VOCALS-REx 2008

Eddy Covariance Measurements of the Sea-to-air Flux of DimethylSulfide (DMS) on board of R/V Ronald H. Brown

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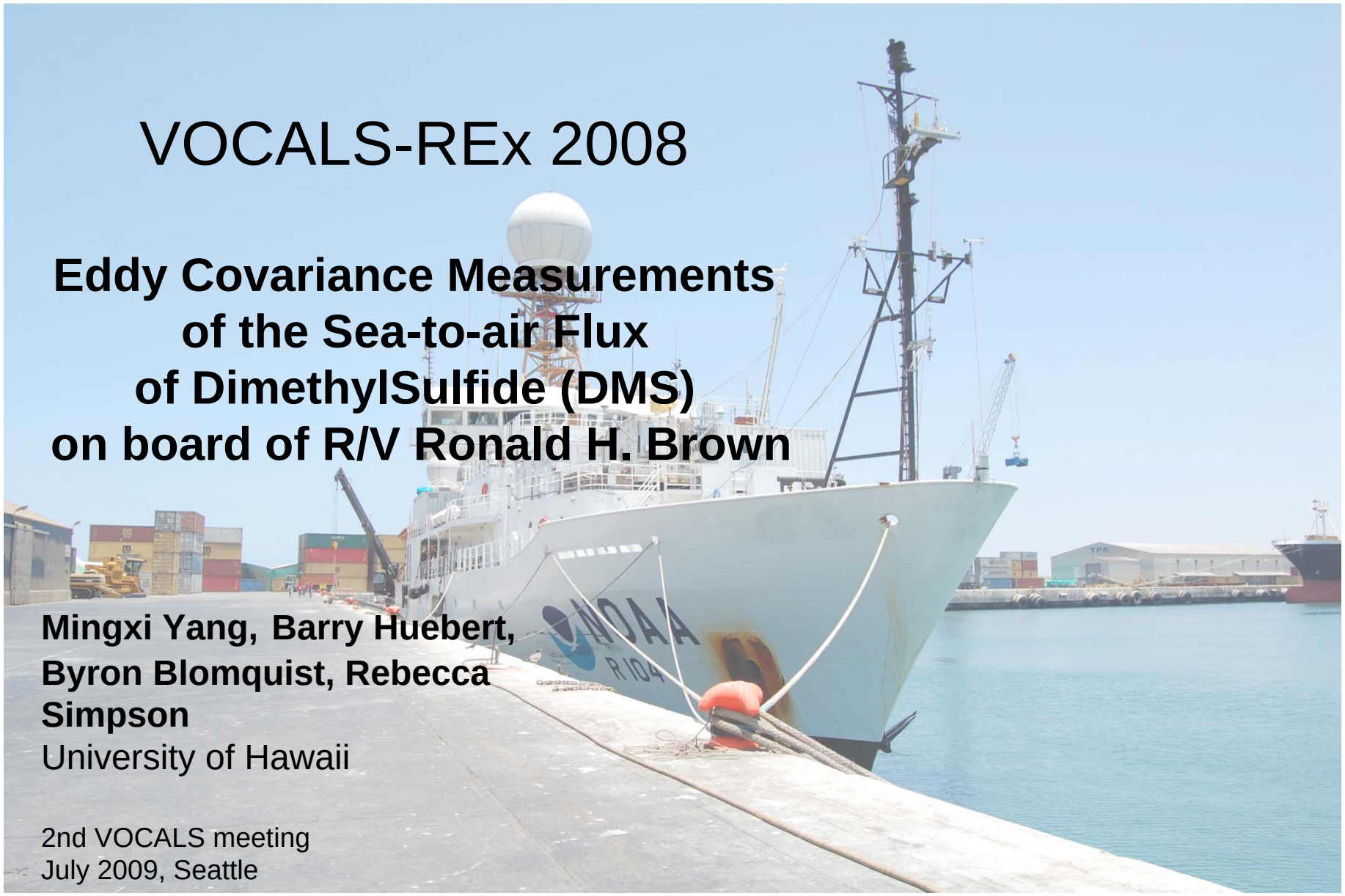
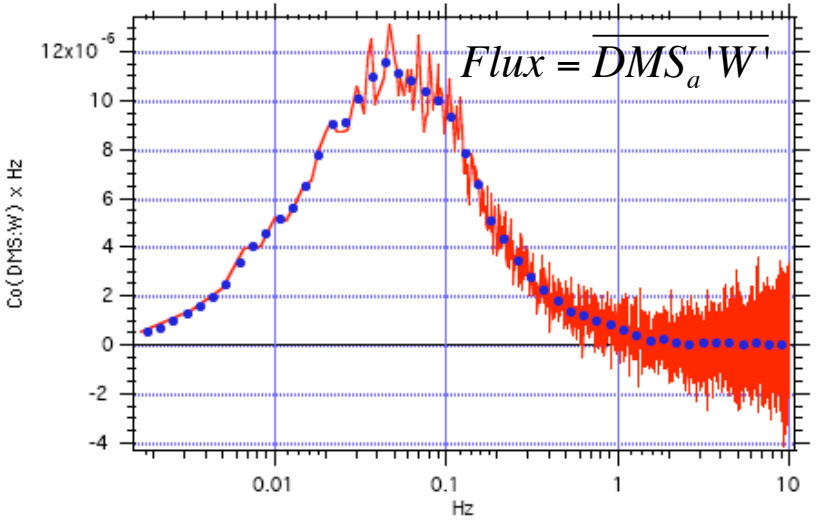


