Possible detection of hydrazine on Saturn’s moon Rhea

Mark Elowitz1*, Bhalamurugan Sivaraman2*, Amanda Hendrix3, Jen-Iu Lo4, Sheng-Lung Chou4, Bing-Ming Cheng4†, B. N. Raja Sekhar5, Nigel J. Mason6

We present the first analysis of far-ultraviolet reflectance spectra of regions on Rhea’s leading and trailing hemispheres collected by the Cassini Ultraviolet Imaging Spectrograph during targeted flybys. In particular, we aim to explain the unidentified broad absorption feature centred near 184 nm. We have used laboratory measurements of the UV spectroscopy of a set of candidate molecules and found a good fit to Rhea’s spectra with both hydrazine monohydrate and several chlorine-containing molecules. Given the radiation-dominated chemistry on the surface of icy satellites embedded within their planets’ magnetospheres, hydrazine monohydrate is argued to be the most plausible candidate for explaining the absorption feature at 184 nm. Hydrazine was also used as a propellant in Cassini’s thrusters, but the thrusters were not used during icy satellite flybys and thus the signal is believed to not arise from spacecraft fuel. We discuss how hydrazine monohydrate may be chemically produced on icy surfaces.

INTRODUCTION

Our knowledge of the geology and surface topography of Rhea, Saturn’s second largest moon, has been greatly advanced by several flybys during the Cassini-Huygens mission. The surface of Rhea is heavily cratered with geomorphological features that indicate possible past endogenic activity, such as large impact craters that have undergone partial relaxation and extensional fault systems that are oriented in the north-south direction (1, 2). Rhea’s surface temperature ranges from about 40 to 100 K (3) and exhibits a high visible geometric albedo of 0.949 ± 0.003 (4). This albedo is consistent with a surface composed mostly of water-ice, a hypothesis supported by measurement of infrared (IR) absorption features (5). Rhea orbits Saturn at a radial distance of about 8.75 Saturn radii with a velocity of 8.5 km s⁻¹, which is much slower than the co-rotating plasma velocity of 86.3 km s⁻¹. Thus, the moon’s trailing hemisphere (centered on 270°W) is irradiated by plasma traveling at ~57 km s⁻¹; E-ring grains also bombard and coat much of the surface of Rhea with a focus on the leading hemisphere (6). Such bombardment, from different sources, may lead to chemical changes in the irradiated surface and subsequent synthesis of a rich surface chemistry (7) while also explaining the chemical composition of Rhea’s tenuous atmosphere of O₂ and CO₂ (8). However, the surface composition of Rhea’s largely remains unknown (9). Here, we address the broad unidentified absorption band centered at 184 nm that was observed in far-ultraviolet (FUV) spectra recorded by Cassini-Ultraviolet Imaging Spectrograph (UVIS) during Rhea’s flybys.

Rhea phase curves derived from Voyager disk-integrated and disk-resolved observations at a wavelength of 0.48 µm show an asymmetry in photometric properties between the leading and trailing hemispheres possibly due to differences in the Hapke roughness parameter between the leading and darker trailing hemisphere (10). An asymmetry between Rhea’s brighter leading and darker hemisphere is also seen in the far ultraviolet (FUV) spectra acquired by the UVIS/FUV instrument on the Cassini spacecraft. A similar asymmetry between leading and trailing hemisphere seen in the phase curve and FUV reflectance spectra is also present in Saturn’s other mid-sized icy satellites, Dione and Tethys (11). Cassini Imaging Science Subsystem (ISS) IR/Green/UV composite maps of Rhea, Dione, and Tethys also show an asymmetry between leading and trailing hemispheres (9). The darkening in the UV on these icy moons is plausibly caused by radiolytic reactions due to a corresponding asymmetry in irradiation by energetic particles of Saturn’s magnetosphere (12). Molecules present in ice phase can be dissociated by such radiation, which can lead to the formation of different molecules that absorb in FUV, including an absorber centered at 184 nm.

