Anonymous Referee #2
Referee comment on "High Homogeneous Freezing Onsets of Sulfuric Acid Aerosol at Cirrus Temperatures" by Julia Schneider et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-319-RC2, 2021

We thank referee #2 for his or her thoughtful comments and feedback. Please find below our responses and suggestions for the manuscript revision, with the referee comments in black, our answers in green, and suggested changes or additions to the manuscript in blue.

General Comment

This manuscript titled "High Homogeneous Freezing Onsets of Sulfuric Acid Aerosol at Cirrus Temperatures" by Schneider et al. reports homogeneous freezing of aqueous sulfuric acid aerosol using the AIDA chamber at conditions relevant for cirrus cloud regime. The highlight of this study is the significant deviation of the onsets of ice formation from the water activity criterion (WAC) (Koop et al., 2000) at temperatures below about 200 K. The manuscript is well-written, clearly discussing the uncertainties in their measurements and the underlying assumptions.
However, the discussion/conclusions rely entirely on a single type of aerosol particle, with the absence of any physical interpretation for deviations from WAC, and in-depth discussion of mixing time of investigated aerosol particle at temperatures below 200 K. Nonetheless, I recommend the manuscript for publication after the authors have addressed the following major questions and revised the manuscript accordingly.

Major Comments

1. Kinetics of sulfuric acid aerosol particles at cirrus temperatures: I understand that not much work has been done related to the activation kinetics and particle phase at such low temperatures. The authors handled this issue with their test for kinetic limitations at 197 K. However, the majority of their ice supersaturation results still comply with the WAC at around that temperature. I am curious why a similar kinetic test was not considered at, e.g. 190 K, where the deviations are suggested to be significantly higher?

Please note that the experiment shown in Fig. 5 was started at 197 K, but the actual ice onset temperature was observed at a lower temperature of about 193 K with a corresponding ice saturation ratio of about 1.95. Therefore, this experiment was done in the temperature regime, where the observed deviation in onset supersaturation is already significant and we consider it as representative for the other experiments with significant deviation. We are planning more AIDA homogeneous freezing experiments with other solutes and will take up your suggestion to carry out more experiments with variable pumps speeds or rate of pressure change.

2. Thermodynamic equilibrium: Based on previous work (Williams and Long, 1995), it won't be surprising that these sulfuric acid aerosol particles are extremely viscous at such low temperatures, likely approaching glassy state (if not already). This could really slow down the mixing. This casts some doubts on whether the particles attained thermodynamic equilibrium condition. I strongly recommend adding a discussion related to this issue and the implications it can have on the presented results and its interpretation.

We agree that the particle phase and the related mixing time at low temperatures are an important aspect to consider in the measurements of this study. We also discussed those aspects in the response to major comment 3 of referee #1, which is attached below:

As a first step, we included the start relative humidity with respect to ice $RH_{i,0}$ and liquid water $RH_{w,0}$ to the tables in the appendix, as suggested.

To illustrate the start conditions and the sulfuric acid phase at the ice onset, we adapted Fig. 4 in Koop, 2004 and added the data points for the start composition of the sulfuric acid aerosol particles in wt% H_2SO_4 for our experiments and the data points for the composition at ice onset (see Fig. 1 in this response, red and blue dots). The weight percentage composition was determined by using Model I of the E-AIM model with the measured temperature and relative humidity inside the chamber. As described in the referee's comment above, a glassy state of the sulfuric acid aerosol particles in our experiments would strongly influence the observed freezing process due to slow mixing processes. In Fig. 1, we can see that all experiments are above the conditions of ultra-viscous and glassy particles, according to the phase diagram in Fig. 4 in Koop, 2004 and the references therein. Especially the glass transition conditions given in this figure are at significantly lower temperatures compared to our starting and ice onset conditions. This is supported by our SIMONE measurements, which show an increasing signal in the forward scattering intensity between pump start and ice onset, for all the experiments. This increase shows that the aerosol particles are able to take up water and dilute. Only from this observation, it cannot be completely ruled out that an enhanced viscosity at low temperatures decelerates the water uptake, so that it might be not sufficient to maintain particles in thermodynamic equilibrium with the environment. For the investigation of a potential decelerated water uptake, we refer again to the experiment shown in Fig. 5 in the manuscript. In this experiment, the relative humidity was kept nearly constant above Koop2000 for about five minutes. Firstly, we observe no ice formation in this time period, which indicates that the AIDA ice onset is not higher than the Koop2000 line due to a delayed ice detection. Secondly, the forward scattering intensity of the SIMONE instrument (dark red line, third panel) shows a significant increase in the first two minutes of constant pumping, which is related to the water uptake of the particles. In the following five minutes of constant relative humidity (slower pumping), no further uptake of the particles is observed. To illustrate this more precisely, we added a second panel to Fig. 5 in the manuscript (see Fig. 2 in this response), which shows a direct comparison of the relative humidity and the forward scattering intensity on a smaller scale. In this panel, we clearly observe the particle diluting in the first two minutes of the experiment, which generally decreases the particles' viscosity. After that period, the particles could have continued taking up water, but no further increase in the forward scattering is observed, as soon as the pump speed is lowered. Rather, the forward scattering intensity follows nicely the slight variations in the relative humidity, but a decelerated or delayed water uptake is not indicated.

