DMS IN THE REMOTE, TROPICAL MBL

3RD PASE WORKSHOP

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Scalar Budget Equation

$$\underset{I}{\frac{\partial c}{\partial t}} + \underset{II}{\nabla \cdot (\boldsymbol{u}c)} = \underset{III}{-L}$$

- I – Storage (rate of change of concentration.
- II – Flux Divergence
- III – Chemical source/sink

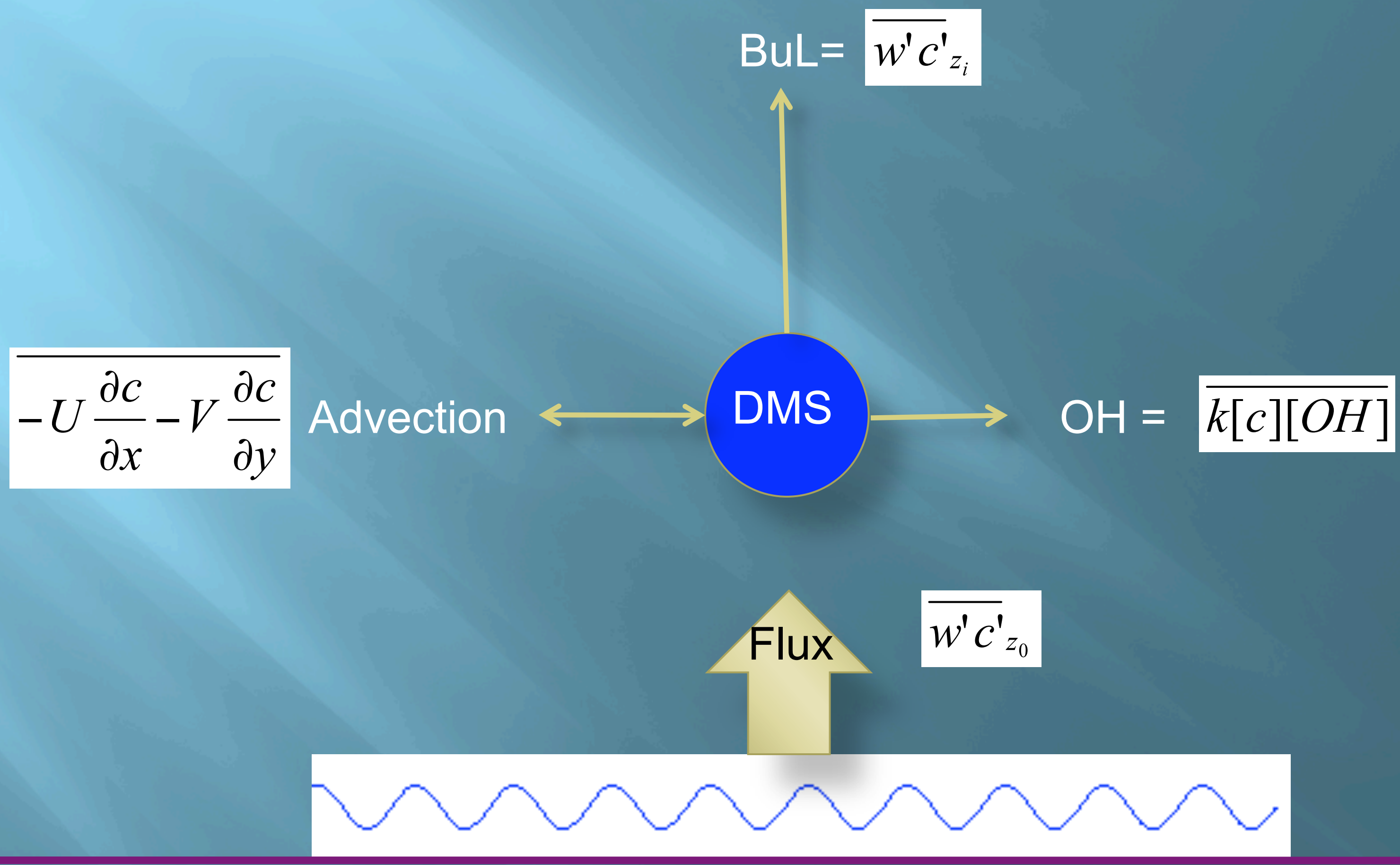
Budget Simplifications

- Flow is incompressible: $\nabla \cdot \boldsymbol{u} = 0$
- Turbulence is horizontally homogeneous: $\frac{\partial}{\partial x}(\overline{u'c'}) = 0; \frac{\partial}{\partial y}(\overline{v'c'}) = 0$

Simplified Budget Equation:

$$\frac{\partial C}{\partial t} = -U\frac{\partial C}{\partial x} - V\frac{\partial C}{\partial y} - \frac{\partial}{\partial z}(\overline{w'c'}) - L$$

DMS From the sea surface up!



Estimating the budget terms

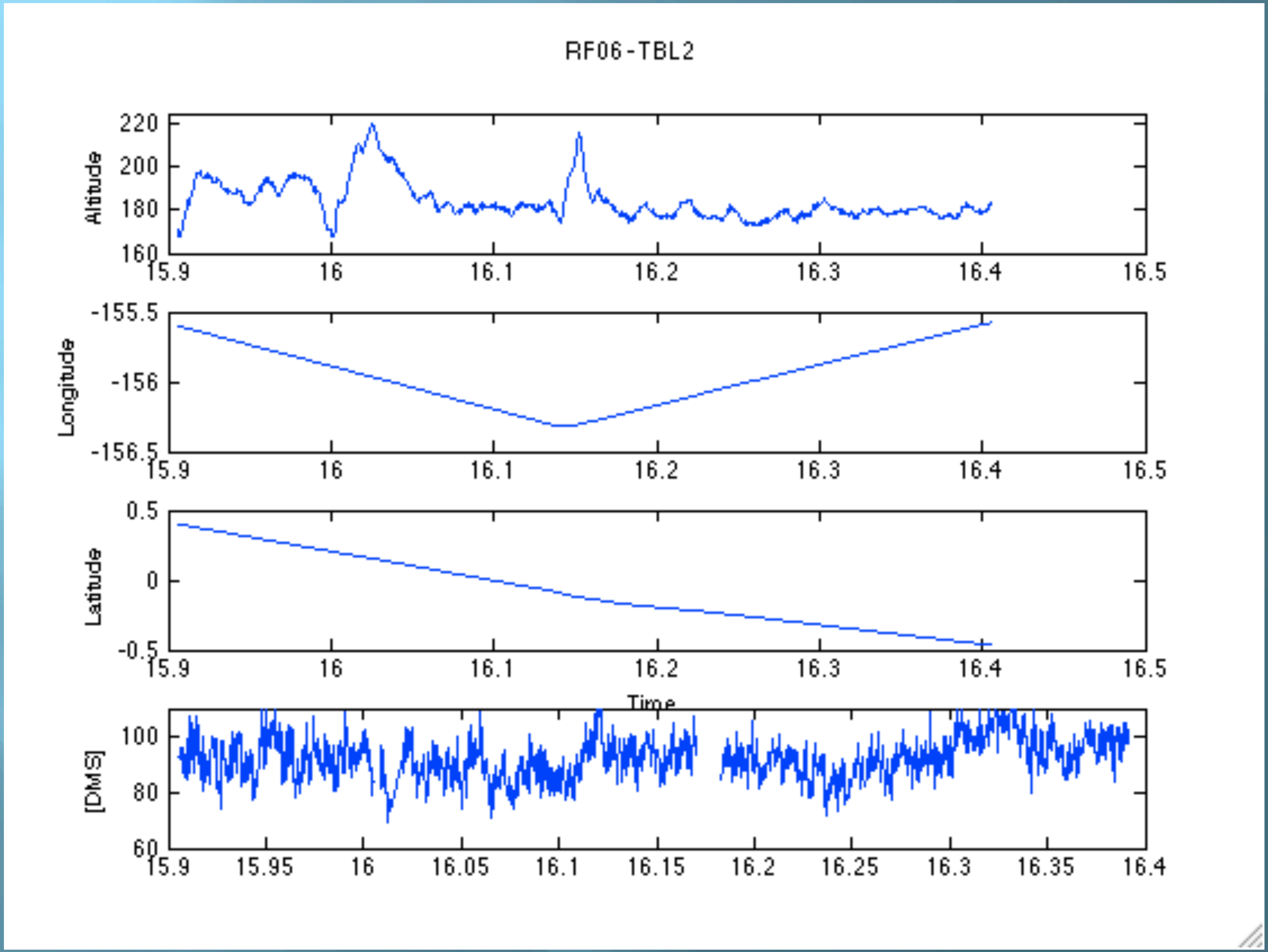
Actual DMS concentration is a complex function of position and time, i.e.