Cassini UVIS observations and the 184-nm band

In this study, we use four Cassini UVIS/FUV disk-resolved observations of Rhea. The locations of the observations are shown in Fig. 1 with the observational geometry outlined in Table 1. The observations sample regions on the leading (yellow and blue slit field of views), trailing (green slit field of view), and anti-Saturnian (cyan slit field of view) hemispheres on Rhea. The UVIS spectra from the four observations of Rhea are shown in Fig. 2. To reduce noise in the UVIS data, a Savitzky-Golay smoothing filter was applied to each UVIS spectrum (13, 14). Tests using Savitzky-Golay smoothing procedure showed it to perform better than basic smoothing filters based on box averaging and running means. All spectra are dominated by water-ice as indicated by the steep spectral slope centered near 165 nm, with a strength that depends on the abundance of water-ice and a wavelength that is dependent on the size of the ice grains and degree of minor contaminants. However, spectra also show a broad, weak absorption feature from approximately 179 to 189 nm, centered near 184 nm. This absorption feature has been noted in previous observations of other icy satellites [e.g., Phoebe (15)] in the Saturnian system, but, to date, its origin has not been explained satisfactorily (15). Attempts to fit Phoebe’s 184-nm absorption feature, using various mixtures of tholin and ice, water-ice and carbon, water and kerogen, and water plus hydrogen cyanide (15), have proven to be unsuccessful. Roeyer et al. (16) discuss the 184-nm feature and point out its appearance, in some (but not all) observations of Mimas, Enceladus, Tethys, and Rhea. Therefore, in this work, we explore explanations for the broad absorption seen over the wavelength range of ~179 to 189 nm in UVIS spectra of Rhea.

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1Department of Physical Sciences, The Open University, Milton Keynes MK7 6AA, UK. 2Atomic Molecular and Optical Physics Division, Physical Research Laboratory, Ahmedabad 380 009, India. 3Planetary Science Institute, Pasadena, CA 91106, USA. 4National Synchrotron Radiation Research Center, Hsinchu, Taiwan. 5B-1, Indus-1, Atomic and Molecular Physics Division, Bhabha Atomic Research Centre at RRCAT, Indore, India. 6School of Physical Sciences, The University of Kent, Canterbury CT2 7NH, UK. *Corresponding author. Email: elowitzt@earthlink.net (M.E.); bhala@prl.res.in (B.S.) †Present address: Department of Medical Research, Huaiilen Tzu Chi Hospital, Buddhist Tzu Chi Medical Foundation, Huaiilen 970, Taiwan.
RESULTS

Using laboratory spectra to model Cassini UVIS observations

During the course of this work, we measured laboratory spectra of several molecular species (see table S1) and their mixtures to derive optical constants for use in Hapke modeling of mixtures. Limited phase cover observations did not permit a satisfactory derivation of Hapke parameters needed to produce model spectra. Instead, attempts to use limited phase angle coverage resulted in a wide range of Hapke parameters that fit UVIS phase curve data equally well within statistical error of the data. Solutions based on Hapke model results can be nonunique when photometric measurements are too sparse. In other words, the phase angle data available for Rhea are too limited in their coverage to constrain Hapke parameters (e.g., coherent backscatter opposition, asymmetry of the phase function of the ice grains, and macroscopic roughness parameter). As a result of these aforementioned problems, an alternative methodology was used to obtain Hapke parameters.

Since Rhea and Dione share similar geomorphology, based on Cassini high-resolution ISS imagery, and both mid-sized icy satellites have O$_2$/CO$_2$ exosphere (8, 17), it is assumed that these two satellites have similar compositional and photometric properties. Both Rhea and Dione exhibit brighter leading hemispheres and show very little limb darkening at low phase angles (18). Brighter hemispheres are most likely caused by a deposition of pure water-ice from Saturn’s E-ring (19) and both Rhea and Dione have similar photometric properties (based on similarities in the shape of the phase curves) and orange/violet color ratios, implying similar surfaces. Therefore, we chose to use Hapke parameters derived for Dione’s leading hemisphere (11). The asymmetry parameter for the width of the forward and backward scattering lobes, and the relative amplitude of the scattering lobes is denoted by $b$ and $c$, respectively, where $b = 0.35$ and $c = 1.0$. The roughness parameter (in radians) is denoted by $S$. The terms $h$ and $B_0$ characterize the shadow-hiding opposition surge, where $h = 0.457$ and $B_0 = 0.95$.

Resultant model spectra of hydrazine monohydrate (N$_2$H$_4$.H$_2$O) and trichloromethane (CHCl$_3$) below a layer of water-ice approximately 0.125 μm thick are shown in Fig. 2 with the UVIS spectra. Spectral models using N$_2$H$_4$.H$_2$O, CHCl$_3$ below water-ice, and CCl$_4$ below water-ice were created using laboratory absorbance measurements and Hapke theory (15). The amount of non–water-ice component was 5%. For reference, a model of pure water-ice is also shown in the comparison plots (Fig. 2).

