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Article

**Keywords:** Pluto, haze, atmospheres

**DOI:** https://doi.org/10.21203/rs.3.rs-588999/v1

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A bimodal distribution of haze in Pluto's atmosphere

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Abstract

Pluto, Titan, and Triton make up a unique class of solar system bodies, with icy surfaces
and chemically reducing atmospheres rich in organic photochemistry and haze
formation. Hazes play important roles in these atmospheres, with physical and chemical
processes highly dependent on particle sizes, but the haze size distribution in reducing
atmospheres is currently poorly understood. Here we report observational evidence that
Pluto's haze particles are bimodally distributed, which successfully reproduces the full
phase scattering observations from New Horizons. This result suggests a dimensional
transition in organic haze formation, and indicates that both oxidizing and reducing
atmospheres can produce multi-modal hazes.

Introduction

Hazes are common in the atmospheres of solar system bodies and exoplanets (Seinfeld
\& Pandis 2006, Wilquet et al., 2012, Sing et al. 2016, Orton et al. 2016) and they play
an important role in atmospheric composition, dynamics, and radiative transfer. Most
of these processes are highly dependent on the haze particle sizes, which usually follow
bimodal/multimodal distributions in chemically oxidizing atmospheres, as observed in
the atmospheres of Earth and Venus (Seinfeld \& Pandis 2006, Yue et al. 2009, Levy et
al. 2013, Wilquet et al., 2009, Wilquet et al., 2012), resulting from multiple atmospheric
processes behind haze formation and evolution (Seinfeld \& Pandis 2006, Gao et al.
2014, Lavvas et al. 2010). In contrast, the modality of haze particle distributions in
reducing atmospheres is currently unknown. Previous observations of such hazes were
interpreted using unimodal haze models for simplicity (Rages \& Pollack 1992,
Tomasko et al. 2008, Cheng et al. 2017). More physical haze models typically
simplified the haze formation process, generating unimodal hazes as well (Gao et al.
2017, Ohno et al. 2021), though bimodal hazes were also seen in some simulations
(Lavvas et al. 2010). More constraining observations are therefore needed to determine
whether hazes in reducing atmospheres are unimodal or multi-modal. Here we report
observations of a bimodal distribution of haze particles in a chemically reducing
atmosphere after ruling out a number of unimodal distribution hypotheses. We show
that full phase scattering observations of Pluto's haze from New Horizons constrains
its distribution to be bimodal, which inspires revisits of Titan's detached haze and the
haze on Triton, as well as the improvement of numerical simulations of organic hazes
in reducing atmospheres.

Pluto's atmosphere is similar to those of Titan and Triton in terms of composition, with
N\textsubscript{2} being the dominant component, percent-level abundances of CH\textsubscript{4}, and smaller
amounts of CO (Gladstone et al. 2016, Lellouch et al. 2017, Young et al. 2018). This composition also resembles that of the Early-Earth atmosphere and therefore has significance in the context of astrobiology (Catling & Zahnle 2020). The surface atmospheric pressure of Pluto is similar to that of Triton and equivalent to Titan’s atmosphere above 400km (Hinson et al. 2017, Young et al. 2018, Gladstone & Young 2019). Therefore, atmospheric processes are expected to be similar in this pressure region on these celestial bodies. Investigation of physical processes on one of these worlds thus has significant implications for the others, as well as for chemically reducing atmospheres in general.

The existence of haze in Pluto’s atmosphere was confirmed by the New Horizons spacecraft during its flyby in July 2015 (Stern et al. 2015, Gladstone et al. 2016, Cheng et al. 2017), together with a recent occultation (Person et al. 2021). Originating from photolysis of CH₄, N₂, and larger organic molecules in Pluto’s upper atmosphere driven by solar ultraviolet radiation, haze particles grow through coagulation of smaller particles as they sediment downwards (Gao et al. 2017, Lavvas et al. 2021), which is similar to processes in Titan’s upper atmosphere (Lavvas et al. 2013). Due to the unexpectedly low atmospheric temperature, however, condensation and/or sticking of gaseous species have considerable impacts (Wong et al. 2017, Luspay-Kuti et al. 2017, Lavvas et al. 2021), which resemble that in Titan’s lower atmosphere and on Triton (Anderson & Samuelson 2011, Ohno et al. 2021). The haze also controls Pluto’s energy budget through radiative heating and cooling of the atmosphere and by altering the surface color (Zhang et al. 2017, Grundy et al. 2018). Understanding the morphology of the haze particles, which determines how they interact with condensing/sticking gases and radiation at visible and infrared wavelengths, and sheds light on haze growth in reducing atmospheres, is therefore critical. Haze in Pluto’s lower atmosphere (<50km) has significance in this context, as particles in this region are representative of their final formation stage, which allows investigations of their origin and formation pathways.

