We thank the two anonymous reviewers for their careful reading and good comments. Below we provide point-by-point answer to comments and action taken to improve the manuscript.

Anonymous Referee #1

This paper presents analysis of a potentially very important process in improving our ability to resolve cloud activation potential. It is becoming increasingly apparent that evaporative loss or increased condensation of semi-volatile compounds not only effects measured behaviour using current instrumentation, but also creates a disconnect between measured behaviour and processes occurring in the atmosphere. This concise paper goes someway to addressing questions that allude to these effects and should be published for wider consumption. Before publication, i would like some comments to be addressed.
Introduction:

General comments:
I would request the use of the Henrys law coefficient definition is consistent throughout the paper. There is use of 'effective Henrys law' and standard Henrys law. I would have to agree with the discussion phase of the paper by Compernolle et al 2014 regarding this issue as there are multiple definitions :http://www.atmos-chem-phys-discuss.net/13/25125/2013/acpd-13-25125-2013-discussion.html.

It is the effective Henry's law coefficient that determines mass transfer between gas and aerosol phase. We will emphasize this in the manuscript.

'2 Methods' line 3 - Following on from the above, is it not effective Henrys law coefficient? Dosnt a Henrys law coefficient restrict its use to infinite dilution? In my mind this dosnt necessarily catch the trajectory of condensation as a function of increasing water vapour saturation.

This is true. To catch the evaporation/condensation we need to use effective Henry's law coefficient and to avoid the infinite dilution limit we have used EQUISOLV to calculate non-ideal liquid phase thermodynamics.

'2.1 Experiments' 'For moderate semivolatile gas concentrations, the water flow rate...is large enough to absorb all the gas' Are you referring specifically to nitric acid here. For organics, for example, over which volatility range would you expect this to hold true? Also, what is a 'moderate' semivolatile gas concentration?

We have made simple calculations for nitric acid, but the results can be generalized quite easily. Typical water flow rate on the wall is Qw=60E-6 L/min and sample air flow rate is Qs=0.5 L/min. When the mixing ratio of a semivolatile vapor is x, the total amount entering the CCN counter is Qs*x*P/(R*T), where P is chamber pressure, R is the gas constant and T is temperature. The concentration of the vapor after complete dissolving into the water on the walls is c=Qs/Qw*x*P/(R*T). Therefore, the ratio of the partial pressures of the semivolatile at the inlet and after dissolving is Pinlet/Pdiss=H*R*T*Qw/Qs, where H is the effective Henry's law constant. For nitric acid (the regular H=2.1E5 mol/kg/atm), the ratio is 2.1E5 mol/kg/atm*1 kg/L*(0.08205746 L*atm/K/mol)*298 K*60E-6/0.5=616, which means that nitric acid vapor pressure can decrease by two orders of magnitude without significantly reducing the effectiveness of the water on the walls to act as a sink. As the effective Henrys's law coefficient of a semivolatile must be greater than 2e7 mol/kg/atm (see below) to have an effect on activation, water on the walls can be considered as a perfect sink for condensable vapor. In the case of evaporation (e.g. ammonium nitrate), the amount of semivolatiles in the particles is negligible due to the low volume of the particles, so the water on the walls can easily absorb all volatile vapors.

'3 results' Please state the source for HNO3 diffusion coefficient. When modelling loss to the wetted walls, are you using a fixed Henrys law coefficient?

Value given for diffusion coefficient can be found from Durham and Stockburger (1986). However, to be exact, we have used temperature dependent value in simulations, that is based on measurement data and having a slightly higher value at 298K. We will correct this and add a reference to Kokkola et al (2003).

We have assumed walls to act as a perfect sink, so the vapor pressure on the wall is assumed to be zero.

'The 100ppt is a reasonable concentration for background air, but the effect on evaporation rates is small because this concentration is well below the equilibrium value.' - If the concentration (of a vapour) is well below the equilibrium value then wouldnt this encourage evaporation? Or am i missing something here?

The idea was that 100 ppt is such a low concentration that it will not make a big difference for evaporation rate if the concentration of 10ppt or 100ppt is used as the surface pressure on particles is much higher than 100ppt. We will reformulate this.

page 8422 line 20 'Aerosol acidity and hygroscopicity are more important than the mass fraction of soluble compounds.' Isnt this a circular argument? If the mass fraction of soluble compounds is large (presumably referencing water solubility) then this increases hygroscopicity and thus nitric acid uptake. Why are you decoupling the two, is this related to effects on thermodynamics?

