Authors' response to reviewer 2 comments on manuscript titled "Fast time response measurements of particle size distributions in the 3-60nm size range with the Nucleation Mode Aerosols Size Spectrometer", submitted to AMT 24th January 2018

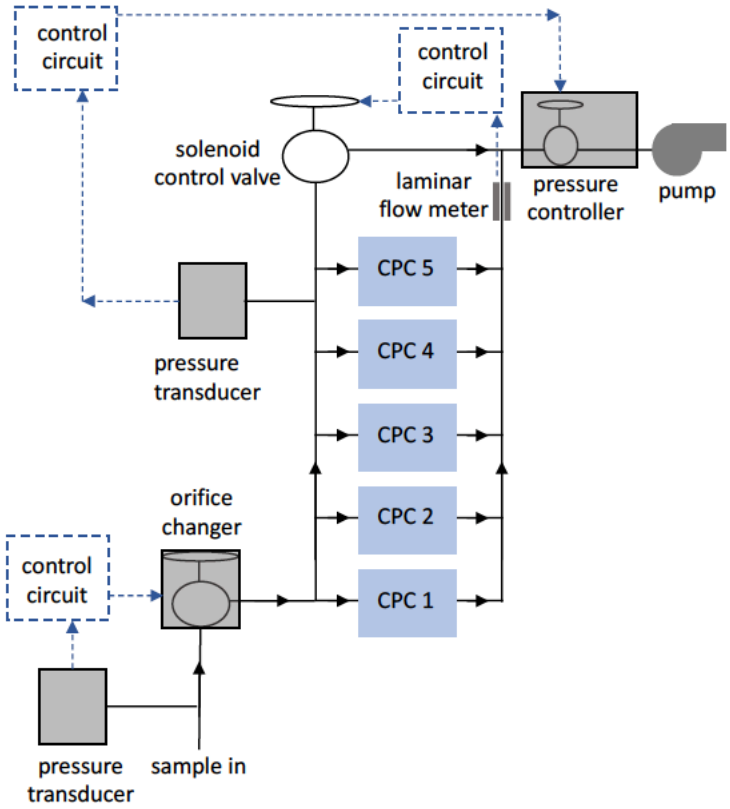
The authors would like to thank the reviewer for their considered and positive evaluations of the manuscript. Our responses are detailed below, with the reviewer comments in normal text and our response in italics.

1. Figures 1 and 2: I don't understand how flow velocity is maintained constant in each of the CPCs. I had assumed that critical flow orifices (CFOs) were installed on the exhaust of each unit (as is done in standard TSI3010 or TSI3772 counters) and that system pressure was controlled by varying flow through a bypass line. However, although it shows a solenoid control valve dangling in space above the column of counters, Figure 1 seems to indicate that system pressure is controlled using the valve located just upstream of pump; varying flow through this valve would result in varying the flow velocity through the entire bank of counters. Can you please clarify? If the flow through each CPC isn't controlled by a CFO or pump, how do you know that sample flow is split equally between the five CPCs? Do you measure total flow through each counter?
   The NMASS CPC design is similar to that of a TSI3025, which includes a valve to regulate the ratio of aerosol sample to sheath flow; how is the flow split maintained in the NMASS CPCs? If the ratio is dictated by flow resistances instead of valves, how does it change as the filters get dirty? How sensitive is the CPC detection efficiency to variations in flow velocity through the condenser region?

   The flow through the CPCs is determined by the pressure drop across the CPC saturator filter (see fig 2) and the solenoid valve on the bypass flow. The pressure drop across the capillary of each CPC is measured continuously. Calibrations were done in the lab to relate the pressure drop to the flow through the capillary and this used to calculate the flow for each CPC continuously during operation. This was indeed unclear in the original manuscript and a full explanation has now been added (p9 line 32 to p10 line 2). The new text reads:
   "The flow through each CPC is determined by the pressure drop across the filter in the saturator (see Fig. 1) and the proportional control valve. The pressure drop across each capillary is continuously measured during operation, as shown in Fig. 1. Calibrations were done to relate these pressure drops to a volumetric flow, and it is these flows that are then used to determine the concentration in each channel from the number of particles counted."

   The flow across each CPC capillary is constantly monitored. If the flow at a given upstream pressure drops by above 10%, the filter is changed to avoid possible effects on the CPC detection efficiency.

