Response to Anonymous Reviewer #2:

We thank the reviewer for helpful comments. Our responses are given below in **bold**.

Comments:

1. Page 14122, lines 21-29: "A remarkable feature of Fig. 1d that could not be studied in previous work is that the change in elemental composition occurs at the onset of NPF before the mode diameter reaches the NAMS size range." I do not fully understand why the authors find this observation remarkable. A few more sentences may help. It also seems to me that the sulfur and nitrogen mole fractions start to increase around 06:00 AM on both days which is several hours before the onset of NPF, even though it is hard to tell from Fig. 1 exactly when NPF starts. Did you observe similar diurnal trends in mole fractions on days with no NPF?

    The observed change in nanoparticle composition before the mode diameter has passed through the NAMS size range is remarkable because 1) it shows how the composition of the condensational sink changes at the onset of NPF and 2) it illustrates how changes in chemical composition and size distribution of ambient nanoparticles are not necessarily correlated. These points will be emphasized more strongly in the revised manuscript.

    With respect to non-event days, we did not have sufficient time resolution to observe diurnal trends with high time resolution as we did on event days. However, Fig. 2d shows average particle composition for event periods and several other non-event periods. Comparing average nanoparticle composition during event periods to average nanoparticle composition during other non-event periods shows that the average composition is essentially the same during non-event periods, whereas it changes substantially during event periods (more S and N, less C). We also note that in a more recent campaign, we were able to make highly time-resolved measurements on non-NPF days and found substantial increases in inorganic components during the daytime, though the exact identities of those components differed from those measured on event days (Bzdek et al., 2013). We will enhance the discussion of this topic in the revised manuscript.

2. Page 14122, lines 16-17: "The carbon mole fraction is anti-correlated with the other elements, which has been observed in urban locations as well." Do the authors have an idea why this is? The fact that the sulfur mole fraction in Fig. 2b drops in the evenings; is this an effect of the sulfur actually decreasing or only because the carbon increases? If the sulfur decreases, may it be evaporation or is it air mass related? And why is the carbon increasing at night?

The carbon mole fraction is anti-correlated with the other elements (i.e. S and N) because carbon is associated with carbonaceous (organic) components of the aerosol whereas S and N are mainly associated with inorganic components (sulfate, ammonium, and possibly nitrate). Thus, this observation indicates that during NPF there is a relative enhancement in the inorganic composition of the aerosol. The drop in S elemental mole fraction and coincident increase in C mole fraction in Fig. 2b-c is a result of gas phase sulfuric acid decreasing substantially and the carbonaceous matter becoming more processed, resulting in an increased C mole fraction relative to the S mole fraction. We note that although the *relative* concentrations of S and C are anti-correlated, the *absolute* amounts of particulate C and S increase substantially during NPF, as there is a substantial increase in nanoparticle mass. We will add discussion about this to the revised manuscript.

3. Page 14125: The authors refer to measurements with AMS and TDCIMS during the NPF events. These measurements should be mentioned in the methods section even if they are only complementary measurements.

   In the revised manuscript we will introduce these instruments in the Methods section.

4. Page 14125, lines 25-30: The authors write "It is possible that the acidity of the aerosol at 20 nm could be explained by an ammonia concentration that is on the same order as the sulfuric acid concentrations. However, ammonia concentrations are typically thought to be two orders of magnitude higher than sulfuric acid concentrations (Kirkby et al., 2011)." What are the typical ratios between ammonia and sulfuric acid during NPF events based on earlier campaigns at this site in Hyytiälä?

   In Hyytiälä, ammonia is typically more abundant than sulfuric acid by about two orders of magnitude. In the revised manuscript, we will clarify this point.

5. Page 14126, line 21: The authors use the term "Carbonaceous matter" and the term "organic" in Fig. 3. It is better to be consistent, since carbonaceous matter includes both organics and black carbon. "Carbonaceous matter is also used on line 19 in the abstract.

   We delineate "carbonaceous" and "organic" matter by referring to the carbon (and associated oxygen) content analyzed by NAMS as "carbonaceous", as we know nothing about the molecular components of this matter. However, when we discuss possible molecular identities, we refer to these species as "organic". NAMS cannot distinguish the various forms of carbon in a particle other than estimation of an oxygen to carbon mole ratio. In the revised manuscript, we will ensure consistency in the use of "carbonaceous" vs. "organic".

6. Page 14122, line 4: The "Draxler and Rolph, 2013" reference should be added to the reference list.

   We will add the reference to the revised manuscript.

7. Fig. 2: Were the eight NPF events in Fig. 2 the only NPF events throughout the whole campaign from 21 March to 24 April? If not, why were these events specifically chosen? Please give some information here.

   The eight events shown were the complete set of NPF events measured by NAMS in the campaign that resulted in particle growth to >20 nm diameter.

8. Fig. 1d: If the sulfuric acid concentration has been calculated using Eq. (2) which depends on the measured growth rate, how was the sulfuric acid concentration then estimated for instance between 12:00 AM and 12:00 PM on 18/4/2011 when there is no growth event in Fig. 1a and therefore hard to estimate any growth rate?

   The sulfuric acid shown in Fig. 1d is an experimental measurement using a chemical ionization mass spectrometer (CIMS) and therefore does not involve Eq. (2) or any assumptions about particle growth. The measured sulfuric acid concentration is compared to a calculated value only during the NPF events when particles are clearly growing, which is the time period during which Eq. (2) applies. We will clarify this in the revised manuscript.

9. Page 14122, lines 18-19: "During NPF, the sulfur and nitrogen mole fractions are larger and the carbon mole fraction is smaller than immediately before or after each event." Did the sulfur and nitrogen mole fractions have similar diurnal trends on days with no NPF?

   As mentioned in the response to comment #1, sufficient time resolution did not exist to observe diurnal trends on non-event days other than morning/evening as shown in Fig. 2d. Fig. 2d indicates that on non-event days the average composition is clearly different from the composition on event days and that the composition on non-event days is similar to average composition during other non-event periods. This observation argues against a diurnal trend in S and N mole fractions of similar magnitude to that observed on event days. A more recent campaign resulted in acquisition of data that permits highly time-resolved nanoparticle composition measurements on non-event days and the results do indicate substantial diurnal trends in composition on non-event days (Bzdek et al., 2013).

References:

Bzdek, B. R., Horan, A. J., Pennington, M. R., DePalma, J. W., Zhao, J., Jen, C. N., Hanson, D., Smith, J. N., McMurry, P. H., and Johnston, M. V.: Quantitative and Time-Resolved Nanoparticle Composition Measurements During New Particle Formation, Faraday Discuss., doi: 10.1039/C1033FD00039G, 10.1039/c3fd00039g, 2013.