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Vertical Mixing and Methane Photochemistry in the Atmosphere of Uranus: Analysis of Voyager UVS Occultation Experiments

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This document is intended to be the final report for NASA basic research grant NAGW-1504, entitled "Vertical Mixing and Methane Photochemistry in the Atmosphere of Uranus: Analysis of Voyager UVS Occultation Experiments". In January 1986, the Voyager 2 spacecraft flew past Uranus and obtained one solar occultation (ingress) and three stellar occultations (Y-Peg ingress and egress, and v-Gem glancing) with the ultraviolet spectrometer (UVS) instrument [Broadfoot et al., 1986]. A modeling analysis of the occultation results was presented by Herbert et al. [1987]. The Herbert et al. paper attempted to cover the occultation results in all aspects, which resulted in a rather broad-brush approach to the treatment of the UVS lightcurves that probed the Uranian stratosphere. In view of the significance of measurements of hydrocarbon abundances for the determination of atmospheric structure (in particular, the strength of eddy mixing) and the characterization of the observed hazes [Pollack et al., 1987], we undertook a more careful reanalysis of the UVS lightcurves probing the stratosphere (wavelengths > 130 nm). The solar occultation in particular proved to be remarkably "clean" (i.e., possessing a relatively large signal-to-noise ratio S/N so that a model-independent analysis was possible [Bishop et al., 1990]. In fact, the analysis presented in Bishop et al. [1990] would appear to be unique among analyses of UVS occultations obtained throughout the voyages of Voyager 1 & 2, in its confrontation of measurement vs. photochemical models (as opposed to relying on photochemical models in the analysis itself). This is in part due to the relatively high S/N of the data and in part to the clarity of the stratosphere itself, reflecting a relatively simple photochemistry.

Details of the analysis are given in Bishop et al. [1990]. Briefly, it was possible to use the UVS solar occultation lightcurves  $I/I_0$  at wavelengths > 153 nm to characterize the background atmosphere over the same altitude range as where hydrocarbon abundances were determined using lightcurves at shorter wavelengths, illustrated in Figure 1 (where altitude is with respect to the 1 bar radius determined by RSS occultation measurements [Lindal et al., 1987]). The longer wavelength lightcurves (see Figure 1, top panel) are dominated by  $H_2$ Rayleigh scattering, so that Abel inversion of the lightcurves yielded directly the profile of H<sub>2</sub> number density as a function of altitude. Prior to inversion, a new "optimal" filtering procedure was applied to the data, which removed the effects of instrument noise and the finite width of the solar disk (effective diameter ~82 km as seen from the spacecraft). The implied total number density profile (assuming a helium mixing ratio of 0.15) is shown in Figure 2a, along with the RSS profile at lower altitudes. At wavelengths 145–153 nm (Figure 1, central panel), acetylene  $(C_2H_2)$  photoabsorption constitutes a major source of opacity, identified by the strong distinctive 3R-X Rydberg transitions at 151.9 and 147.8 nm. The 145-153 nm lightcurves were inverted to obtain C<sub>2</sub>H<sub>2</sub> number densities, using the longer wavelength results to correct for the H<sub>2</sub> Rayleigh scattering opacity. Proceeding next to shorter wavelengths (135–145 nm, bottom panel of Figure 1), ethane ( $C_2H_6$ ) densities were similarly extracted from the observed opacities (with perhaps some contribution from  $CH_4$ ), after adjustment for  $C_2H_2$  and  $H_2$  opacities.



