

## Abundances of ammonia and carbon disulfide in the Jovian stratosphere following the impact of comet Shoemaker-Levy 9

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**Abstract.** Radiative transfer-scattering models were used to fit the Hubble Space Telescope UV spectroscopic observations of Jupiter taken 2.5 hours after the impact of fragment G of comet P/Shoemaker-Levy 9. They yield abundances for NH<sub>3</sub> and CS<sub>2</sub> of  $(0.25-1.3) \times 10^{16} \text{ cm}^{-2}$  and  $(0.43-1.1) \times 10^{15} \text{ cm}^{-2}$ , respectively, at unit optical depth level in the part of the atmosphere above the G-impact site. This range in the abundances is caused by the uncertainty in the composition of the aerosol material and the atmospheric pressure level where the aerosol unit optical depth in the UV occurs. The upper limits of H<sub>2</sub>S, PH<sub>3</sub>, C<sub>2</sub>H<sub>2</sub>, and C<sub>6</sub>H<sub>6</sub> at the unit optical depth level are  $1.2 \times 10^{16} \text{ cm}^{-2}$ ,  $3.3 \times 10^{14} \text{ cm}^{-2}$ ,  $1.0 \times 10^{17} \text{ cm}^{-2}$ , and  $3.3 \times 10^{14} \text{ cm}^{-2}$ , respectively. The abundance of NH<sub>3</sub> and the upper end of the CS<sub>2</sub> abundance range are now found to be each approximately a factor of two lower than was reported previously [Noll *et al.*, 1995], and H<sub>2</sub>S is not detected.

### Introduction

The chemical nature of a portion of Jupiter's atmosphere was modified following the impact of comet Shoemaker-Levy 9 (SL9) in July, 1994. Spectroscopic monitoring of the impact region of fragment G (43.66S, 26.8W) with the Hubble Space Telescope's Faint Object Spectrograph (HST/FOS) G190H and G270H gratings covered a range from 1600Å to 3200Å with an effective resolution of 3.3Å and 4.6Å, respectively, for the two gratings. The HST/FOS aperture had a diameter of 0.86 arcsec corresponding to about 3250 km at Jupiter. Many new constituents in the stratosphere of Jupiter were identified [Noll *et al.*, 1995]. The present paper is based on improved reflectances, and, for the first time, it takes into account explicitly and rigorously the effects of aerosol scattering and the aerosol material on the abundance and location of the derived gas species. It also includes the relevant details of a radiative transfer-scattering model used to determine the abundances of the constituents. As a result of these improvements, we are able to place more stringent constraints on the abundances and locations of ammonia (NH<sub>3</sub>) and carbon disulfide (CS<sub>2</sub>) and are able to

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derive the upper limits of hydrogen sulfide (H<sub>2</sub>S), phosphine (PH<sub>3</sub>), acetylene (C<sub>2</sub>H<sub>2</sub>), and benzene (C<sub>6</sub>H<sub>6</sub>) in the Jovian stratosphere above the G-impact site. The data relevant to these species are mainly in the 1700-2200Å range; therefore we confine the analysis in this paper to the HST/FOS G190H grating data taken on July 18, 1994, 2.5 hours after the impact of fragment G on Jupiter.

### Modelling of Reflectance Spectra

The reflectance spectrum is given by  $R = I/F$ , where  $I$  is the intensity and  $\pi F$  is the Spacelab 2 Solar Ultraviolet Spectral Irradiance Monitor (SUSIM) solar flux [VanHoosier *et al.*, 1988] scaled to Jupiter's heliocentric distance and convolved with the instrument response function. In the case of the observations,  $I$  is Jupiter's observed flux which includes the diffusely reflected solar spectrum. The FOS spectra are reduced following STScI's pipeline procedure, corrected for Jupiter's filling the aperture, and converted to intensity by division of the aperture solid angle. Division by the convolved SUSIM spectrum canceled out the solar lines and continuum variation. In correcting for the grating scattered light [Rosa, 1993], we subtracted a constant background from the observed count rate with the value chosen to flatten the continuum below 1900Å. The propagated statistical error ( $\sigma_\lambda$ ) for the observations does not include the ~10% error in the absolute calibration of the FOS. Given the photoabsorption cross sections of the species identified in the spectra (cross sections are taken from Chen *et al.* [1991] and Wu *et al.* [1989] for C<sub>2</sub>H<sub>2</sub> and PH<sub>3</sub>; Lee *et al.* [1987] for H<sub>2</sub>S; Schmitt and Brehm [1966] for C<sub>6</sub>H<sub>6</sub>; Chen *et al.* [1994] for NH<sub>3</sub>; Rabalais *et al.* [1971] and Xu and Joens [1993] for CS<sub>2</sub>) and the Rayleigh scattering cross sections of H<sub>2</sub> and He [Dalgarno and Williams, 1962; Allen, 1973], we used the following method to obtain fits of the reflectance. The monochromatic radiative transfer equation for diffuse radiation is given by:

$$\mu \frac{dI}{dz} = -\sum_i n_i (\sigma_i^{\text{abs}} + \sigma_i^{\text{scat}}) I + \sum_i n_i \sigma_i^{\text{scat}} \int_{\Omega'} \frac{p_i(\Omega, \Omega')}{4\pi} I(\Omega') d\Omega' + \sum_i n_i \sigma_i^{\text{scat}} p_i(\Omega, \Omega_0) \frac{\pi F}{4\pi} \exp\left(-\frac{\tau}{\mu_0}\right) \quad (1)$$

where  $\mu, \mu_0, n_i, \sigma_i^{\text{abs}}, \sigma_i^{\text{scat}}, p_i, \Omega, \Omega_0$ , and  $\tau$  are the cosine of the earth zenith angle (53.7°), cosine of the solar zenith angle