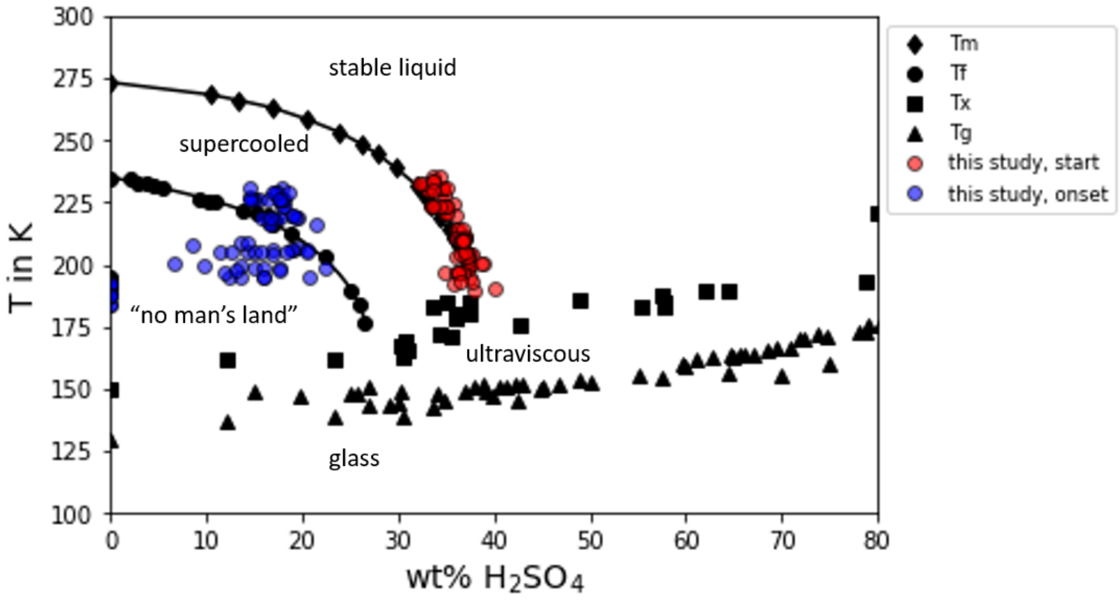

Fig 1: Phase diagram of sulfuric acid adapted from Koop (2004). The phase diagram of sulfuric acid as given by Koop (2004) and references therein is shown and complemented by the calculated weight percentage concentrations of the sulfuric acid aerosol particles

before the start of the AIDA experiments of this study (red dots) and at the observed ice onset during the experiments (blue dots).