Thus, it proved possible to derive partial mixing ratio profiles for  $C_2H_2$  and  $C_2H_6$  spanning roughly one atmospheric scale height that were entirely independent of modeling procedures. These are shown as the data points in Figures 3a,b. Bishop et al. [1990] went on to use the H<sub>2</sub> Rayleigh scattering results to construct an empirical stratosphere model in combination with the model defined at deeper levels (pressures > 2 mbar) by the Radio Science Subsystem (RSS) occultation results [Lindal et al., 1987] and with the UVS thermosphere model (pressures < 0.1 µbar) from Herbert et al. [1987]; this model is shown in Figure 2b as the composite profile with an upper temperature of 130 K. This stratosphere p-T model was then used in calculating 1-D photochemical models of equatorial stratospheric hydrocarbon abundances on Uranus for comparison







with the UVS results. The observed acetylene mixing ratios were found to be consistent with values of the eddy mixing coefficient at the methane homopause in the range 103-104 cm<sup>2</sup> s<sup>-1</sup>, depending on the manner with which the eddy mixing coefficient K (which parameterizes the strength of vertical mixing driven by mechanisms other than molecular diffusion) was assumed to vary with background atmospheric number density. For the "conventional" dependence on the inverse square root of the total number density,  $K_{\rm h} = 1-2 \times 10^3 \text{ cm}^2 \text{ s}^{-1}$  is indicated, as shown in Figure 3a. However, the fall-off with decreasing pressure of the observed mixing ratio profile as seen in this Figure is less steep than the theoretical profiles; furthermore, the measured C<sub>2</sub>H<sub>6</sub> mixing ratios are much larger than the theoretical values (Figure 3b). These two pieces of information suggest that the one-dimensional picture of vertical mixing encapsulated by the eddy mixing coefficient used in these models is too simple-minded. Ethane is more stable in a photochemical sense than acetylene, in that the timescales for photochemical change in  $C_2H_6$  abundances in the stratospheres of the giant planets are longer than for  $C_2H_2$ . Thus, a large-scale motion of the atmosphere imposed on localized mixing (e.g., a general circulation as proposed by Flasar et al. [1987]) could perturb hydrocarbon mixing ratios away from a steady state model in the manner suggested by the observations; for example, a slow upward movement in the equatorial region could "lift" ethane mixing ratios as shown in Figure 3b while keeping acetylene mixing ratios closer to their photochemical equilibrium values (Figure 3a). Obviously, further work is warranted.

The work funded under this grant was nominally for two years. The work outlined above and presented in *Bishop et al.* [1990] basically represents the results of the first year of work. We decided, however, to postpone the work we had planned pertaining to Uranus for the second year, in order to focus our efforts on analyzing the UVS occultation results from the Neptune encounter, in particular the solar occultations of the Neptune atmosphere at wavelengths > 125 nm (analogous to the Uranus work). We felt justified in this, not only in view of the timeliness for the Neptune encounter (closest approach occurred on 25 August 1989), but also in that a postponement in further Uranus work would allow us to later carry out a more careful comparative analysis utilizing both Uranus and Neptune datasets. There have been difficulties with the Neptune work, however, particularly in the preparation of a "final" set of occultation data. This has slowed the appearance of the results of our analyses in publication form, although results have appeared in the form of conference presentations.

The Neptune solar occultation lightcurves (both ingress and egress occultations were acquired [*Broadfoot et al.*, 1989]) at wavelengths longer than hydrogen Lyman- $\alpha$  (121.6 nm) probe the stratosphere as on Uranus, but indicate a markedly different stratosphere. On Uranus, weak eddy mixing was the order of the day, and the UVS lightcurves at wavelengths 130–170 nm all probed the stratosphere above the CH<sub>4</sub> photochemical peak, resulting in considerable overlap in altitude of the lightcurves. On Neptune, however, the lightcurves transecting the stratosphere fall into 3 groups. At wavelengths < 130 nm, the lightcurves