( $58.8^\circ$ ), number density, absorption cross section, scattering cross section, scattering phase function, direction vector, direction of the sun, and optical depth, respectively, with the summation including both gases and aerosols. Dividing (1) by  $F$ , gives us an equation for  $R$  which we solved using a two stream approximation [Chandrasekhar, 1960] for an atmosphere with multiple layers. Comparisons with four streams and a standard adding-doubling code [Hansen *et al.*, 1974], showed the two-stream approximation, in which Rayleigh scattering is equivalent to isotropic scattering, to be adequate in modelling the spectra to about 10% [Toon *et al.*, 1989]. Parameters for each layer are the total column density, the mixing ratio of each constituent, the aerosol scattering and absorption optical depths, and the asymmetry factor (the degree of forward scattering) of the aerosol scattering phase function. The asymmetry factor was determined from the scattering phase function calculated by assuming the aerosols to be made up of Mie particles. Restricting the number of layers to one with a lower boundary placed in the troposphere, assuming a scattering optical depth for aerosols, and an index of refraction gives us a set of free parameters that can be used to fit the spectra. Fits were achieved by using the Simplex method to minimize the Chi-square error, given by

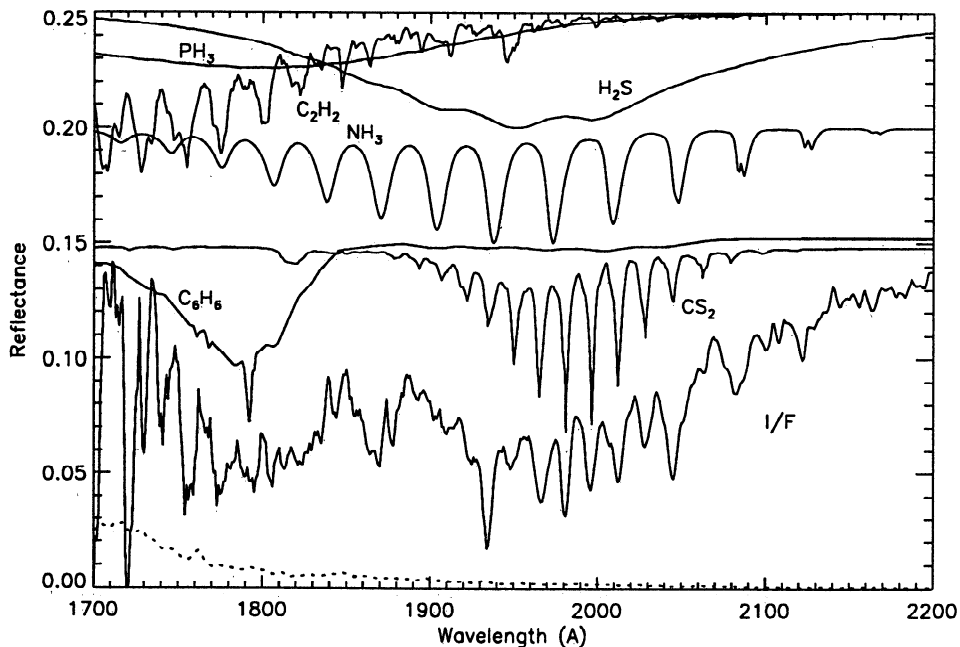
$$\chi^2 = \sum_{\lambda=1}^N \frac{(R - R_\lambda)^2}{N\sigma_\lambda^2} \quad (2)$$

where  $R_\lambda$ ,  $R$ ,  $\sigma_\lambda$ , and  $N$  are measured reflectance, calculated reflectance (sampled at  $1\text{\AA}$  and smoothed to the effective resolution of the measurements), the propagated statistical error of the observations, and number of wavelength intervals, respectively.

## Data Analysis

The HST/FOS short wavelength spectrum, particularly from  $1700\text{\AA}$  to  $2200\text{\AA}$ , spans the range in which previously known Jovian constituents, particularly  $\text{NH}_3$ ,  $\text{PH}_3$ , and  $\text{C}_2\text{H}_2$ , have their spectral signatures. In addition, we predicted the possible presence of  $\text{H}_2\text{S}$  in the stratosphere; it is either dredged from Jupiter's troposphere or formed in the reentry shock from comet and/or Jupiter's sulfur. Figure 1 shows the  $1700\text{-}2200\text{\AA}$  reflectance spectrum for the G-impact site on July 18 along with the inverted and normalized cross sections of various molecules suspected of contributing to the observed behavior. Also shown in the figure is the propagated statistical error (lower dotted curve). Below  $\sim 1800\text{\AA}$  the signal to noise ratio becomes low, mainly due to grating scatter, and the data are not reliable.