**Results**

Observations of Pluto’s haze have been obtained by multiple instruments onboard the New Horizons spacecraft, with wavelength coverage from the ultraviolet (UV) to infrared (IR). Pluto’s haze possesses a bluish color, which suggests Rayleigh-type scattering by particles with radii smaller than visible wavelengths. However, the haze also has strong forward scattering, which is an indication of large particles (Gladstone et al. 2016). Given these properties, the haze particles are thought to be fractal aggregates—highly porous and randomly shaped ~μm particles consisting of small ~10nm spherical monomers, similar to those in Titan’s atmosphere (Tomasko et al. 2008). However, the non-negligible backscattering of Pluto’s haze in the observations is inconsistent with a haze composed purely of fractal aggregates (Cheng et al. 2017). The backscattering characteristics are similar to those of Triton’s haze, which is thought to arise from the combination of surface reflection and low-altitude clouds (Rages & Pollack 1992). Several size distributions (log-normal, bimodal, power-law) together with surface reflection have been proposed to reproduce Pluto’s haze observations obtained from different instruments (Gao et al. 2017, Cheng et al. 2017, Kutsop et al. 2021), but degeneracy is significant, as they are separately constrained using observations from different instruments. The backscattering has especially not been well investigated, and a combination of full phase observations covering a wide
wavelength range is necessary. Here, we conduct a joint retrieval using observations
obtained by all instruments onboard New Horizons that observed Pluto’s haze and show
that the backscattering of Pluto’s haze originates from an additional mode in the haze
size distribution. We further conclude that a unimodal distribution of haze cannot
reproduce the observations, and a bimodal distribution of haze is the simplest feasible
solution, which is also the only one among a number of proposed scenarios that can
currently be constrained.

Four New Horizons instruments measured the optical properties of Pluto’s haze. These
observations include (1) UV extinction obtained by the Alice UV spectrograph (Stern
et al. 2008) through solar occultation, (2) scattered light at a single visible wavelength
at high and low phase angles captured by a wide-band camera, the Long Range
Reconnaissance Imager (LORRI, Cheng et al. 2008), (3) forward scattered IR spectra
measured by a spectral imager, the Linear Etalon Imaging Spectral Array (LEISA,
Reuter et al. 2008), and (4) scattered light at four narrow visible and near-IR wavelength
bands at high and low phase angles by a narrow-band camera, the Multispectral Visible
Imaging Camera (MVIC, Reuter et al. 2008). Observations obtained by the MVIC
panchromatic mode are not included due to data coverage and calibration issues
(Methods). Among these currently available observations of Pluto’s haze, we select the
ones with the highest quality and good resolution (>5km), which are summarized in
Table 1 and Figure 1.

We focus on the lower 50km of Pluto’s atmosphere above the surface, which includes
its thin troposphere and an overlying thermal inversion. Haze in this region was
observed by all aforementioned instruments, and therefore its optical properties are the
best constrained. The morphology of haze particles in this region is representative of
the final stage in the shaping of haze particles through microphysical processes in
Pluto’s atmosphere, just before they sediment onto the surface. The pressure level (0.5-
1Pa) of this region is comparable to 400-600km altitude in Titan’s atmosphere where
the detached haze layer is imaged (West et al. 2018), and to altitudes of <50km in
Triton’s atmosphere (Tyler et al. 1989).

