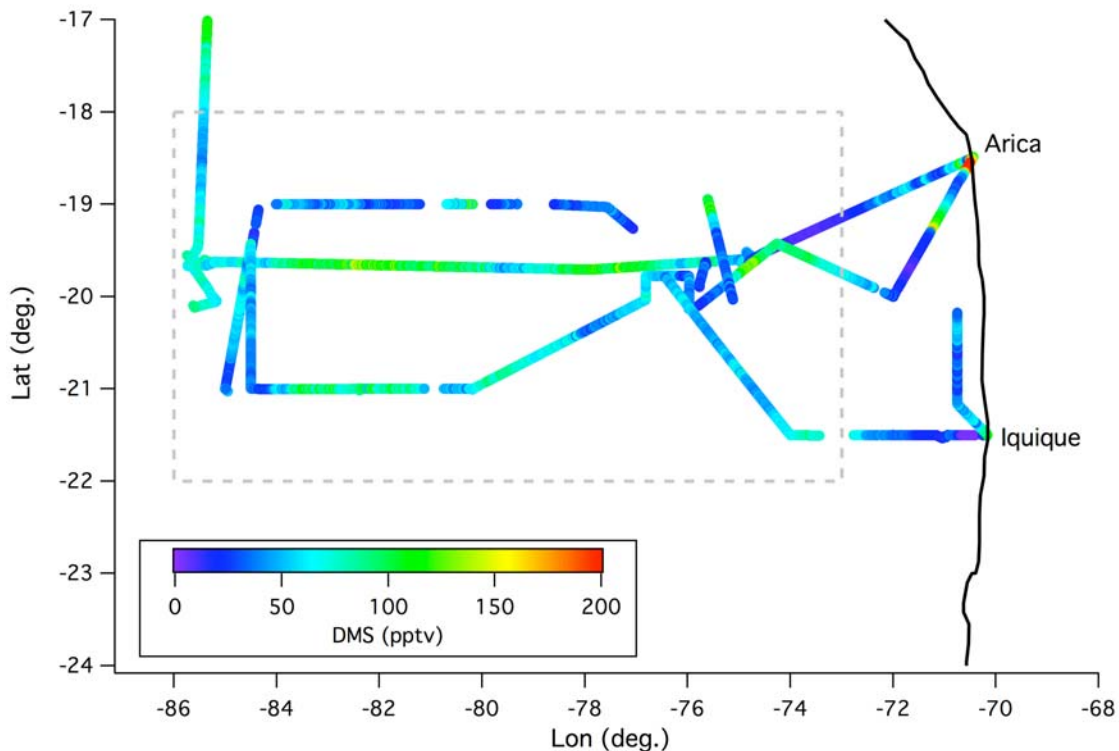


Much appreciation for the helpful *comments from I. Faloona*, here are our responses:

*The work involves heavy averaging in space and time to derive vertical profiles and steady-state, diel DMS behavior over an area of ~600,000 km<sup>2</sup>. The overbar in Equation (3) does not represent only a time-average, as the authors state, but a wide spatial average as well. The idea of such an average of the Eulerian derivative in Eq. (3) might be made more clear if a map of the "VOCALS region" and the locations of sampling were presented to the reader. For example, the authors decide to neglect mean advection in the budget because this wide area, long-term average DMS gradient is found to be perpendicular to the mean wind. But it's the gradient just upwind of the "VOCALS region" that might systematically present a contribution to the average DMS budget, and it is likely that such a strong, persistent gradient does indeed exist in the nearshore environment. A question like this that naturally arises in the reader's mind may be more tractable if a map of the region were made available with all the data collection points contained therein.*

We agree that including a map of the "VOCALS region" will be instructive. See below:



In this figure, the color-coding represents the atmospheric DMS concentration; the "VOCALS region" is defined within the dashed line. We can see that while there were a couple of isolated pockets at the coast where DMS was high (e.g. near Arica and Iquique), the corresponding areas are quite small and thus unlikely to cause a large horizontal gradient. Overall, instead of a clear trend, we observe patchiness in atmospheric DMS that is likely related to ocean eddies and mesoscale structures.

*On p. 16275 (l. 23-25) the authors state that "With an average inversion height of 1.3 km in the VOCALS region, this [DMS] vertical gradient increased significantly with boundary layer depth." I do not see any physical reason why the DMS vertical gradient should be directly proportional to MBL depth. In fact, taking the vertical derivative of the decoupling relationship that the authors use indicates that the very opposite is true: viz., the vertical gradient of concentration,  $C(z)$ , is inversely proportional to MBL depth,  $z_i$ .*

This is a problem of our poor wording initially. We meant to say that "With an average inversion height of 1.3 km in the VOCALS region, the difference between DMS concentration at the surface and that just below the inversion increased with boundary layer depth."

*One particular concern in reading this was the circumvention of the strong diurnal modulation of the inversion height unique to these stratocumulus topped MBLs. The reason is that the shortwave heating of cloud top acts to stabilize the MBL and dramatically reduces TKE to drive entrainment (and promotes 'decoupling'). On the other hand, longwave cooling at cloud top during the night feeds the energetics of entrainment.*

*That is why the DYCOMS-II results (Faloona et al, 2005) show we of about 1 mm/s for the daytime flight, and ~ 6 mm/s for the night flights.*