Picture taken by R. Simpson

DMS Sea-to-air Flux

DMS_w: GC

DMS_a: MS



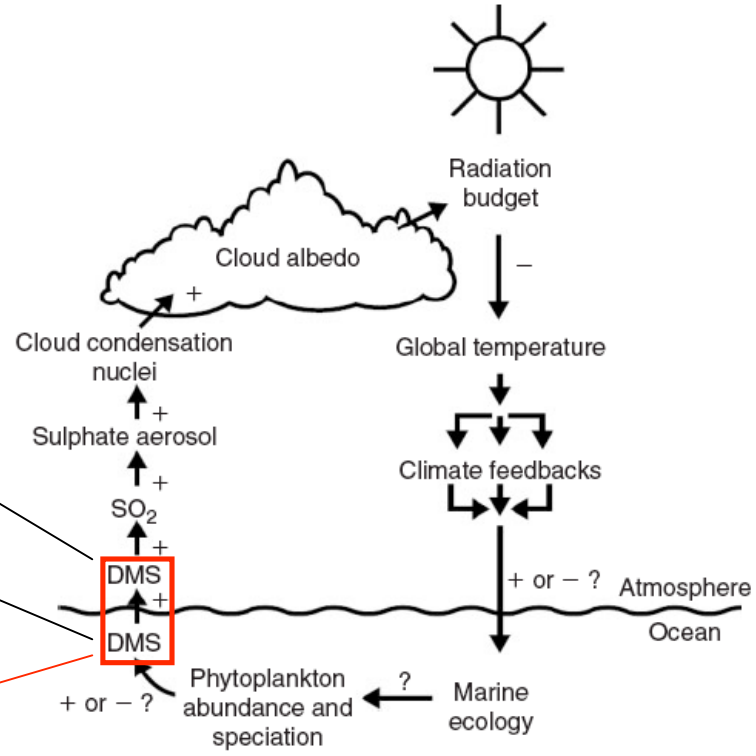
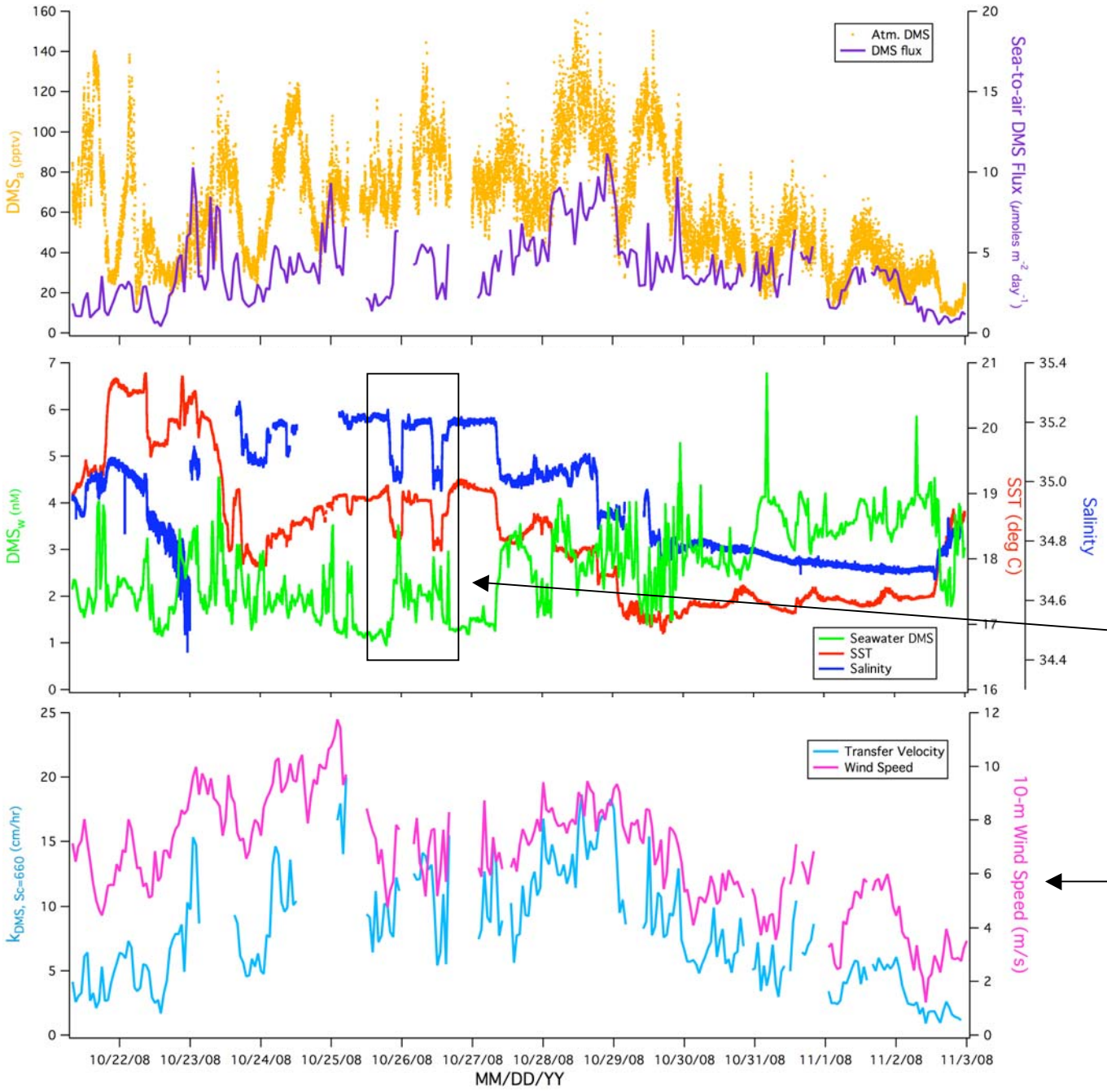


