Rose et al. show in their paper measurements on new particle formation (NPF) at a mountain site (Puy de Dome) in central France. Measurements were made during February 2012 with instruments including a NAIS and a PSM. Neutral as well as charged NPF rates as well as growth rates were derived from the measurements at particle sizes of 1.5 and 3 nm, respectively. Since temperatures were quite low during the measurement period the sampling site was located in the free troposphere (FT) for certain periods, which was concluded from LIDAR measurements. One of the main conclusions from the paper is that neutral nucleation dominates in the FT and in the interface between the planetary boundary layer and the FT.

The only major comment I am having concerns the reported concentrations of sulfuric acid ([H_2SO_4]). After revision of the manuscript regarding this aspect, I recommend its publication.

Reported sulfuric acid concentrations were not measured directly but derived by an indirect method, which is summarized in the following and also in the manuscript (sections 2.2.3 and 2.3.1). During a different field campaign (not related to the current study) in Hyytiälä an APi-TOF and a CI-APi-TOF were operated in parallel. While the CI-APi-TOF can measure the neutral sulfuric acid concentration directly, the APi-TOF measures the naturally charged ions. Based on the measurement of $NO_3^-(HNO_3)_{0-1}$ and $HSO_4^-(HNO_3)_{0-1}$ ions the sulfuric acid concentration can be estimated (Eisele, JGR, 1989). Based on their earlier measurements the authors derived a calibration constant that relates the measured ion signals by the APi-TOF to the sulfuric acid concentration. This calibration constant for the APi-TOF was then used during the measurements at Puy de Dome between January 30 and February 6 to derive H_2SO_4 concentrations. From these a scaling factor was derived for an empirical equation that relates the global radiation, sulfur dioxide concentration, relative humidity and condensation sink to the sulfuric acid concentration. This equation was then used to calculate the [H_2SO_4] from their equation (2) during the discussed measurement phase (between February 10 and February 29). Why, am I repeating this methodology here? Because it shows that the [H_2SO_4] is derived around 2 corners and therefore uncertainties are introduced, which can be substantial. It is not clear whether the same calibration factor for the APi-TOF can be used for the Hyytiälä and the Puy de Dome measurements. Furthermore, the measurement period over which the scaling factor was derived is quite short and therefore it is unknown whether the proxy can be used for all conditions during the later phase of the campaign. Therefore, I highly recommend adding some discussion on the uncertainties of the [H_2SO_4]. Mentioning the average bias between the concentrations derived from the proxy and the ones from the APi-TOF (end of section 2.3.1) is not sufficient because a potential systematic error from the APi-TOF derived concentrations is ignored.

Further comments:

page 3, line 2: „occurs at a size of 1.5 nm ±0.4 nm“

page 4, line 5: “Millikan”

page 5, line 7: The first sentence of this section is misleading because it sounds as if the paper would discuss the ion chemical composition. In line 26 (same page) it is then said that the APi-TOF data was not directly used. It would be better to say this in the very beginning. It is also not clear why the resolution of the instrument is being discussed and why equation (1) is necessary.

page 6, line 2: add a hyphen between “APi” and “TOF”

page 6, line 7: better to use “performed” than “achieved” page 9, line 20: Cluster concentrations starting at diameters above 1 nm were used to derive $J_{1.5}$. Is a cluster with a diameter of 1 nm below or above the critical size? What would be the error if it was below the critical size?

page 11, line 28: insert "." after "sub-periods"

page 12, line 6: "positive ions"

page 12, line 16: "Fig. 3" instead of "Fig. 2"?

page 12, line 24: "depends on"

page 14, line 7: To my knowledge Kirkby et al. (2011) did not report that ternary (H_2SO_4-H_2O-NH_3) ion-induced nucleation prefers the positive pathway but stated that the positive route was observed at a high ammonia mixing ratio, whereas the negative pathway was also observed at a rather low NH_3 concentration.

page 14, line 15: "… the IIN rates …"

page 15, line 5: remove comma before "(2010)"

page 15, line 28: "in Fig. 5"

page 29, line 6: "row" instead of "raw"

page 33, Fig. 3: It might be better to use a log-scale for the y-axes of this plot

page 34, Fig. 4f: log-scale would be better to show the [H_2SO_4]