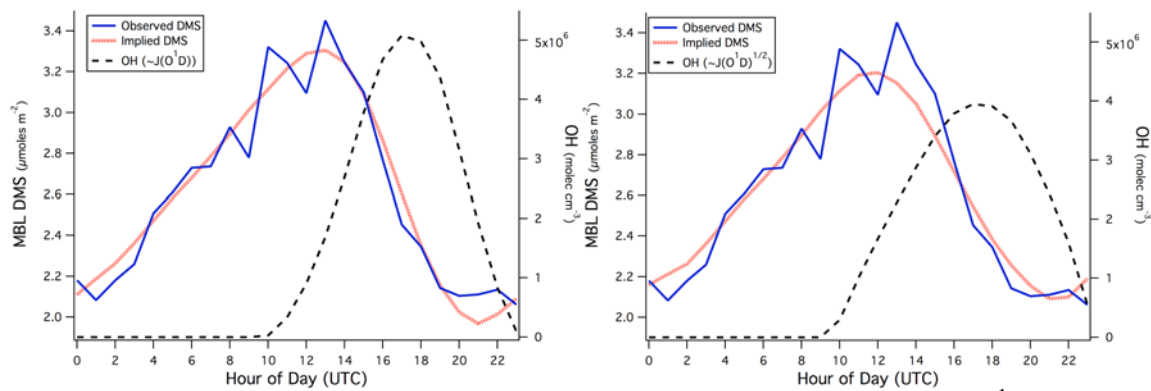
*This effect can be clearly seen in Figure (6): the MBL top descends under subsidence during the day (because the entrainment is much weaker) and rises again after sunset (because of the additional source of TKE from cloud-top radiative cooling.) A rough estimate of the MBL growth after sunset (Fig. 6) shows that it rises about 200 m in about 6-7 hours - approximately 8 mm/s. This MBL growth rate is the net, in excess of subsidence, which is most likely of the same order. Likewise, the depression during the day appears at a similar rate (~200m/8hr). Thus, when estimating a mean, long-term entrainment by looking at an average nighttime budget, the large variability needs to be considered. In other words, it is highly unlikely that you would obtain an entrainment estimate from data throughout the night that would match the average over the entire diurnal cycle.*

The diurnal variability in the entrainment velocity is definitely a cause of concern. However, as the reviewer pointed out, in this stratocumulus environment entrainment tends to be the stronger at night than during the day. Therefore the nighttime entrainment velocity we were able to estimate from the DMS diurnal cycle is probably higher than day during the day, which would render the daytime entrainment flux even less important relative to the photochemical destruction term.

*As for the comparison of the authors' OH estimate to the expected abundance, the use of OH observations from rural northeast Germany (Holland et al., 1998; Hofzumahaus et al., 1998) seems distinctly misguided (p. 16280). The referenced OH studies took place in mildly polluted continental regions where NO<sub>x</sub> levels were most likely much higher than in the MBL of the Humboldt Current upwelling system. And if the NO<sub>x</sub> levels were low*

(<25 pptv) during VOCALS as expected, then the steady state OH should be more proportional to the square root of the O<sub>3</sub> photolysis frequency, rather than linearly dependent as the authors claim. The reason it was linear in the Holland et al. (1998) study was because of the moderately polluted NO<sub>x</sub> levels. Intermediate relationships between J\_O<sub>3</sub> and OH are discussed in Berresheim et al. (2003). But this issue really begs the nagging question of how much NO<sub>x</sub> was in the VOCALS region during these measurements. There is, in fact, no effort made to establish the photochemical setting of this experiment whatsoever, thus the ‘accuracy’ of the technique in estimating OH remains by and large unsubstantiated. Are there no CO, O<sub>3</sub>, or NO<sub>x</sub> observations from this experiment that could be used to slightly constrain the photochemical conditions? The works of Tan et al. (2001) and Olson et al. (2001) show that the sources and sinks of OH in the region are fairly simple.

We agree with the reviewer that comparisons to a polluted atmosphere in Europe might not be appropriate. CO and O<sub>3</sub> were measured aboard the C130 during VOCALS-REX and those data should be available soon. As to whether OH should follow a linear or square root relationship with O<sub>3</sub> photolysis frequency (J(O<sup>1</sup>D)), both scenarios are tried and shown below.



The left panel has OH linearly proportional to shortwave flux (and hence J(O<sup>1</sup>D)); the right panel has OH proportional to (J(O<sup>1</sup>D))<sup>1/2</sup>. Note that the diurnal average OH concentration is unchanged. It appears that with a square root dependence, implied DMS does not decrease as rapidly as observed upon sunrise because the OH distribution is too broad. While the actual exponent for J(O<sup>1</sup>D)) might not be unity, judging from the DMS diurnal cycle alone, the power is closer to one than to 0.5.

Of course this result does not say anything about halogen radicals whose concentrations may also be proportional to J(O<sup>1</sup>D)). A quick calculation shows that a diurnal average concentration of just over 1 pptv of BrO alone will be enough to cause the same amount of DMS oxidation (or about 4 pptv if BrO also scales with shortwave).

Furthermore, the widespread climatologies of OH published by Davis et al. (2001) are not very representative of the stratocumulus topped MBLs which, judging from their high albedos, most likely reduce the surface actinic fluxes substantially (e.g., Mauldin et al., 2001; Lefer et al., 2003). The airborne data from all the NASA GTE programs are archived and available and would most likely yield specific HO<sub>x</sub> observations of much greater geographical and/or seasonal relevance to the VOCALS study.

Thanks for the recommendation. The actinix flux during VOCALS was approximately 20% lower than in the tropical Pacific, judging from observations made during the Pacific Atmospheric Sulfur Experiment (PASE). We will have a look at the data you listed above.

*Finally, the authors need to reconsider the meaning of 'zonal averages' used throughout. Typically I have found that a zonal average applies to variables that are averaged along lines of constant latitude and thus lose their dependence on longitude (e.g., Fig. 5).*

We will use 'average along the 20S line' instead of 'zonal average' to avoid this confusion. Thank you for pointing it out.