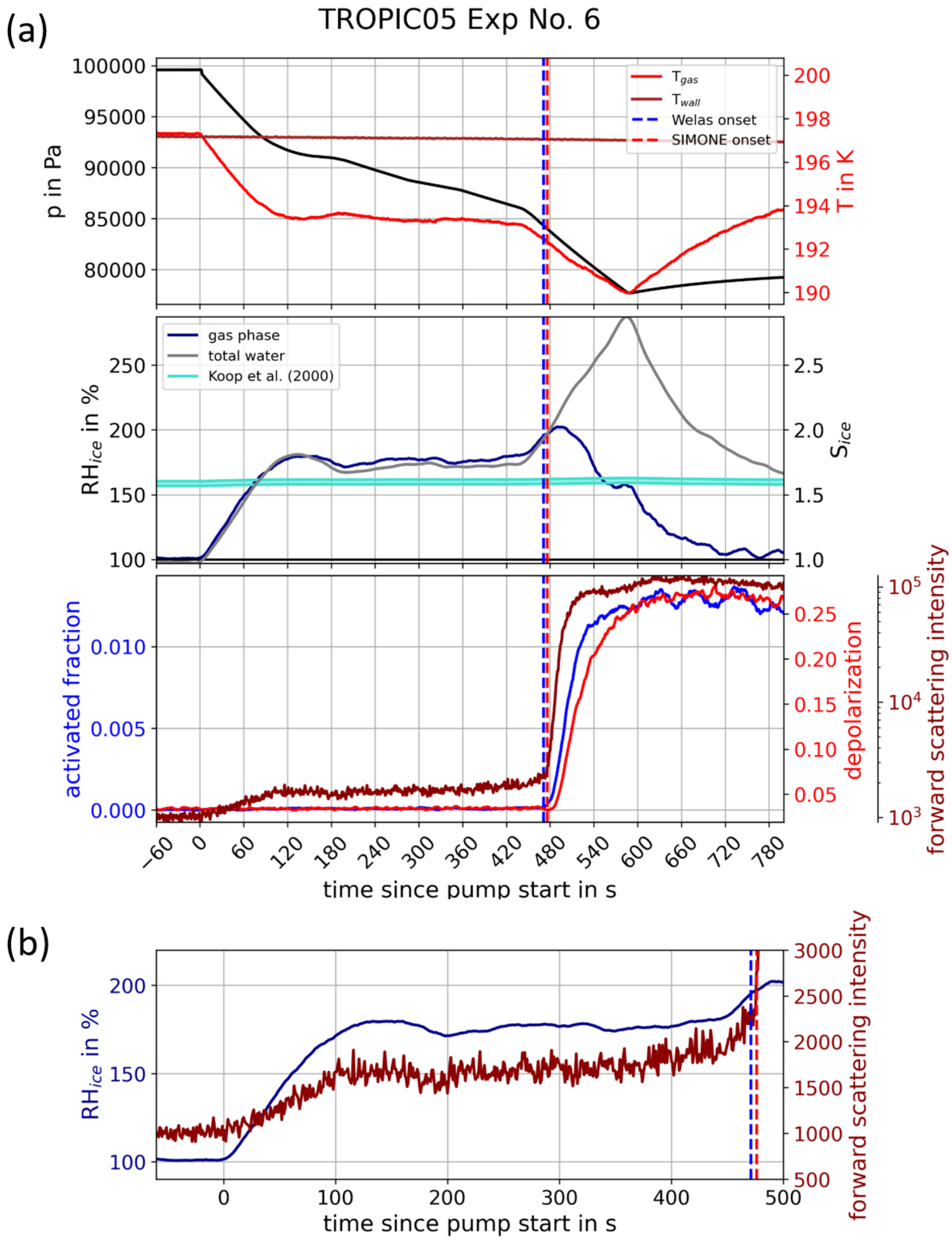

Fig. 2: Investigation of kinetic limitations with respect to water uptake in the AIDA chamber. Panel (a): The figure is composed in the same way as Fig. 3. For the experiment shown, started at about 197K, the pump rate was controlled in such a way that the relative humidity with respect to ice stayed relatively constant for about 5 minutes at about 170%, hence above the homogeneous freezing threshold suggested by Koop et al. (2000). For details on this experiment, see Sect. 3. Panel (b): Enlarged view

of the relative humidity and the forward scattering intensity at the beginning of the experiment.

To our knowledge, studies or measurements of the viscosity of sulfuric acid aerosol particles at very low temperatures are not available in the literature. Measurements and suggested fits for viscosity in dependence on temperature and H_2SO_4 weight percentage concentration by Williams and Long (1995) extend to 200 K and are given for any concentration between 30 and 80 wt%. Fig. 1 in Williams and Long (1995) shows that the viscosity is strongly increasing with decreasing temperature. As the AIDA experiments have weight percentage concentrations < 30% at the ice onset, we can only use the Williams and Long parameterization to determine the viscosity before experiment start when wt% > 30% and for experiments starting at temperatures > 200 K. In Fig. 3a in this response, we show the calculated viscosities η using the Williams and Long parameterization for the AIDA experiments at experiment start (black dots) with start temperatures > 200 K. The viscosities for experiments starting at temperature < 200 K are shown in the grey shaded box, as they need to be treated with caution. In addition, we used these viscosities to determine the diffusion coefficient D of water molecules in the sulfuric acid solution aerosol particles using the Stokes-Einstein-equation (red triangles). With the diffusion coefficients, the diffusion length D_L for a water molecule on a time scale of 6s is calculated and shown in Fig. 3b in comparison to the measured mean diameter of the H_2SO_4/H_2O aerosol particle populations. The diffusion length is significantly larger than the mean particle diameter in all experiments, which indicates that the diffusion of water in the aerosol particles is fast enough to keep them in thermodynamic equilibrium with the environment. Note again that these calculations are only strictly valid for experiments starting at temperatures > 200 K and only for the experiment start conditions.

