## Dear editor,

we would like to reply to your three comments in the following paragraphs.

## Comment 1

As mentioned by referee #2, I agree that you cannot just exclude data for application of x\_therm that possess the same quality as the other data points. You may question your underlying model (x\_therm) that does not capture this particular effect. Also, the exclusion of these data is just given in the figure caption and is not mentioned/discussed in the main text which is not appropriate. This is particularly crucial since you emphasize that this is a case study for a new deposition ice nucleation description. I assume you would like to aim for a general validity of your approach. You can also not argue to use a different x\_therm function for this specific data point (since it is for same particles and same experiment). This further weakens your novel approach. I understand that even without these data your actives sites are only one order of magnitude off, so they are still reasonably represented. However, this is only known after the fact, may be coincidental for this specific case, and may be not generally valid (again, since you advertise this as a new method). In principle, leaving out data, would speak against the general validity of this approach, or?

We agree that indeed it would have been better to include all data points and we stated this omission of data points more clearly in the manuscript. However, we would also like to explain our selection of data points by referring to a study which observed a similar ice nucleation behavior close to water saturation and high temperature. Hiranuma et al. (2014) investigated the ice nucleation properties of hematite for a temperature range between 195 and 237 K, which overlaps with our temperature range.



Figure 2. The constant  $n_s$  magnitudes are joined by lines (blue) representing "isolines" of hematite freezing profiles in the *T* and RH<sub>ice</sub> space. The interpolated isolines are equally spaced at every order of magnitude from  $10^{12} \text{ m}^{-2}$  (top) to  $10^9 \text{ m}^{-2}$  (bottom). Experimental trajectories of AIDA expansion experiments with hematite particles are shown as red dotted lines. The data indicated with green color on the water saturation line represent the previously reported results of immersion freezing (Hiranuma et al., 2014). The subpanel shows a magnified section of *T* (-35 to -45 °C) and RH<sub>ice</sub> (110–120%) space with equidistant  $n_s$  spacing (every quarter magnitude). The error bars at  $n_s$  of  $10^{12} \text{ m}^{-2}$  are from welas.

It was found that ice nucleation initiated by hematite particles was observed to be strongly temperature dependent between 223 and 237 K, but not between 223 and 213 K (Fig. 2 in Hiranuma et al., 2014). Also, at temperatures below 213 K, deposition nucleation was observed to dependent on temperature again. These observations can be reconciled by the hypothesis that at higher temperatures and closer to water saturation, there are certain microphysical processes at the particle surface and/or perhaps even within the bulk phase which occur in addition to pure deposition nucleation. One of these mechanisms could be pore condensation freezing (Marcolli et al., 2014). This would mean that ice nucleation at temperature above 223 K would be governed by deposition nucleation and some additional contribution by pore condensation processes. Consequently, this would justify using separate x\_therm approaches for temperatures above and below 223 K because the curvature of the n\_s isolines above 223 K could be captured better by a linear x\_therm approach. For future studies, however, we strongly recommend to investigate deposition nucleation down to temperatures close to 200 K.

# We will add a comment reflecting these additional clarifications on p.6:

"Note also that xtherm as a linear function in humidity and temperature is assumed to be only strictly valid between 226 and 250 K. Other studies show that the ns isolines for deposition nucleation caused by materials such as hematite are strongly temperature dependent between 223 and 237 K, but not between 223 and 213 K (Hiranuma et al., 2014). Thus, these results suggest that different xtherm or other approaches might be needed within different temperature regimes. Also, deposition nucleation close to water saturation may coincide with pore condensation freezing (Marcolli et al., 2014)."

#### Comment 2

What is the source of this saturation and temperature parameterization? It somehow reminds one of the water activity criterion where also RH or RH\_ice and freezing temperature where combined into one parameter. However, in that case it was based on rather strict thermodynamic considerations derived from a phase diagram and the parameter is physical. This is not clear here, in addition to exhibiting a unit problem. The average temperature in the chamber is Kelvin, you cannot just make it unit less, or? Application of active sites approach is fine, but one could play devil's advocate saying "the active sites approach cannot capture the experimental data. So, a made-up parameterization is used to somehow get modelled/fitted active sites in agreement". You should elaborate here a bit more and make a case why this underlying active sites parameterization may be something one should consider for future deposition ice nucleation analyses. This new approach, I assume, is the main objective of this paper. As is, it significantly weakens your work.

