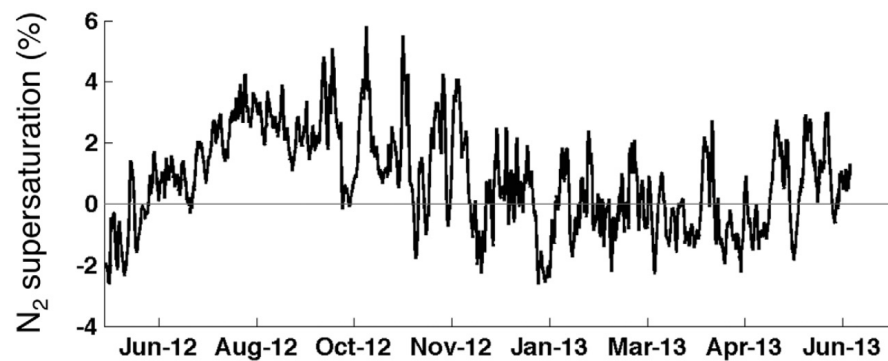
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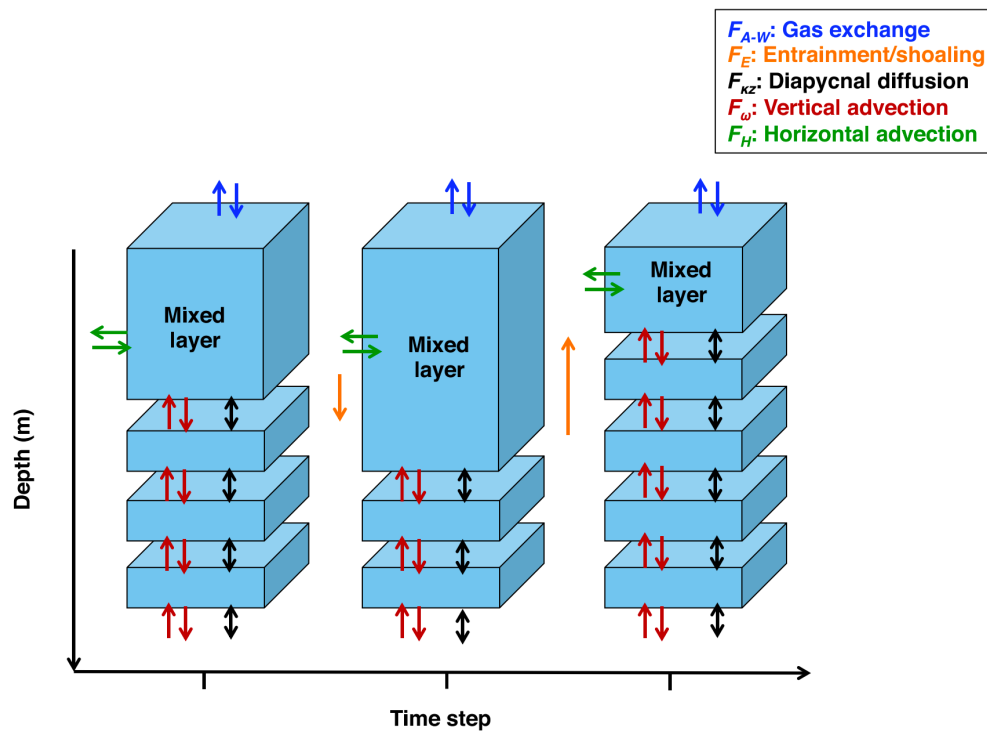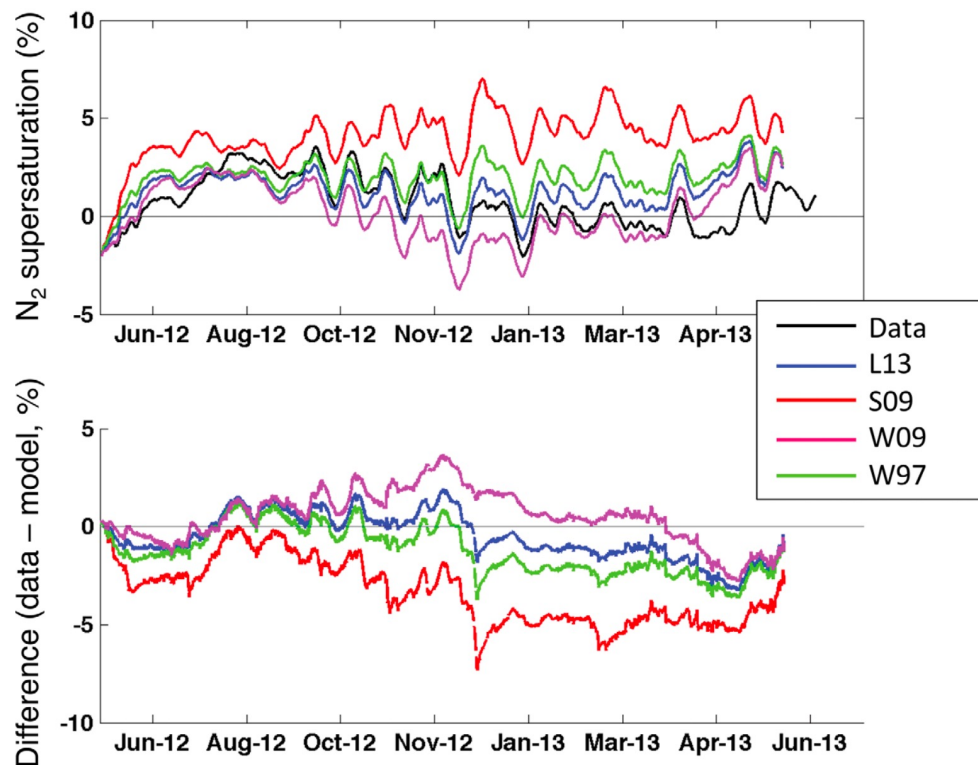# Experimental methods: Upper ocean mass balances