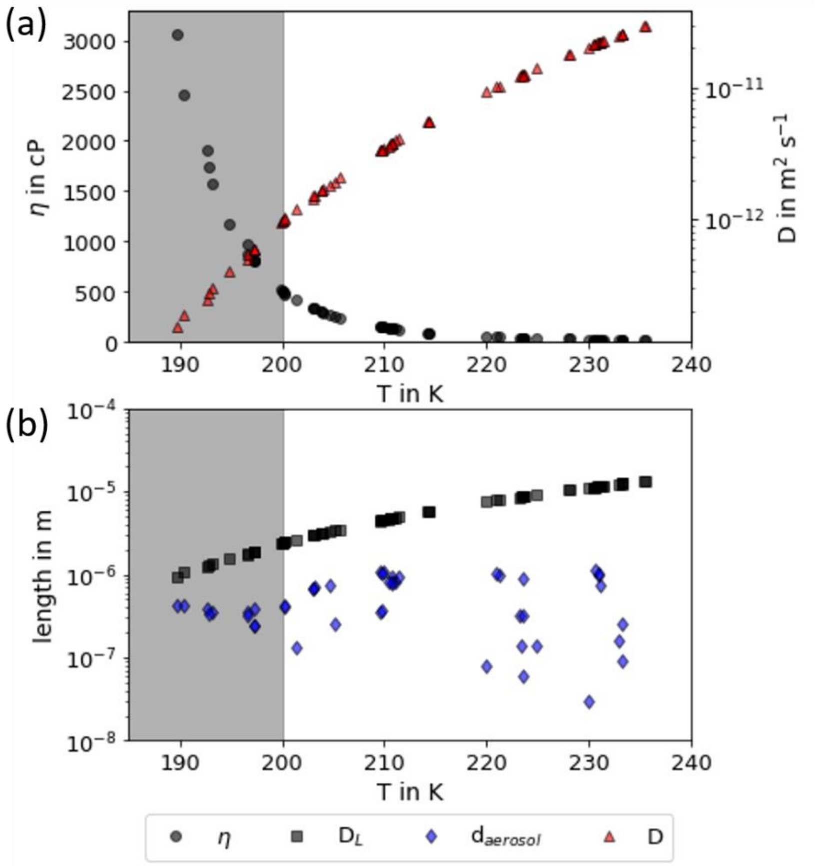

Fig 3: Viscosity and diffusion coefficients for the start conditions of the AIDA experiments. Panel (a): Viscosity (black dots) and diffusion coefficients (red triangles) calculated using the parameterization of Williams and Long (1995) and the Stokes-

Einstein-equation. Panel (b): Comparison of the mean particle diameter of the sulfuric acid particle population (blue diamonds) to the diffusion length of water molecules (black squares) on a time scale of 6s. Note that the shown data points represent the starting conditions of the AIDA experiments. As the parameterization of Williams and Long (1995) is only valid for T > 200 K, the data points of experiments at lower temperatures are displayed in a grey shaded box.

We added a similar discussion to Section "3. Results and discussions" and added the Figs. 1 and 3 to the appendix, as additional information, and changed Fig. 5 in the manuscript by adding panel b (see Fig. 2 in this response). Changes in the manuscript are as follows:

For lower temperatures, the viscosity of sulfuric acid increases with decreasing temperature (Williams and Long, 1995) and it can even undergo a transition to a glassy state (Koop, 2004). An enhanced viscosity or a glassy state may reduce the water uptake of the aerosol particles and therefore may delay or inhibit particle growth and dilution. Such effects were discussed to e.g. inhibit homogeneous freezing of secondary organic aerosol particles at T < 200 K (Fowler et al., 2020). For aqueous sulfuric acid solutions, Koop (2004) summarized the conditions for transitions to ultra-viscous and glassy states in dependence on temperature and aerosol composition. To get information about the phase state of the H_2SO_4/H_2O aerosol particles in the AIDA experiments, we used Model I of the E-AIM model (Clegg et al., 1992; Carslaw et al., 1995; Massucci et al., 1999; Wexler and Clegg, 2002; Clegg and Brimblecombe, 2005) to calculate the sulfuric acid weight percentage for the temperature and humidity conditions before experiment start and at the observed ice onsets (see Tables B1, B2 and B3). For all experiments, the calculated weight percentages are above the values for a transition to ultra-viscous or glassy particles according to Koop (2004) and references therein (see Fig. D1). In particular, the glass transition occurs at significantly lower temperatures compared to the AIDA starting and ice onset conditions. This is also supported by the aerosol particle forward scattering intensity measurements with the SIMONE instrument, which show an increasing signal between pump start and ice onset for all the experiments. This increase shows that the aerosol particles take up water and dilute, which would not be expected for glassy particles. To calculate the viscosity of the H_2SO_4/H_2O aerosol particles in the AIDA chamber, we applied the parameterization by Williams and Long (1995). This parameterization is only valid for temperatures > 200 K and for a H_2SO_4 weight percentage between 30 and 80 wt% and can therefore only be applied to the starting conditions of AIDA experiments with T_0 > 200K (see Fig. D2, data points for experiments with T_0 > 200K are displayed in a grey shaded box). The Stokes-Einstein-equation was then used to calculate the diffusion coefficient of water molecules in the aqueous sulfuric acid aerosol particles. From this, we then calculated the diffusion length of water molecules in the aerosol particles on a time scale of 6 seconds, which is much larger than the mean aerosol diameter. This indicates that the water diffusion is still fast enough to keep the aerosol particles in thermodynamic equilibrium.

[…] As soon as the relative humidity is kept at a constant value, no further increase in the forward scattering intensity was observed. **Fig. 5b shows the relative humidity and the forward scattering intensity at the beginning of the experiment. The forward scattering intensity clearly increases during the first two minutes of the experiment, which is caused by water uptake as a result of increasing relative humidity, and results in a decreasing viscosity of the particles. As soon as the relative humidity is controlled to an almost constant value by the reduction of the pump speed, no further increase in the forward scattering intensity is observed. The signal even follows the slight variations in the relative humidity, and a delayed water uptake is not indicated.** This supports our assumption that the aerosol particles can well follow thermodynamic equilibrium conditions during an AIDA experiment at typical pump rates and related rates of cooling and relative humidity increase.

Figure descriptions in the appendix:

Figure D1. Phase diagram of sulfuric acid adapted from Koop (2004). The phase diagram of sulfuric acid as given by Koop (2004) and references therein is shown and complemented by the calculated weight percentage concentrations of the sulfuric acid aerosol particles before the start of the AIDA experiments of this study (red dots) and at the observed ice onset during the experiments (blue dots).

Figure D2. Viscosity and diffusion coefficients for the start conditions of the AIDA experiments. Panel (a): Viscosity (black dots) and diffusion coefficients (red triangles) calculated using the parameterization of Williams and Long (1995) and the Stokes-Einstein-equation. Panel (b): Comparison of the mean particle diameter of the sulfuric acid particle population (blue diamonds) to the diffusion length of water molecules (black squares) on a time scale of 6s. Note that the shown data points represent the starting conditions of the AIDA experiments. As the parameterization of Williams and Long (1995) is only valid for T > 200 K, the data points of experiments at lower temperatures are displayed in a grey shaded box.

Figure description of Fig. 5:

Figure 5. Investigation of kinetic limitations with respect to water uptake in the AIDA chamber. **Panel (a):** The figure is composed in the same way as Fig. 3. For the experiment shown, started at about 197 K, the pump rate was controlled in such a way that the relative humidity with respect to ice stayed relatively constant for about 5 minutes at about 170%, hence above the homogeneous freezing threshold suggested by Koop et al. (2000). For details on this experiment, see Sect. 3. **Panel (b): Enlarged view of the relative humidity and the forward scattering intensity at the beginning of the experiment.**

3. Physical interpretation of the deviations from WAC: There is no attempt made at understanding the microphysical picture underlying the reported deviations from WAC. It is unclear why sulfuric acid system would behave in such a fashion. I recommend that the authors state this lack of understanding clearly in the manuscript.