The observed line-of-sight (LOS) integrated quantities (optical depth and I/F) are first
converted to local optical properties assuming spherical symmetry using the Abel
transform for noisy data (see Methods). We then apply light scattering models and our
retrieving algorithm to these local quantities (Methods). We consider the scattering
effects of fractal aggregates, spherical haze particles, and surface reflection in ten
scenarios (Table A2, Methods), which are chosen based on a balance between the
number of observations and the degree of freedom. The values of the parameters under
each scenario and their uncertainties are obtained using the Markov chain Monte Carlo
(MCMC) approach (Mackay 2003, Foreman-Mackey et al. 2013, Methods). We
parameterize the morphology of fractal aggregates using three quantities, the fractal
dimension ($D_f$), monomer radius ($r_m$) and number of monomers in each aggregate ($N_m$),
which follow the relationship $N_m=(R_a/r_m)^{D_f}$ with aggregate effective radius ($R_a$). The
fractal dimension describes the porosity of the aggregate and relates the change in size
of the aggregate with its change in mass. Aggregates with larger $D_f$ are more compact.
In the nominal forward model, we use the complex refractive indices of “tholins”, the
laboratory analogues of Titan’s haze (Khare et al. 1984), for Pluto’s haze, motivated by
similar atmospheric compositions between Pluto and Titan. The higher fraction of CO
and organic ice condensation in Pluto’s atmosphere may influence these optical
properties (Lavvas et al. 2021, Jovanović et al. 2021, Ohno et al. 2021), but our
sensitivity study shows that differences in refractive indices would not significantly
change the observables, and therefore would not affect our interpretation (Methods).
We compute the optical properties of aggregates by adapting a light scattering model
that was used for Titan’s haze (Tomasko et al. 2008) and well validated (Liu et al. 2019),
and we use Mie theory (Wiscombe 1979) to compute scattering from spherical particles
and monomers (Methods).

As the simplest assumption, a monodispersed population of ~1μm fractal aggregates
consisting of ~20nm monomers can reproduce the UV extinction, visible forward
scattering, and the slope of the IR forward scattering spectra (Figure 1). The aggregate
effective radius is two times larger than those estimated in previous works (Cheng et al.
2017). However, as backscattering is orders of magnitude less intense than forward
scattering for large fractal aggregates (Figures 1 and 2), the monodispersed aggregates
scenario underestimates the observed backscattering by a factor of ~3 (Figure 1b),
which is equivalent to a LOS I/F difference of ~5*10^-3 in the visible scattering
configuration in the lower 50km. Moreover, tests of other possible unimodal scenarios
(log-normal, power-law or exponential distribution of two-dimensional aggregates or
spheres) that can be sufficiently constrained by observations suggest that unimodal
distributions of either aggregates or spheres cannot reproduce the scattering intensities
at the observed phase angles in the visible and the forward scattering spectrum in the
IR with the given UV extinction (Methods, Figures A7 and A8). This is an indication
of additional scattering sources.

Surface reflection was proposed as another scattering source (Cheng et al. 2017), as a
combination of surface reflection and low-altitude clouds successfully explains the
large backscattering observed for Triton’s haze (Rages & Pollack 1992). However,
secondary scattering, sunlight first reflected off of the surface and then scattered once
by the haze particles into New Horizons’ instruments, is not sufficient in the case of
Pluto. We conduct a quantitative estimation by adapting the Hapke model (Hapke 1981),
which was used to study Triton’s surface (Hillier et al. 1990 and 1991), to simulate
Pluto’s surface reflection (Methods). An upper limit of <2.5*10^-3 is derived for the I/F
contribution from secondary scattering at the observation wavelength and phase angles
when assuming that the incident and emission vectors are in the same specular plane
(Figure A6). Therefore, secondary scattering is not sufficient to bridge the gap between
the observed backscattering and that predicted by the monodispersed aggregates
scenario.

We found that an additional population of small particles scattering in the Rayleigh
regime could explain the large backscattering, as their forward and backward
scatterings are comparable in strength, so that the total backscattering of the haze could
be considerable when small particles are mixed with large aggregates (Figure 2).
Results of the joint retrieval under this scenario show that a combination of large two-
dimensional aggregates and small spheres can reproduce all the observations (Figure
1). The two types of haze particles have comparable UV extinctions while the
aggregates dominate the forward scattering and spheres dominate the backscattering at
visible wavelengths (Figure 2). Vertical profiles of the retrieved parameters of the
bimodal distribution are almost constant between 50 and 15km (Figure 3), indicating
that the haze particle morphology does not change in this region. The larger-size
population consists of ~1μm two-dimensional aggregates with ~20nm monomers
(Figure 3b and 3e), while the smaller-size population consists of ~80nm spheres (Figure 3b). The number density of the aggregates is ~0.3cm$^{-3}$, within an order of magnitude of previous estimations (Cheng et al. 2017), while that of the spheres is around ~10cm$^{-3}$ (Figure 3d). Although the number densities differ by two orders of magnitude, the total masses of these two populations are almost the same (Figure 3f). The mass of the aggregates is slightly greater, but within a factor of 2.