Reviewer has made a good point, and this is related to thermodynamics. The results with different mass fractions of ammonium sulphate and insoluble compound are presented because the relative change in hygroscopicity was the same for all composition ratios, but this was not exactly the case with "model compound" having different hygroscopicities. We will add more discussion why this is presented.

Page 8424 'The effect seen on particles' CCN activity is naturally dependent on the compounds, and, especially, the Henry's law coefficient..' It would be great if this analysis could be extended to other semi-volatiles. I fully appreciate the difficulty in this but the reader might not. There is a continual reference to the importance of Henrys law coefficients (standard and effective). Is the model reliant on this value or could the use of saturation vapour pressures and activity coefficients in dilute solutions be used, or would you recommend otherwise. Presumably, for organic compounds, this would be

a huge source of error?

We will add a section into the manuscript where we discuss more about the effect of Henry's law coefficient. We will do this with a hypothetical compound with properties of the nitric acid molecule and effective Henry's law partitioning coefficient between 1e5 mol kg-1 atm-1 and 1e11 mol kg-1 atm-1. The high value represents nonvolatile material that evaporates too slowly inside the CCN column to affect the activation of particles and the lower value represents semivolatile aerosol that has higher volatility than that of nitric acid. It turns out that the evaporation will become efficient when coefficient is smaller than ~5e9 mol kg-1 atm-1. On the other hand the condensation will be playing a role after the coefficient exceeds 2e7 mol kg-1 atm-1. These values naturally depend on the assumed gas phase concentration at the top of the CCN column. When effective Henry's law coefficient is between 2e7 mol kg-1 atm-1 and 5e9 mol kg-1 atm-1 particles may grow or shrink based on the chemical composition and gas phase concentration. However, to get effective Henry's law coefficient, thermodynamics is needed. With organics we would also need to take account the phase of particles. Thus the evaporation of organics is not within our scope.

Also, could the authors please note whether the analysis could be extended to multi-component evaporation/condensation?

Absolutely. Actually in the case of ammonium nitrate, we already have evaporation of both ammonia and nitrate. We will add discussion to the end of manuscript.

Minor comments:

Abstract: line 10: How big 'a' fraction line 14 - please specific whether this is in the instrument

We will add that this refers to the concentration in the instrument

References

Durham, L., and L. Stockburger, Nitric acid-air diffusion coefficient: Experimental determination, Atmospheric Environment (1967), Volume 20, Issue 3, 1986, Pages 559-563, 1986

Kokkola H.: Model studies on the evolution of fogs and clouds in the presence of semi-volatile gases. Ph.D. thesis, Report Series in Aerosol Science, Finnish Association for Aerosol Research, Helsinki, 106 pp, 2003

Anonymous Referee #2

Review:
This manuscript presents a model case study of how evaporation of ammonium nitrate particles and one particular semi-volatile compound would influence the measured CCN concentration in the DMT CCN counter. This is not reflected in the title and abstract, which are much more general and suggest a combined thorough modeling and measurement study (e.g. line 4-5 of abstract). Only at the end of the manuscript it was clear to me that no experiments on the partitioning of nitric acid would be presented. The title suggests a more general study of maybe more CCN counters, more semi-volatile compounds, or a quantification of the effect in terms of volatility etc.. In the case of what is actually presented, I think it would be appropriate for a technical note, but not for a full paper (even for a technical note, the scope is relatively narrow). I would therefore suggest to re-submit this manuscript as a technical note, with an updated title and abstract that more reflect what is actually done in the paper. To have a full paper, either the scope of the modeling study should be wider (e.g. use hypothetical compounds with a range of volatilities etc…), or experimental results for the nitric acid studies should be added.