closely resemble one another, exhibiting half-light points in the 0.3 µbar region where the methane photolysis peak lies, while those at longer wavelengths (135-155 nm) probe progressively deeper into the stratosphere and those at wavelengths > 155 nm closely resemble one another with half-light points near 0.2 mbar. Thus, eddy mixing coefficients of the order 107 cm<sup>2</sup> s<sup>-1</sup> near the methane photolysis peak are suggested, and the characterization of hydrocarbon abundances over an extensive pressure interval (0.1 µbar to 0.1 mbar) is possible with the UVS data. We have finally settled on the tack of presenting the shorter wavelength (125-138 nm) results first, which are dominated by methane photoabsorption, and then to present the results at longer wavelengths in a subsequent publication. The shorter wavelength results (for both ingress and egress occultations) have been submitted for publication and we are currently winding up the revision. We anticipate presenting comparative ingress-egress results at longer wavelengths at the upcoming special meeting on the Voyager Neptune and Triton encounters in Tucson in January 1992. Here a summary of the shorter wavelength results for the ingress occultation will be given.





In Figure 4, the UVS lightcurves used in the analysis are shown, as  $I/I_o vs$ . altitude above the 1 bar level as placed by RSS measurements [Lindal et al., 1990]. These data are too noisy to permit use of the filtering-inversion technique used with the Uranus dataset, so that the model-dependent procedure of lightcurve simulation has been adopted. The main uncertainty in modeling the CH<sub>4</sub>dominated lightcurves is in specifying the background atmosphere, in that these lightcurves probe pressures in the information gap between the pressure regimes mapped by the RSS (pressures > 1 mbar [Lindal et al., 1990]) and UVS (pressures  $< 0.03 \mu bar [Broadfoot et al., 1989]$ ) occultations (the latter at wavelengths in the H<sub>2</sub> Lyman-Werner band systems). In Figure 5a we show fitted lightcurves (identified by UVS channel center wavelength) using a model atmosphere consistent with what is known about the Neptune stratosphere structure from RSS, IRIS [Bezard et al., 1991], and UVS sources, and from ground-based sources [Hubbard et al., 1985, 1987; French et al., 1985]. The photochemical models for the simulated lightcurves are characterized in terms of the adopted profile for eddy mixing. We illustrate here (Figure 5b) models wherein K is constant with height in the altitude range probed by the lightcurves (model "b", solid curves in Figure 5a) and models wherein K increases in inverse proportion to the square root of the atmospheric number density (model "a", dashed curves in Figure 5a). (Note: the simulated lightcurves include opacity arising from H<sub>2</sub> Rayleigh scattering and photoabsorption by  $C_2H_2$ ,  $C_2H_4$ , and  $C_2H_6$ , but at these pressures and wavelengths methane is the dominant source of opacity.) Also shown are models in which the methane mixing ratio in the lower stratosphere is held to saturation-limited ("cold-trap") values (3  $\times$  10-5, models "s"), based on the cold tropopause temperatures (50-55 K) revealed by the Voyager RSS occultations and IRIS soundings [Lindal et al., 1990; Bezard et al., 1991], and models in which methane is present in amounts suggested by pre-Voyager analyses of ground-based IR brightness observations (3 × 10-3, models "h") [Orton et al., 1987, 1990; Bezard et al., 1991]. The strength of eddy mixing near the CH<sub>4</sub> photolysis peak in these models was varied to try to yield the correct placement of the half-light points over the 125-138 nm wavelength interval (over which methane photoabsorption cross sections vary appreciably). Model-data lightcurve comparisons are shown in Figure 5c for the "s" and "h" models. It can be seen that the saturation-limited lightcurves (dashed curves) are too laid-back and are not consistent with the UVS data over the 125-138 nm wavelength interval, being too widely spaced; the models suggested by the ground-based IR results (models "h", solid curves), on the other hand, are too steep and are too closely spaced. The uncertainties in atmospheric structure have been explored and are not enough to compromise the conclusion suggested by the results shown in these figures. Namely, the UVS lightcurves (both ingress and egress) indicate that methane is present in the Neptune stratosphere in amounts  $1-5 \times 10^{-4}$ , in excess of what would be expected using simple cold-trap arguments. The ground-based IR brightness results are also called into question. Indeed, the IR brightness results are extremely sensitive to the assumed temperature profile, and are possibly plagued with calibration uncertainties [Orton et al., 1990]. By combining these UVS analysis results with the ground-based IR brightness measurements, it should now be possible to turn the IR analysis procedure around and use the IR observations to determine the temperature structure in the lower stratosphere.