Our retrieved size distribution of Pluto’s haze stemming from consideration of all observations from the UV to the IR resolves the degeneracy between previous interpretations (Gao et al. 2017, Cheng et al. 2017, Kutsop et al. 2021), and provides observational evidence of bimodally distributed haze particles in chemically reducing atmospheres, which indicates multi-mode haze formation processes therein. The population of larger particles is likely the result of dimensional change during haze particle coagulation, as sizes and dimensions of these two haze particle populations agree well with models of Titan’s haze at ~500km (~0.5Pa) where a transition from spheres to aggregates is artificially introduced (Lavvas et al. 2010). Although the exact physical condition that triggers the dimensional transition is unknown, the resulting unimodal distribution of two-dimensional aggregates in Titan’s lower atmosphere (<200km), simulated with microphysical formation processes, have scattering properties that agree well with observations. The fact that dimension transition has now been inferred on two separate worlds at similar pressures gives clues to the possible physical processes involved in dimension transition.

In the numerical simulations of Titan’s haze (Lavvas et al. 2010), the modeled haze at the dimensional transition region (~0.5Pa) follows a bimodal distribution similar to that retrieved here for Pluto, including ~1μm two-dimensional aggregates and small spheres with radii on the order of tens of nanometers. The spherical particles are transported from higher altitudes, where all particles follow a unimodal distribution. When these particles sediment to the transition pressure level, they begin coagulating into two-dimensional fractal aggregates. The cross sections of the aggregates are much larger than equivalent-mass spheres, so the number density of the larger-size population increases rapidly through coagulation once the dimensional change initiates, which results in a loss of medium size particles. The aggregates stop growing at ~1μm due to Coulomb repulsion. In the case of Titan, the smaller-size population continues coagulating with large ~1μm aggregates below the dimension transition region, ultimately being subsumed into the large fractal aggregates population. In contrast, the atmosphere of Pluto is much thinner with a surface pressure of ~1Pa. As such, the haze particles reach Pluto’s surface in the middle of dimensional transition and the bimodal distribution is maintained. Moreover, the sharp decrease of the atmospheric temperature from ~100 to 40K in the lower 15km (Hinson et al. 2017) could slow down or freeze the process of coagulation and may introduce another dimensional change with rapid condensation of gaseous species (Lavvas et al. 2021) and possibly the collapse of porous fractal aggregates, which is suggested by the significant increase of fractal dimension of the larger-size aggregate population below 15km altitude (Figure 3a).

The similarity of our results to those of Titan’s detached haze at ~500km motivates a revisit of observations thereof to better understand dimension transition in haze formation. Images taken with the Imaging Science Subsystem (ISS) onboard the Cassini spacecraft would provide fruitful observations over a number of viewing configurations and different seasons (West et al. 2018). Backscattering observations of
the detached haze layer would contain information about a second population of small spheres. Reanalysis of observations of Triton’s haze is also crucial in light of our results. Two types of spherical particles, hazes and clouds, were proposed at different latitudes to explain the observed scattering obtained by Voyager 2 wide-angle camera, together with surface reflection (Rages & Pollack 1992). Haze particles are assumed to be spheres, but fractal aggregates are more likely given the formation mechanism (Ohno et al. 2021, Lavvas et al. 2021). Moreover, the observed large backscattering may be due to a second population of small particles, which could contribute orders of magnitude more to the observed I/F than the secondary scattering of surface reflected light.

