Reply to Referee 1:

We'd like to thank the reviewer for her/his comments which helped to greatly improve the manuscript. In the following, we provide replies to the reviewer's comments (typeset in bold).

**This manuscript examines biogenic and anthropogenic SOA surrogate particles for their ice nucleating ability. The SOA particles were photochemically generated in the Manchester aerosol chamber (MAS) and then transferred to the Manchester Aerosol and Ice Cloud Chamber (MICC) where ice nucleation was probed between -20 C to -28.6 C at water saturation mimicking mixed-phase cloud formation conditions. Reference ice nucleation experiments employing ammonium sulfate and kaolinite particles were conducted. Under probed conditions only kaolinite particles initiated ice nucleation. The authors present a study of increasing interest, i.e. if and how organic, in particular, secondary organic aerosol (SOA) particles form ice in the atmosphere. This is an important topic and I am in support that new experimental results should be published.**
We are happy about and thank the reviewer for this positive assessment.

**However, I find that this manuscript lacks discussion of recent literature on ice nucleation and diffusion of SOA particles to set the new results in the right context. SOA ice nucleation or diffusion has been studied by several groups in recent years (among others, Wang et al., 2012, Ignatius et al. 2016, Mohler et al., 2008, Charnawskas et al., 2017, Price et al., 2015, Wagner et al., 2017, Lienhard et al., 2015, Kanji et al., 2017, Ladino et al., 2014, recent review by Knopf et al., 2018). These papers should be present in introduction and may be further discussed in other sections of the manuscript.**
We gave the introduction a major overhaul and also revised the discussion to include more references and compare to the referenced works.

**Furthermore, the SOA generation procedures may vary among this and other studies. This should be mentioned/discussed in places.**
We agree that the SOA generation procedures can vary substantially among different studies. We mentioned this fact and included mentioning/discussion in the respective sections (i.e. mainly introduction and discussion).

**I find the supplemental material should be better implemented within the main text. As is, there are some notes to it, but the supplement has a lot of important information. I feel the bounce experiments would be better situated in the main text, also to be more visible, but I leave this to the authors.**
Following the reviewers suggestion, we moved the bounce experiments into the main text as new Section 3.1 and slightly adapted the text due to its new position.

**The figures in text and supplement reporting ice nucleation experiments should also include the supersaturation of ice, Sice. This is crucial information missing.**
Unfortunately, the TDL system was malfunctioning during the experiments, thus only the CR4 provided humidity measurements. However, the CR4 can only sample at ambient pressure, which means, there are no humidity measurements during cloud activation runs. We provide initial $S_{ice}$ (i.e. measured just before a cloud activation run) for each cloud activation run. Due to the phase changes from water vapour into the particle phase, simulation of $S_{ice}$ would yield high uncertainties; therefore, we do not provide these. However, we included the initial $S_{ice}$ measurements in the overview in Table 1.

**I recommend that the abstract states explicitly the particle systems investigated for ice nucleation.**

We added: "These are namely alpha-pinene, heptadecane, and 1,3,5-trimethylbenzene."

**p. 2, l. 10-15: other studies mentioned above in general comment should be mentioned.**

We have rewritten the introduction, and now it includes references to other studies as the ones you have mentioned above.

**p. 3, l. 24-25: The 33 m transfer line. You show data later but please elaborate on particle losses due to diffusion, gravitational settling etc. What is the flow speed and pressure in this transfer line? Since this is a new experiment, it would be beneficial to know these parameters.**