Within our study, we wanted to test whether the ice nucleation active surface site density approach, which has successfully been used to describe immersion freezing, can also be used to describe deposition nucleation. Temperature dependent exponential functions (with T in Kelvin, i.e. being made unitless as well) have proven to be a suitable tool to describe immersion freezing. Analogously, we wanted to develop a description for deposition nucleation which hugely depends on the aerosol surface properties which makes expressing the ice nucleation efficiency by the surface site density a reasonable approach. In order to support the applicability of the ice nucleation active surface site density approach (i.e. the proportionality between ice crystal concentration and aerosol surface area), we also varied the aerosol size distribution of the ATD dust particles. For all cases, we get a good agreement between the ice fractions normalized by the total aerosol surface. Based on these very promising results, we strongly advise to test the applicability of the n<sub>s</sub> approach with regard to other aerosol types. We fully agree that it would be highly desirable to build a deposition nucleation/pore condensation parameterization based on physical processes. However, we do not have an idea yet which exact physical processes contribute to the observed ice nucleation properties.

# Comment 3

I think your statement "First, the most important reason is that classical nucleation theory assumes that the temperature dependence is much weaker than we actually observed (compare to calculations presented in Fig. A1 in the review by Hoose and Möhler (2012))." needs more clarification. I feel it is misleading to use your Fig. A1 in this way, if I am not missing something. In the AIDA experiment you change T and RH at once. If you would draw in Fig. A1 the AIDA trajectory for the experiments, you will find that as T decreases a bit, RH increases, and Jhet increases by orders of magnitude. In fact, this trend is in agreement with Fig. A1 which shows dramatic increases of Jhet as RH increases (in agreement with measurements of my group and others). I can imagine that your second reason, the measurement uncertainties during the nucleation phase, are the culprit for this problem. Since you likely need to go via frozen fraction to derive Jhet, you will be very sensitive to uncertainties in frozen fraction making Jhet analysis difficult. To be fair, this is not the fault of CNT, but the "fault" of the experiment and data analysis, something we sometimes cannot change for the specific data/experiment. As is, the criticism would be that it appears that one approach is inferior to the other which is likely not true. If so, it should be clearly stated that the data with their uncertainty do not allow to apply CNT properly (or as you would like to, see your review article) for a fair comparison with your novel approach.



Fig. A1. Deposition nucleation rates calculated from classical nucleation theory, with different formulations of the kinetic factor  $A_{dep}$ : (a) Fletcher (1958), (b) Chen et al. (2008), and (c) Barahona (2011). The red lines are valid for  $f_{het} = 0.0005$ , and the blue lines for  $f_{het} = 0.01$ . From bottom to top, the isolines correspond to  $j_{dep} = 1$ ,  $1 \times 10^{6}$ ,  $1 \times 10^{12}$ , and (beyond the plotting range for the blue lines in **b** and **c**)  $1 \times 10^{18} \text{ m}^{-2} \text{ s}^{-1}$ . In (b) and (c), a desorption energy  $\Delta g_d$  of  $6.5 \times 10^{-20} \text{ J}$  was used.

We would like to emphasize here that the main result that we took from our study is not that classical nucleation theory is in general not suitable to describe deposition nucleation. However, our ensemble of expansion runs could not be described with a common mean contact angle which does not depend on temperature. The reason for this observation could be the contribution by pore condensation (see proposed addition to the manuscript).

Fig. A1 (from Hoose and Möhler, 2013) shows different heterogeneous ice nucleation rates as inferred from classical nucleation theory. These nucleation rates are calculated for two different contact angles (corresponding to  $f_{het}$ =0.0005 and  $f_{het}$ =0.01). These nucleation rates can be converted into an "ice nucleation active surface site density equivalent" by assuming a fixed  $\Delta t$ . This would mean that classical nucleation theory for deposition nucleation predicts that  $n_s$  isolines basically straight lines in the T-RH space at temperatures below 233 K and show an upward slope with decreasing temperature. In Fig.7 of our work similar  $n_s$  isolines are depicted which are in contradiction to classical nucleation theory. Therefore, at this point we compare measured ice nucleation active surface sites to  $n_s$  equivalents.