The discovery of the small-spheres haze particle population has significance in improving our understanding of the radiative processes in Pluto’s atmosphere, and therefore Pluto’s energy budget. Pluto’s unexpectedly low atmospheric temperature was well explained by efficient radiation by ~1μm haze particles and effective collision between these particles and gas molecules (Zhang et al. 2017). Solar energy absorbed by gases is transferred to haze particles rapidly through collision and radiated away from Pluto, which is proposed as the dominant energy pathway in Pluto’s atmosphere. However, compared to the ~1μm aggregates, smaller particles with radii on the order of tens of nanometers have shorter radiative relaxation timescales but much longer collisional heat-transfer timescales with gases. Compared to ~1μm aggregates, the ratio of these two timescales for ~80nm particles differ by 2-3 orders of magnitude, which leads to the radiation timescale being smaller than the collision timescale. In other words, the radiative cooling of smaller particles can be so efficient that the heat-transfer between the particles and gases through collision is not sufficient to keep their temperatures the same. This could result in the small particles being cooler than the ambient atmosphere. The larger-size particles are still the key medium transferring heat between gases and particles, but the dominant factor of radiative cooling needs reevaluation. The more efficient radiative cooling of smaller particles may result in another peak and/or a steeper slope in the mid-infrared emission spectrum of Pluto than previously predicted (Zhang et al. 2017), which can be further investigated by future missions, e.g., through the Mid-Infrared Instrument on the James Webb Space Telescope (Glasse et al. 2015).

Discussion

Condensation of gaseous species onto haze particles may influence their evolution in the atmosphere and their interactions with visible and infrared radiation. In the lowest 50km of Pluto’s atmosphere, where the pressure varies between 0.1-1Pa, haze particles are expected to contain an organic ice component, similar to the predictions for Triton’s atmosphere (Lavvas et al. 2021). In contrast, Titan’s atmospheric temperature is higher than 150K at the same pressure level, which would inhibit gas condensation. Therefore, the hazes in Pluto’s and Triton’s atmosphere may have larger scattering and less absorption than “tholins”. However, such gas condensation processes could not simultaneously reproduce the large forward and backward scattering with the given UV extinction observed at Pluto. With the inclusion of gas condensation, numerical simulations show that fitting the UV extinction results in an underestimation of both the forward and backward scattering I/Fs at visible wavelengths by a factor of 2-3 (Lavvas et al. 2021). Another scattering source is still necessary. To test the influence of organic ice, we conducted a sensitivity study of two-dimensional aggregates with
different optical properties (Methods). This represents an upper limit for the extent to which ice condensation or other organic composition can influence the observables. The result shows that the difference in scattering intensity at the wavelengths and phase angles considered here due to the different refractive indices is negligible. The observed quantities, especially the phase functions of fractal aggregates, are not sensitive to the difference in optical constants between organic ice and “tholins”. A second component of small spheres is still required to explain the observed backscattering of Pluto’s haze. Thus, the influence of gas condensation is not large enough to alter our inferred pathway of haze formation.

As Pluto’s atmosphere is in sublimation/deposition equilibrium with surface N\textsubscript{2} ice (Gladstone et al. 2016, Young et al. 2018), its surface pressure is expected to vary significantly over seasons (Bertrand et al. 2018, Johnson et al. 2021). Small changes of the atmospheric temperature (e.g., a few K), driven by the eccentric orbit (e~0.25), can result in orders of magnitude differences in surface pressure. Numerical simulations show that Pluto could have a minimum surface pressure between 10^{-3}-0.3\text{Pa} on its current orbit near aphelion (Johnson et al. 2021), which is large enough to maintain CH\textsubscript{4} photochemistry and therefore haze formation (Young et al. 2018). However, the dimensional transition region of Pluto’s haze (0.5-1\text{Pa}) will move downward and finally disappear when Pluto moves farther from the Sun, which may result in a significant difference in the size of haze particles depositing onto Pluto’s surface over seasons. Also, the CH\textsubscript{4}/CO ratio could become smaller when the atmospheric temperature decreases, so the CO content is expected to be larger in Pluto’s haze when Pluto moves away from perihelion. Thus, while understanding the morphology of haze particles and its influence on Pluto’s system is important, the current interpretation is only representative of New Horizons’ flyby in 2015. As Pluto’s atmosphere evolves, so will the haze distribution and composition, allowing future observations and missions to capture different stages of organic haze evolution near Pluto’s surface.
Table 1. Summary of Pluto’s haze observations.