The flow speed and pressure in the transfer line are governed by the pressure (difference) in MICC and MAC. At the beginning of a transfer, the pressure in the transfer line close to MAC will be close to the pressure in MAC, while at the end close to MICC, it will be rather at MICC pressure. As the pressure difference reduces over time (when MICC is filling) the flow speed decreases. A typical flow speed time series for a refill from MAC after a MICC cloud activation is shown below.
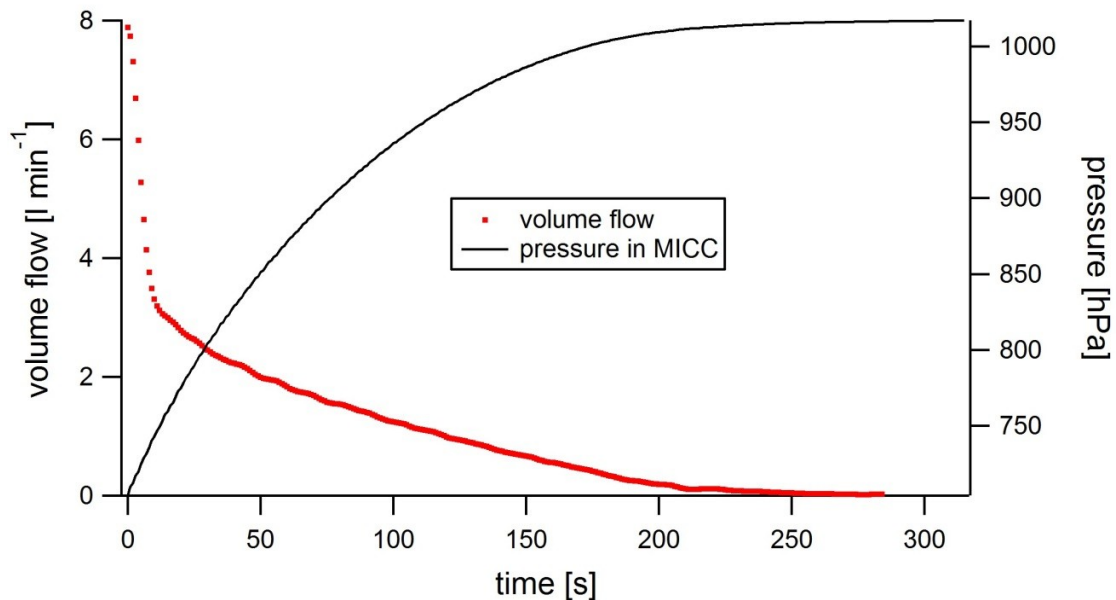


Figure: Volume flow in transfer pipe during refill of MICC from MAC after a cloud activation.

The comparison between DMPS size distribution measurements from MAC just before transfer and SMPS size distributions measured in MICC just after the transfer can be used to infer how severe particle losses during transfer are: The figure below shows the respective size distributions for the different SOA experiments. In case of the alpha-pinene SOA, no significant change in the main particle mode diameter is apparent, while for the first heptadecane experiment and the TMB experiment growth in the main particle mode happened, possibly in MICC due to the colder temperatures there compared to Mac temperature which fosters the condensation of the organic vapours onto the particles.
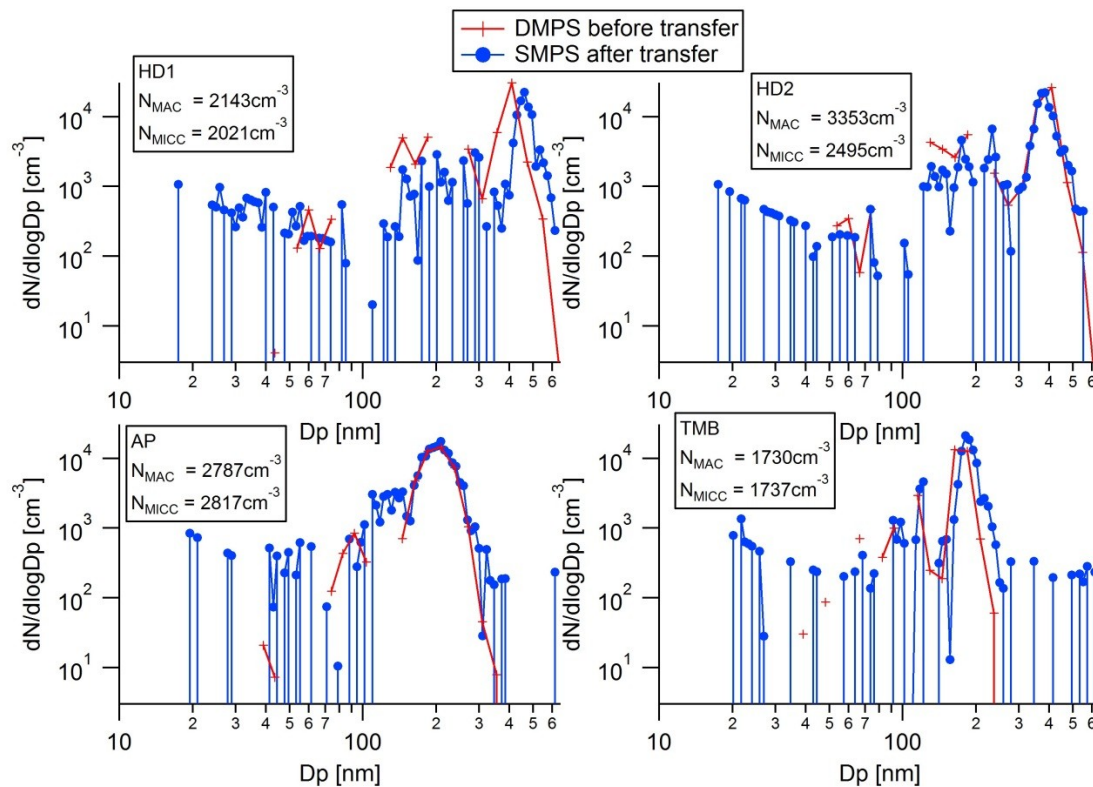
Figure: Comparison of size distributions just before transfer in MAC and just after transfer in MICC.

