Dear Editor:

Enclosed please find the manuscript "Dancing on Water: The Choreography of Sulfur Dioxide Adsorption to Aqueous Surfaces" by Eric S. Shamay, Kevin E. Johnson, and Geraldine L. Richmond, submitted for consideration for publication in *The Journal of Physical Chemistry B.*

The scientific community's understanding of the behavior of simple, inorganic gases at an aqueous interface is currently sparse as such systems have remained largely unexplored. Great advances have been made in studying gases at air-water interfaces over the last few years, however much can still be learned in the study of the binding geometries and specific interactions that occur with the water surface. Computational analysis of molecular dynamics simulations is well suited for the study of narrow interfacial regions due to its ability to address microscopic environments and to probe the origins of specific phenomena that are otherwise impossible through current experimental techniques.

In this study we have shown that sulfur dioxide gas molecules affect the structure and hydrogen bonding of water at the air-liquid interface through reorienting and binding with the water surface. We augment a recent experimental sum frequency spectroscopic study by our group and strengthen the conclusions of that work while also building a more complete microscopic picture of the interfacial region. The alteration of water in comparison to the interfacial environment without sulfur dioxide gas is quite drastic. We find that increasing concentration of adsorbing gas molecules affects the interfacial aqueous layer differently than at lower concentrations, and that adsorbing gas molecules undergo a reorientation relative to the water surface during transit towards and into the interface. Our work will be of interest to the readership of *The Journal of Physical Chemistry B* because it contributes significantly to our understanding of how the subtle interplay between water and an adjacent gas uniquely alters the interfacial region and molecular geometries and orientations within the air-liquid interface, a subject that has implications in many important processes such as gas adsorption and transport into aqueous systems.

I am the corresponding author and may be contacted at the above address, by e-mail (richmond@uoregon.edu), telephone (541-346-4635), or fax (541-346-5859). All authors have seen and approved the submission of this manuscript, and we look forward to the results of the review.

Thank you for your consideration.

Sincerely, Geraldine Richmond

Richard M. and Patricia H. Noyes Professor