- N_2 supersaturation in the N. Pacific is purely due to physical processes
- If we can parameterize or ignore non-air-sea flux terms, we can solve for air-sea gas exchange

Emerson and Bushinsky, 2016

Experimental methods: Upper ocean mass balances

F_{A-W}: **Gas exchange**
F_E: **Entrainment/shoaling**
$F_{\kappa z}$: **Diapycnal diffusion**
F_ω: **Vertical advection**
F_H: **Horizontal advection**

- N_2 supersaturation in the N. Pacific is purely due to physical processes
- If we can parameterize or ignore non-air-sea flux terms, we can solve for air-sea gas exchange

$$\frac{d(h[C])}{dt} = F_{A-W} + F_H + F_\omega + F_E + F_{\kappa z} \pm J_C$$

Bushinsky et al., 2016

Experimental methods: Upper ocean mass balances

Emerson and Bushinsky, 2016

- N_2 supersaturation in the N. Pacific is purely due to physical processes
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$$\frac{d(h[C])}{dt} = F_{A-W} + F_H + F_\omega + F_E + F_{\kappa z} \pm J_C$$

0

- By testing different air-sea flux parameterizations we can find the one that minimizes differences from obs.
- Additional work then used this approach to tune the bubble injection terms

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Ocean observation derived gas transfer coeff.
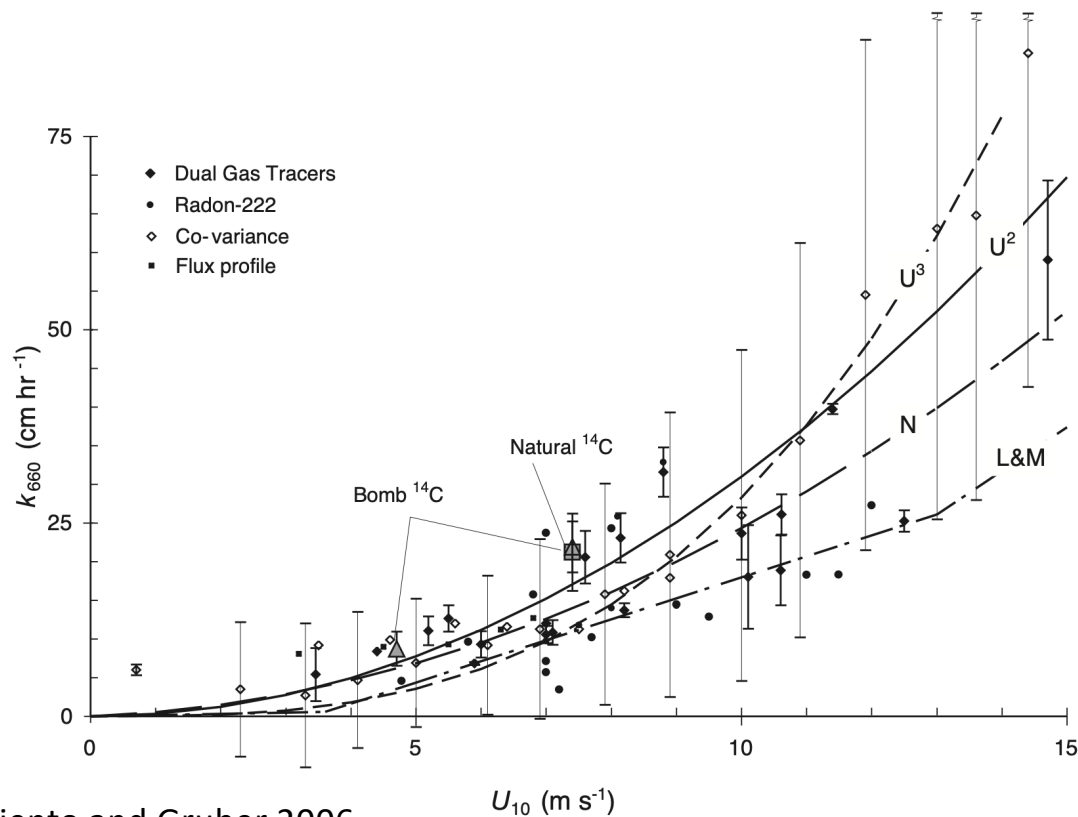
- Wide range in experimental observations
  - Does k vary according to U^2? U^3?

- Some of this reflects uncertainty in observations
- Some of this is the fact that wind speed alone cannot fully capture the wind-wave dynamics
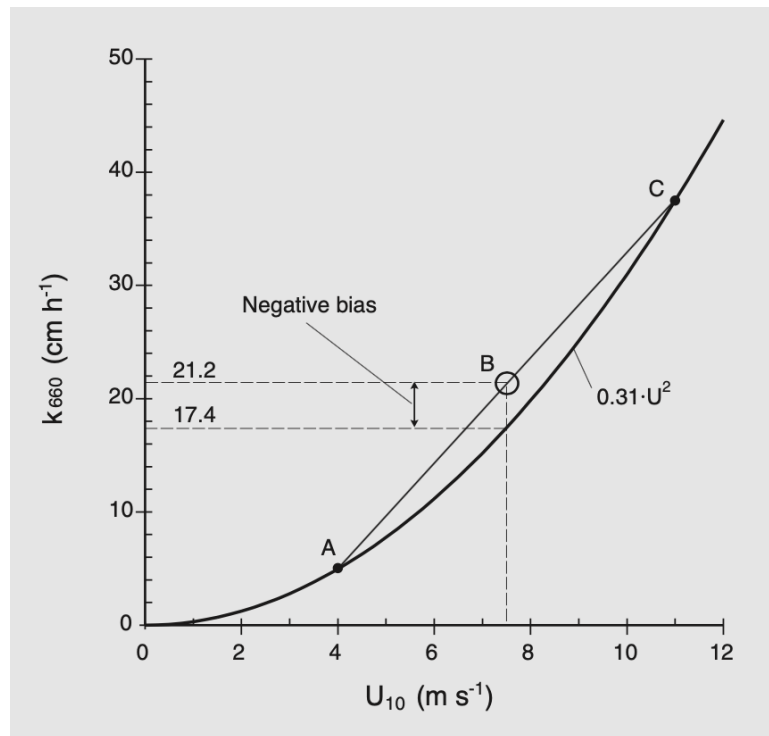
Sarmiento and Gruber 2006
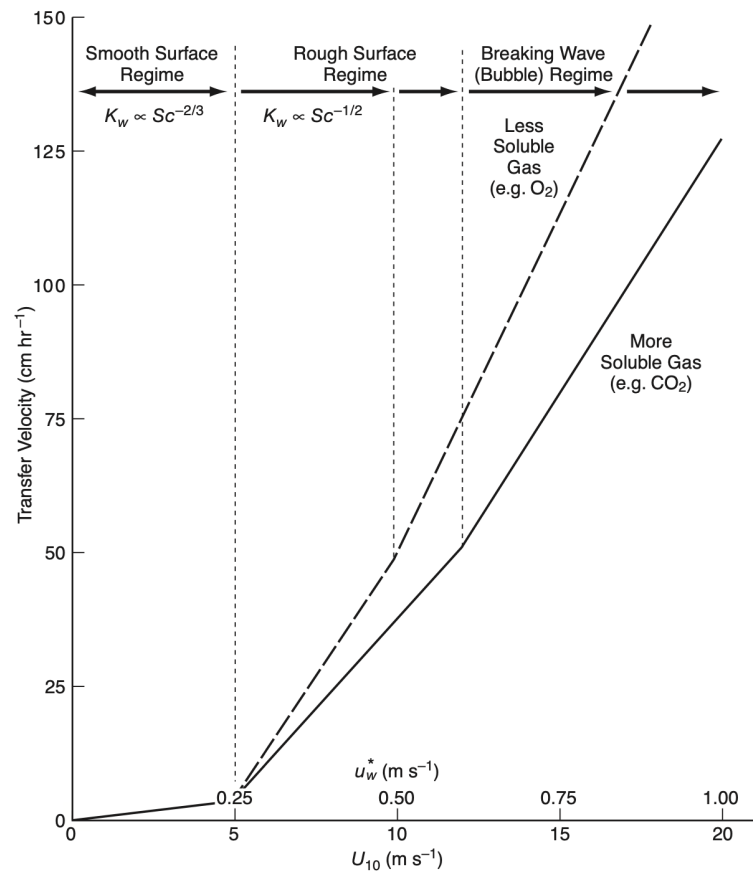
- Idealized relationship between the mass transfer velocity and wind speed at 10 m