**p. 5, l. 4-8: Here, I would give the bounce experiments more exposure. "Bouncy" is not really a physical parameter, is it possible to use phase state definitions, such as semi-solid, solid etc.?**

We moved the section about the particle bounce measurements into the main manuscript at the end of the introduction part of section 3 and modified it slightly to adapt to the new position.

We agree with the reviewer regarding the phase state definitions and use phase state definitions (semisolid or liquid) instead.

**p. 5, l. 19: Here, the reader learns the first time the NOx is involved in SOA formation. This can be different from above mentioned studies. What does this mean for SOA composition, viscosity etc.? This could be important but is not discussed.**

The effect of $NO_x$ on SOA viscosity is not studied (at least the authors are not aware of any publications on this). It should be noted though, that also the O:C of produced SOA was relatively low in these experiments. As shown in Pajunoja et al. (2015) the particle viscosity (or bounce behaviour) at elevated RHs depends strongly on O:C. This is because the particle hygroscopicity (i.e. the particle phase water content at certain humidity) increases with O:C. Hence, not only the $NO_x$ might affect the composition or viscosity, but also the oxidation conditions more generally.

The use of $NO_x$ in these experiments was to help initiate the photochemistry, and the work did not include a systematic investigation of the effect of $NO_x$ on particle composition or properties.

We added this discussion to the manuscript.

**p. 5, l. 26 – p. 6, l. 8: Some details are not entirely clear to me: The SOA from MAC flows into evacuated MICC. Then MICC is filled with gas. Do you expect losing**

Correct, the evacuated MICC is refilled from MAC until ambient pressure is reached. Thus, there is no further dilution of the aerosol population other than by the air that remained in MICC prior to the transfer (it is not possible to evacuate MICC to vacuum, but the lowest pressure is 200hPa). As stated in the text, about $8m^2$ air from MAC (which holds the aerosol) is diluted by the remaining $2m^2$ of filtered air in MICC. The dilution leads to a tendency for components to evaporate. The transferred air cools in MICC and cooling leads to a tendency for the components to condense. Thus, there are opposing tendencies and any semi-volatile component in the aerosol will have a tendency to transfer between phases accordingly. The rate at which the components follow this tendency may possibly be influenced by the changes in condensed phase properties with temperature, since an increase in viscosity towards or across the glass transition may lead to a decrease in condensed phase diffusion and decrease in rate of bulk equilibration.

Effects of dilution and cooling can be investigated to a certain amount by comparing the SMPS and CPC measurements from directly following the transfer of air from MAC to MICC (when MICC temperatures were above the target temperature) and the repeated measurements once the target temperature was reached. For example, looking at the SMPS size distributions given in Figures 3 and 4 (and others in the supplement), which shows the size distribution directly "after transfer" in red and the two size distributions sampled when the target temperature was reached "S1/S2 before evacuation" in blue colours, no significant changes are apparent. Thus, it seems that enough organic vapour remained in the gas phase in MAC prior to transfer such that particles would not evaporate/shrink when entering MICC even if amounts of the vapour are lost to condensation on the chamber walls.

We added some explanation to the manuscript.