Fig. 7. n<sub>s</sub> trajectories derived from experimental runs (see Table 1) - the arrow indicates decreasing temperature and increasing RH<sub>iem</sub> during expansion experiments (see Fig. 4); colored lines correspond to isolines of the fitted INAS density (log<sub>10</sub>(n<sub>s</sub>)) from Fig. 6 - symbols indicate ice nucleation active surface site densities derived from experimental studies by other authors (same color code as for isolines)

This does not mean that classical nucleation theory per se is not suitable for describing deposition nucleation . However, our observations illustrate the need for a better understanding of ice nucleation processes occurring directly at the particle surface.

We will add a comment reflecting these additional clarifications on p.10: "Additionally, the contribution of pore condensation freezing to deposition nucleation observed close to water saturation might lead to difficulties with applying classical nucleation theory directly."

Best regards,

Isabelle Steinke on behalf of the co-authors

# Answer to "Anonymous Referee #1"

# Dear referee,

thank you for the detailed reading and commenting of our manuscript. In the following, we would like to reply to the points that you mentioned.

# 1) Deposition nucleation as homogeneous or immersion nucleation in pores and cavities

The recent paper by Marcolli [ACP, 14, 2071-2104, 2014] presents a pretty compelling case that what is typically interpreted as deposition nucleation is, in fact, condensation of water into cavities or pores, followed by freezing in the immersion mode or homogeneous freezing, if the temperature is low enough. Lines 1 through 5 on pg. 18511 of this paper are consistent with that hypothesis. Higher RH with respect to ice is required at higher temperatures while freezing begins at low RH for the colder runs.

This remark refers to our observations that at 250 K ice nucleation occurs at relative humidity values between 112 and 125%, whereas for temperatures below 235 K ice nucleation is already observed slightly above saturation with respect to ice. Indeed, pore condensation freezing could contribute to the observed total ice nucleation. In our setup, deposition nucleation and pore condensation cannot be measured separately because we did not investigate the dynamics of freezing processes at the particle surface. Therefore, it is not possible to estimate the individual contributions of the two freezing mechanisms. However, it is a very valid point that pore condensation freezing should be mentioned. In the future, small scale experiments investigating the influence of different surface structures on freezing processes might be able to capture pore condensation freezing as opposed to deposition nucleation.

# We propose to add the following lines on p. 18522, l.21:

"It should be noted that the observed freezing thresholds could also be partly explained by a freezing mechanism other than deposition nucleation, namely pore condensation freezing. Pore condensation freezing was proposed by Marcolli et al. (2014) as an explanation for freezing below water saturation. Note, however, that in our experimental setup we cannot clearly distinguish between these freezing mechanisms and thus make the assumption that ice nucleation is mostly caused by deposition nucleation.

# 2) Why is a parameterization for Arizona Test Dust in the deposition mode necessary?

Why is this parameterization a valuable addition to the literature, especially considering that measurements of ATD in the deposition mode have already been made. I think the authors could have made the case for these measurements and this analysis a bit more forcefully

We agree that ATD is only a starting point for many experimental studies investigating the ice nucleation properties of natural mineral dusts. It is planned to develop similar temperature and humidity dependent ice nucleation active site density parameterizations for natural mineral dusts in the future. The advantage of ATD is that it is available in large quantities to all interested laboratories and that the results from different instruments can thus be more easily intercompared than results for natural dusts. Our data set differs from previous studies because we have investigated deposition nucleation by ATD particles over a wide range of temperature and humidity conditions. Within the AIDA cloud chamber temperature and humidity can be measured simultaneously and with very small measurement uncertainties. Additionally, we can control the cooling rates, i.e. the time scale, within the AIDA cloud chamber. Thus, being able to study the impact of variations in temperature, humidity and cooling rate is a very unique feature of AIDA cloud chamber studies.

The ATD experiments presented in this work are supposed to be only a first step in rigorously investigating deposition nucleation in order to gain a better understanding of the factors which are relevant for deposition nucleation. In the future, more extensive deposition nucleation studies with natural aerosols are planned. These experiments can then be analyzed with the proposed INAS density approach in order to inform ice nucleation parameterizations.