As far as we know, our study is the first one where it is actually shown by means of modelling and experiment how a semivolatile evaporates from particles and condenses on the wetted walls in the CCN column of DMT CCN counter, and how this affects the measured CCN activity of particles. Evaporation of ammonium nitrate has been found to affect aerosol measurements with several other instruments (Bergin et al 1997, Cheng and Tsai 1997, Shang and McMurry 1992, Hering and Cass 1999, Kecka and Wittmaack 2005, Hua et al 2011), and here we show that the same issue exists with DMT CCN counter. Ammonium nitrate is chosen as thermodynamics for it is well known, and it is climatically more relevant than any other semivolatile compound found from the atmosphere what comes to the effect on aerosol hygroscopicity during cloud droplet formation. Based on literature some organics have similar effect, but their availability is limited and the exact thermo-physical properties are often lacking. It is possible, like reviewer suggests, to use hypothetical compounds with range of Henry's law coefficients (for range of volatilities) and we will add discussion to the manuscript on the range of Henry's law coefficient where evaporation from or condensation into particles need to be taken into account.

Based on this, we will not re-submit the manuscript as a technical note. Instead we will change the title to be "Ammonium nitrate evaporation and nitric acid condensation in DMT CCN counters**"**, and also rewrite the abstract to be more descriptive to avoid confusion with the manuscript content.

In case of re-submission as a technical note several important points should be addressed.

1) The goal of the nitric acid model study is not very clear: For example in the conclusions is stated that "typical concentrations in the atmosphere are such that that concentrations of semi-volatile gases are too low…to affect the CCN studies" (page 8426, line 2-5), whereas in the introduction it is stated "we show that DMT CCN counters can be used to assess the effect of nitric acid on CCN activity" (page 8416, line 7-9) It sounds like two goals are mixed together: (1) To study what the effect would be in the ambient atmosphere, and (2) to make an initial evaluation if the CCN counter could be used for an experimental study in the lab, where high concentrations of nitric

acid could be used to study the effect of nitric acid partitioning on CCN concentrations. It is ok to make both points, but clearly separate them and make them explicit. From the results it seems to me that at atmospheric concentrations the wall losses in the CCN counter are too large to see a much of effect of semi-volatiles on CCN concentrations. But it might be possible to make high concentrations of nitric acid in laboratory experiments, so that atmospherically relevant concentrations could be left in the air flow to interact with the activating droplets. If this indeed were the points the authors were trying to make, this should be stated much more clearly.

The Reviewer has understood the idea of manuscript correctly. We will make these different goals more clear throughout the manuscript.

2) It is not very explicit throughout the manuscript if the nitric acid concentration mentioned is the concentration in the inlet, of in the CCN counter at point of activation. It would make things much more clear to also mention how much nitric acid is left over at the point of activation that then causes the reduction in critical diameter, e.g. at page 8422, line 5ff, and line 15ff; page 8423, line 15-16) but also other places. Or how much nitric acid was lost to the walls. Otherwise, for non-experts it can sound like actually 13 ppb of nitric acid would only have a small effect on CCN activation. For example, you could show how much nitric acid went into the particle and how much to the walls, before the particle was activated. Especially for the comparison with Laaksonen (page 8422), it would be good to mention this explicitly, otherwise it looks just like a discrepancy between different models.

We will improve this and add more discussion on how much nitric acid is left at point of activation. Especially the comparison to Laaksonen et al is now misleading and we will add explicit explanation why the nitric acid effect is much smaller with the CCN counter. However, at the same time it needs to be emphasized that it is not the amount of nitric acid left in the gas phase at point of activation that defines the strength of effect but the concentration prior to activation and actually the history since the sample entered into the CCN column.

3) Please have your paper proofread by a native speaker, as some formulations are quite cumbersome and there are problems with definite and indefinite (no) articles.

We will proofread the article. If needed, the article will go through copy-editing process before publication in AMT.

4) If you use the general term CCN activity in abstract etc please define it first

We will rewrite the abstract and define CCN.

References

Bergin M. H., J.A. Ogren, S.E. Schwartz, and L.M. McInne: Evaporation of Ammonium Nitrate Aerosol in a Heated Nephelometer: Implications for Field Measurements, Environ. Sci. Technol., 31 (10), pp 2878–2883, 1997.

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Zhang X., and P. H. McMurry, Evaporative losses of fine particulate nitrates during sampling Atmospheric Environment. Part A., 26, 18, 3305–3312, 1992.

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Kecka, L., and K. Wittmaack: Effect of filter type and temperature on volatilisation losses from ammonium salts in aerosol matter, Atmospheric Environment, 39, 22, , 4093–4100, 2005

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