The exact RH trajectories are not known. We do have information about the initial RH in MAC and the RH in MICC after transfer. Due to the not-ambient pressure it is not possible to measure the RH during transfer.

MICC walls are ice coated from the humidity that entered the chamber during the cleaning cycles. Thus, any humidity in excess of $Rh_{ice}$ = 100% would condense onto the walls after the transfer. However, this process will take some time, following a rough calculation assuming a gradient in saturation ratio of 0.15 over 0.5m (half the diameter of MICC), molecular diffusion of water vapour is really small and the supersaturation would stay there for maybe 1.5 hours. There will be turbulence in the chamber too (from the air stream filling the chamber), which should reduce this time, and the presence of particles (that have their own water content, eventually subsaturated wrt liquid water) will prolong the diffusion, too. Generally, RH can have an impact on the phase state of the SOA particles. From the bounce measurements presented here, we can estimate that even at low RH (and thus at high RH as well) the SOA particles from heptadecane precursor will be liquid, while TMB is in a semi-solid phase even at high RH.

**p. 6, l. 30-31: Here and Fig. 4 case: Are activated droplet sizes what would be expected from Kohler theory and diffusional growth?**

Generally, yes! However, the size distribution will be broader than theory predicts due to non-uniformities in the temperature and humidity fields in the chamber (see also reply to comment "p. 8, l. 16-21" below).

**p. 7, l. 27-35: For this discussion it is crucial to know also the temperature and $S_{ice}$ values during measurement of the activated fraction. At this point the discussion is confusing and one wonders about these results. Maybe at fast pumping speed, i.e. at high $S_{ice}$, the activated fraction of HD is not as sensitive compared to lower pumping speed and thus lower $S_{ice}$?**

We modified Figure 6 which now also includes the temperature measurements during the cloud expansions and the initial $S_{ice}$. Due to the failure of the TDL system, we unfortunately, only have humidity measurements at ambient pressure, thus, not during the cloud expansion experiments. We included statements about the temperatures and initial $S_{ice}$ in the manuscript.

"In the slower pump speed experiment, however, the second heptadecane run shows a higher activated fraction than the first run, though initial ice supersaturation are the same and tepmeratures in both runs are within 0.2°C. Thus, the aerosol becomes more efficient at activating to droplets. The first run here exhibits lower activated fractions as the fast pump speed runs, the second run peak activated fraction is about the same as in the fast pump speed runs. The α-pinene slow pump speed experiment shows the opposite behaviour, the second cloud run has slightly lower activated fractions as the first. The same is true for the TMB slow pump speed runs. However, the initial temperatures differ by about 0.7°C resulting in a less strong temperature drop during expansion, and also the initial ice supersaturation differs."

**p. 8, l. 16-21: Is it possible to make this speculative discussion a bit more quantitative?**

We expanded this discussion:

"... it will take time for the cloud to extend downwards to the bottom and be sampled, see also discussion in Möhler et al. (2003) for similar effects in the AIDA cloud chamber.

The observed cloud particle size distribution is wider than the simulated one. This could be due to effects of non-uniformity in temperature and humidity that lead to broadening of the size distribution. For example, as the chamber walls will stay at close to their initial temperature while the chamber centre cools adiabatically during an expansion, wall heating effects can create a temperature gradient within the chamber (warmer towards the walls and colder towards the chamber centre), which will induce a humidity gradient as well. The incoming air stream causes mixing which on the one hand side can reduce this gradient, but on the other side can induce inhomogeneous temperature and humidity fields leading to variation in the activation and growth of the cloud particles."

**p. 8, l. 29-31: I find this too simplified and feel it needs more discussion. Please look at studies mentioned above in general comment.**

We made significant changes to this section of the discussion, where we refer to other studies, comparing those results and conditions used during SOA generation with our experiments.

**p. 9, l. 1-5: I find this needs more discussion. Please look at studies mentioned above in general comment. SOA particles are produced in different ways, different temperatures are probed etc.**

We expanded the discussion and made significant changes, see comment above.

**p. 9, l. 12-13: Different heptadecane properties due to different VOC/NOx ratio applied?**

As mentioned above, the effect of $NO_x$ on SOA viscosity is not studied. It is difficult to speculate on this given the lack of any specific work on the issue. Also, given the relatively small difference in VOC/ $NO_x$ ratios in our experiments, it is probably unlikely that this would be the main reason behind the difference in the bounce behaviour.

