

## Abstract

Sulfur dioxide is an important air pollutant that is emitted into the air from anthropogenic and natural sources. High SO<sub>2</sub> mixing ratios are known for their serious health implications, as the gas is oxidized to sulfuric acid within the human lung. Even at much lower mixing ratios SO<sub>2</sub> can affect forest ecosystems negatively. The gas is efficiently accumulated on leaves (dry deposition) and acid is formed in situ in the presence of water, which results in acid rain. Due to these environmental consequences, SO<sub>2</sub> emissions are increasingly regulated and have decreased since the 1990s in most countries around the world. It is important to study and understand the global sulfur cycle primarily to the large impact of sulfate aerosol formed on the global aerosol radiative forcing  $-0.41 \pm 0.2$  W/m<sup>2</sup>, which currently offsets between 10-36% of the CO<sub>2</sub> induced warming. The 50% uncertainty in this global forcing estimate of anthropogenic SO<sub>2</sub> emissions, is largely due to uncertainties associated with the treatment of the SO<sub>2</sub> oxidation to sulfate in global climate models. In the study I will discuss important empirical observations pertaining to the aqueous phase oxidation of SO<sub>2</sub> in wintertime fog in the NW-IGP. The study looks at visibility reduction due to presence of fog and classifies days with a visibility below 2 km throughout as foggy. For those foggy days, I investigate the changes of SO<sub>2</sub> mixing ratios that coincide with simultaneous changes in the aerosol liquid water content to quantify the effective reactive uptake of SO<sub>2</sub> (g) and NH<sub>3</sub> (g) into the aerosol liquid phase. I have found that at our site SO<sub>2</sub> is fully neutralized and has been converted to (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>. Therefore I have predicted that the fog water pH should be close to neutral which well correlates with the experimental pH measured during winter 2015-16. Out of 12 global models, 4 models fix pH in the Northern Hemisphere and the others treat it with an equation valid from pH 3.5-5.5. Five models participate in IPCC-AR5 but no model is capable of treating aqueous phase chemistry of SO<sub>2</sub> when pH of the system goes beyond 5. As India and China contribute to 40% of the total global SO<sub>2</sub> budget, it becomes more important to include all sinks of SO<sub>2</sub> to estimate correct budget. These are densely populated areas with large portion of land use being agricultural thus having high ammonia emissions from this sector which affects the lifetime and forcing of sulfate. Thus amount of aqueous phase sulfate oxidation is underestimated which needs to be taken care of, especially when the systems are neutral as it would help to improve the global budget of SO<sub>2</sub> and total radiative forcing due to sulfate.