Figure from the International Geosphere–Biosphere Programme (IGBP) Report number 50

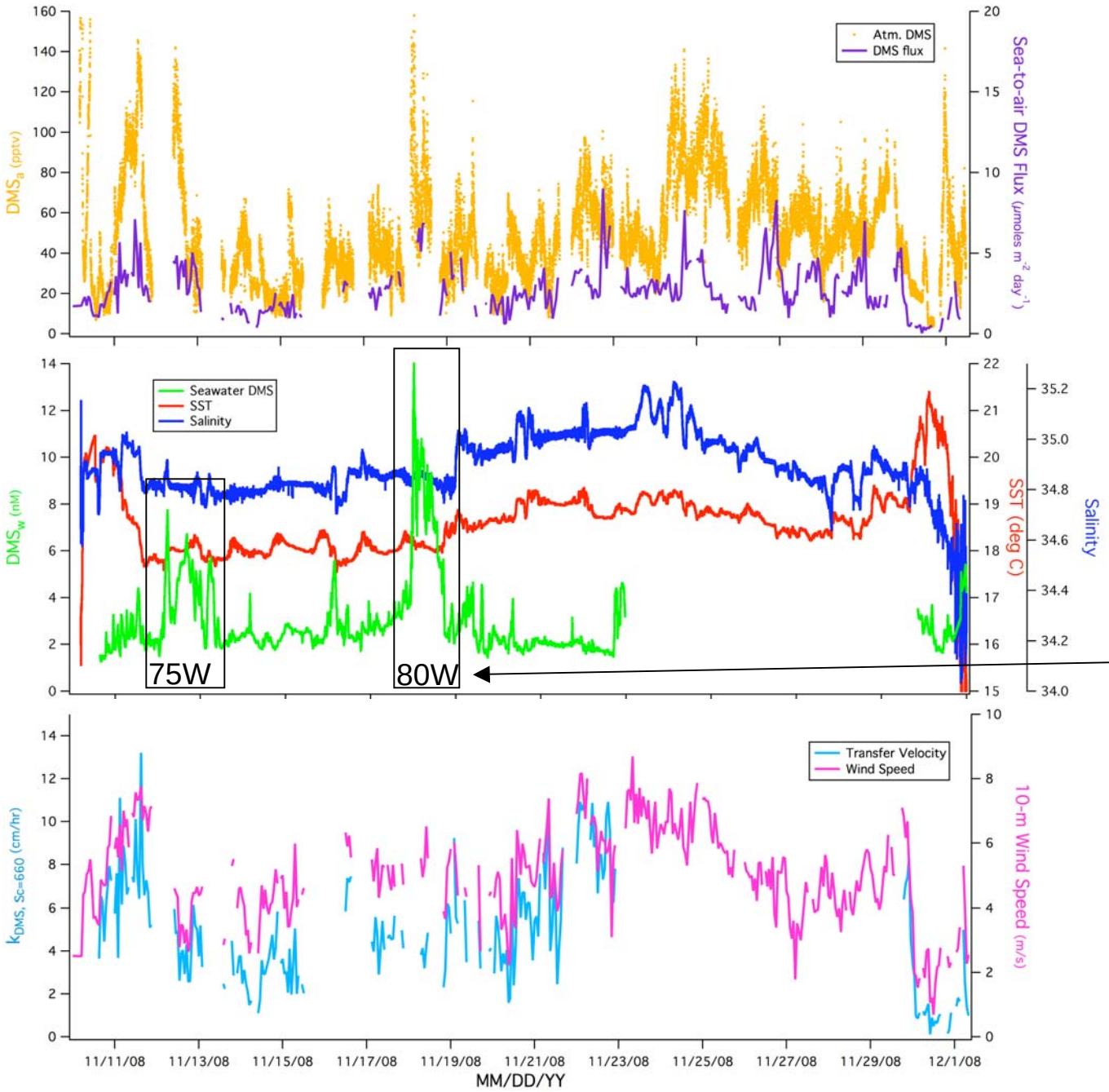


Leg 1

$$\frac{\text{Flux}}{(DMS_w - DMS_a)} = k_{DMS}$$

Localized DMS_w peaks associated with eddies

k_{DMS} largely followed wind speed



Leg 2

High DMS_w were observed in isolated pockets even away from the coast