$$F_{A-W} = F_S + F_C + F_P \qquad \text{mol m}^{-2}\text{ s}^{-1}$$

Total air-water flux (F_{A-W}) is a combination of diffusive flux, completely dissolved bubbles (F_C) and partially dissolved bubbles (F_P)
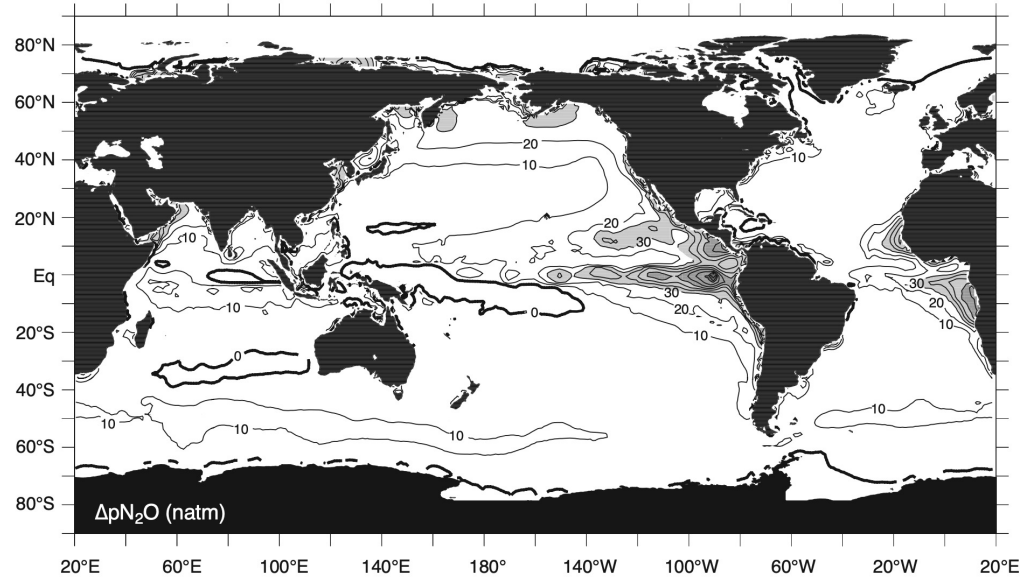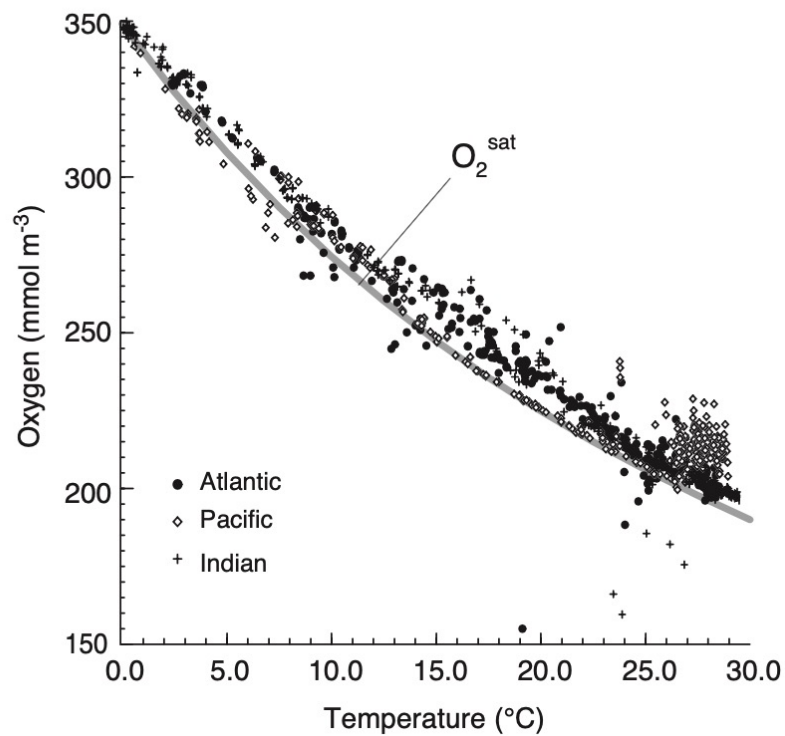
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Be careful with average winds...