We propose that the following sentences are added to the discussion section: "The ATD experiments and modeling studies presented in this work are supposed to be a first step in rigorously investigating deposition nucleation over a wide temperature and saturation range in order to gain a better understanding of the factors which are relevant for deposition nucleation. This knowledge was then used to develop a metric which can be easily employed for the comparative analysis of other deposition nucleation studies." (p.18524, l.26)

# 3) Minor point

*Pg. 18512, lines 24-25: "...T represents the numerical value of the prevalent temperature..." What is a prevalent temperature? Is that the average temperature in the chamber? This is a bit confusing to me.* 

Thank you for this remark – we will change "prevalent temperature" to "average temperature within the cloud chamber".

# Answer to "Anonymous Referee #2"

# 1) Applicable range of parameterization

The manuscript states that the parameterization of ns is valid between 226 and 250 K. In Figure 6, the experimental data starting at 223 K was excluded. Does that mean we need another parameterization for temperature lower than 226 K? Why the parameterization did not include the data at lower temperature? One of the purposes of parameterization is to use it in modeling studies for wider ranges of conditions, including temperature. As for the current parameterization (eqn. 16), the observed ns are within the 2 orders of magnitude. If include the data, it may go up to 3 or 4 orders of magnitude. If consider this variation/uncertainty, how would this affect on the results of the box model simulation? In other words, any sensitivity test in box model simulation to include uncertainty of the parameterization (two constants in eqn. 16)?

The experimental data starting at 223 K was excluded because we observed that the humidity at which the ice nucleation onset is observed does not change much between 223 and 226 K. Thus, since the temperature dependence becomes less important we assume that ice nucleation should ideally be described by a different function for  $x_{therm}$ . Nevertheless, Fig.6 also demonstrates that ice formation can largely be described by our proposed ns function even below 226 K. Also, we would like to highlight that our study is mainly intended to provide a case study for investigating deposition nucleation properties of aerosol particles. The sensitivity of the two parameters in the ns equation can be roughly estimated from the experimental data. The individual experimental ns trajectories (see Fig.6) show that the two coefficients in the ns equation can be maximally varied such that the ns value itself varies by roughly one order of magnitude. For the first coefficient (a1,b1) this variation directly would be caused by a change within one order of magnitude, whereas for the second coefficient (a2, b2) this variation translates into a change within +/-15%.

We would expect similar results from box model calculations investigating parameterizations taking this sensitivity into account, i.e. only variations within approximately one order of magnitude.

# 2) Estimation of the time dependence from classical nucleation theory

To investigate the time dependence, this study proposed another two equations (eqn. 19 and 20) for parameterization. These three (including previous one, eqn. 16) equations and three corresponding parameterizations were obtained for the same data set. When do the simulation for a reasonable range of pre-settings (temperature/RH/time), isn't it expected similar simulation results? To confirm the time dependence conclusions derived from box model simulation using ns parameterizations, one possible way is to do the same simulation using parameterizations based on classical nucleation theory (CNT).

The simulations that were performed for the three different parameterizations were intended to demonstrate how the ice crystal concentrations would develop if conditions were applied which differ from those which were investigated experimentally. Therefore, we chose starting conditions with very low aerosol concentrations and a wide variation of cooling rates. And even though the time dependence and thus the difference between the parameterizations was relatively small, we still wanted to estimated the variation of the predicted ice crystal concentrations in different scenarios.

We agree that extracting the time dependence by using classical nucleation theory might appear to be more promising in order to estimate the time dependence. However, there are three reasons why there are difficulties with this approach. First, the most important reason is that classical nucleation theory assumes that the temperature dependence is much weaker than we actually observed (compare to calculations presented in Fig. A1 in the review by Hoose and Möhler (2012)). Secondly, in the experiment the nucleation phase is associated with larger measurement uncertainties than later phases of the experiment. Thirdly, at different temperatures a variation in the contact angle is expected which adds another degree of complexity (Welti et al., 2012). These measurement uncertainties are caused by fluctuations in the observed ice crystal concentration at the beginning of ice formation. These two factors contribute significantly to the observed variation in contact angle distribution parameters which are estimated from the experimental data. Looking forward, using contact angle distribution parameters with large uncertainties will translate enormous deviations of the predicted ice crystal concentrations. Also, classical nucleation theory is apparently not able to describe deposition nucleation by ATD particles.