First of all, we would like to state that we present here experimental results after careful discussion and uncertainty analysis, with direct measurements of the temperature and ice saturation ratio at the point of homogeneous freezing onset for aqueous sulfuric acid aerosol particles. That our data deviates from those derived from the WAC-based parameterization by Koop et al. (2000) is an experimental result, and does not require any physical reasoning. Physical reasoning is important and may be helpful when discussing possible reasons for this disagreement, and some are included in the discussion in section “3. Results and discussion”, but unfortunately we do not yet have a definite explanation for the observed deviation.

We therefore included the following statement into Section “3. Results and Discussion” to emphasize that we are not questioning the underlying physical principles of the WAC, but rather its application to the AIDA parameter space at low T:

Finally, we do not have a solid explanation for the deviation of the H_2SO_4/H_2O homogeneous freezing thresholds observed in AIDA experiments from the WAC-

based homogeneous freezing lines. This deviation may be related to uncertainties in the formulation of physicochemical properties at low temperatures, which are required for the conversion between the Koop2000 parameter space (a_w-T-space) and the AIDA parameter space (S_{ice}-T-space), as described above.

4. Atmospheric models: Sulfuric acid aerosol particles certainly dominate in the stratosphere, however, there are several studies showing the presence of various other components such as organics and inclusions of aluminum and silicon (Murphy et al., 2014), with even more variety of components present in the upper troposphere. While the presented study focuses only on sulfuric acid aerosol particles. Keeping this in mind as well as the comments stated in 1-3 above, I find the authors' suggestion to use their parameterization in atmospheric models over WAC a bit far fetched. I do not agree with this suggestion and recommend removing this part. In addition, the authors should state this caveat in their abstract, discussion and conclusions. There needs to be more work done on other atmospherically relevant aerosol particles at such low temperatures and high ice supersaturation conditions, to establish whether the deviations from WAC reported here are universal or not.

We understand that a replacement of the WAC and the application in atmospheric models should not be suggested, as it is also discussed in the response to major comment 4 of referee #1. We changed the related sections in the manuscript, as follows:

Abstract:

Based on **the**~~our~~ experimental results **of our direct measurements**, we suggest a new fit line **to formulate** ~~as a parameterization for~~ the onset conditions of homogeneous freezing of sulfuric acid aerosol particles **as an isoline for nucleation rate coefficients between $5 \cdot 10^8$ $cm^{-3}s^{-1}$ and 10^{13} $cm^{-3}s^{-1}$**. ~~As a next step, we propose the new parameterization to be implemented in atmospheric models as an improved version of the WAC-based parameterization from Koop et al. (2000). The new~~ **The potential significant impacts of the higher** homogeneous freezing thresholds, **as directly observed in the AIDA experiments under simulated cirrus formation conditions,** ~~may have significant impacts~~ on the **model** prediction of cirrus cloud occurrence and related cloud radiative effects **are discussed**.

Results and Discussion:

Based on the AIDA results and the discussion above, we ~~suggest a new parameterization~~ **provide a new fit line** for homogeneous freezing **of H_2SO_4/H_2O aerosol particles directly measured as function of** S_{ice} and T.

~~In a next step, this fit is constrained to the well-known homogeneous freezing temperature of pure water droplets, so that when applied to atmospheric models it matches the freezing point of pure water droplets (see Fig. 6c and d, red line).~~ (The section about the constrained fit was completely removed.)

~~We suggest to use the constraint fit with the coefficients a = −0.75±0.04 and b = 263±8 K(see Eq. 1 and 6c and d, red line) for use in atmospheric models.~~ **Applying** ~~T~~**t**his **new freezing threshold for cloud formation processes in the atmosphere,** ~~would then result in a shift of~~ the homogeneous freezing onset in cirrus formation **would be shifted** to ice saturation ratios of about 2.0 at temperatures around 185 K. A higher homogeneous freezing onset may explain the high ice saturation ratios **occasionally** reported for low temperatures in the upper troposphere in some field studies (Jensen et al., 2005; Lawson et al., 2008; Krämer et al., 2009; Krämer et al., 2020). **However, it needs to be considered that the fit line only describes homogeneous freezing of H_2SO_4/H_2O aerosol particles under laboratory conditions. Other aerosol species,**

which could be relevant for homogeneous freezing processes in the atmosphere, are not taken into account. Application to atmospheric conditions therefore needs to be done with caution.