Examination of the modeled spectra in Fig. 2 clearly shows that N$_2$H$_4$.H$_2$O or simple chloromethane molecules could explain the weak, broad absorption seen between approximately 179 and 189 nm. To
compare the relative strengths of absorption, we remove the continua of each spectrum to show four normalized spectra of Rhea. These results are shown in Fig. 3. We do not see any significant variation in the band strength with observation or location on Rhea.

**DISCUSSION**

To determine which chemical compound (N₂H₄·H₂O or simple chlorine compounds) may be responsible for producing weak absorption from ~179 to 189 nm, we explore possible sources and sinks of each molecular species. Results of the modeling imply that the best agreement between the derived observational UVIS spectra and the modeled spectra occurs (i) with a chlorine-containing ice layer located just below a (~125 nm) layer of water-ice or (ii) when hydrazine is mixed with water in the form of a monohydrate. H₂O and CCl₄ are immiscible, because CCl₄ cannot form hydrogen bonds to water. On Rhea, there might be a source for CCl₄ (e.g., endogenic), with, at a later time, a fresh layer of water-ice being delivered on top of the CCl₄ from Saturn’s E-ring. Since E-ring material could coat both hemispheres of Rhea, it would explain why CCl₄ below water-ice is seen in all UVIS spectra analyzed in this research. We obtain an equivalent thickness of water-ice of ~1.2 × 10⁻¹⁰ m/year or about 1.2 × 10⁻⁴ µm/year. Since UV reflectance spectroscopy is sensitive to only the upper few micrometers, we see that a layer of chloromethane compounds beneath a deposited water-ice could be readily detectable.

However, chlorine compounds are somewhat difficult to explain via chemical pathways, and their origin on Rhea would require the presence of an internal ocean layer or exogenic delivery by micrometeoroids and/or asteroids that contain chlorine. It should also be noted that CHCl₃ and CCl₄ have melting points of 209.7 and 250.2 K, respectively. Thus, if these compounds exist deep in the interior of...
Rhea, they would depress the freezing point of water-ice increasing
the probability of an aqueous layer. Chlorine-based salts (e.g., NaCl)
have been detected in plumes of Enceladus (20), which provide evi-
dence for an internal ocean; however, it is not likely that chlorine
compounds could migrate to the surface of Rhea through cracks in
the ice shell (assuming Rhea is partially differentiated) owing to the
great depth of such a liquid layer as determined through numerical
models (21). The only other possible source of chlorine is via exogenic
delivery by chondritic asteroids over the history of Rhea. Condensed
chlorine compounds on Rhea’s surface could be redistributed to
other regions of the satellite by sputtering induced by charged
particles from Saturn’s magnetosphere. This could, in turn, explain
wide distribution of chlorine compounds as sampled by UVIS ob-
servations. Distribution of the observed chlorine species across
Rhea’s surface, in theory, should be determined by velocity distribu-
tion of sputtered molecules. If chlorine is delivered to the surface of
Rhea, then it might be possible for free radical halogenation to occur via
irradiation of UV photons and/or charged particles. This process
could lead to molecular compounds with increased chlorination.

The main unresolved questions on this scenario is whether there are
any barriers to these reactions at the lower temperatures en-
countered on the surface of Rhea. Furthermore, there is no evidence of
CH4 trapped within the ice matrix on Rhea. To synthesize CCl4,
the hydrogen in CH4 has to be replaced by Cl and HCl is the by-
product in every step of replacing one hydrogen atom. Since HCl is also
produced in the production of simple chloromethane com-
 pounds, it should also be present in the surface ice on Rhea. However,
comparisons of the UVIS observations presented in this paper with
laboratory thin ice measurements of deuterium chloride (DCl)
(Fig. 4) suggest that no HCl is present on Rhea. Furthermore, pro-
duction of CO2 is also unlikely since it would require the presence of
CCl4. For these reasons, the focus of the remainder of this paper
will be exploring the possible sources of N2H4·H2O on Rhea.

