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Atmospheric Chemistry and Physics Discussions

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 #3

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This paper presents a detailed budget of sulfur dioxide and sulfate in the marine boundary of the south east pacific Ocean. The analysis is carefully carried out and presented in a clear way that can easily be followed by the reader. This is to be commended and serves as a blueprint for how budgeting of this type may be performed in future studies. The results indicate that a simple sulfate budget in the MBL cannot reproduce the diel cycle of sulfate observed in the region during VOCALS. The authors show that this is due to decoupling of the upper, cloud-filled BL from the surface layer during the day, whilst at night closer coupling occurs. Cloud processing of SO2 provides a large fraction of the MBL sulfate production and hence the diurnal sulfate cycle cannot be captured without considering boundary layer dynamics and decoupling.

This is a careful and well considered piece of work and deserves to be published in ACP. I have a few minor comments that the authors should consider in addition to those of the other referees.

Specific minor comments

Page 2875 line 3: sulfate should be written as 2-.

Page 2875 lines 10-11: by what longitude offshore?

Page 2880 line 17: near unit “collection” efficiency

Sections 2 and 3: It seems odd that neither of the 20S overview papers are referred to in either of these sections. Whilst the overview of BL thermodynamics and cloud along 20S is referenced (Bretherton et al 2010) this is specifically in support of the fraction of precip reaching the surface. Neither this paper, nor the chemistry overview of Allen et al is cited in the introductory remarks. Given that the work presented in the paper is directly related to the result in these papers it seems a shame not to discuss the linkages. Notably the longitudinal regions “near shore”, and “remote”. Allen et al in particular provides a back trajectory analysis that also supports the pollution contributions as a function of longitude that should be referenced.

Page 2884 line 20: Similarly Allen et al shows aerosol size distributions from the MBL and FT in the nearshore and remote regions. These should be discussed.

Page 2884 line 24: 150 gm-3?

Page 2884 lines 25-26 “The optical rain detected sporadic precipitation events” This sentence doesn’t seem to make sense

Page 2888 lines 9-13: This assumes that the MBL grows in height commensurate with entrainment. If other dynamical processes occur to maintain boundary layer top at a more constant altitude then a better estimate of the net entrainment of sulphate is the former case involving the difference between the concentrations above and below the

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boundary layer. The authors are right to say that the mass flux into the MBL from above the BL top does not depend on the concentration in the MBL, only on the concentration aloft. However, if BL height is constant the mass of air in the BL can only be maintained with an opposite mass flux to the lower FT and this should be considered. The authors should be clear about whether the BL is growing in response to entrainment or not before favouring one formulation or the other.

Page 2889 line 12: “all of which would be converted to $SO2- 4$ aerosols” all seems a little strong. If production of SO2 from DMS is calculated to be approximately 10 times the entrainment flux (see section below) then around 10% of the concentration in the MBL arises from entrainment, Whilst it is a minor component it isn’t zero.

Page 2891: right bracket missing in equation 8

Page 2893 lines 16-18: It would be instructive if the authors could put uncertainty estimates on the size of the effect of not considering mass transfer limitations.

Page 2898 line 5: Were DMS measurements not made? If they were why cant the data be also shown in fig 13 for comparison?

Page 2898 line 21: “The last term represent” represents

Page 2900 line 11” 40,pptv” remove comma

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