# **Minor comments**

 p.18500, Eqn. 2, although used as a dimensionless parameter, is there any physical reason to directly add T and S<sub>ice</sub> as a "thermodynamic" variable? The term of x<sub>therm</sub> shouldn't be identified as thermodynamic variable.

We will rename x<sub>therm</sub> from "thermodynamic variable" into "temperature and saturation dependent function".

- 2) p. 18500, define Sice.
   We will add on p.18500 I.20: "...with the saturation ratio with respect to ice S<sub>ice</sub>>1..."
- 3) p.18503-18504 and section 3.2.3, about the CNT and data analysis, what are the J values or how the J was used to derived theta?

Which value of surface tension at the ice/vapor interface was used? Please provide the values of the parameters used in CNT analysis.

The Delta *gd* in Chen et al. (2008) (Fig. 2 of the paper) was about 2E-20 J for temperature lower than 223 K. Why the Delta *gd* of 4E-20 J was used here?

Regarding the heterogeneous nucleation rates, we would like to point out that we did not explicitly calculated heterogeneous nucleation rates. However, we implicitly used the heterogeneous nucleation rates J<sub>het</sub> to calculate the ice fractions (eq. 11 from Wheeler et al. (2012)):

$$\frac{N_f}{N_0} = 1 - \int_0^\infty \int_0^\pi \exp\left[-\pi D^2 J_{\text{het}}(\alpha, T, S_{\text{ice}})t\right] f_\alpha(\alpha) f_{\text{num}}(D) d\alpha \, dD,$$

- The surface tension was described as a temperature dependent function according to Pruppacher and Klett 1997 (see eqs. (5–46), (5–47a), and (5–12)).
   We will add on p. 18503 l. 18: "The surface tension was described as a temperature dependent function according to Pruppacher and Klett 1997."
- It is true that the  $\Delta g_d$  as directly inferred from the experimental data for ATD was at roughly 2.0·10<sup>-20</sup>J (Chen et al., 2008). However, in the literature data cited in the study by Chen et al. (2008) for mineral dusts generally higher values were found, e.g.  $8.7 \cdot 10^{-20}$ J for ATD (Gustafsson et al., 2005). Therefore, we slightly adjusted the value of  $\Delta g_d$  used in our calculations. Also,  $\Delta g_d$  has only a limited influence on the predicted ice crystal concentrations compared to contact angle distribution parameters (Chen et al., 2008).
- 4) p. 18505, I.26, define AIDA when it was used for the first time.
   We will add: "...(Aerosol Interaction and Dynamics in the Atmosphere)..." on p.18505
   I.23.
- 5) p. 18508, l.11-14, does this mean that if there is sufficient water vapor and at high RH, particle with soluble materials will become aqueous droplets and could also be detected, especially at higher temperature, e.g. 250 K?
  Yes, the WELAS instruments can detect droplets as well. For immersion freezing experiments, typically the formation of droplets is observed before these droplets are then converted into ice crystals at lower temperatures.

6) p.18508, I.14-17, this manuscript focused on deposition ice nucleation, can you rule out the possibility of immersion freezing at high temperature, i.e., 250 K? If there is very small amount of soluble materials, once it takes up water, it will form aqueous particles then ice will nucleate through immersion freezing. It is not about the subsaturated conditions, it is about what are the soluble components and when it takes up water or deliquesces. We agree that immersion freezing cannot be ruled out completely but ATD particles contain only very small amounts of soluble components and thus droplet formation is very limited below RH<sub>wat</sub>=90% (Vlasenko et al., 2005). Thus we assume that deposition nucleation is the dominant ice nucleation mechanism even at 250 K.

# 7) p.18508, l.18-19, what is "a suitable size threshold"?

With "suitable" threshold we wanted to express that the size thresholds that are used for the analysis of WELAS data are adjusted individually, depending on the aerosol size distribution and the ice nucleation mode. For immersion freezing, larger size thresholds may be used than for deposition nucleation experiments with the same aerosol types because for immersion freezing particles are activated to droplets which are larger than the aerosol particles.

- 8) p.18508, define SIMONE when it was first used in the text.
   We will add: "...SIMONE (Scattering Intensity Measurements for the Optical Detection of Ice)" on p.18508, I.7.
- 9) p.18511, l.22, how is the RHice uncertainty calculated? What are the uncertainties of gas and wall temperatures?