| Instrument | Wavelength (μm) | Altitude Range (km) | Phase Angle (degree) | Reference        |
|------------|----------------|---------------------|----------------------|-----------------|
| Alice      | 0.185          | 0-300               | Extinction           | Young et al. (2018) |
| LORRI      | 0.608          | 0-100               | 19.5                 | Cheng et al. (2017) |
|            |                | 0-50                | 67.3                 |                 |
|            |                | 0-75                | 148.3                |                 |
|            |                | 0-200               | 169.0                |                 |
| LEISA      | 1.235-2.435    | 0-299               | 169.0                | Grundy et al. (2018) |
| MVIC       | 0.624          | 0-50                | 18.2                 | This work       |
|            | 0.492          |                      | 38.8                 |                 |
|            | 0.861          |                      | 169.4                |                 |
|            | 0.883          |                      |                      |                 |
Figure 1. Comparison of different model haze scenarios to observations of Pluto’s haze. Haze observations obtained by instruments onboard New Horizons (shaded areas) are compared to best-fit model results for monodispersed fractal aggregates constrained using all observations (dashed lines with crosses), monodispersed fractal aggregates constrained using all observations except for the backscattering LORRI and MVIC data (dotted lines with squares), and a bimodal distribution of haze particles constrained using all observations (solid lines). Error bars show the 1-σ uncertainties of the bimodal distribution scenario. (a) UV extinction coefficient at 0.185μm measured by the Alice spectrograph during solar occultation ingress (indigo) and egress (light red), taken from Young et al. (2018). (b) Local scattering intensity at 0.608μm derived from LORRI images at four phase angles of 19.5° (pink), 67.3° (light green), 148.3° (dark red), and 169.0° (dark blue), processed using data from Cheng et al. (2017). (c) Local scattering
intensity spectrum as a function of altitude derived from LEISA observations at a phase angle of 169.0°, processed using data from Grundy et al. (2018). (d) Local scattering intensity at the 0.624, 0.492, 0.861, 0.883μm (from top to bottom) wavelength bands derived using MVIC images at three phase angles of 18.2° (red), 38.8° (grey), and 169.4° (brown).
Figure 2. Contributions of fractal aggregates and spheres to UV extinction and scattering intensity in the visible. (a) Observations obtained by the Alice spectrograph during ingress (indigo) and egress (red) of solar occultation are denoted as error bars and compared to the contributions from aggregates (blue) and spheres (orange). (b) Observations obtained by LORRI are denoted as black error bars and compared to the contributions from aggregates (blue shaded areas) and spheres (orange shaded areas). The solid black lines are the total contributions of aggregates and spheres, which are also the sum of the shaded areas. The colored dashed lines represent the ratio of contribution from each component.
Figure 3. Retrieved profiles of haze parameters. Under the scenario of the bimodal distribution, we retrieve the best-fit vertical profiles for the (a) aggregate fractal dimension, (b) monomer/sphere radius, (c) number of monomers in each aggregate, and (d) haze particle number density, with which we derive profiles of the (e) aggregate effective radius, and (f) mass density assuming a material density of 1 g/cm$^3$. The best fit profiles are shown in the solid curves, while the 1-σ uncertainties of their posterior distribution functions are shown as shaded areas, with blue representing the aggregates population and orange representing the spheres population.
Acknowledgements

We thank William M. Grundy for sharing the LEISA data, Michael L. Wong and Xue Feng for improving figure representations, and Yan Wu for comments. P.G. is supported by NASA Hubble Fellowship grant HST-HF2-51456.001-A awarded by the Space Telescope Science Institute, which is operated by the Association of Universities for Research in Astronomy, Inc., for NASA, under contract NAS5-26555. X.Z. is supported by NASA Solar System Workings Grant 80NSSC19K0791.

Author contributions

S.F. conducted the data analysis, performed the calculations, and wrote the manuscript. S.F., P.G., X.Z., and Y.L.Y. conceived and designed the research. P.G. and D.J.A. provided the microphysical model. P.G. and C.L. provided the aggregate scattering model. N.W.K. and C.J.B. contributed to the analysis of MVIC data. J.Y. contributed to interpreting and presenting the retrieval results. L.A.Y. and A.F.C provided insights into interpreting New Horizons observations. All authors contributed to the manuscript writing.

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