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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.***

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This manuscript reports atmospheric observations of various components of the sulfur cycle, its link to the dominant natural source (described in a previous paper by Yang et al 2009), and a modeling exercise to explain or account for the diel variability at 20S during the VOCALS Rex program in the SE Pacific Ocean. The scarcity of previous measurements, air and water, especially air-sea fluxes, in this region alone makes this a valuable piece. The fact that it does not agree with previous diel cycling in a tropical region makes it even more interesting. Sources and sinks for SO<sub>2</sub> and SO<sub>4</sub> are discussed, quantified or estimated in a step-by-step manner that clearly guides the

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reader through the process; I really valued that, regardless of whether one agrees or disagrees. One of the VOCALS Rex hypotheses represented the considerable controversy and disagreement on whether natural vs anthropogenic sources dominate the atmospheric sulfur cycle in this region, with proponents of both sides participating in the campaigns. And, remarkably, in this manuscript. Nice job.

A general comment: Please make sure that every abbreviation is spelled out the first time it is used throughout the text and including the figure legends. Also, please check that all refs are there - the very first one I looked for was not on the list-; I did not cross check the others. Clearly, some sections were moved around and such details not re-checked, suggesting some sloppiness.

I recommend publication, once comments are attended to.

Specific comments:

P2875L12-15 indicate that this is for the offshore region; L15-16, inshore and/or offshore?; L25 update the K&A'99 estimate with Lana et al. GLOBAL BIOGEOCHEMICAL CYCLES, VOL. 25, GB1004, doi:10.1029/2010GB003850, 2011, of 28.1 (17.6–34.4) TgS/yr; does not change the point.

P2876L5 spell out MBL; L28 do you mean that all CCN, as a size fraction, represent a large fraction of the TOTAL aerosol number?

P2877L8 nss SO4 in both cases?; L15 spell out FT; L25 from or towards the SE?

P2878L10 NoveMber; L11 add latitude of Arica;

P2879L16 AMS sulfate concentration? data? missing word; L18 end of sentence, please inser (data not shown)

P2881L8 Benedict et al 2011 missing from bibliography; any others? L18 replace 'to' with 'into' ... 2 regions

P2882L4 showS; L6 SO4 concentration? in fig 2, where is the marker size scale?; L8,

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is RF14 later in the season? any flight characteristic that makes this RF14 different in any way? Just because there was more pollution doesn't make it any less real. There is no later discussion of such events; L13 insert 'on' before 'aircraft'; L18-22 this section on seawater DMS needs references (Yang et al 2009? Hind et al 2011? Other?) and it would benefit from a sentence that indicates this is a brief summary of seawater concentration and fluxes ... OR add references for each of the next statements in this paragraph; L26 fluxES

P2883L5 SO<sub>2</sub> and SO<sub>4</sub> concentrations; L25 what proportion of the sampling time, hence data, were cloud free or POC?; L27 'new particle nucleation' from what? DMS? what is/are possible source(s) for the 30nm particles seen above the cloud layer? Is the difference bw 64ppbv and 74ppbv that huge for only one to be high and related to long distance transport?

P2885L27 very interesting; L29 evokes or invokes?

P2886L2 would be, rather than was- this is a speculation, as indicated later; L21, L23 spell out LHS and RHS, respectively;

P2889L1-3 were the resulting SO<sub>2</sub> concentrations higher because of higher oxidation rates or higher DMS concentration or flux during the day time, ie, faster or more substrate? Is the diel DMS cycle shown in Yang et al 2009; certainly not in this manuscript? From later in the text, it appears that a diel cycle of OH is seen with mid-day max and a range of gamma values. Did observed fluxes (independent of wind speeds??) or atmospheric concentrations show a diel cycle?

P2890L9 if MSA WERE (not was); L11 WERE formed from DMS; L12 please replace 'implies' with ' would imply'; L17 using the same range for gamma?

P2891L12 R? is this the same R as defined later? a different one? make sure that all symbols are defined the first time they are used in the text;

P2894L2 please indicate explicitly why it would be 'unrealistic'; L should not exceed

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P, given that advection is negligible (?) as an alternate source OR unless there is another unaccounted source (?). Even to summarize the arguments made earlier in the discussion; L11-12 whose data? First time these MODIS data are mentioned; L22 what is meant by “qualitatively” in this sentence?;

P2897L29 and is the magnitude of this factor (0.43) comparable to something? too high? too low? what would have happened if a closed system had been chosen? even if unrealistic?

P2898L21 representS

P2902L9 ten?

P2904L18 allowed us TO estimate

Table 1: format last section like others underlining subtitles

Fig 3: pollution, as indicated by xx and yy, or see text for details...

Fig 6: is the difference between day and night for inversion height statistically significant?

Fig 13 how does the implied DMS compare to the measured DMS? Or was it prescribed? Then say so. Same as indicated above.

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