   The line connecting the solenoid control valve to the pump was indeed missing in this figure, as the review later points out. We have replaced this in a new version of Fig 1 (see below).



2. Is Fluorinert hygroscopic? If so, how does the CPC performance change over time as H2O is absorbed into the working fluid? During ATOM, the aircraft often flies through very humid regions do you dry the sample flow and if not, how often do you change the fluid in the counters?

   Flow into the NMASS is always dried to below 20% RH, so we do not expect problems from H_2O being absorbed in the working fluid. This explanation was missing in the original manuscript, and so has been added to p25 lines 22-25, new text reads:
   "For operation on ATom the sample flow is passed through a large diameter Nafion™ dryer before entering the NMASSes. This reduced the relative humidity to below 20%. This ensures that particles measured in the NMASS are classified consistently by dry diameter, and avoids potential problems of particle losses associated with water vapour condensation during flow expansion in the orifice or effects of water vapour on the performance of the CPC working fluid."

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3. Page 9, orifice discussion: Sample temperature will drop as flow is expanded across the orifice, which in very humid cases, may lead to vapor condensation and associated particle losses. Was this effect considered in your experiments and analyses?
   The sample flow is dried before entering the instrument (this explanation was missing from the original manuscript and has now been added to p25 lines 22-25, and page 9 lines 30-31), so the problem of vapor condensation and associated particle losses as the flow expands across the orifice can be discounted as we

do not experience high humidities in the instrument. For operation without a drier in humid areas this would need to be considered. New text reads:
Page 9: "A Nafion drier upstream of the NMASS instrument maintains RH to <20%, elminating the possibility of condensation in the pressure reducer."

Page 25: "For operation on ATom the sample flow is passed through a large diameter nafion™ dryer before entering the NMASSes. This reduced the relative humidity to below 20%. This ensures that particles measured in the NMASS are classified consistently by dry diameter,and avoids potential problems of particle losses associated with water vapour condensation during flow expansion in the orifice or effects of water vapour on the performance of the CPC working fluid."

4. Page 9, line 30: the by pass line is not clearly indicated in Figure 1.
   This bypass line was missing in figure 1 and has now been corrected (see above)

5. Figure 7: the caption is cut off and thus doesn't make complete sense.
   This has now been corrected – figure caption now reads:
   Fig. 7 Counting efficiency of NMASS 1 as a function of particle diameter for particles of different chemical composition: limonene ozonolysis products (diamonds), atomized ammonium sulphate (stars) and dioctyl sebacate (circles). Only three channels are shown here because it was not possible to produce atomized particles small enough for the two channels with the smallest cut-off sizes by atomizing ammonium sulphate or dioctyl sebacate. Counting efficiencies fall with decreasing particle diameter as particles become smaller than the Kelvin activation diameter of each channel. There is no statistically significant sensitivity of counting efficiency to particle composition.

6. Page 20-21 discussion on NPF formation and growth: you might mention Rodney Weber's technique of using an OPC to measure the size of droplets coming out of an ultrafine CPC to infer new particle formation and growth: Weber et al., Measurements of enhanced H2SO4 and 3-4 nm particles near a frontal cloud during the First Aerosol Characterization Experiment (ACE 1), JOURNAL OF GEOPHYSICAL RESEARCHATMOSPHERES, 106 (D20), 24107-24117, 2001.
   We have now included this in our summary of airborne aerosol size distributions measurements on page 4 lines 9-10. New text reads
   "Weber et al. (2001) made airborne measurements using an optical particle counter as the sensor for an ultra-fine CPC to determine the size of the grown droplets and infer the size distribution of 3-10 nm particles that nucleated them."

7. Regarding use of SMPS instruments to study NPF from aircraft: even if they could be operated at fast scanning rates, standard SMPS systems lack sensitivity to nucleation mode particles at low pressure because of the particles's greatly increased electrical mobility. For example, at 100 hPa, a 10 nm particle has about the same electrical mobility as a 3 nm particle at sea level pressure.

   This is an excellent point, which we had neglected in the original manuscript. We have now included it in the discussion of SMPS performance for airborne measurements on page 4 lines 13-14. Text reads:
   "Weber et al. (2001) made airborne measurements using an optical particle counter as the sensor for an ultra-fine CPC to determine the size of the grown droplets and infer the size distribution of 3-10 nm particles that nucleated them. "

8. Page 22, line 14: should be "than" instead of "then"
   addressed

9. Page 22, lines 17-22: repeated text
   addressed