$c = f(x,y,z,t)$

Expanding f in a Taylor series, retaining only first order terms leads to:

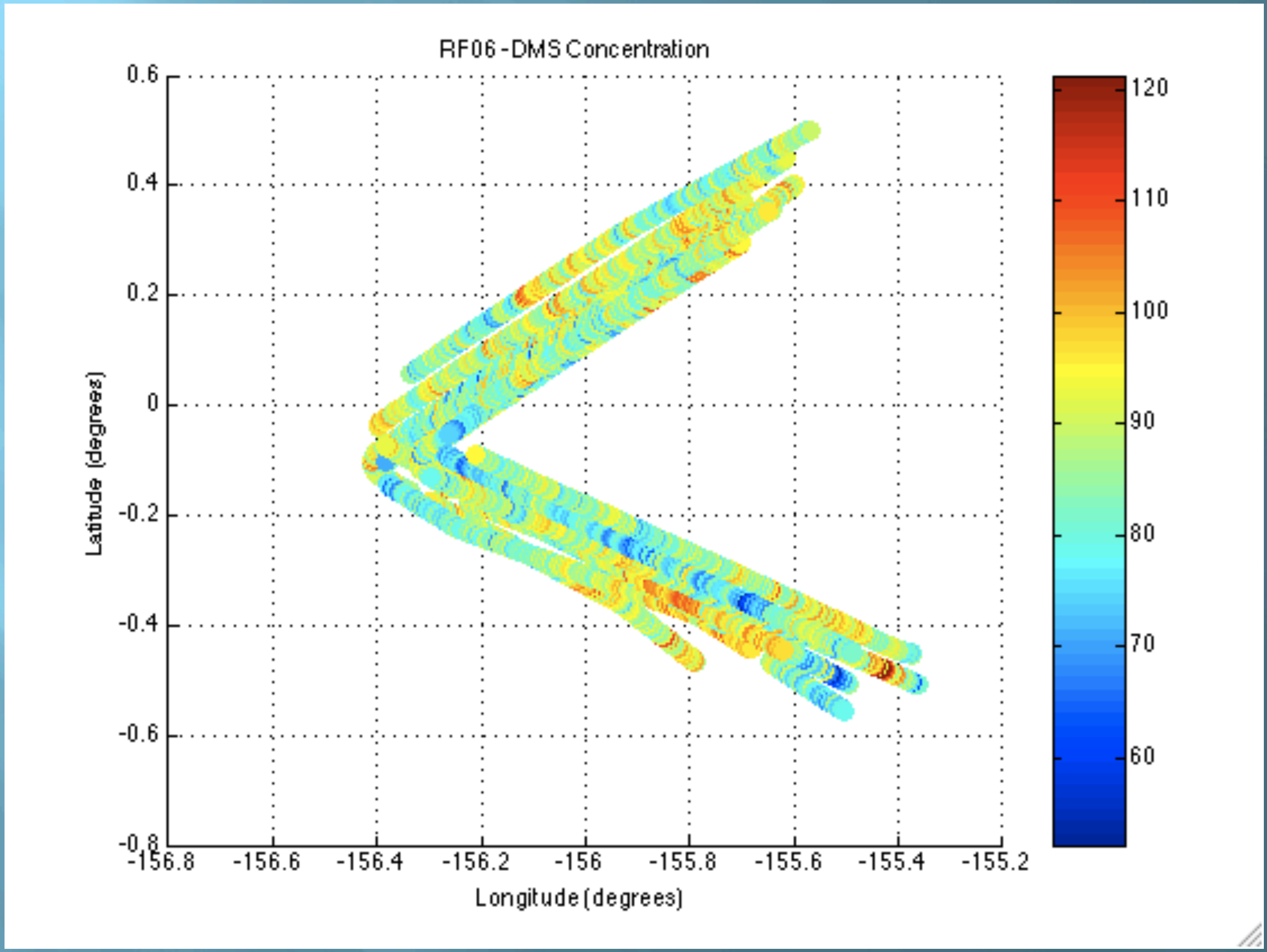
$$c = c_0 + \frac{\partial c}{\partial x}\Delta x + \frac{\partial c}{\partial y}\Delta y + \frac{\partial c}{\partial z}\Delta z + \frac{\partial c}{\partial t}\Delta t$$

RF06 TBL2

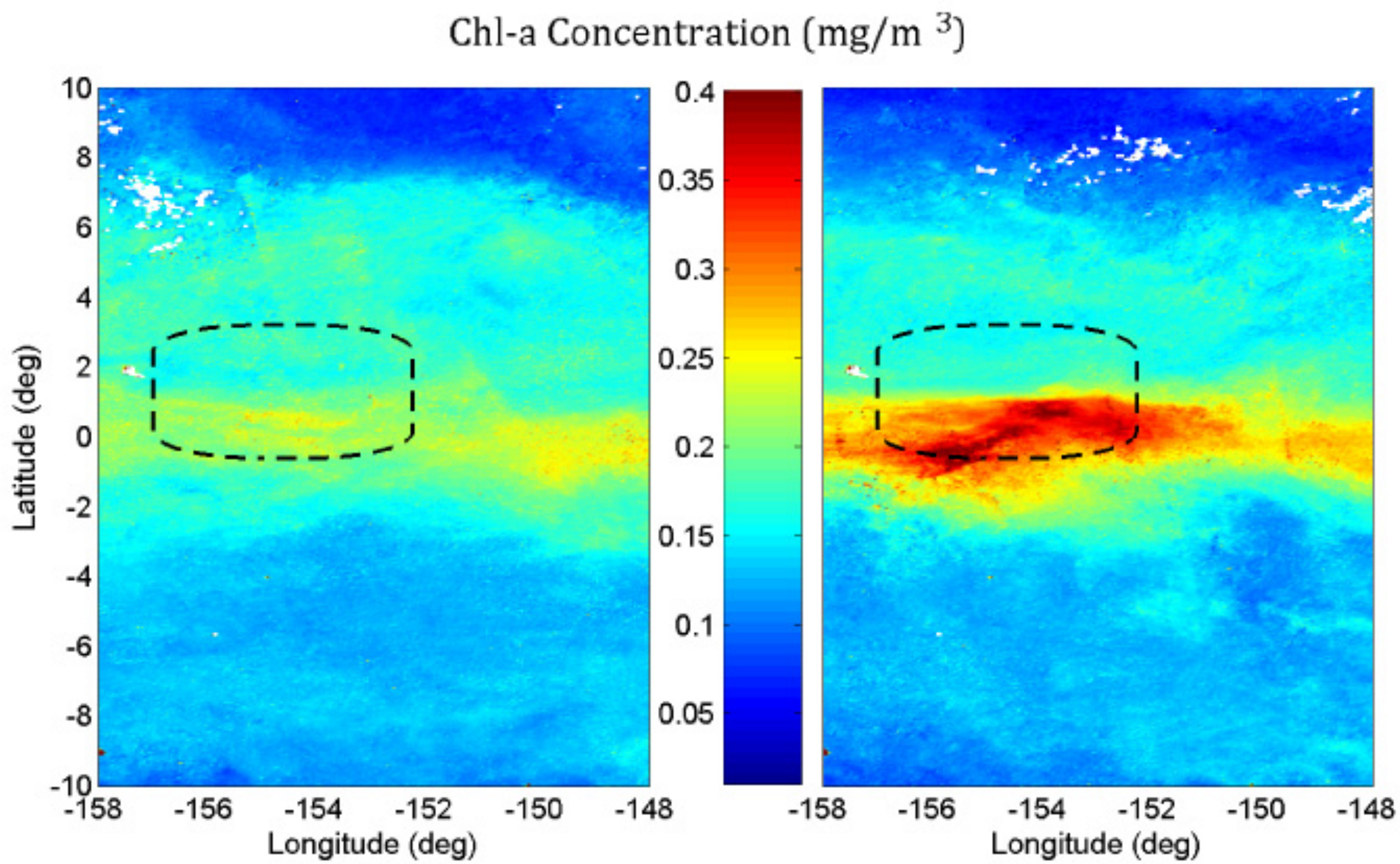


RF06-TBL2
Altitude
220
200
180
160
15.9
16
16.1
16.2
16.3
16.4
16.5
Longitude
-155.5
-156
-156.5
Latitude
0.5
0
-0.5
Time
[DMS]
100
80
60
15.95
16.05
16.15
16.25
16.35

RF06 DMS Concentration

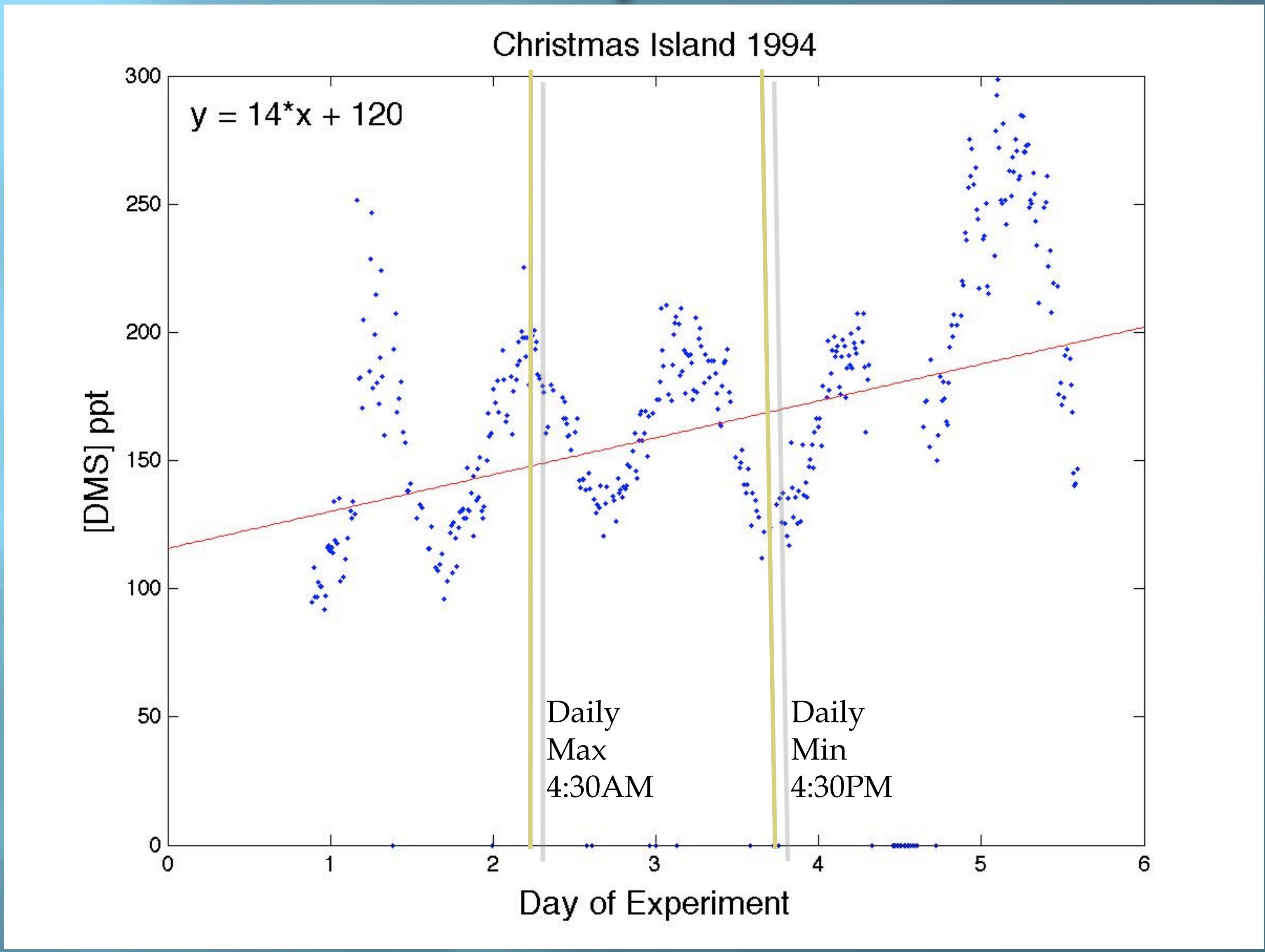


Horizontal Homogeneity?