- Calculating the mean gas transfer velocity based on mean wind speeds can lead to bias

- Make sure you use a parameterization that uses a similar wind averaging to your observations
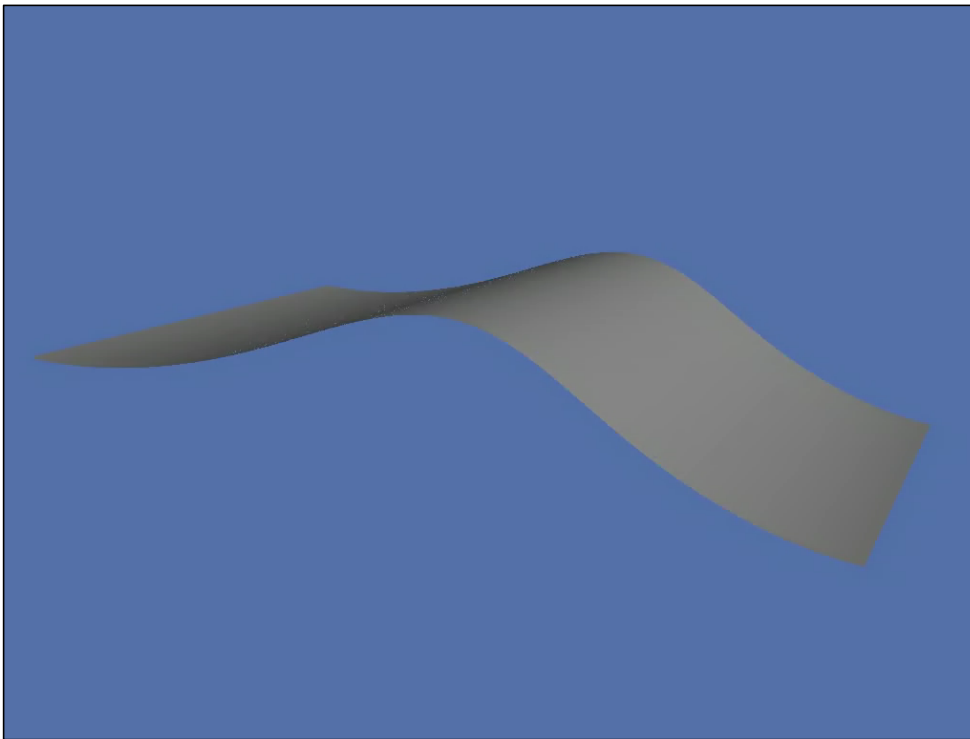
Sarmiento and Gruber, 2006

pCO$_2$ anomalies

ΔpCO_2 (JFM) (μatm)

