The paper by Rose et al. discusses new particle formation (NPF) events, which were observed on a mountain (Puy de Dôme, 1465 m a.s.l.) in central France. The measurements were performed between the 10th and 29th of February, 2012. LIDAR measurements indicate that during this period the height of the boundary layer was mainly below the measurement site. This suggests that during these periods the site was influenced by free tropospheric air. For the data interpretation the measurement period was split into three sub-periods. During the first one (three days long) the measurement site was not completely disconnected from the boundary layer and NPF was observed during three consecutive days. It is noticeable that during this period the condensation sink (CS) was on average $1.4\text{x}10^{-2}$ s^{-1} which indicates a rather high level of pollution. The estimated sulfuric acid concentration was $7\text{x}10^{6}$ cm^{-3}. Back trajectory calculations suggest that air masses originated from Eastern Europe and were most likely influenced by high emissions from domestic heating due to cold temperatures. The second period (two days long) experienced no NPF although it was characterized by a rather low condensation sink ($1.6\text{x}10^{-3}$ s^{-1}) and the highest H_2SO_4 concentration ($6.9\text{x}10^{7}$ cm^{-3}). According to the authors an almost complete disconnection between the measurement site and the boundary layer is responsible for the low condensation sink and the fact that other (most likely organic) ingredients besides sulfuric acid were missing to promote NPF. During the third sub-period (two days long, CS = $2.4\text{x}10^{-3}$ s^{-1} and $[H_2SO_4]$ = $8.6\text{x}10^{6}$ cm^{-3}) NPF was observed although the boundary layer height was similar to the one observed during sub-period two. However, the origin of air masses was such that they could have picked up more pollution on their way to Puy de Dôme according to the authors.

The nucleation measurements were made with the following instruments: A Particle Size Magnifier (PSM) measured the total (i.e. neutral and charged) concentration of particles starting from a size close to 1 nm. The PSM was operated in a stepping mode which allowed the d_{50} cut-off diameter to be varied between approximately 1 and 2.5 nm. A Neutral cluster and Air Ion Spectrometer (NAIS) measured the size resolved concentration of charged ions, clusters and particles between 0.8 and 42 nm. Measurements of both polarities (positive and negative) were performed with the NAIS. From these data and the size distribution measured with a Scanning Mobility Particle Sizer (SMPS) total (neutral and charged: J^{tot}) and charged nucleation rates (J^{+} and J^{-}) were derived for diameters of 1.5 and 3 nm, respectively.

From the observations two main conclusions are derived: (1) neutral clusters show a strong diurnal pattern with a maximum in their concentration around noon during NPF events whereas cluster ions do not show such a clear pattern; this suggests that neutral clusters are mainly responsible for the formation of new particles at Puy de Dôme during February 2012, and (2) sulfuric acid does not seem to be the main species responsible for nucleation and early growth because NPF occurs only when a reasonably high level of pollution is reached, which is supplied from the boundary layer. The second process is termed as “free tropospheric feeding” in the manuscript.

I recommend the publication of this paper as it shows important data for the free tropospheric region where information on NPF is scarce. However, there are several weaknesses of the paper, which will be described in the following. These should be addressed carefully in a revised version. In summary the three main points of criticism are (further details below under “Major comments”): (i) The concentrations of sulfuric acid were not directly measured but were derived from proxies like the global radiation, the condensation sink, the SO_2 concentration, the RH and an empirically derived scaling factor. The level of $[H_2SO_4]$ is crucial for the interpretation that sulfuric acid is not the main ingredient for NPF. However, in the current version of the manuscript it is not explained how exactly the sulfuric acid concentration was derived and what the errors of the reported levels are. (ii) The observed occurrence of NPF is attributed solely to the presence of pollutants at the measurement site. However, other parameters like the temperature and the relative humidity varied as well. Given the limited amount of days (7 in total where 2 days experienced no nucleation) it is challenging to attribute the occurrence of NPF solely to the variation of one parameter (the pollutants). E.g. sub-period 1 (showing NPF) was characterized by the lowest temperatures (average of -14°C) and the highest RH (91%) - both conditions should favor nucleation - whereas sub-period 2 (no NPF) was characterized by higher temperatures (-1 °C) and low RH (29%). (iii) The presented data indicates that it is not always the case that neutral nucleation is about 40 times higher compared to ion-induced nucleation (IIN) as stated in the abstract. During sub-period 3 IIN seems to have a major contribution to the overall nucleation.

Given the uncertainties in the sulfuric acid concentration and the influence of the other parameters (T, RH) the authors need to be careful not to over-interpret their data. A revised version of the manuscript should carefully address these points.

Major comments:

(1) Sections 2.2.3 and 2.3.1: The APi-TOF (Atmospheric Pressure interface-Time Of Flight) mass spectrometer is introduced in this section; however, none of its data is shown, although it is mentioned (end of section 2.2.3) that APi-TOF data was used to derive a proxy for determining the H_2SO_4 concentrations from other parameters like the global radiation, the condensation sink, the RH and the SO_2 concentration. As the values of the sulfuric acid concentrations are crucial for the conclusion that other compounds besides sulfuric acid are required to explain the observed nucleation rates, the evaluation of the $[H_2SO_4]$ deserves much more attention. Several points need to be addressed when reporting H_2SO_4 levels: (i) the empirical factor k was determined from data between January 30 and February 6, which is a rather short time to gain confidence in the derived factor, (ii) it is mentioned that the period between February 10 and February 29 was characterized by unusual cold temperatures, it is therefore questionable if the derived scaling factor is valid under such conditions, and, most importantly (iii) the APi-TOF cannot directly measure the sulfuric acid concentration as it measures only atmospheric ions and not the neutral H_2SO_4 molecules. Previous studies, like the ones cited (Petäjä et al., 2009; Mikkonen et al., 2011) were however deriving scaling factors based on measurements with a chemical ionization mass spectrometer (CIMS), which can measure the neutral $[H_2SO_4]$ and should therefore yield much more reliable approximations. To my knowledge the method of deriving H_2SO_4 values from APi-TOF ion measurements has not been described anywhere in the literature before; therefore a detailed description is required in this manuscript. Furthermore, an evaluation of the deployed methods should be presented, which relies on a side-by-side measurement of an APi-TOF and a CIMS over a certain period.