Consequently, a precise description of homogeneous freezing processes is crucial to understand cloud radiative effects in the present climate as well as in predictions of climate change. ~~The new parameterization suggested here may be used as a replacement of the Koop2000 homogeneous freezing onset lines.~~ This may in particular be relevant for cirrus clouds in the cold tropical tropopause layer
(TTL). (This section was moved from Section "4. Conclusions" to Section "3. Results and Discussion")

Conclusions:

A higher homogeneous freezing onset as derived from our experiments may also explain field observations of high clear-sky supersaturation, which should not occur according to the freezing thresholds predicted by Koop2000 (Jensen et al.,2005; Lawson et al., 2008; Krämer et al., 2009; Krämer et al., 2020). **However, the discussed high freezing onsets only consider homogeneous freezing of H_2SO_4/H_2O aerosol particles under laboratory conditions, without involving other atmospherically relevant aerosol species.** ~~We suggest a new parameterization for the homogeneous freezing onset of H2SO4/H2O aerosol particles, which is based on~~ **A**~~a~~n empirical fit to the AIDA results with the form $\ln(S_{ice}) = a + 1/T \cdot b$ with fit parameters a = ~~−0.75±0.04~~ **−1.40±0.05** and b = ~~263±8~~ **390±10** K **describes the observed homogeneous freezing onsets of H_2SO_4/H_2O aerosol particles in the chamber**. **This fit line is based on direct measurements of the freezing onset conditions at simulated cirrus formation conditions and provides an isoline for homogeneous freezing of sulfuric acid aerosol particles for nucleation rate coefficients between $5 \cdot 10^8$ $cm^{-3}s^{-1}$ and 10^{13} $cm^{-3}s^{-1}$.** ~~This fit is constrained to the homogeneous freezing temperature of pure water at 235K and water saturated conditions at this temperature.~~ **The application of this fit line to atmospheric conditions requires further work on the physical behavior of H_2SO_4/H_2O aerosol particles at low temperatures and on the involvement of other atmospheric aerosol particle types.** Ongoing experiments in the AIDA cloud simulation chamber aim at investigating homogeneous freezing onsets of different solutes and at constraining the descriptions for liquid water saturation pressures to experimental results.

Minor comments:

Figure 4: Could the uncertainties in predictions from Koop et al. (2000) be added in this figure for clarity.

In Koop, 2004, the uncertainty range for the freezing line in the a_w-T-space is given with 5% uncertainty in the water activity. We included this range of uncertainty in Figs. 4 and 6, as a shaded area around the WAC freezing curves.

L6: WAC not a function of size of the aqueous aerosol particles. Please correct.

We changed the sentence in l. 6 as follows:
The WAC describes the homogeneous nucleation rate coefficients only as a function of the water activity, ~~temperature and size of the aqueous aerosol particles,~~ which makes this approach well applicable in numerical models.

L255: Please mention/reference the specific E-AIM models used here and for evaluating the data from other studies shown in Fig. 1

We used the Model I. We added the correct references to this line, as suggested on the E-AIM website:
In cases where water activities or relative humidities were not given by the authors, we used the Extended AIM Aerosol Thermodynamics Model (E-AIM)**, Inorganic Model I** on http://www.aim.env.uea.ac.uk/aim/aim.php **(Clegg et al., 1992; Carslaw et al., 1995; Massucci et al., 1999; Wexler and Clegg, 2002; Clegg and Brimblecombe, 2005)** to transfer given H_2SO_4 concentrations into water activities, which we assume to be equal to relative humidity.

Technical comments:

L8: "...laboratory-based homogeneous..."
Corrected.

L11: "Aqueous sulfuric acid aerosol particles of high purity were generated by..."
Corrected.

L47: "...to be equal to relative humilities."
Corrected.

L80: "...and the potential deviation from..."
Corrected.

L107: "...pump starts..."
Corrected.

L122: a period missing at the end of the sentence
Corrected.

Fig3. Caption: "...AIDA chamber as a function of time since pump start ..."
Corrected.

Fig6. Caption: "The fit shown in panel (a) is constrained..."
This sentence was removed completely due to other referee comments.

L353-54: "...freezing onset may explain the high ice saturation ratios..."
Corrected.

References

Koop, T., Luo, B., Tsias, A., and Peter, T.: Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, Nature, 406, 611-614, 2000.