ΔpCO_2 (JJA) (μatm)

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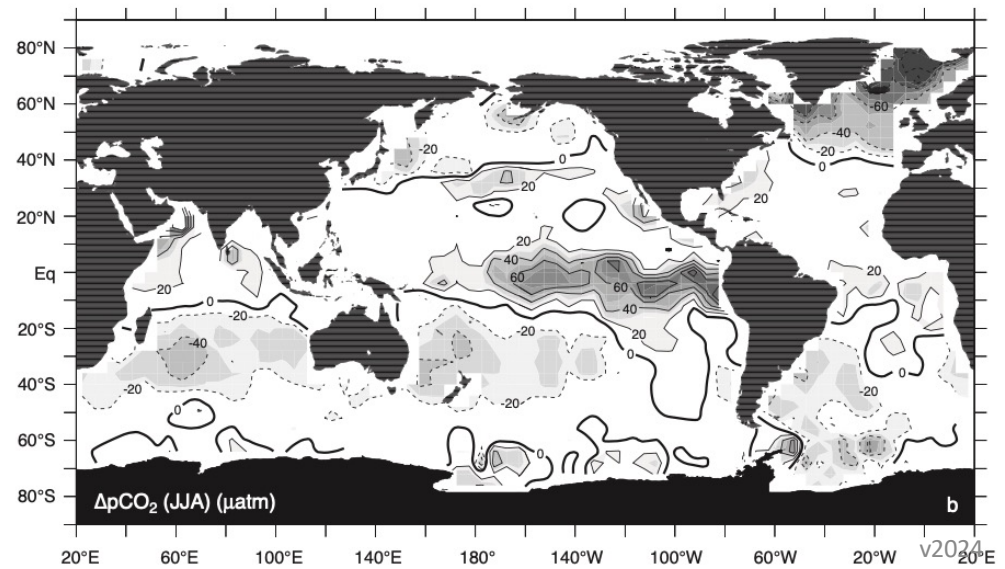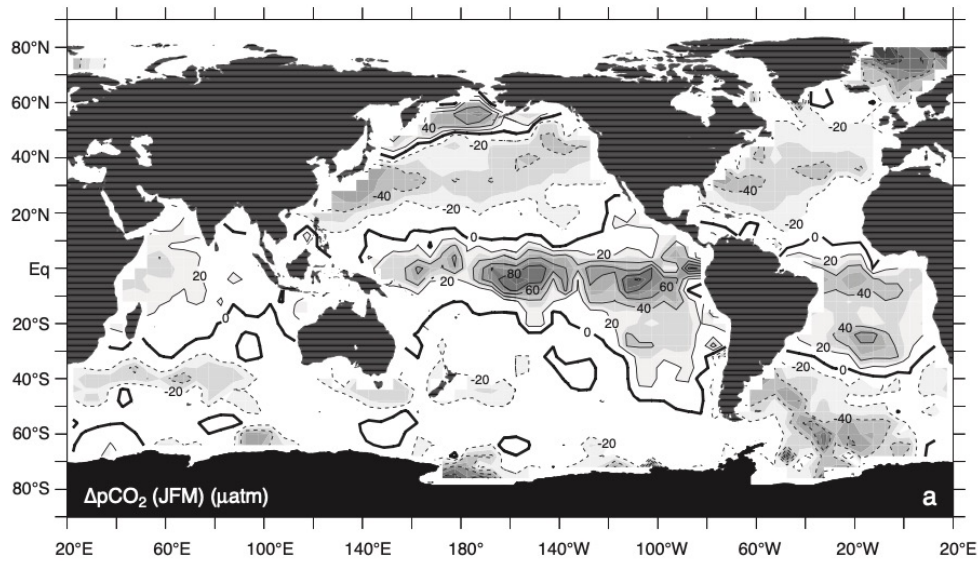
Oxygen and N₂O anomalies

Numerical model of breaking waves

- Physical model of breaking wave dynamics
- Goal is to understand breaking wave behavior beyond more wind = more waves
- As wave models become available, this may add another possible parameterization term that can reduce the spread in current estimates.

Deike et al. (2017)

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