The production of N2H4·H2O is easier to explain than the pres-
ence of any chlorine-containing derivatives as it can be produced in
chemical reactions involving water-ice and ammonia, or possibly
delivered from Titan’s nitrogen-rich atmosphere where the com-
 pound could be synthesized. The possibility of contamination of the
UVIS data by a hydrazine propellant from Cassini spacecraft was
considered but we feel is highly unlikely, since the hydrazine thrusters
were not used during icy satellite flybys. Furthermore, there are nu-
merous observations of all icy Saturnian satellites that do not show
the 184-nm feature. For instance, several disk-integrated spectra ac-
quired from Tethys show no indication of absorption near 184 nm (Fig. 5).
Furthermore, a calibration UVIS/FUV spectrum of the star epsilon
CMa was obtained during the T41 I occultation on 23 February 2008
(22). The spectrum shows no indication of a broad absorption feature
between ~179 and 189 nm. Thus, the presence of such a 184-nm fea-
ture is confirmed as being specific to Rhea surface observations by
the Cassini spacecraft and is not a result of contamination from
hydrazine fuel on the UV spectrometer.

Irradiation of ammonia by charged particles from Saturn’s mag-
netosphere may induce dissociation of NH3 molecules to synthesize
N2H2 and N2H4. Lyman alpha photons with an energy of 10.2 eV
(121.6 nm) can also dissociate NH3, leading to the production of
N2H4 (23). The possibility of hydrazine synthesis in Titan’s atmo-
sphere has also been investigated in laboratory experiments carried
out by Zheng et al. (24). The team performed laboratory experi-
ments to investigate synthesis of hydrazine from irradiation of NH3
ice over a temperature range of 10 to 60 K, where two NH2 radicals
from NH3 dissociation synthesize N2H4. Initial IR spectrum could
not identify absorption bands due to N2H4 spectra overlapping NH3
features. However, during the warm-up phase of the experiment,
hydrazine absorption features became visible in the IR spectrum.
The experiment conducted by Zheng et al. (24) showed that strength
of hydrazine absorption bands increases as ammonia sublimes.

The source of NH3 could be primordial, incorporated into the
interior of Rhea during its formation and brought to surface during
a period when Rhea had endogenic activity. Evidence for past re-
 gional endogenic activity on Rhea is apparent in Cassini ISS imagery
in the form of morphological features, including relaxation of large
impact craters, curvilinear grabens, and orthogonal ridges. In con-
trast, transfer of the ammonia observed in plumes of Enceladus to
Rhea’s surface is unlikely due to Rhea’s distance from Enceladus
and the instability of NH3 to photo- and radiolytic dissociation. If
Rhea is a uniform mixture of rock and ice (undifferentiated), then
minor amounts of ammonia may be present over a large percentage
of its surface. This would explain why hydrazine appears to be present
over a geographically wide area as sampled by the UVIS observations.
However, NH3 is unlikely to survive on the surface of Rhea indefi-
nitely. Furthermore, there are several processes that can reduce
production rate or prevent synthesis of hydrazine within the upper
surface ice layer of Rhea.

Since water-ice is most likely to host NH3 ice, the probability of
UV photons, with energy 10.2 eV (121.6 nm) or less, destroying NH3
decreases because of the strong absorption of Lyman-alpha photons
by the water-ice matrix. This scenario applies for the case where
NH3 is located beneath water-ice, and the photon path length is short
enough to allow sampling by UV/IR spectroscopy. Other impurities,
such as hydrazine or chlorine, may also be shielded against destruc-
tion by UV photons beneath a thin layer of water-ice. Lower-energy
(<6.05 eV or <205 nm) radiation, however, can penetrate further
into the surface ice layer, with the potential to destroy any underly-
ing impurities, such as NH3, chlorine, and hydrazine compounds.
Instability due to longer wavelength photons may explain why no

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**Fig. 4.** Deuterium chloride spectrum compared with a Rhea FUV spectrum. Error,
±6% for the observational data and ±5% for the experiment data, not added to
spectra for clarity.
N₂ has been detected on Rhea over the wavelength range sampled by Cassini UVIS instrument. Ammonia could, in theory, be more stable if it exists on icy satellites in the form of a hydrate mixture—i.e., ammonia in the form of a hydrate will minimize energy in thermodynamic equilibrium. Concentration of any hydrazine synthesized in chemical reactions between NH₃ and H₂O will depend on several factors, including dissociation rate of NH₃ by energetic electrons from Saturn’s magnetosphere and localized heating due to micrometeoroid bombardment. Localized heating will tend to bring fresh NH₃ to the surface, where it is susceptible to destruction by radiolysis. NH₃ sublimation due to localized heating can also occur since NH₃ is more volatile than water-ice. For these reasons, we look for an exogenic source of hydrazine. A possible exogenic source of hydrazine would be from another moon of Saturn with an atmosphere capable of synthesizing the chemical compound.