Sea-to-air DMS Flux

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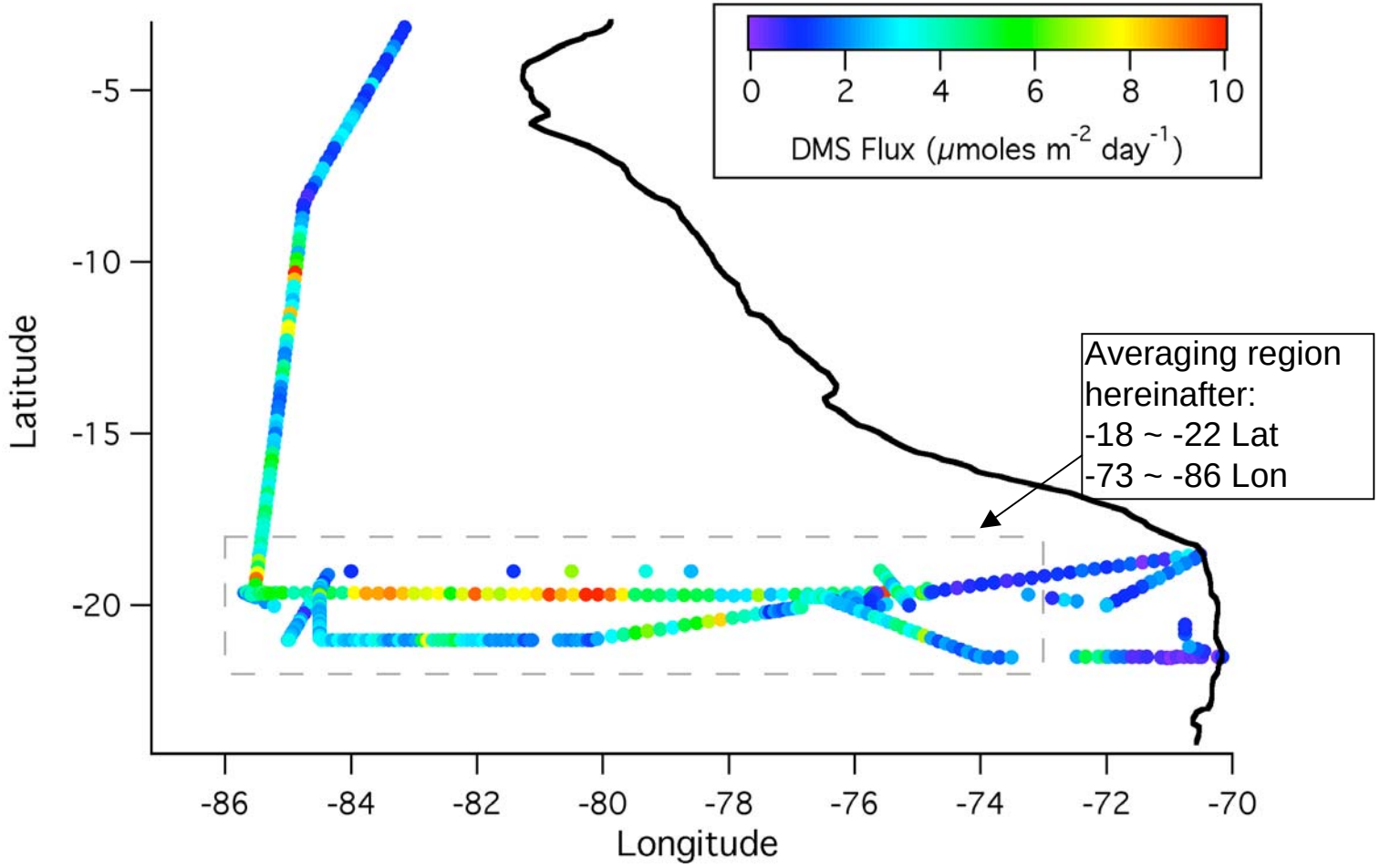
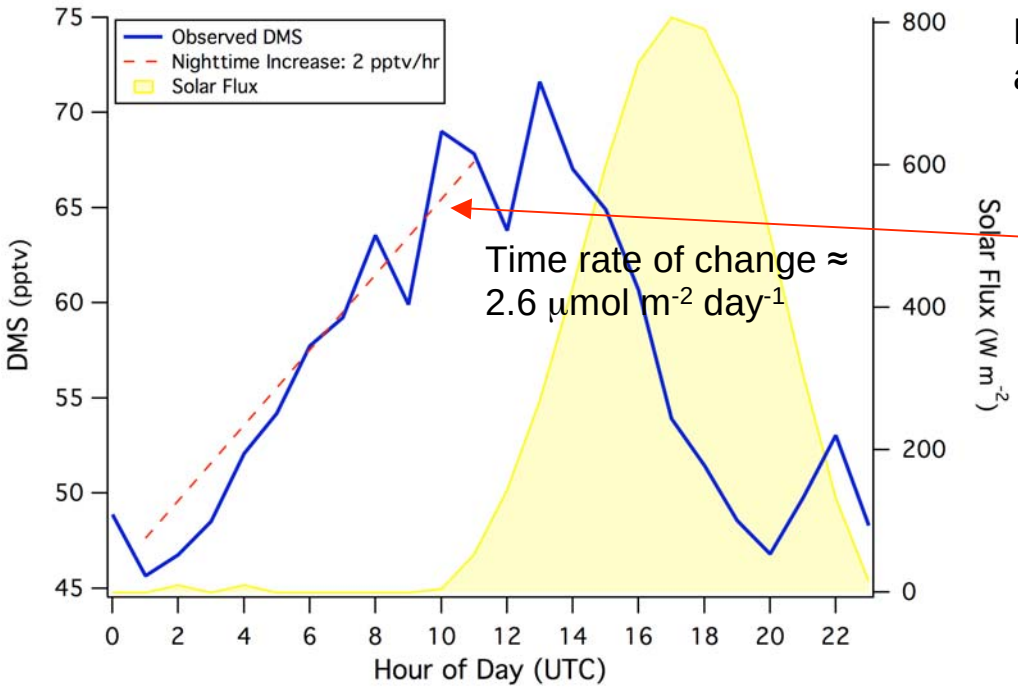
The Natural Source of Sulfur to the VOCALS Region



DMS showed Clear Diurnal Cycle
- allowing us to solve for entrainment velocity (ω_e)



Time rate of change ≈ 2.6 μmol m^{-2} day^{-1}

Horizontal advection small

No chemical production

Nighttime oxidation small

$$\frac{\partial \langle \overline{S} \rangle}{\partial t} + \langle \overline{u} \rangle \frac{\partial \langle \overline{S} \rangle}{\partial x} - F_0 + F_{z_i} = \langle P \rangle - \langle L \rangle$$

