Referee 1: we thank Referee 1 for his comments and suggestions.

Main comment: 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 NO3 –(HNO3)0-1 and HSO4 –(HNO3)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 H2SO4 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 [H2SO4] 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 [H2SO4] 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 [H2SO4]. 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.

Reply 1: Similar comments were addressed in a previous review. It is true that the empirical factor was obtained from a rather short period, but during which atmospheric conditions were very similar to the ones observed during the studied period. Especially, between 30 January and 6 February, temperatures were very low, on average -11.8°C. We clearly believe that the use of this factor gave us the opportunity to work with a proxy adapted to the site and especially to the unusual conditions observed in February 2012. We also strongly believe that the use of this proxy is much more reliable than the use of any other proxy that would be derived from other location/time periods.

A potential systematic error on the sulfuric acid concentration calculation from the Api-Tof due to a unadapted use of the PUY scaling factor is of course possible, and would affect the absolute values of calculated sulfuric acid concentrations, but not their variability. In the present paper, we focus on relative changes in the sulfuric acid concentration from one period to the other, and do not actually need to be accurate with the proxy. We now mention the possible error on the sulfuric acid concentrations in the paper, and clearly state that it is only a proxy.

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

Reply 2: correction done.

Comment 3: page 4, line 5: “Millikan”

Reply 3: correction done.

Comment 4: 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.

Reply 4: It is true that the first part of section 2.2.3 does not significantly contribute to the reader comprehension, and was thus removed.

Comment 5: page 6, line 2: add a hyphen between “APi” and “TOF”

Reply 5: correction done.

Comment 6: page 6, line 7: better to use “performed” than “achieved”

Reply 6: changed.

Comment 7: page 9, line 20: Cluster concentrations starting at diameters above 1 nm were used to derive J1.5. 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?

Reply 7: The smallest particles include both clusters and large molecules, and the exact critical size cannot be known (Kulmala et al., 2013). This is the reason why different J, such as $J_{1.5}$ and J_3, are used to describe nucleation events. This last point is now mentioned in Section 2.3.3.

Comment 8: page 11, line 28: insert “.” after “sub-periods”

Reply 8: correction done

Comment 9: page 12, line 6: “positive ions”

Reply 9 : correction done.

Comment 10: page 12, line 16: “Fig. 3” instead of “Fig. 2”?

Reply 10: correction done.

Comment 11: page 12, line 24: “depends on”

Reply 11: correction done.

Comment 12: page 14, line 7: To my knowledge Kirkby et al. (2011) did not report that ternary (H2SO4-H2ONH3) 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 NH3 concentration.

Reply 12: Precision regarding ammonia mixing ratio was added: “In particular, the occurrence of ternary nucleation involving sulphuric acid at high ammonia mixing ratios, with typical path on positive way, could explain the excess of positive ions, at least on event days.”

Comment 13: page 14, line 15: “… the IIN rates …”

Reply 13: correction done.

Comment 14: page 15, line 5: remove comma before “(2010)”

Reply 14: correction done.

Comment 15: page 15, line 28: “in Fig. 5”

Reply 15: correction done.

Comment 16: page 29, line 6: “row” instead of “raw”

Reply 16: correction done.

Comment 17: page 33, Fig. 3: It might be better to use a log-scale for the y-axes of this plot

Reply 17: y- axes were changed.

Comment 18: page 34, Fig. 4f: log-scale would be better to show the [H2SO4]

Reply 18: log scale was used for H_2SO_4

Referee 2: we thank Referee 2 for his comments and suggestions.

Comment: I do not agree with authors attributing the period 3 to NPF in free troposphere. AT the same time around 9 - 10 AM when nucleation begin to happen also NO2 values begin to increase, CS and RH shows also show increase. All this is an indication that the air at sampling site is influenced by different air mass. In this case most likely by boundary layer air. I would like to ask authors to show detailed figures for period 3 showing temporal evolution of RH, CS, NO2, clusters and new aerosol particles to clearly show if the presence of new particles happens before other parameters start to change their values or not. Comparing average values can be misleading.

Reply : As suggested, a new figure (Fig. 3 shown below) was added to the manuscript to highlight the temporal evolutions of cluster concentration, RH CS and NO$_2$. This figure is focused on the 28th, to show that on this particular day the total cluster concentration starts to increase ~30 minutes before the other parameters listed before, suggesting that nucleation could be initiated in the free troposphere. Similar analysis conducted on the 29th led to conclude that, in contrast, the process was most probably triggered at the interface between the BL and the FT. The manuscript was thus modified and conclusions were balanced according to the previous observations.

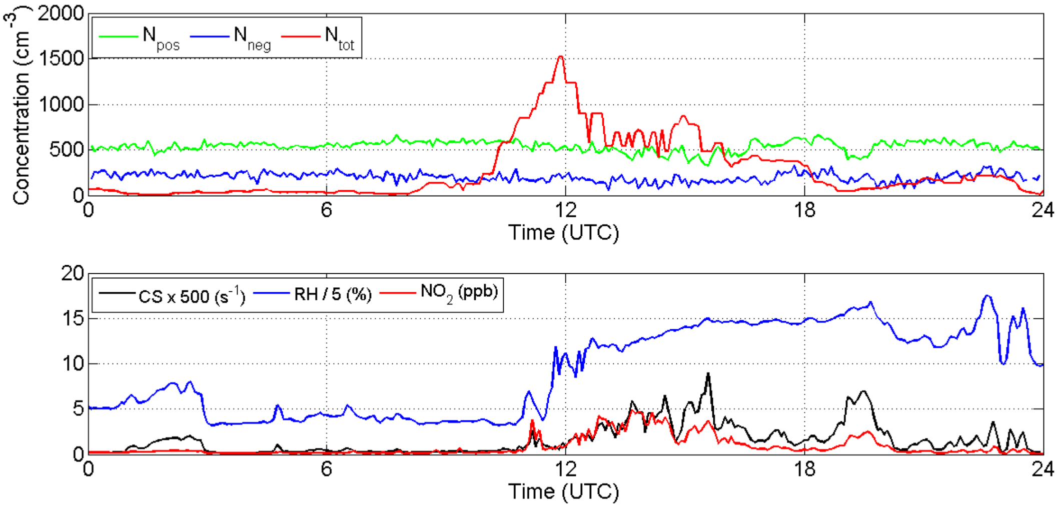

Fig. 3. Diurnal variation of the cluster concentrations (upper panel) and BL tracers (lower panel: condensation sink (CS), relative humidity (RH) and NO$_2$) observed at the PDD on the 28th of February.