**p. 9, l. 14-15: Not necessarily higher supersaturation are needed but longer times. The need of larger supersaturation may be "apparent", i.e. disequilibrium between gas and condensed phase.**

We rephrased:

"Thus, it takes a longer time for these viscous SOA particles to take on water vapour and grow due to diffusion limitations."

**p. 9, l. 15-16: Indeed, different temperature in chambers or sampling lines can affect particle properties. See e.g. discussion in Knopf et al. (2018) and Ignatius et al. (2016).**

True, we expanded the discussion by adding "Indeed, an effect of different temperatures in the chambers or sampling lines had been found and discussed by e.g. Ignatius et al. (2016) and Knopf et al. (2018)." to the end of this possible explanation.

**p. 9, l. 28-30: Above mentioned literature may enhance this discussion.**

We expanded "Thus, the phase state of the aerosol particles (represented by their bounciness) could play a role in the onset of activation, as hinted in measurements by e.g. Ignatius et al. (2016); Ladino et al. (2014); Wagner et al. (2014)."

Furthermore, the discussion has been modified substantially, and we have referenced and discussed above mentioned literature in various places throughout the discussion now.

**p. 10, l. 5-6: Please refer to, e.g., Wagner et al. (2014, 2012) articles.**

We added at the end of this passage:

"Pre-cooling of SOA particles has the potential to increase the aerosol particles' ability to act as INP (e.g. Ladino et al., 2014; Wagner et al., 2014). Thus, in the respective second cloud activation runs, there could have been a higher chance of ice formation. However, apparently the minimum temperatures reached during the first cloud runs here were not cold enough for such a pre-activation of the aerosol particles and hence, no ice had formed."

**p. 10, l. 16-18: Heptadecane is more viscous and therefore the activated fraction is lower?**

Actually, based on the bounce measurements heptadecane is less viscous than alpha-pinene and TMB (mentioned p9, l13, see also bounce figure in supplement or Saukko et al., 2012). The heptadecane SOA behaves as liquid droplets even at dry RH.

**Figures: 3-5 and similar ones in supplement: specific for figures in main text: particle images are not described in caption. Panel indicators are missing. As stated above, please include Sice. It is confusing to have a legend in third panel that includes definitions for other panels. Please split legend to corresponding panels.**

The figures have been altered as suggested. As mentioned before, we unfortunately can only state the initial $S_{ice}$ for these experiments. Due to the formation of cloud particles, i.e. phase change of vapour to particles, a calculation of $S_{ice}$ would be hampered by high uncertainties. The initial $S_{ice}$ is now included in Table 1 (alongside initial $S_{water}$).

**Figure 5: What do you mean by "eating up"? A Wegener-Bergeron-Findeisen process? Please change expression.**

We rephrased: "The data show formation of ice in a second mode, and the decrease and almost disappearance of particle numbers of smaller drops over time (Wegener-Bergeron-Findeisen process)."

**Table 2: You mean MAC and not "aerosol chamber"? "Amount injected": unit? Please elaborate. Have aqueous solutions been injected? What is the mass? The mole fraction or other information is needed. Units missing for gas species. The mass difference, last column, between pinene and TMB is correct?**

MAC is the synonym for the Manchester Aerosol Chamber, thus, both would be correct. The gas species are in $nmol\,mol^{-1}$, we added that information to the table. The VOCs were injected as high purity chemical liquids into a heated glass bulb where it immediately evaporates. A continuous flow of nitrogen then flushes the gas phase VOCs into the aerosol chamber. (See description in original manuscript p3 l7-9 and p5 l18/19.) We now expanded: "To prepare the system for injection of relevant gases for particle formation in MAC the volatile organic compound (VOC) injection glass bulb is heated and continuously flushed with nitrogen. The precursors for the SOA are injected into the glass bulb in form of high purity liquids, where they evaporate immediately. The vapourised VOCs and $NO_x$ are then injected during the last filling of the MAC air bag."

As we use pure liquids, their mole fraction is unity. To avoid confusion we changed the way we state the amount of VOCs injected: As we calculated the amount of liquid for the volume injection, taking into account the chamber bag volume, we now state the "nominal VOC mass" that should enter the chamber as gas. We also made a reference to the text, Sect. 3.1 Experimental Design.