Sea-to-air flux measured by EC ≈ 3.2 μmol m^{-2} day^{-1}

Entrainment flux ≈ (3.2 - 2.6) = 0.6 μmol m^{-2} day^{-1}

No DMS above the inversion

$$F_{z_i} = \omega_e \{ [DMS_{z_i^-}] - [DMS_{z_i^+}] \}$$

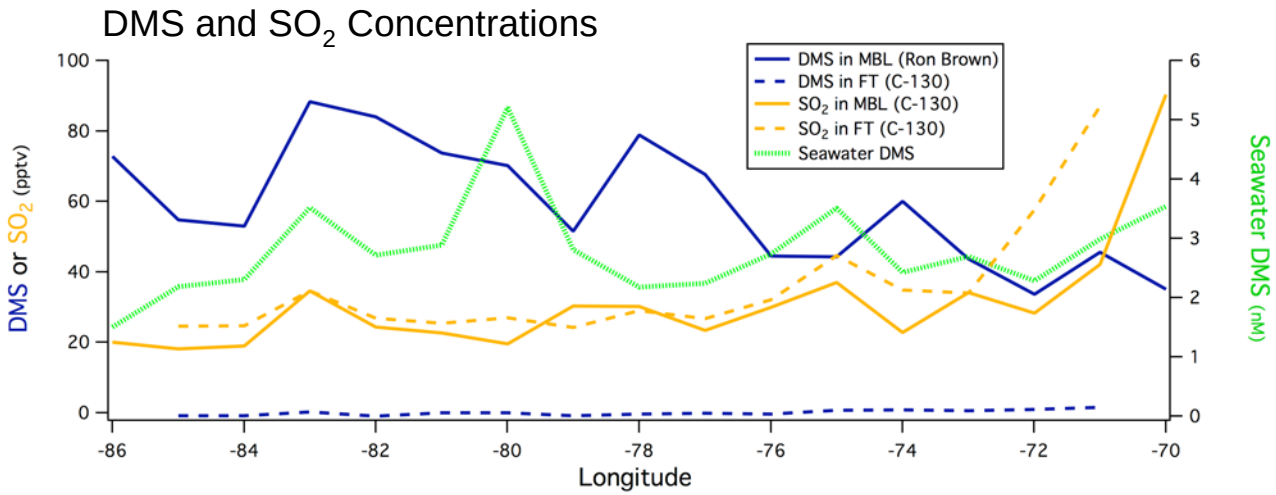
ω_e ≈ 4 mm sec^{-1}

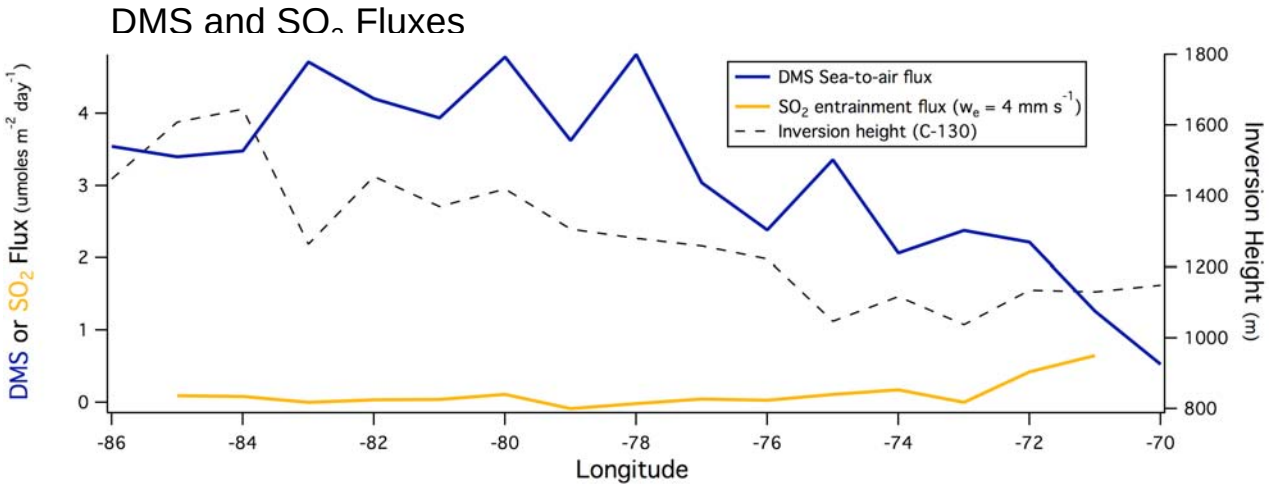
(agrees well with Wood and Bretherton 2004; Caldwell et al. 2005)

VOCALS-average DMS shows clear diurnal cycle, with maximum just after sunrise (built up from air-sea exchange) and minimum just before sunset (OH oxidation)
- this implies limited mesoscale instabilities

What was the Main Sulfur Source for nss-Aerosols during VOCALS?