Saturn’s largest satellite Titan orbits at approximately 20 Saturnian radii (exterior to Rhea) with an orbital period of 15.95 days. Titan’s thick atmosphere is dominated by N₂ with minor amounts of hydrocarbons including CH₄. If N₂ or N₂H₄, since 4H does not add a significant atomic mass to the hydrazine molecule) can escape from Titan’s atmosphere, then it is possible that Rhea could accumulate it as it orbits Saturn. Rough estimates show that Jean’s escape mechanism is not sufficient to eject hydrazine from Titan’s atmosphere. Thus, an alternative ejection mechanism is required to eject N₂H₄ from Titan’s atmosphere. Sputtering is a nonthermal process that is an effective escape mechanism on small solar system bodies (e.g., Titan and Mars) that have no inherent magnetic fields. Experiments conducted by De La Haye et al. (25) lead to a sputtering rate for N₂ on Titan of ~3.6 × 10²⁸ atomic mass units/s (59.78 kg/s). Thus, over a geological timespan of 4 billion years, the amount of N₂ ejected from Titan’s atmosphere due to sputtering would be ~7.5 × 10¹⁸ kg or 5.6 × 10⁻⁵ Titan masses. This is equivalent to the mass of some of the more massive asteroids. It is also important to note that this is an upper limit estimate that assumes that N₂ or N₂H₄ has existed in Titan’s atmosphere over the last 4 billion years and that it does not get destroyed before being accumulated onto Rhea’s surface. We should, however, note that Rhea’s surface temperature is not supportive to harbor pure N₂ or CH₄ to remain on the surface; however, they may be present in low concentration in water-ices but because of the low absorption cross section of the N₂ and CH₄ absorption band coinciding with the water absorption below 145 nm (26), we are unable to conclude the presence of these molecules in the FUV spectra. Future research work on the details associated with the production, ejection, and transfer of hydrazine from Titan’s atmosphere are, therefore, needed to assess whether satellite-to-satellite transfer of materials can explain the possible presence of N₂H₄H₂O on Rhea, and why the feature appears in the spectra of some satellites sometimes, and not others (e.g., (16)).

The first detailed geochemical survey of Rhea’s icy surface in the FUV region indicates the possible presence of chloromethane compounds under a layer of water-ice or the presence of a N₂H₄H₂O complex. The presence of CCl₄ or CHCl₃ beneath a thin layer of water-ice is more difficult to explain, and we therefore suggest that N₂H₄H₂O is the more likely candidate for the observed UV spectral feature at 184 nm, since it can be produced by chemical reactions induced by irradiation processes on molecular ices containing water and ammonia. If ammonia is present within the icy upper layer on Rhea, then the source of N₂H₄H₂O might be indigenous in nature. It is also possible that hydrazine could be synthesized in the atmosphere of Saturn’s largest moon Titan and transferred to Rhea over geological timespans.

**MATERIALS AND METHODS**

**Observations and data reduction**

The Cassini UVIS is a spatial-spectral instrument (1024 spectral channels and 64 spatial elements) used to collect UV data over two spectral channels. The extreme UV and FUV channels cover the wavelength ranges of 56 to 118 nm and 110 to 190 nm, respectively, providing spectral resolutions of 0.24 and 0.48 nm. The entire slit length is 64 mrad. The Rhea spectra discussed here were collected using the FUV channel, and we focus on the ~150- to 190-nm region for the geochemical study of Rhea’s surface since shorter wavelengths did not have sufficient signal-to-noise ratio (SNR) to allow chemical analysis.

Cassini UVIS/FUV datasets for the Rhea 2007, 2010, and 2011 targeted flybys were extracted from the NASA PDS archive. One constraint used in selecting the datasets is that the instrument footprint is completely filled with Rhea’s surface. Another constraint is selecting the highest signal-to-noise spectra over the leading and trailing hemispheres of Rhea to provide the best chance of fitting model spectra. A table of the Cassini UVIS observations presented in this paper is listed in Table 1.