Since we injected precursor gases with very different characteristics, the resulting aerosol differs not only by mass but also by e.g. their nucleation and growth characteristics and losses to the walls. TMB is much harder to produce and therefore, masses are smaller than for alpha-pinene, which contrary nucleates much faster. There was indeed an error in the TMB mass, but still the TMB mass is much smaller than e.g. the alpha-pinene mass.

**Table 3: How can mass in MICC be larger than in MAC (mass_DMPS vs. mass_SPMS)?**

Due to the differing size ranges of the DMPS and SMPS, the mass retrieved from these instruments can be different even if sampled from the same aerosol population. As the SMPS starts measuring at 13.8nm (vs 40nm DMPS) it will add mass from the small particles. If you look at the comparison of size distributions (figure above in the reply to your comment on the transfer pipe), in the α-pinene and TMB cases also larger particles are observed by the SMPS, which are absent in the DMPS measurements. These contribute to the SMPS mass accordingly.

**Supplement: p. 1: change "cooking" to "processing" or other terminology.**

Changed to: "...for any experiment performed on specified aerosol photochemically produced in the aerosol chamber."

**Figure S4: Please include pinene SOA bounce fraction.**

Since the bounce measurements shown here were not from the same experiments but from a previous experiment, there were unfortunately, no measurements of pure alpha-pinene SOA. However, for example Saukko et al. (2012) and Pajunoja et al. (2015) did show by flow tube measurements that alpha-pinene is semi-solid. We state more clearly now in the main text that the bounce measurements and the ice nucleation

measurements were not made in the same experiment but the bounce measurements were part of an earlier experiment using the same set up as our measurements.

**p. 4: Change "ingredients" to "species" or "compounds", etc.**
Changed to: "...all chemical substances as in a normal SOA experiment were used, without the actual precursor."

**What is the difference between S17 and S19 and S18 and S20 experiments? Maybe additional text is necessary?**
Yes, it seems additional text should be provided. There were two experiments with ammonium sulfate aerosol, with two cloud activation runs each. Thus, the difference between S17 and S19/S18 and S20 is the actual experiment, S17/S18 are cloud activation runs from experiment 2, S19/S20 from experiment 9 (see also Table 1 in main manuscript). We included a reference to the respective experiment (and the table) for all cloud activation run figures in the supplement.


**Technical corrections:**
**I suggest throughout manuscript and supplement to change the expression "cloud evacuation" to "cloud activation experiment" or something along those lines.**
We changed according to the reviewer's suggestion.

**p. 7, l. 25: Change language. Avoid the term "sister run".**
Changed to: "...the already shown heptadecane [...] and dust [...] runs accompanied by the respective other runs in the same experiments, plus a further heptadecane experiment...".

**p. 8, l. 26: Maybe use "employed" or "applied" instead of "used".**
Changed to "employed".

**p. 9, l. 8: Avoid "kicks in". Change language.**
Changed to "commences".

**p. 9, l. 30: Exchange "than" with "as".**
Done.

**p. 9, l. 32: Instead of "sucking" use" pumping" or "evacuating".**
Changed to "pumping".

**p. 10, l. 6-7: It feels there is an error in this sentence.**
It is not clear where the reviewer suspects an error. However, we added some information to the previous sentence and hopefully this will resolve the misunderstanding?
"Generally, cloud processing is thought to increase the efficiency of activation into cloud particles (e.g. Hoose et al., 2008), through changing the internal chemical structure and/or composition of the aerosol particles. However, as the experiments reported here exhibit pure SOA aerosol, and we expect no other organic and inorganic material (or vapours) in the chambers, cloud processing here will only change the aerosol mass, not aerosol chemistry."

**p. 10, l. 30: Exchange "no" for "not".**
Done.

**p. 11, l. 22: Exchange "bump" to "maximum" or similar.**

Rephrased sentence:

"The sharp increase in cloud particle numbers at 20 seconds (i.e. the major activation) coincides with the departure in the corrected temperature curve from the adiabatic curve."

*References:*

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Wagner, R., Möhler, O., Saathoff, H., and Schnaiter, M.: Enhanced high-temperature ice nucleation ability of crystallized aerosol particles after pre-activation at low temperature, J. Geophys. Res. Atmos., 119, 8212-8230, doi:10.1002/2014JD021741, 2014.