

Response to Creamean's comments:

Wu et al. presented an interesting observation of two different NPF events at a midmountain site in Germany. I find these observations strikingly similar to a study we conducted at a midmountain site in the Sierra Nevada Mountains in California presented by Creamean et al.<sup>1</sup> We observed two different periods of NPF events during the 2-week winter study; with contribution of amines and sulfate to the growth of the newly-formed particles. Further, our study presented direct measurements of the newly-formed ammonium sulfate particles as being CCN-active, thus highlighting the broader impact on cloud formation in mountain regions. Wu et al. speculated the contribution of organics in addition to sulfuric acid to their observed NPF events. A discussion about the contribution of amines was briefly presented, citing studies conducted by Smith et al. (2010) and Barsanti et al. (2009), both excellent references for the case of amines. I wanted to make the authors aware of the similarities between their observations and those presented in Creamean et al. Both sites appeared to be fairly remote, midmountain locations. The growth rate (5.0 nm/h) was within the range of our calculated growth rates (2-8 nm/h) and similar to those calculated by Lunden et al.,<sup>2</sup> which were 1-7 nm/h at another fairly remote, mid-mountain site. Wu et al. also suggest ammonium may contribute to condensational growth via neutralization of sulfuric acid, although the concentrations of ammonium were quite low and only contributed to 18% of particle volume. We also suggest ammonium likely did not contribute greatly to particle growth due to a lack of a correlation between particles with sulfate and ammonium (see Figure 4 in Creamean et al.). We did however see a strong correlation of sulfate with amine species (Figure 4 in Creamean et al.), in addition to an increase in particles containing amine species throughout the course of the NPF events (Figure 3 in Creamean et al.). Our mass spectrometric technique (aerosol time-of-flight mass spectrometry or ATOFMS) enables us to examine the size and chemical composition of individual ambient particles, therefore, we observed amines and sulfate present within the same individual particles during the NPF events in the Sierra Nevada Mountains. Although the Wu et al. allude to the fact that organics could be responsible for particle growth, and that amines may contribute, more emphasis could be placed on amines due to the recent observations presented in Creamean et al. and the similarity between the sites. Amines are stronger bases than ammonium and have been shown to displace ammonium in sub 3-nm clusters by Bzdek et al.,<sup>3</sup> which might be a useful point for Wu et al. to address. Overall, the observations presented by Wu et al. were intriguing. However, the authors might benefit by comparing observations with Creamean et al. due to the similarities that existed between the two different studies.

### **Response:**

We greatly appreciate that J. Creamean read the manuscript and give this constructive suggestion. This suggestion was adopted in the revised manuscript.

### **Modifications in the MS:**

“On average, the volume fraction ( $\epsilon_{(\text{NH}_4)_2\text{SO}_4}$ ) contributed by  $\text{H}_2\text{SO}_4$  condensation is between 4.9~10.1% during the NPF event on 12 October. By comparing the soluble fraction ( $\epsilon_{\text{soluble}}=58.5\pm 2.9\%$ ) derived HTDMA, one can find that only a minor soluble fraction can be explained by  $\text{H}_2\text{SO}_4$  condensation. This

may indicate that some other soluble chemical compounds contributing to the particle growth. Recently, amines in forming organic salts were proposed to be a pathway for contributing to atmospheric nanoparticle growth (Smith et al., 2010; Barsanti et al., 2009). Amines are stronger bases than ammonium and have been shown to displace ammonium from ammonium sulfate (Bzdek et al., 2010; Qiu et al., 2011). Smith and coworkers (2010) have shown that alkylammoniumcarboxylate salts account for 20–50% of the mass of freshly nucleated particles in locations that include Atlanta, Mexico City, Boulder, and Hyytiälä, while sulfates account for only about 10%. A field measurement performed at a fairly remote mid-level mountain site (The Sierra Nevada Mountain in California), which is similar to our study showed a lack of a correlation between particles with sulfate and ammonium during the NPF events (Creamean et al., 2011). Creamean et al. (2011) found that amines and sulfate present within the same newly formed individual particle, implying amines could be responsible for particle growth. Moreover, laboratory measurements of alkylammonium—carboxylate salt nanoparticles showed only slightly lower hygroscopicity than ammonium sulfate nanoparticles (Smith et al., 2010). Therefore, amines may partly explain the missing source of soluble fraction.”