Each UVIS raw spectrum is converted to geophysical units by applying a unique set of calibration matrix coefficients to the data (27). All UVIS observations are corrected for background signals from the Cassini spacecraft radioisotope thermoelectric generators. The SNR of the calibrated spectra is increased by summing over a given number of spatial elements. Each calibrated spectrum is then converted to I/F (reflectivity) by dividing by a solar spectrum (scaled to the heliocentric distance of Rhea) obtained from the SOLSTICE.
instrument on the SORCE spacecraft (28) for the given date of the UVIS observation, corrected for sub-solar longitude. The I/F spectrum can be mathematically expressed by

\[ \frac{I}{F} = \frac{P}{(S/\pi)} \]

where \( P \) is the calibrated UVIS spectrum and \( S \) is the solar flux spectrum (scaled to the heliocentric distance of Rhea on the date of the observation), where \( S = \pi F \). Each I/F spectrum is smoothed using a running mean method to reduce noise and allow easier comparisons with laboratory reference spectra.

The average SNR of the UVIS data presented in this work was calculated using a statistical method that is robust with respect to the presence of any outliers. The method does not depend on assumptions and/or decisions made by the user, since the signal is the median of the reflectance, and the noise calculation is based on the standard deviation of a Gaussian assuming a normally distributed variable. An advantage of this method is the simplicity used in the algorithm for calculating the SNR, which outweighs the slight increase in precision used by methods that depend on full information about the specific sensor. Since the method is generic in nature, the SNR calculated for spectra can be compared across different instruments. According to the Spectrum Data Model of the International Virtual Observatory Alliance, the statistical method used to calculate the SNR of the UVIS data is becoming the standardized method of computing the SNR of spectral data. For the UVIS observations presented in this paper, the average SNR is approximately 8.8 (near 180 nm). This SNR is approximately the same for all observations.

To determine the location of the UVIS instrument footprints on Rhea’s surface, JPL NAIF SPICE (Space Planet Instrument Camera-matrix Events) kernels were used to calculate the relative positions of the Cassini spacecraft and Rhea, the orientation of the spacecraft and Rhea, and instrument pointing data at the time of each observation. The projection of the UVIS/FUV slit on Rhea’s surface was plotted using the “Geometry” software, developed by the Cassini UVIS instrument team. The latitude and longitude calculated by the Geometry IDL code is used to produce an image of Rhea’s surface with a projection of the UVIS instrument footprint for the observations presented in this paper. The “Environment for Visualization of Images” software package was used to produce the final UVIS/FUV slit field-of-view plots.

**Experimental methodology**

Experiments simulating Rhea conditions were carried out in an ultrahigh-vacuum chamber housing a cold finger on which a lithium fluoride (LiF) window was attached. The chamber pressure went down to 5 x 10⁻⁹ mbar while temperatures down to 10 K were achieved at the LiF window. This entire system was attached to the end station of the 03A1 High Flux beamline at the National Synchrotron Radiation Research Center (NSRRC), Taiwan. Vacuum UV (VUV) light in the 110- to 200-nm range was used to probe the molecular ices that are formed at low temperatures. To obtain the photoabsorption spectra of simulated molecular ices, a spectrum of the LiF window, \( I_o \), was recorded at 10 K. Then, after sample deposition, another spectrum was recorded, \( I \). The photoabsorption spectrum was then obtained using the Beer-Lambert law.

Molecules from liquid samples of hydrazine monohydrate, trichloromethane, tetrachloromethane, and water were extracted into the gas phase before the gases were allowed into the chamber to form molecular ice films. In the case of layered ices, trichloromethane and tetrachloromethane were first deposited, respectively, above which water molecules were deposited, whereas in the case of mixed ices, a 1:1 mixture of trichloromethane/tetrachloromethane with water was prepared in the gas phase before deposition. All the samples were deposited at 10 K and were later warmed to higher temperatures (mostly covering Rhea diurnal temperatures) for spectral recording. Apart from the molecules given above, we have compared the known VUV spectra of various astrochemical ices (table S1) (26, 29-38) with that of the Rhea spectra.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/7/4/eaba5749/DC1

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Mark Elowitz, Bhalamurugan Sivaraman, Amanda Hendrix, Jen-Iu Lo, Sheng-Lung Chou, Bing-Ming Cheng, B. N. Raja Sekhar and Nigel J. Mason

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