First Two Weeks of PASE | Last Two Weeks of PASE

Time Dependence



DMS Rose by 14 ppt/day – 70 ppt total!

Methodology

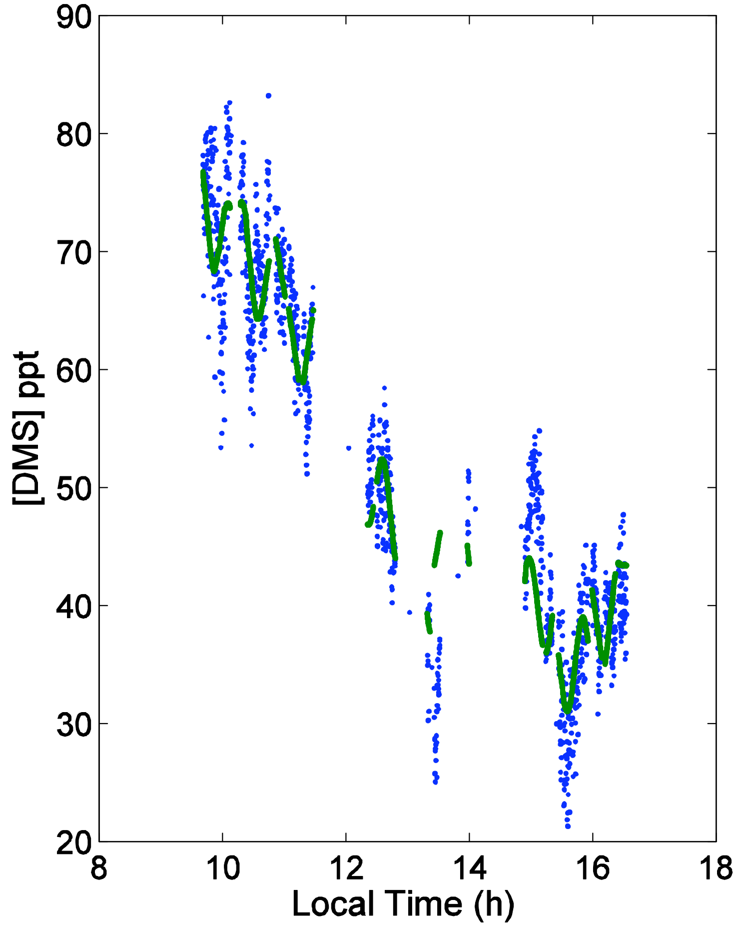
- Assumption:

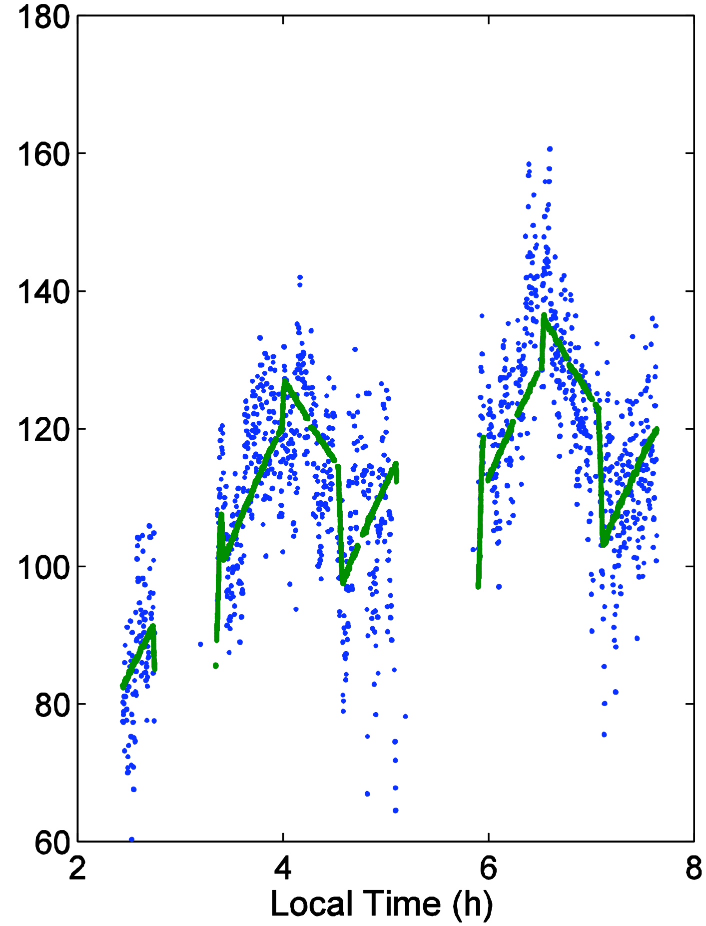
$$c = \beta_0 + \beta_1 x + \beta_2 y + \beta_3 z + \beta_4 t + \beta_5 \cdot \sin(\frac{2\pi(t-\phi)}{24})$$

- Find the coefficients (β_n) such that the sum of the squares of the deviations between [DMS] calculated and actual is a minimum.

- Passed 1-second averages of *c,x,y,z,t* into Matlab's multiple linear regression solver to estimate the coefficients and the uncertainty in those coefficients.

DMS Fit Accuracy





RF02: R^2 = 0.82

RF02: R^2 = 0.44

Vertical Flux Profile

- Take the vertical derivative of budget equation:

$$\frac{\partial}{\partial z}\left(\frac{\partial C}{\partial t}\right) = -\frac{\partial}{\partial z}\left(U\frac{\partial C}{\partial x} - V\frac{\partial C}{\partial y}\right) - \frac{\partial^2}{\partial z^2}\left(\overline{w'c'}\right) - \frac{\partial L}{\partial z}$$

- Switch the time and height derivatives and assume advection is constant with height. Given the small variability of OH in the MBL, assume the last term is zero also. This leads to the assumption of linearity in the flux profile, i.e. $\frac{\partial^2}{\partial z^2}\left(\overline{w'c'}\right) = 0$