- Zonal Averages along 20°S (except RF-14 from the C-130)

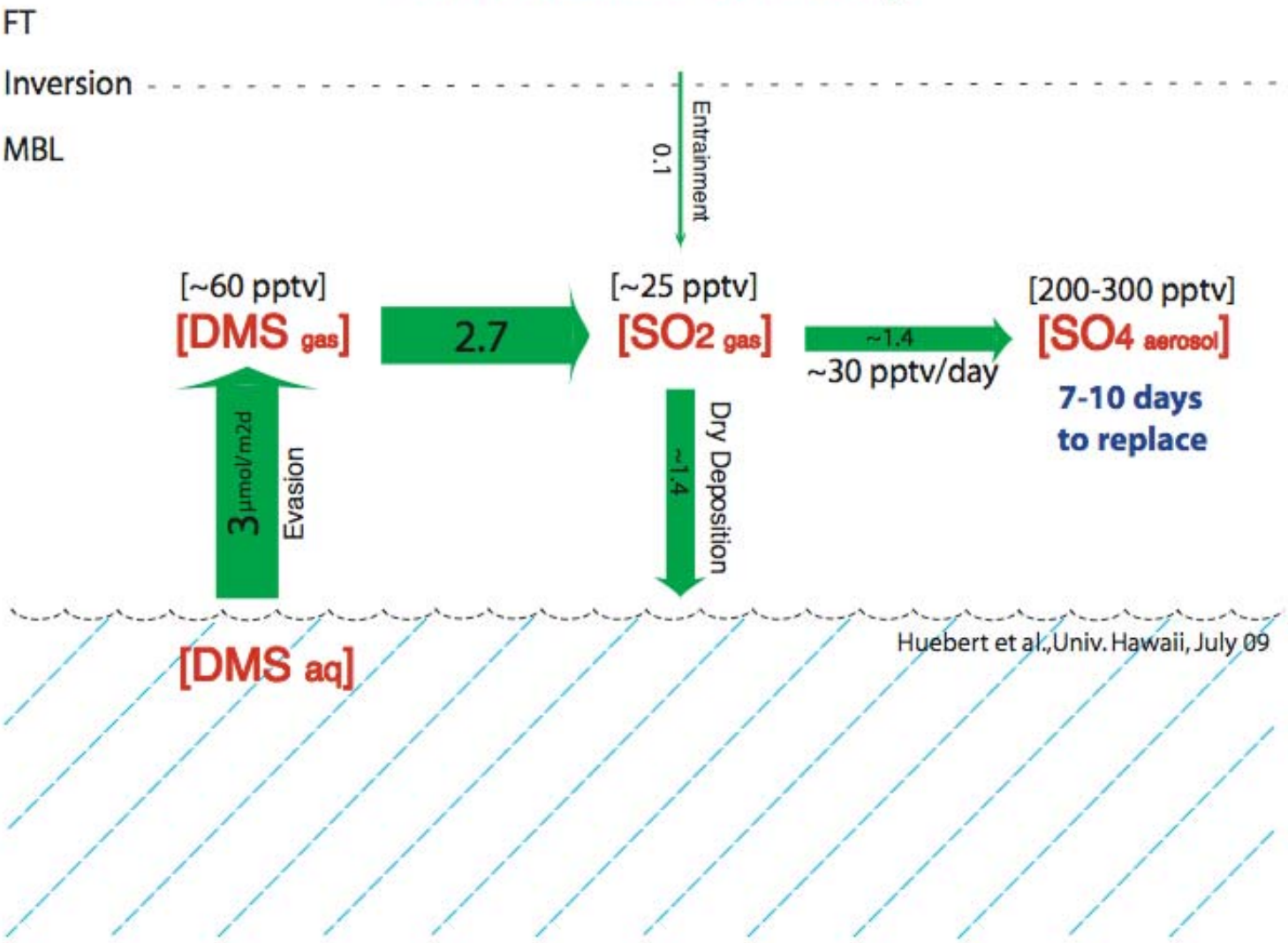




Away from the coast,
DMS Sea-to-air flux >> SO_2 entrainment flux

While there were instances of off-shore transport, typically anthropogenic SO_2 had virtually no impact on VOCALS MBL Sulfur