 $RH_{ice}$  is derived from the absolute water vapor concentration as measured with the TDL (tunable diode laser) absorption spectroscopy and the water vapor saturation pressures with respect to ice at a certain temperature (Murphy and Koop, 2005). The deviation from the calculated saturation vapor pressures (Murphy and Koop, 2005) was less than 3% during the Aquavit campaign (Fahey et al., 2013). The measurement uncertainty regarding the well mixed cloud chamber is approximately  $\Delta T$ =0.3 K which translates into an overall uncertainty of  $\Delta RH$ =5%.

10) p.18516, l.3-11 and Fig. 7, at 233 K, the RHice onsets are more than 10% lower than Kohler et al. (2010) and Welti et al. (2009), does that mean only the large particles nucleated ice in this study (polydisperse particles, see surface distribution in Fig.2)? How do these RHice onsets compare to the ice nucleation data by Knopf and Koop (2006). As assumed correctly by the referee, larger particles will initiate ice nucleation first. Thus, particle size distributions including larger particles (d<1µm) will indeed show ice nucleation onsets which are shifted towards lower relative humidities. This trend is also visible for our set of ice nucleation thresholds (see Fig. 4).

A comparison with the study by Knopf and Koop might not be appropriate because of differences in the experimental methods. Knopf and Koop (2006) used ATD solutions whereas in our experiments the dust was dry dispersed. Also, the range of humidities at which ice nucleation was observed by Knopf and Koop (2006) was very large. At 250 K, for example, ice nucleation occurred between RH<sub>ice</sub>=105% and RH<sub>ice</sub>=115%. So, even

though we observed ice nucleation at similar conditions, it would be difficult to draw solid conclusions from this comparison.

11) p.18516, l.15-16, "devations" should be "deviations"? This statement didn't explain the deviation. If the ATD used in these studies are from the same source, the ice nucleation efficiency (RHice thresholds) by nature should be very similar and so the INAS at the same temperature. Does the statement in l.15-16 imply that the INAS parameterization provided here is only valid or limited to AIDA experiments? Then, how this parameterization can be applied for atmospheric application? Is there any other possible explanation for these deviations, what is the difference in surface area compared to the cited studies?

We do not think that the INAS density approach is only applicable to AIDA results. We would like to point out that even though the particle diameters were given for the studies used for comparison, the aerosol surface area was not explicitly measured. Thus, some difference might as well come from differences between real and estimated aerosol surface areas. The differences between the results observed for different experimental setups highlight the necessity to achieve very small measurement uncertainties regarding temperature, relative humidity and aerosol surface area. "Devations" will be changed into "deviations."

- 12) *p.18518,I.18-19, the manuscript didn't provide sufficient proof to support this statement.* We would like to point out that there is a strong temperature dependence for deposition nucleation occurring between 235 and 250 K, because the ice nucleation onsets for similar experimental conditions vary strongly (see Fig.4). This strong temperature dependence is to our knowledge not reflected in current classical nucleation theory formulations.
- 13) Table 1: It would be nice see the RHice threshold for each experiments. We can add the valued displayed in Fig.4. Thresholds in Fig. 4 are defined as humidity values at which ice crystal concentrations exceeding aerosol background concentrations were observed.
- 14) Figure 5. please add description for the error bars showing in the figure. We will add the following sentence to the caption: "The error bars represent the measurement uncertainties in  $n_s$  with  $\Delta n_s / n_s \approx 35\%$  and  $x_{therm}$  with  $\Delta x_{therm} / x_{therm} \approx 5$ ."
- 15) Since the parameterization is only valid for temperature above 226 K, it is misleading showing the blue solid line for 220 K. Where is the grey dashed line in the figure? The experimental results for 220 K and the two dashed lines (above and below the actual parameterization) are shown to illustrate that even though the x<sub>therm</sub> formulation fits best between 226 and 250 K, deviations between experimental results and parameterization are within one order of magnitude even below 226 K.
- 16) Any simulation at 250 K? Do they show similar results?We conducted selected simulations at 250 K. These simulations yielded conceptually similar results. However, we are only presenting results for 235 K, because for

atmospheric observations at 250 K, generally, there is a higher probability to find contributions by several ice nucleation modes. At 235 K, in contrast, deposition nucleation should be the dominant ice nucleation path for heterogeneous ice nucleation.

# References

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