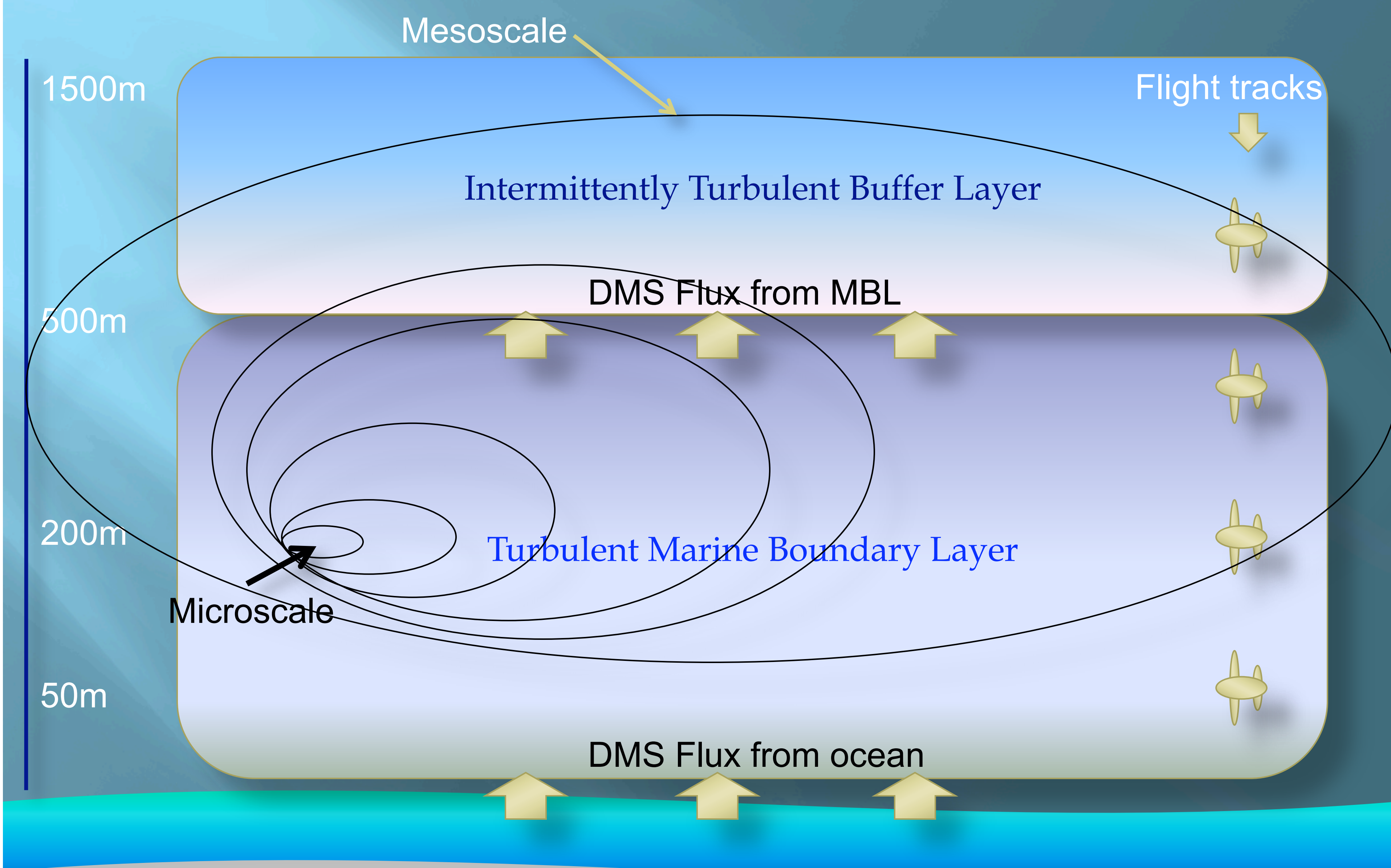
Vertical Flux Calculation

$$Flux = \overline{w'c'}$$

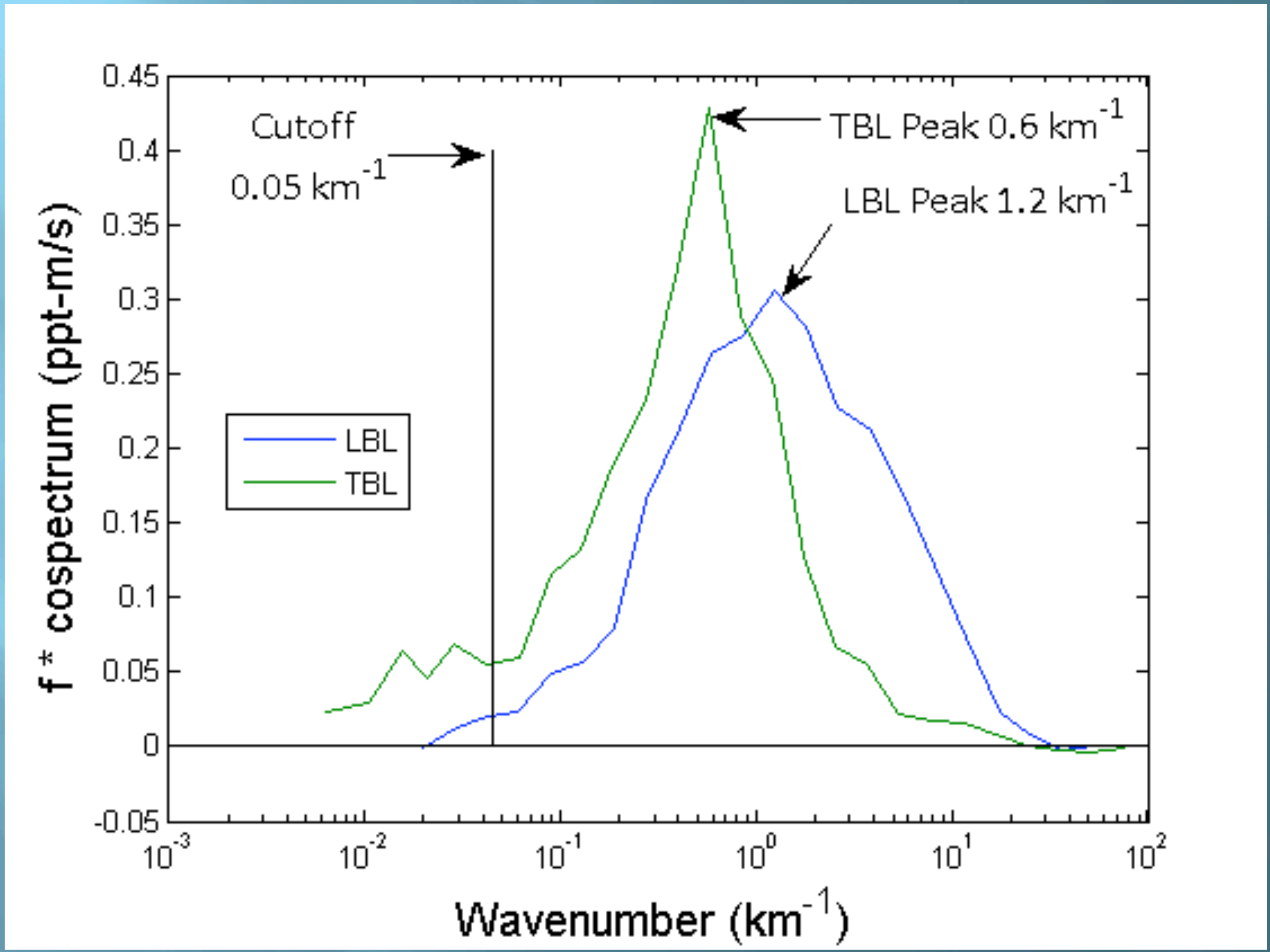
$$w' = w - \overline{w} \qquad c' = c - \overline{c}$$

What is the mean vertical wind?

Scales of Motion

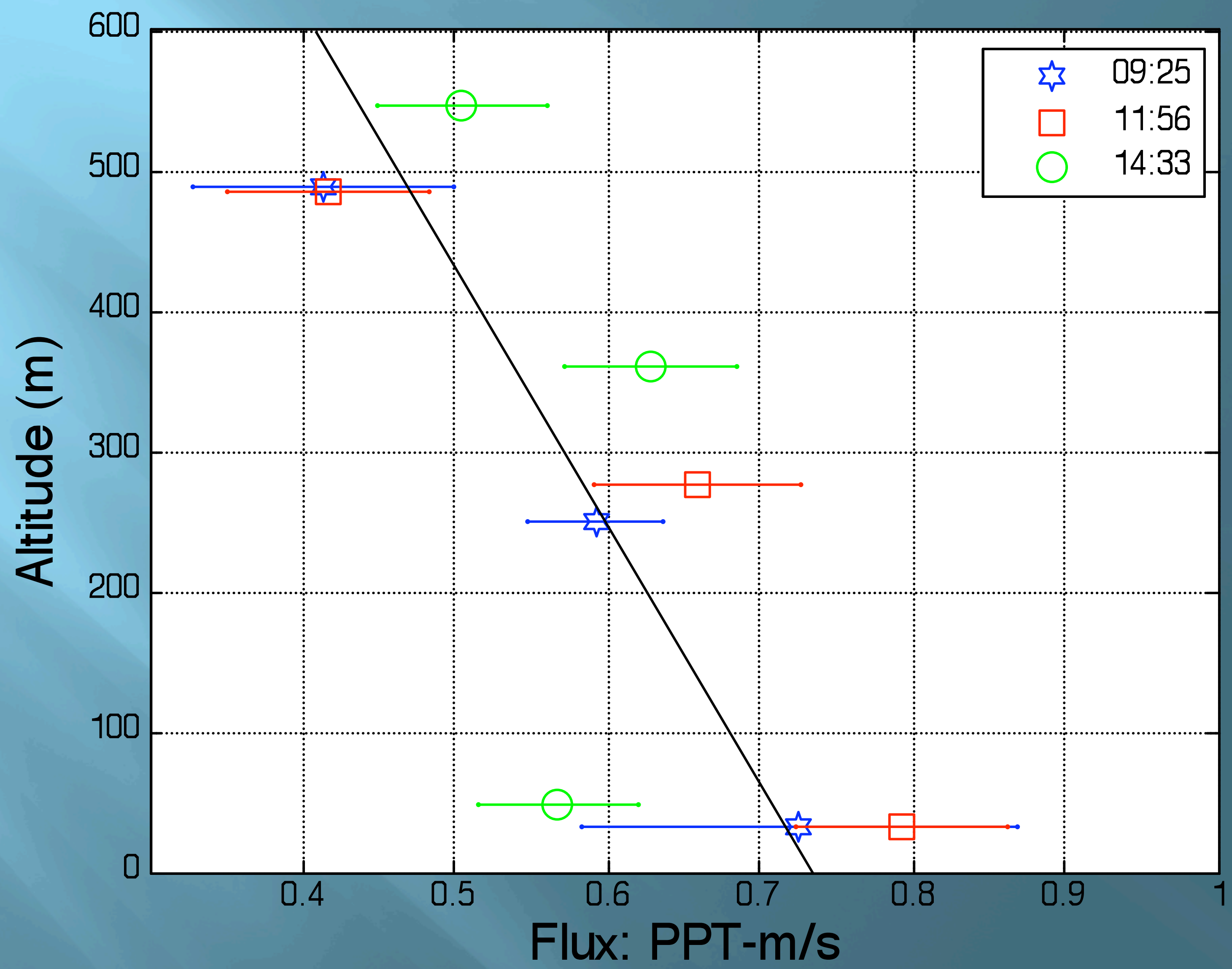


Cospectra of DMS and W



Cutoff corresponds to length scale of 20 km.

RF03 Vertical DMS Flux Profile



DMS Budget

RF	Obs ppt/hr	Div ppt/hr	Adv ppt/hr	Chem ppt/hr	OH	Net Error
2	-5.2	0.9	0.5	-6.5	-7.0	0.4
3	-6.0	1.9	-0.8	-7.1	-7.0	0.4
5	-4.7	4.1	2.0	-10.9	-10.8	0.7
8	-5.6	0.8	-0.5	-5.8	-5.5	0.4
11	-5.9	1.3	2.3	-9.5	-6.4	1.4
12	-5.9	1.5	-1.1	-6.3	-7.0	0.5
14	-7.7	-0.8	-1.2	-5.7	-10.7	0.5
Avg	**-5.8**	**1.4**	**0.2**	**-7.4**	**-7.8**	**0.6**
6	-0.8	-0.2	1.8	-2.4	-2.2	0.6
13	-1.6	3.2	-2.5	-2.2	-3.4	0.7
Avg	**-1.2**	**1.5**	**-0.4**	**-2.3**	**-3.4**	**0.6**

Dark Territory?