VOCALS Sulfur Chemistry

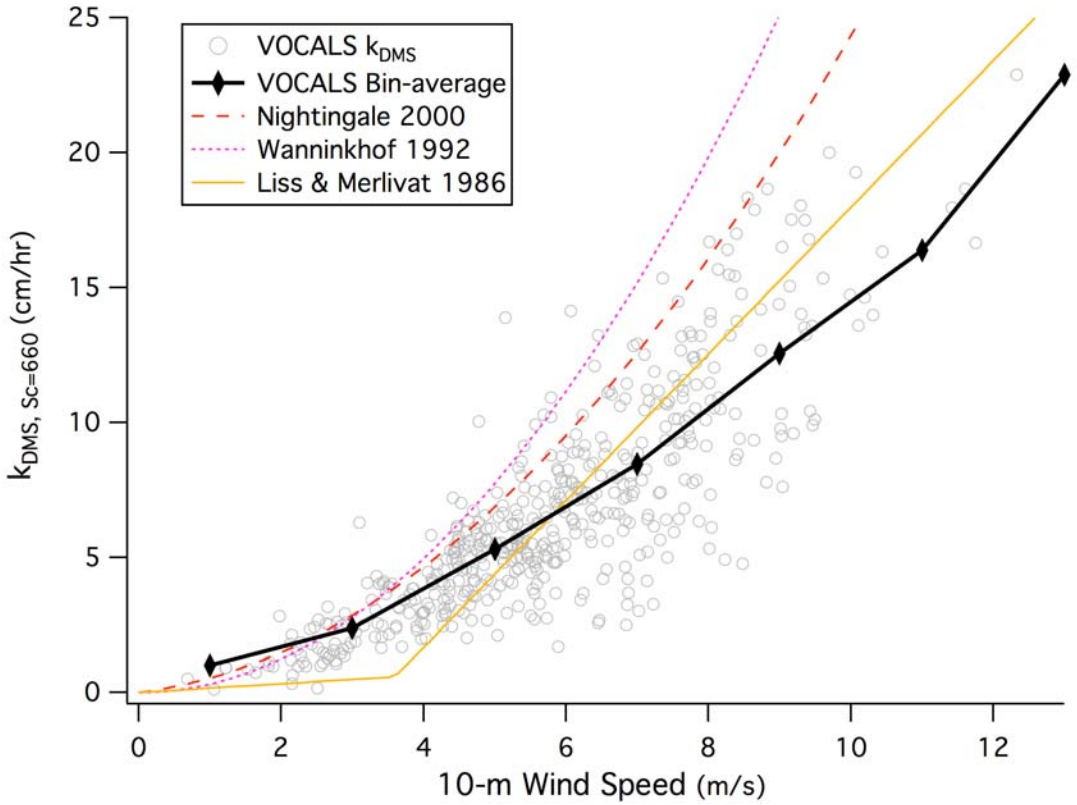


Air-Sea Transfer Velocity of DMS

$$k_{DMS} = Flux/(DMS_w - DMS_a/\alpha)$$

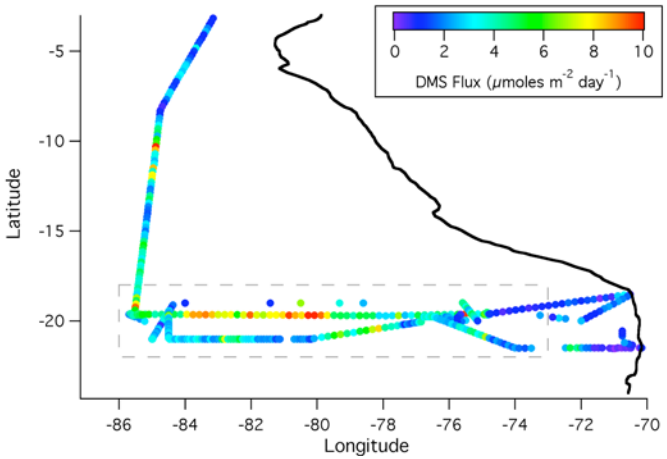
$$k_{660} = k(\frac{660}{Sc})^{-1/2}$$

k_{DMS} : transfer velocity
DMS_a: atm. DMS
DMS_w: seawater DMS
α : solubility coefficient
Sc: Schmidt number



Oft-used wind speed based parameterizations over-estimate k_{DMS}, with Liss & Merlivat (1992) coming the closest to observations (except in very low winds)

Conclusions



Coupled Ocean-Atmosphere-Land Hypothesis #2: By changing the physical and chemical properties of the upper ocean, upwelling has a systematic and noticeable effect on aerosol precursor gases and the aerosol size distribution over the SEP.

Synthesis: DMS concentrations were not strongly elevated near the coastal upwelling zone. However, photochemical destruction of DMS seemed to be dominant source of new sulfate far offshore, dominating SO_2 entrainment from the free troposphere.

- DMS concentration and sea-to-air flux averaged to be 60 pptv and 3 μmol m^{-2} day^{-1}, respectively (both were higher offshore)
- Away from the coast, DMS emission from the ocean was the dominant sulfur source in the MBL, not entrainment of pollution-derived SO_2
- It would take 7~10 days to refill to the typical sulfate concentration from DMS oxidation alone
  - Probably why POCs persist for so long

Acknowledgements

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