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## *Interactive comment on* "Atmospheric sulfur cycling in the Southeastern Pacific – longitudinal distribution, vertical profile, and diel variability observed during VOCALS-REx" *by* M. Yang et al.

## Anonymous Referee #1

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## General comments:

"Atmospheric sulfur cycling in the Southeastern Pacific – longitudinal distribution, vertical profile, and diel variability observed during VOCALS-REx" presents a comprehensive observation-based analysis of offshore atmospheric sulfur budgets and their diel cycles in the Southeast Pacific. The paper represents a critical step in the integration and synthesis of REx observations to quantify the sources of offshore oxidized sulfur and the sulfate aerosols that influence stratocumulus over the region. Exceptional detail is provided in the calculation of chemical transformation, deposition, and aerosol thermodynamic rate constants and their uncertainties, which is essential for support-

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ing the very important conclusion that biotic sources of DMS dominate the offshore sulfur cycle. They also make this article a very important resource for the evaluation of chemical transport and aerosol-cloud modeling in the region and in other marine environments. The article provides an updated template for future chemical constituent budgeting from multi-platform in-situ observations, and therefore represents a method-ological contribution beyond its rich dataset and important findings.

The paper is clearly written and complete, and minor revisions are recommended. One area requiring additional analysis is that only diel average values and uncertainties are generally presented, due to incomplete episodic data. It would be helpful if the authors could directly compare the variability and uncertainties in the SO2 and SO4 budgets for those times and sources/sinks where daily or hourly terms can be estimated. Even if incomplete, this would help put the uncertainties in perspective, and would help address how synoptic variability affects the relative importance of the sources and sinks presented.

## Specific comments:

page 2882, line 10: "Because the aircraft usually took off from Arica in the early morning, reached 80 âLij 85° W at around sunrise, and returned to shore in the afternoon, spatial and temporal biases are inherent." It may be helpful to discuss how these biases affect Figure 3 and other results.

page 2885, line 25: if the O3/SO2 increase after sunset is due to boundary layer recouping, what is the most likely source of the higher concentrations in the descending air? a) between LCL and top of the MBL b) cloud level c) free troposphere, entrained into MBL d) all of the above, to varying degrees

This will necessitate a more sophisticated calculation than Lilly (1968), but the available C-130 vertical profiles should support it. If quantified through observations, this would be an important finding in itself, and may have implications for other findings. How would you tell which layer was the source for SO2? For O3? Are they the same? Look

at relative rates, and use the time series of observations to address the variability. Minor comments:

page 2884, line 25: "The optical rain [gauge] detected" page 2885, line 19: SI units: 11 knots = 5.66 m/s

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