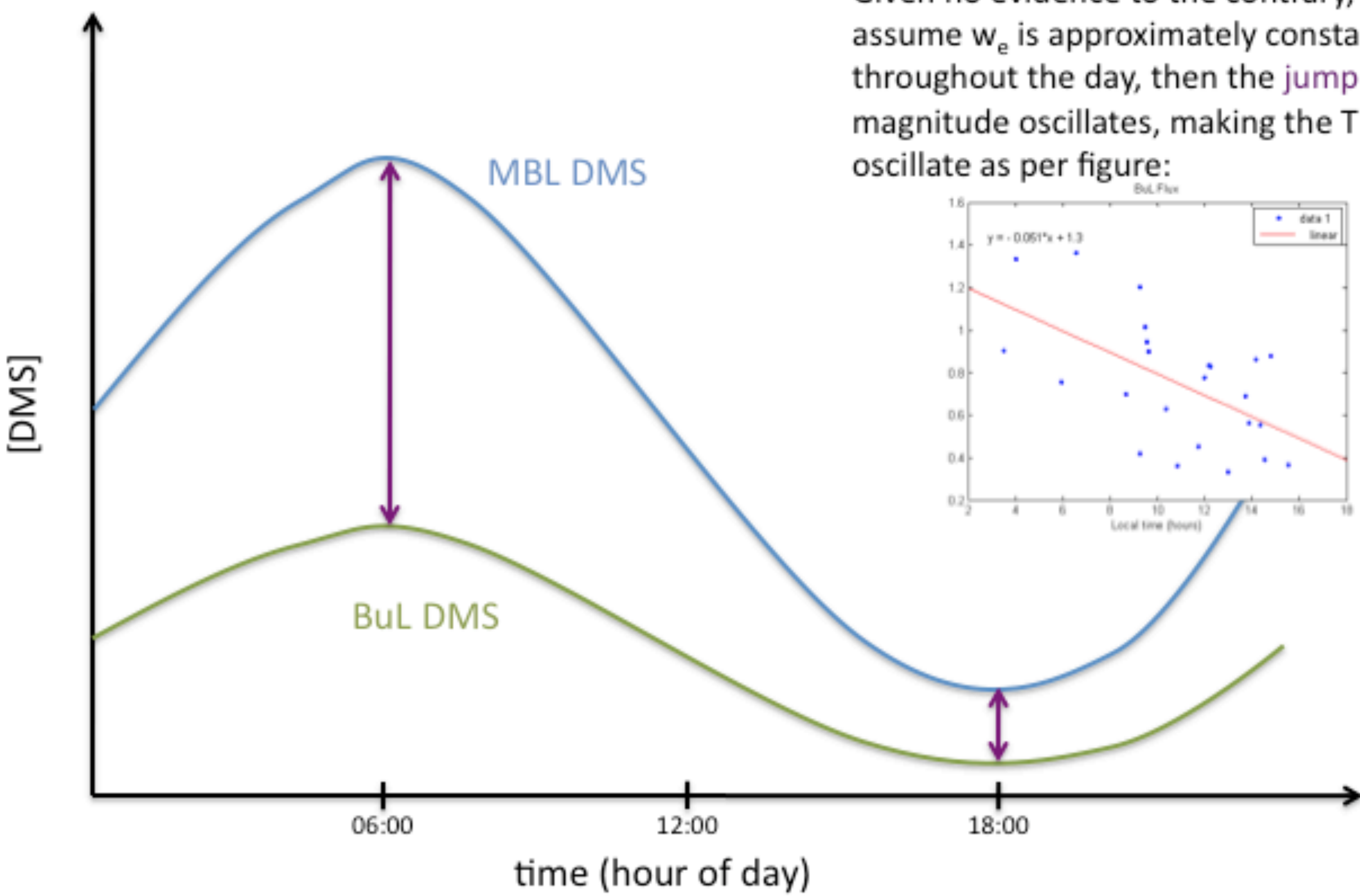
- Using the average daytime loss rate of ~6 ppt/hour, an average day of photochemistry will consume ~50 ppt of DMS.

- Assuming vertical flux divergence (~1.4 ppt/hour) is the only source of DMS, and that the buildup occurs over the 12 hours of darkness, we can only account for ~20 ppt during the entire night.

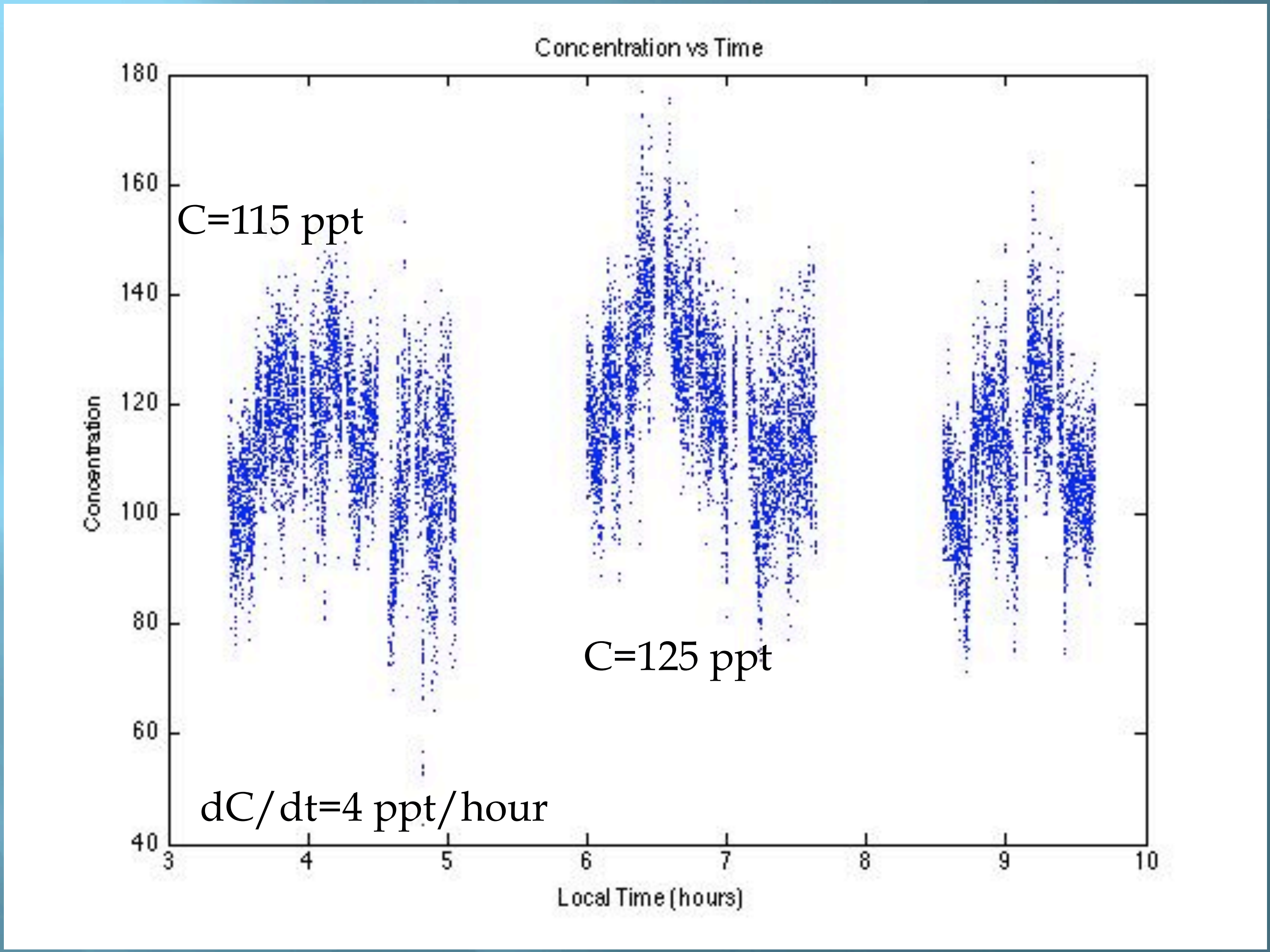
- Assuming that the system is in or near steady state, we are unable to account for more than half of the nighttime buildup of DMS.

GRAPHIC OF DIURNAL OSCILLATION OF THE DMS ENTRAINMENT FLUX

Given no evidence to the contrary, we assume w_e is approximately constant throughout the day, then the jump magnitude oscillates, making the TBL flux oscillate as per figure:



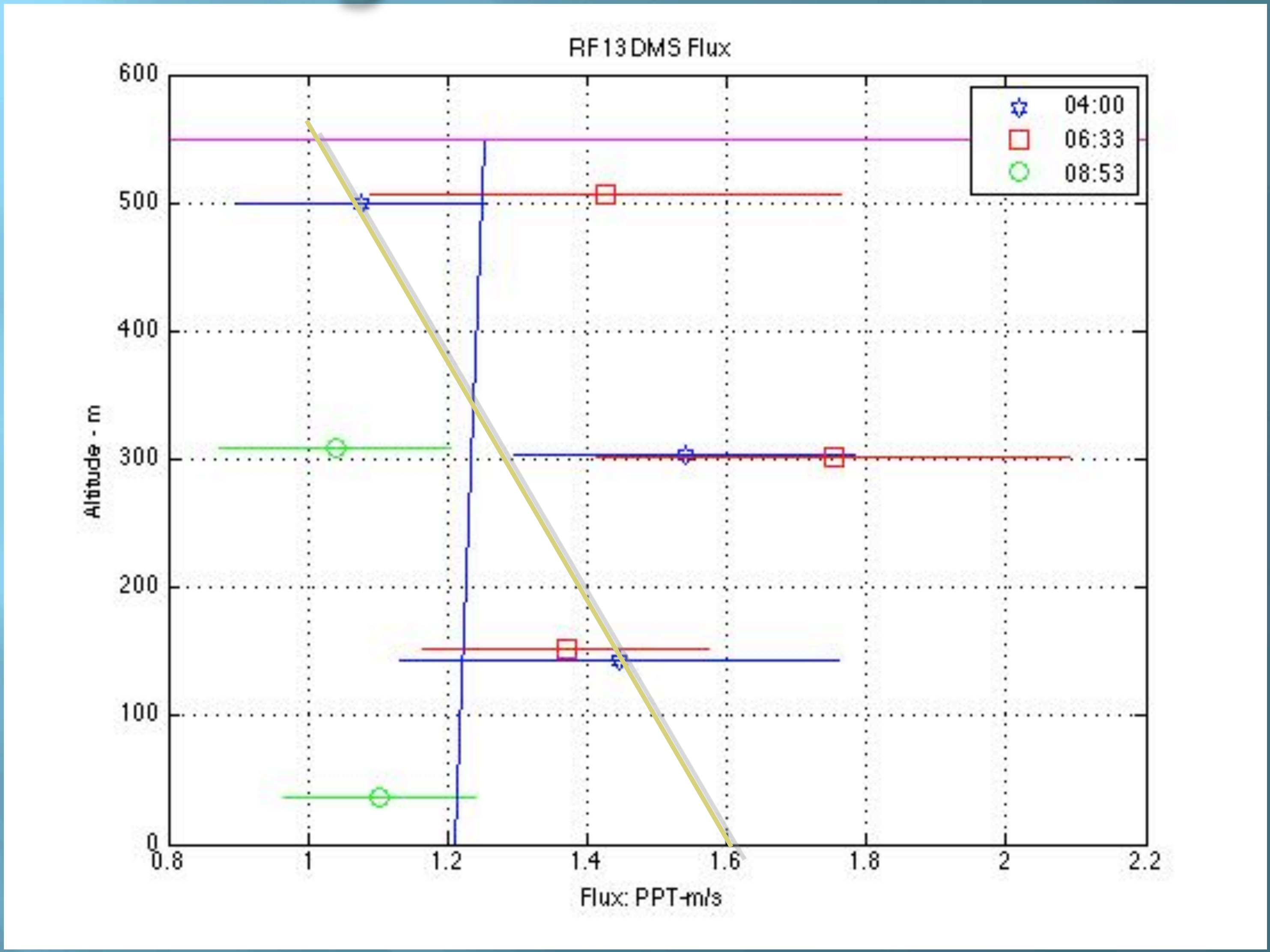
Nighttime Return



Nighttime Return

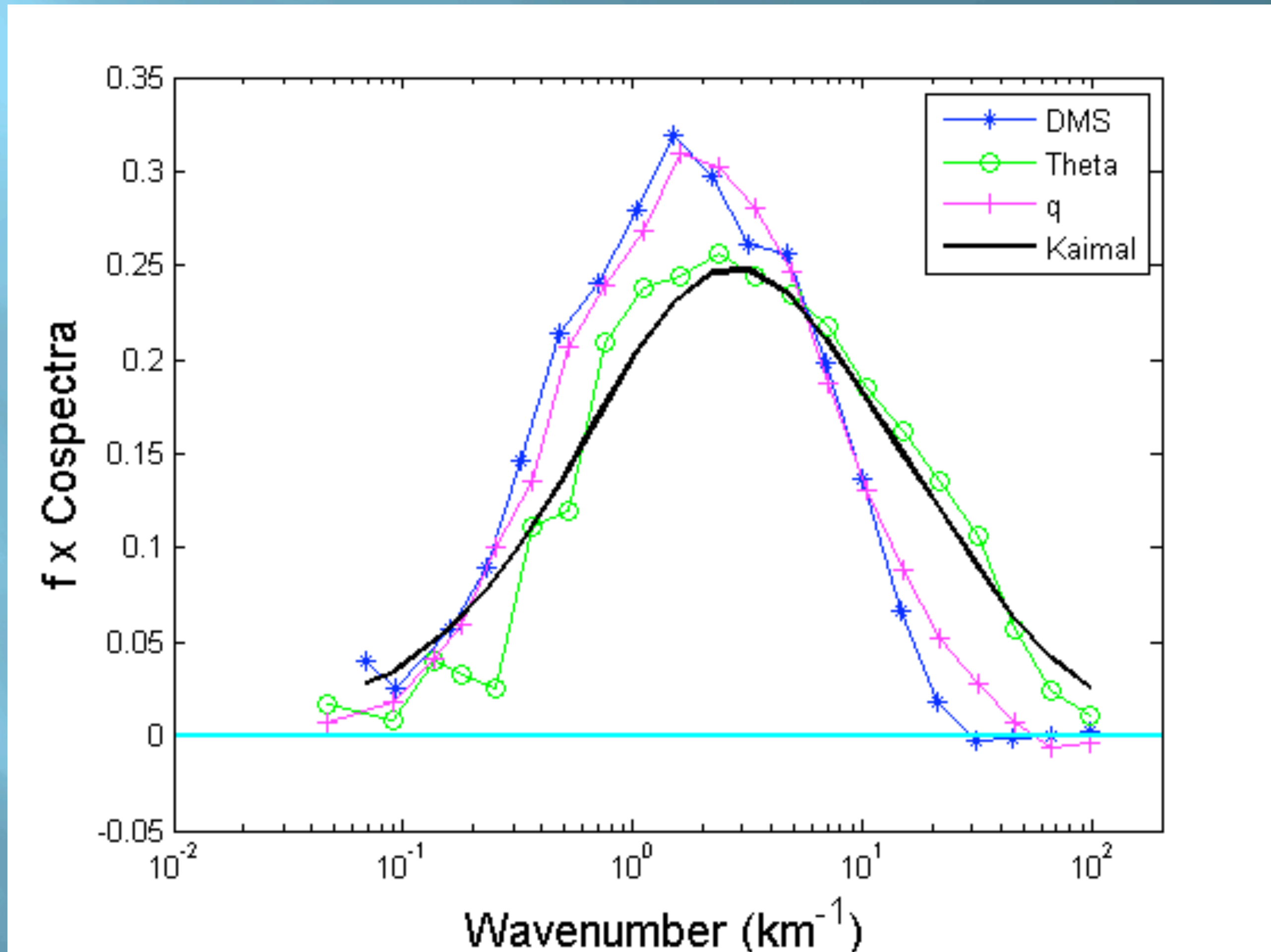
- From the 1994 Christmas Island data, the daily maximum occurs around 4:30AM.
- During PASE night flights, our first MBL leg occurred between 3AM and 4AM – right at the end of the increase.
- RF06 shows no significant nocturnal increase (stack #2 is 2 ppt higher than stack #1)
- RF13 does show a significant increase between stacks 1 and 2, nearly 10 ppt (4 ppt/hour)

Nighttime Return



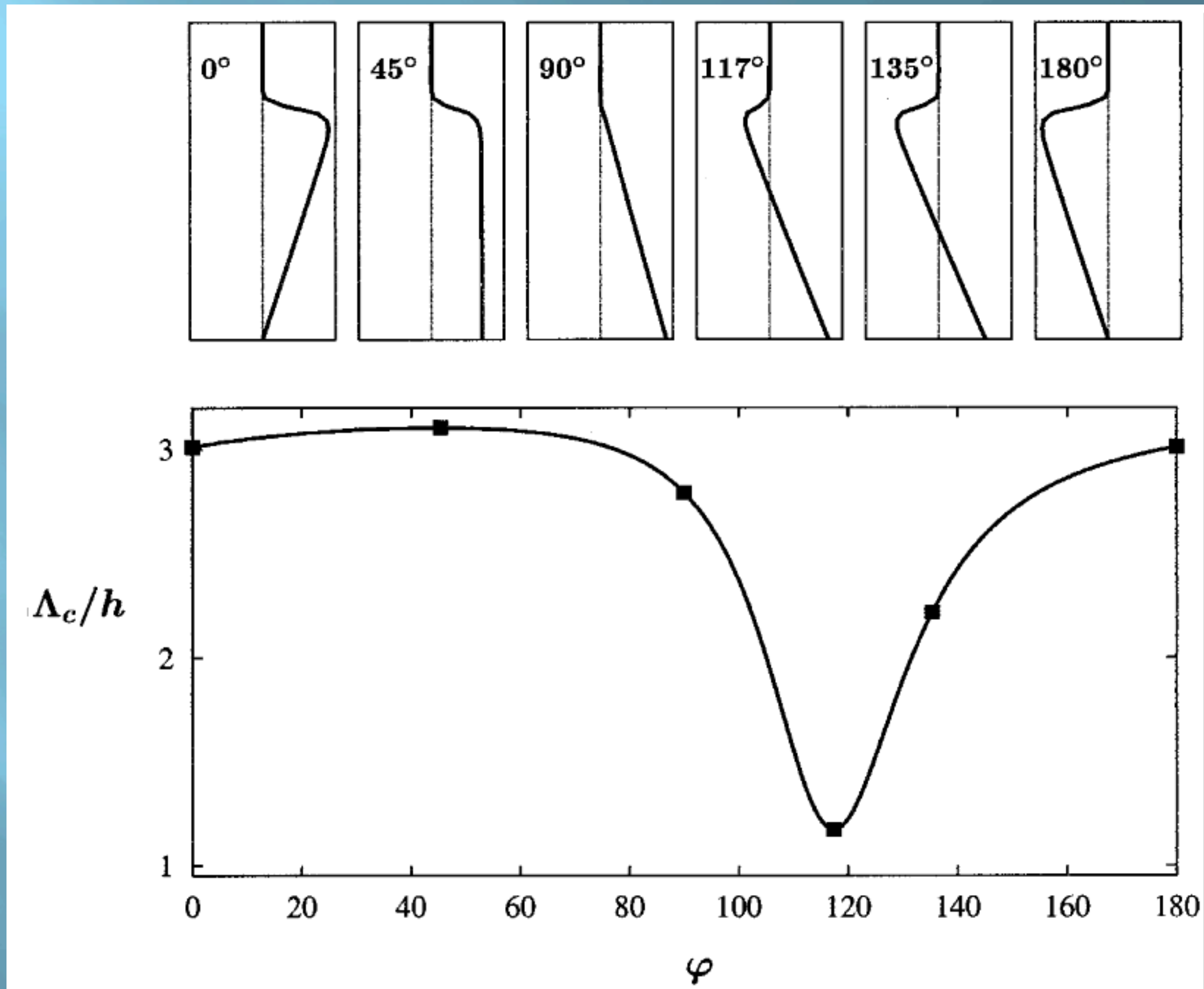
Stack #1 Flux Divergence = 0.6 ppt-m/s ÷ 550m × 3600 s/hr= 4 ppt/hr

Kaimal Cospectra



- Θ behaves much like the Kaimal curve
- DMS & water vapor are shifted toward larger scales