These results on narrowing the range of uncertainty in methane mixing ratios in the lower stratosphere and the strength of eddy mixing near 0.3 µbar are not dependent on photochemical details; basically, methane is transported upward from the troposphere and is photolyzed by Lyman- $\alpha$  near the 0.3 µbar level. (*Note*: a large fraction of the incident Lyman- $\alpha$  flux driving the CH<sub>4</sub> photodissociation at Neptune comes from the local interstellar medium (LISM), which is included in the modeling using LISM Lyman- $\alpha$  intensity measurements obtained by the UVS near the time of the Neptune encounter.) The UVS lightcurves at longer wavelengths are dominated by stable product C<sub>2</sub> hydrocarbons (acetylene, ethylene, ethane) and H<sub>2</sub> Rayleigh scattering, and unraveling these separate sources of opacity requires extensive photochemical modeling. Preliminary results have been presented at the 1991 Spring AGU Meeting, and completion of this aspect of the analysis of the Neptune UVS solar occultation datasets will occupy our efforts in the near future.

In conclusion, the work supported by this grant has resulted in the development of extensive capabilities in the analysis of UVS absorptive lightcurves, independent of other centers (e.g., Tucson), and the application of these capabilities to the Voyager UVS datasets from Uranus and Neptune has provided significant findings regarding the stratospheres of these planets. In particular, the direct comparison between photochemical models and UVS measurements accomplished by our efforts is unique, and helps guarantee that the information returned by the Voyager 2 spacecraft is being utilized to the fullest extent possible. Future work on the Uranus dataset will focus on melding our work with results from other investigators and will hopefully lead to our being able to address the hypothesis of latitudinal variations in a direct manner. Future work on the Neptune datasets will permit us to investigate the question of latitudinal variations on that planet, and to conduct a thorough comparison with Uranus.

Lastly, I would like to thank the American people for supporting this work.

PUBLISHED PAPERS deriving from support under this grant:

"Reanalysis of Voyager 2 UVS Occultations at Uranus: Hydrocarbon Mixing Ratios in the Equatorial Stratosphere", by J. Bishop, S. K. Atreya, F. Herbert, and P. Romani, *Icarus* 88, 448–464 (1990).

SUBMITTED PAPERS deriving from support under this grant:

"Voyager 2 UVS Solar Occultations at Neptune: Constraints on Stratospheric Hydrocarbon Abundances and Comparison with Photochemical Models", by J. Bishop, S. K. Atreya, P. N. Romani, F. Herbert, and B. R. Sandel, submitted to J. Geophys. Res.

PRESENTATIONS AT MEETINGS deriving from support under this grant:

"Reanalysis of Voyager 2 UVS Solar Occultation Data: Uranian Hydrocarbon and Eddy Mixing Distributions", J. Bishop, S. K. Atreya, F. Herbert, and P. Romani, 1989 DPS Meeting, abstract: *Bull. Am. Astron. Soc.* **21**, p. 918 (1989).

"Hydrocarbon Abundances in the Neptune Stratosphere from the Voyager UVS Ingress Solar Occultation", J. Bishop, S. K. Atreya, F. Herbert, B. Sandel, and P. Romani, 1990 DPS Meeting, abstract: *Bull. Am. Astron. Soc.* 22, p. 1104 (1990).

"Hydrocarbon Abundances in the Neptune Stratosphere: Latitudinal Variations from Voyager UVS Solar Occultations", S. K. Atreya, J. Bishop, and P. N. Romani, 1991 Spring AGU Meeting, abstract: EOS 72, p. 185 (1991).

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