In summary, in order to report any $[H_2SO_4]$ values and use them for the interpretation of the data the methods used to evaluate the sulfuric acid concentrations need to be presented in much greater detail and the authors need to convince the reader that their derived concentrations are accurate enough to be used at all. Detailed error estimates are required as well.

(2) Nucleation rates are known to depend strongly on temperature and also on relative humidity in some systems. However, these effects are neglected from the discussion of the observed nucleation rates. Instead, the nucleation rates are discussed mainly in terms of the sulfuric acid concentration and the degree of pollution (condensation sink and black carbon concentration) during NPF. If one compares the temperatures (T) and the relative humidities (RH) of the different sub-periods, at least qualitatively the variation of T and RH can also explain the occurrence of NPF. In my opinion the amount of data is too small to disentangle the effect of all involved parameters unambiguously. This should be explicitly stated.

(3) I am not completely convinced, that neutral nucleation dominates to the extent the authors suggest. Taking the data from Table 1, one can calculate the fraction of the ion-induced nucleation (IIN) from the ratio of the ion-induced formation rates ($J_{IIN,1.5} = J_{1.5}^{+} + J_{1.5}^{-}$) and the total formation rates ($J_{1.5}^{tot}$). These fractions are rather low for the first three events (<5%) (all belonging to sub-period 1) but are 49% and 19% for the last two events (both belonging to sub-period 3). If one takes into account the low temperatures during sub-period 1 (-14 °C) and compares this to the temperature during sub-period 3 (+5 °C) another possibility, consistent with the observations, would be that the neutral nucleation pathway dominated due to the lower temperatures (and maybe higher RH). In their revision, the authors should therefore discuss such a possibility. The fact that IIN is compatible with neutral nucleation during sub-period 3 can also be concluded from the data in Table 3 where the concentration of charged nuclei is much higher than the neutral one. Since during sub-period 3 the site was influenced by free tropospheric air (in contrast to sub-period 1) it would be possible that IIN is rather important under these conditions.

Other comments:

(4) page 18356, line 15: following the arguments given in comment (3) 40 times higher neutral nucleation rates are an extreme case because the fraction of IIN can also reach ~50% during observations made in this study; this statement should therefore be revised

(5) page 18358, line 21: remove extra dot before the word “and”

(6) page 18359, line 5: “Milikan diameters”

(7) page 18359, line 12: “ensures”

(8) page 18359, line 17: the word “further” should be removed

(9) page 18360, line 4: replace the word “sampling” by “activation of particles”

(10) page 18361, line 7: maybe better to use “deriving” instead of “defining”

(11) page 18361, line 25: Which other trace gases besides SO_2 were measured? Could these measurements give further insight into the origin of air masses?

(12) page 18363, line 4: there is something wrong with the unit of k, it should be $m^2\ W^{-1}\ s^{-1}$

(13) page 18364, line 18: The particle concentration in the size range between 1 and 2.5 nm could also include sub-critical particles (the authors mention that the critical size is somewhere close to 1.5 nm in the introduction). If the PSM was operating in scanning mode it should also be possible to provide a number concentration for particles between 1.5 and 2.5 nm. Do the authors have evidence that a size of 1 nm is above the critical diameter?

(14) page 18364, equation (5) and page 18365, equation (6): Shouldn't the denominator in the third term on the RHS of the equations contain the width of the size bin $N_{1\text{-}2.5}$, which is 1.5 nm instead of 1 nm?

(15) page 18366, lines 27 and 28: "ion concentrations"

(16) page 18367, line 16: remove the word "at"

(17) page 18367, line 22: "maxima" instead of "maximum"

(18) page 18372, lines 21 to 23: I do not agree with that statement. Figure 6b clearly shows that the nanoparticle concentrations decrease when the condensation sink (CS) exceeds ~$7\text{x}10^{-3}$ s^{-1}. It rather seems that there is a range where the CS has no clear effect; however, for large values it has an effect as the concentrations decrease by about one order of magnitude as CS increases from $7\text{x}10^{-3}$ to $3\text{x}10^{-2}$ s^{-1}.

(19) page 18380, line 5: "third row" instead of "second raw"

(20) page 18381, line 8: "in the table"?

(21) page 18382, line 2: "indicated in the table"

(22) page 18383, line 3: "calculated" instead of "calculating"

(23) page 18384, line 3: "shaded" instead of "shading"

(24) page 18385, figure 3: It is surprising that all Js are positive throughout the whole day (from 8 to 18 UTC). In fact, $J_{3,tot}$ is always larger than 0.4 cm^{-3} s^{-1} even in the early morning and in the late evening when there is no sunlight anymore. What is the explanation for this observation?

(25) page 18387, line 2: "close" instead of "closed"

(26) page 18388, figure 6a: red/magenta circles seem to be missing for the high H_2SO_4 values; the word "no" seems to be missing in the last row of the legend