Jonker et al. 1999

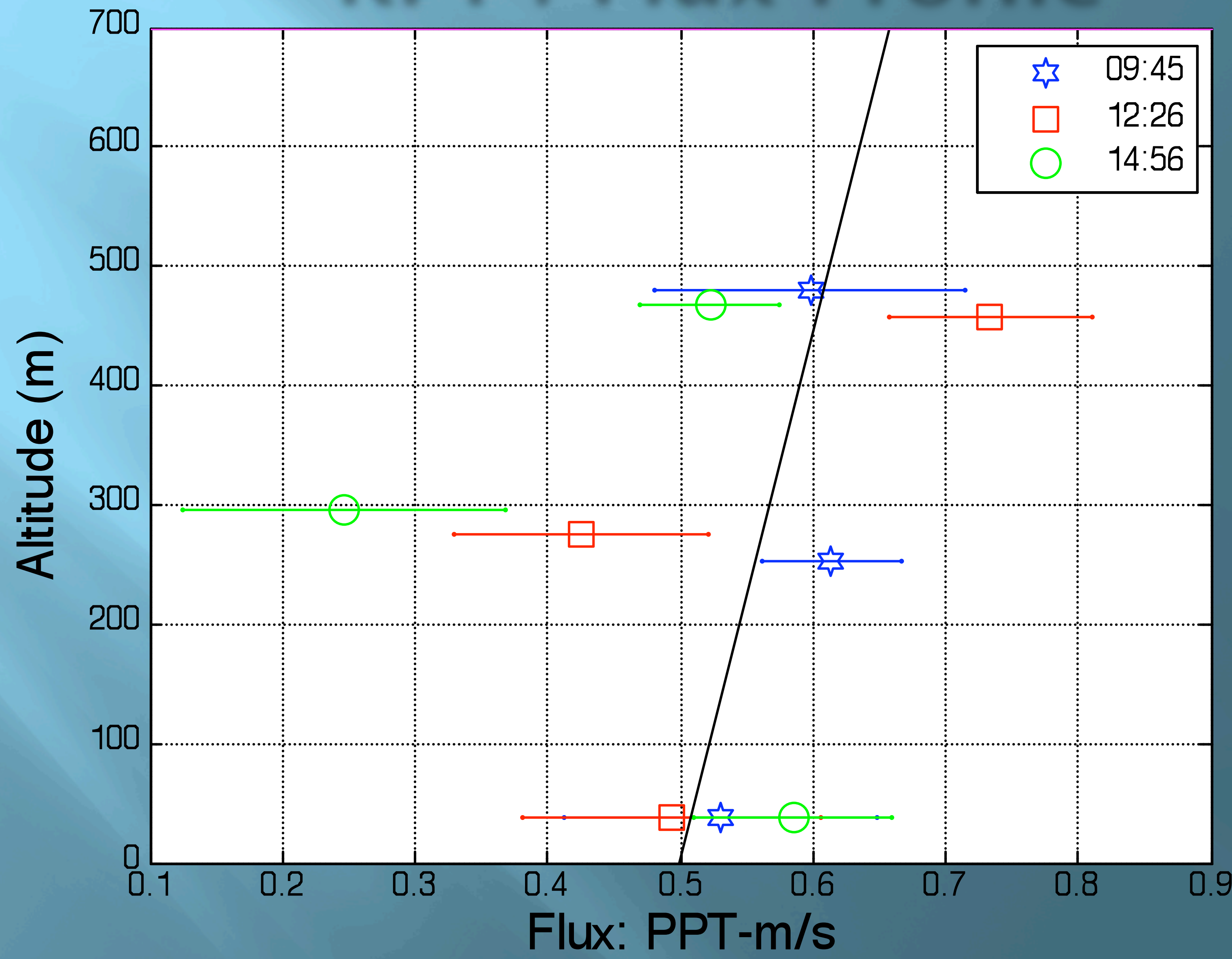


- LES simulation shows similar results, for θ (φ~135) the peak occurs at smaller scales than it does for DMS (φ~60).

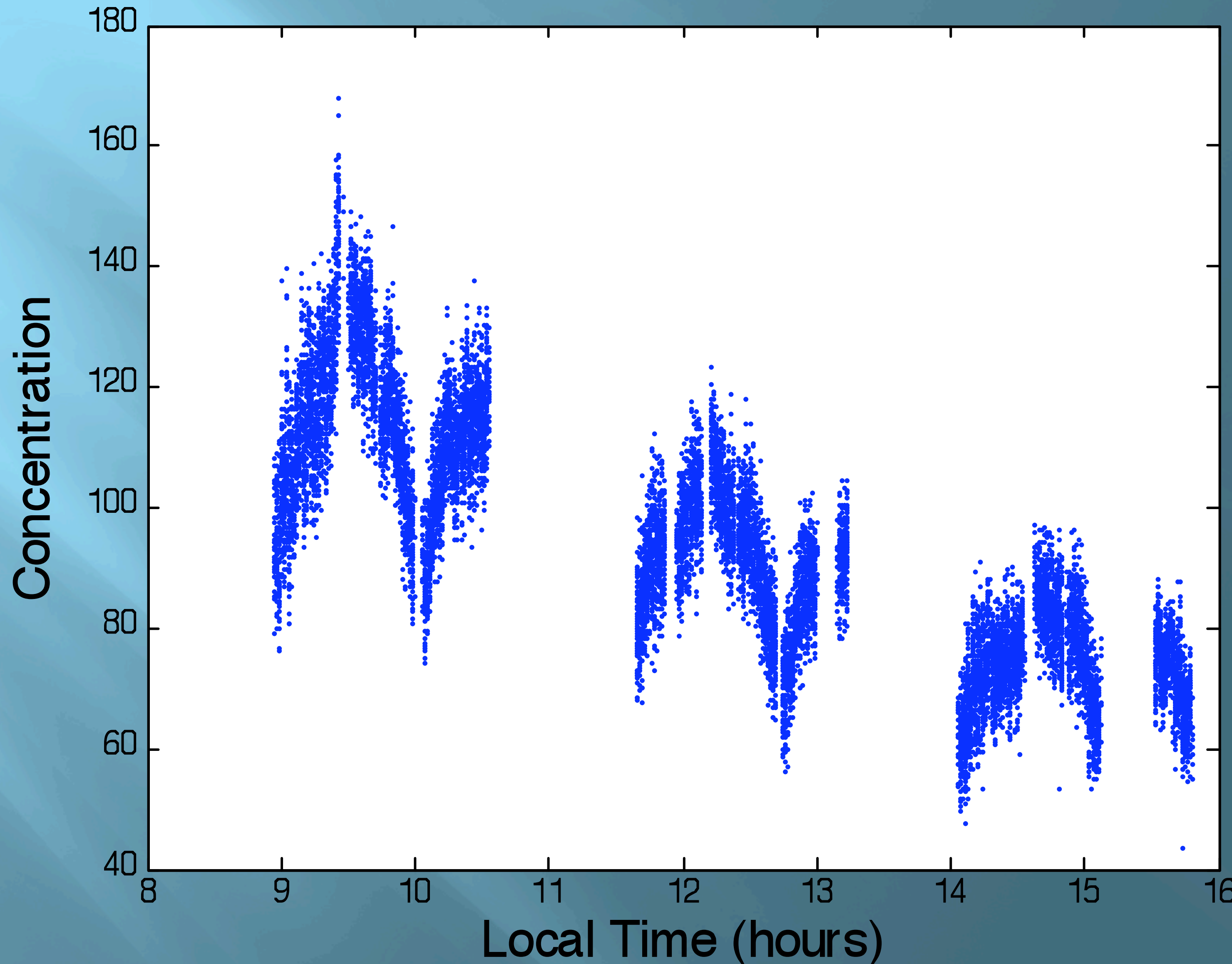
The Mystery Flight!

Observed Decrease	-7.7 ppt/hr
Flux Divergence	-0.8 ppt/hr
Advection	-1.2 ppt/hr
Chemical Loss	-5.7 ppt/hr
Predicted OH Loss	-10.7 ppt/hr