Murphy, D. M., Froyd, K. D., Schwarz, J. P., and Wilson, J. C.: Observations of the chemical composition of stratospheric aerosol particles, Q. J. R. Meteorolog. Soc., 140, 1269-1278, https://doi.org/10.1002/qj.2213, 2014.

Williams, L. R., and Long, F. S.: Viscosity of Supercooled Sulfuric Acid Solutions, J. Phys. Chem., 99, 3748-3751, 10.1021/j100011a050, 1995.

References in author’s response:

Carslaw, K. S., Clegg, S. L., and Brimblecombe, P.: A thermodynamic model of the system HCl-HNO3-H2SO4-H2O, including solubilities of HBr, from <200 to 328 K, J. Phys. Chem., 99, 11 557–11 574, https://doi.org/10.1021/j100029a039, 1995.

Clegg, S. L. and Brimblecombe, P.: Comment on the “Thermodynamic dissociation constant of the bisulfate ion from Raman and ion interaction modeling studies of aqueous sulfuric acid at low temperatures”, The Journal of Physical Chemistry A, 109, 2703–2706,https://doi.org/10.1021/jp0401170, 2005.

Clegg, S. L., Pitzer, K. S., and Brimblecombe, P.: Thermodynamics of multicomponent, miscible, ionic solutions. Mixtures including un-symmetrical electrolytes, The Journal of Physical Chemistry, 96, 9470–9479, https://doi.org/10.1021/j100202a074, 1992.

Jensen, E. J., Smith, J. B., Pfister, L., Pittman, J. V., Weinstock, E. M., Sayres, D. S., Herman, R. L., Troy, R. F., Rosenlof, K., Thompson, T. L.,Fridlind, A. M., Hudson, P. K., Cziczo, D. J., Heymsfield, A. J., Schmitt, C., and Wilson, J. C.: Ice supersaturations exceeding 100% at the cold tropical tropopause: Implications for cirrus formation and dehydration, Atmos. Chem. Phys., 5, 851–862, https://doi.org/10.5194/acp-5-851-2005, 2005.

Koop, T., Luo, B., Tsias, A., and Peter, T.: Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, Nature, 406, 611–614, https://doi.org/10.1038/35020537, 2000.

Koop,T.: Homogeneous ice nucleation in water and aqueous solutions, Z. Phys. Chem., 218,1231–1258, https://doi.org/10.1524/zpch.218.11.1231.50812, 2004.

Krämer, M., Schiller, C., Afchine, A., Bauer, R., Gensch, I., Mangold, A., Schlicht, S., Spelten, N., Sitnikov, N., Borrmann, S., De Reus, M.,500and Spichtinger, P.: Ice supersaturations and cirrus cloud crystal numbers, Atmos. Chem. Phys., 9, 3505–3522, https://doi.org/10.5194/acp-9-3505-2009, 2009.

Krämer, M., Rolf, C., Spelten, N., Afchine ,A., Fahey, D., Jensen, E., Khaykin, S., Kuhn, T., Lawson, P., Lykov, A., Pan, L.L., Riese, M., Rollins, A., Stroh, F., Thornberry, T., Wolf, V., Woods, S., Spichtinger, P., Quaas, J., and Sourdeval, O.: A microphysics guide to cirrus – Part 2: Climatologies of clouds and humidity from observations, Atmos. Chem. Phys., 20, 12 569–12 608, https://doi.org/10.5194/acp-20-12569-2020, 2020.

Lawson, R. P., Pilson, B., Baker, B., Mo, Q., Jensen, E., Pfister, L., and Bui, P.: Aircraft measurements of microphysical properties of subvisible cirrus in the tropical tropopause layer, Atmos. Chem. Phys., 8, 1609–1620, https://doi.org/10.5194/acp-8-1609-2008, 2008.

Massucci, M., Clegg, S. L., and Brimblecombe, P.: Equilibrium partial pressures, thermodynamic properties of aqueous and solid phases, and Cl2 production from aqueous HCl and HNO3 and their mixtures, The Journal of Physical Chemistry A, 103, 4209–4226,https://doi.org/10.1021/jp9847179, 1999.

Wexler, A. S. and Clegg, S. L.: Atmospheric aerosol models for systems including the ions H+, NH+4, Na+, SO2−4, NO−3, Cl−, Br−, and H2O, J. Geophys. Res. Atmos., 107, https://doi.org/10.1029/2001JD000451, 2002.