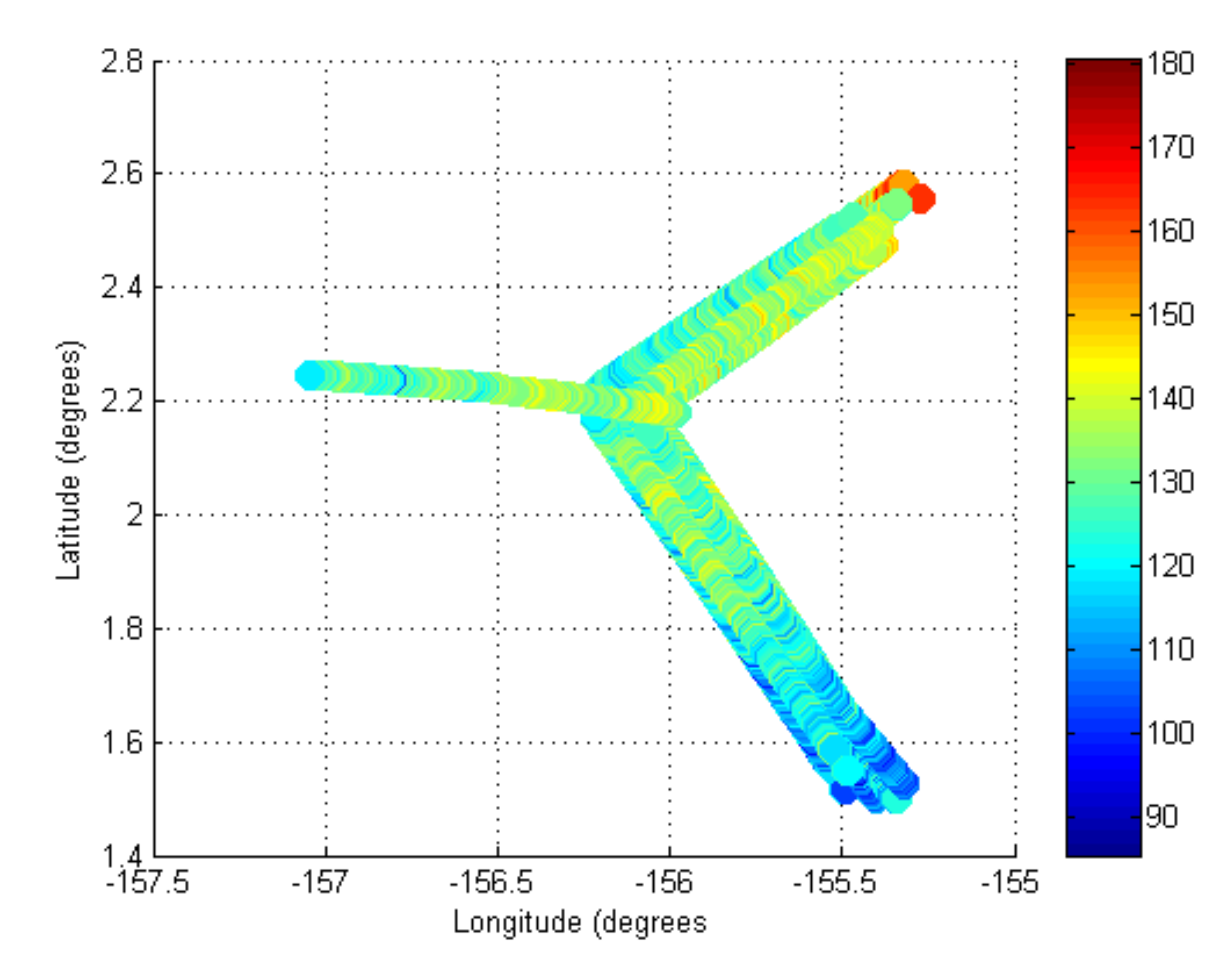
RF14 Flux Profile



RF14 DMS Time Series



RF14 Horizontal Gradient



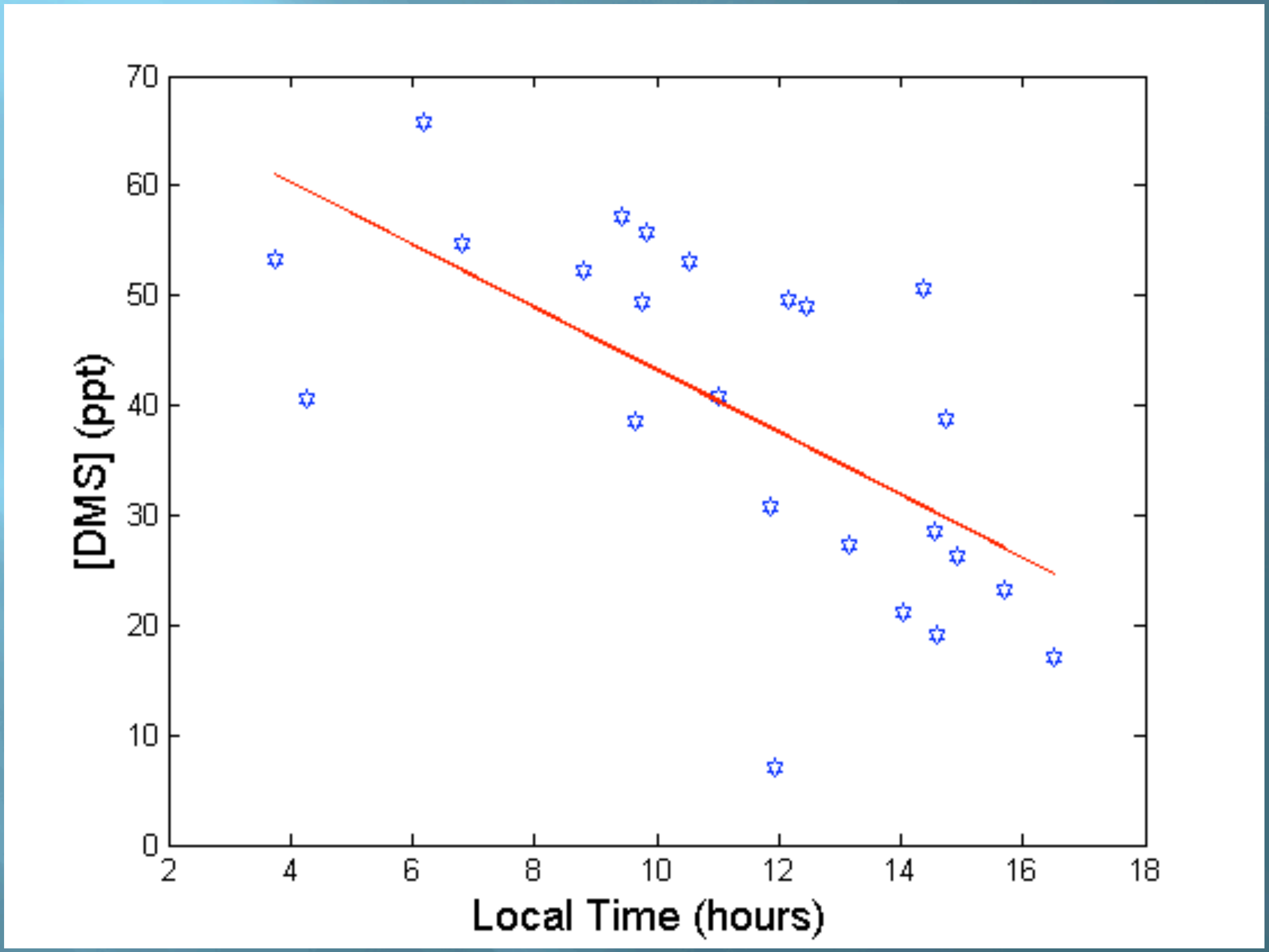
RF14 Assumptions

- Flux profile not linear
- Air mass far from steady state
- Recently travelled over very productive water
- Horizontal gradient difficult to measure
- MBL height varied throughout the flight

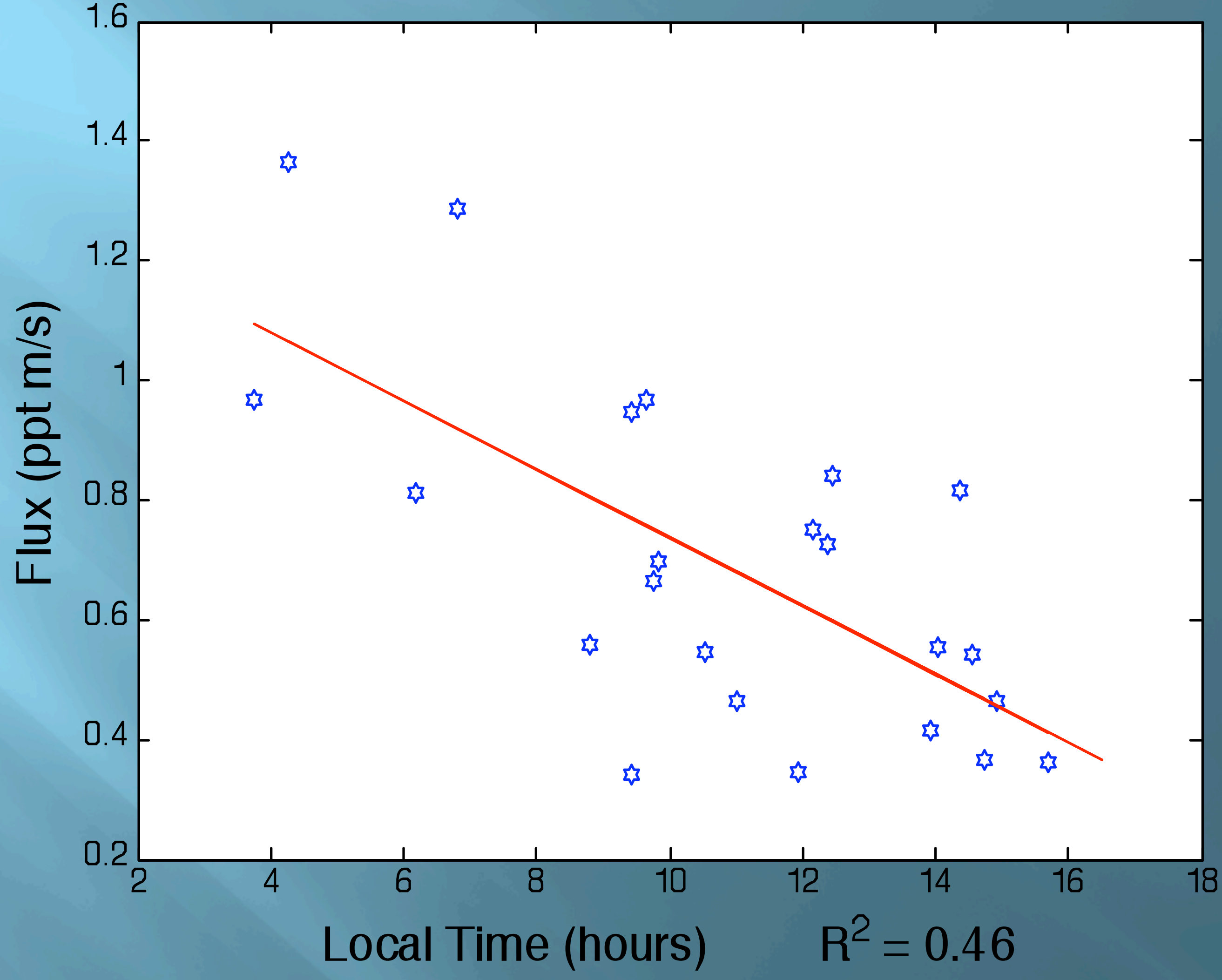
Flux Profile Evolution

- Assuming $\overline{w'c'}_{z_i} = v_e \cdot ([c_{BuL} - c_{MBL}])$
- Throughout the day, DMS in both layers is reduced by the same fraction.
- The magnitude of the difference decreases throughout the day.
- We expect the BuL flux to increase throughout the day.

BuL - MBL



DMS Flux into BuL



DMS flux into BuL decreases throughout the day

Conclusion

- With the exception of RF14, the budget closure was excellent! More night flights!!
- The budget closes without the need to invoke halogen chemistry.
- During the flight hours of PASE, ~80% of the DMS entering the MBL from the surface is passed to the BuL. Need more night flights!!