Simulations of the reflectance spectra were first done using a standard Jovian model atmosphere [Lindal *et al.*, 1981; Atreya *et al.*, 1981] considering only Rayleigh scattering and a single absorber. Although no good fits to the observed reflectance spectra were obtained in this manner, an examination of the absorption cross sections and the correlation with the observations allowed us to reject systematically many constituents as potential candidates, and narrowed the list of possible candidates to explain the behavior in the  $1750\text{-}2200\text{\AA}$  range to  $\text{NH}_3$ ,  $\text{CS}_2$ ,  $\text{H}_2\text{S}$ ,  $\text{PH}_3$ ,  $\text{C}_2\text{H}_2$ , and  $\text{C}_6\text{H}_6$ . In addition, aerosols (dust, condensed matter) are expected to contribute to the absorption and scattering of light. From the UV-IR imaging observations soon after the impact of the fragments, West *et al.* [1995] find that aerosols with mean radius  $0.15\text{-}0.3\ \mu\text{m}$  are present to altitudes where the pressure is  $0.3\ \text{mb}$ , and they are mixed with the gas scale height down to a pressure  $>200\ \text{mb}$ . In



**Figure 1:** Reflectance spectra ( $I/F$  where  $\pi F$  is the solar flux at Jupiter) and cross sections (labeled by species name) showing where features coincide. The cross sections (shown on a linear scale with arbitrary units to fit the plot) are inverted since the transmission of light through the atmosphere is proportional to this quantity. The dotted curve at the bottom is the propagated statistical error for the reflectance. The effective resolution of the reflectance is  $3.3\text{\AA}$ .

the UV, the majority of locations at intermediate optical depth ( $\tau \sim 1$ ) are best fit by the model with aerosols between 0.3 and 10 mb [West *et al.*, 1995]. In our radiative transfer-scattering calculations, we considered two possible scenarios:

(1) The aerosols result in extreme forward scattering of light as their radii are comparable to the wavelength being observed. In this case the aerosols are treated as any other gaseous absorber and the scattering of light back out of the atmosphere is due to Rayleigh scattering by the atmospheric H<sub>2</sub> and He. At 2000Å, the unit optical depth for the atmosphere in our model occurs at  $8 \times 10^{24} \text{ cm}^{-2}$  ( $\sim 67$  mb), whereas the Rayleigh scattering unit optical depth is at  $2.1 \times 10^{25} \text{ cm}^{-2}$  ( $\sim 178$  mb). In this scenario, the NH<sub>3</sub> and CS<sub>2</sub> abundances are found to be  $10^{16} \text{ cm}^{-2}$  and  $1.3 \times 10^{15} \text{ cm}^{-2}$ , respectively, above the unit optical depth level in the atmosphere.

(2) The aerosols participate in the scattering of light as well as absorption, a more likely scenario than the one above. Here we considered several subsets to represent the possible range in the aerosol unit optical depth level -  $\tau = 1$  at 0.3 mb or at 10 mb - and in the composition of the aerosol material, expressed in terms of the imaginary part of the index of refraction,  $k$ . We assumed the following values of  $k$ : 0.02-0.03 which was inferred for the debris in the UV, 0.5 at 2000Å for HCN polymer (both from West *et al.*, [1995] Fig. 2), and 0.001. The real part of the index of refraction was assumed to be 1.4 [West *et al.*, 1995]. The calculated abundances of NH<sub>3</sub> and CS<sub>2</sub> above the aerosol unit optical depth level are given in Table 1 (note that although the calculations include scattering by H<sub>2</sub> and He, the Rayleigh scattering optical depth at 2000Å is  $< 0.07$  for  $p < 10$  mb). A representative fit of the radiative transfer-scattering model to the data is shown in Fig. 2 for the  $\tau = 1$  at 0.3 mb,  $k = 0.03$  case. Similar fits are obtained for the other cases in Table 1.

There is a reasonable agreement between the reflectance

spectra and the radiative transfer scattering model for the NH<sub>3</sub> and CS<sub>2</sub> spectral features. The largest discrepancies occur at 1935Å, and in the lack of reproducibility of bands in the 2140-2200Å region and below  $\sim 1880$ Å. A number of possibilities exist to explain these discrepancies: S<sub>2</sub>, which was identified at longer wavelengths [Noll *et al.*, 1995], has several electronic transitions at wavelengths less than 2000Å, e.g.  $X^3\Sigma_g^- \rightarrow C^3\Sigma_u^-$ ,  $X^3\Sigma_g^- \rightarrow D^3\Pi_u$ ,  $a^1\Delta_g \rightarrow g^1\Delta_u$ , [Huber and Herzberg, 1979] that could contribute but cross section data do not exist; the laboratory cross section data for the various species could be inaccurate for the Jovian environmental conditions after the impact; and there could be other as yet unidentified minor contributors to the opacity. The resolution of these issues requires new laboratory and theoretical studies.

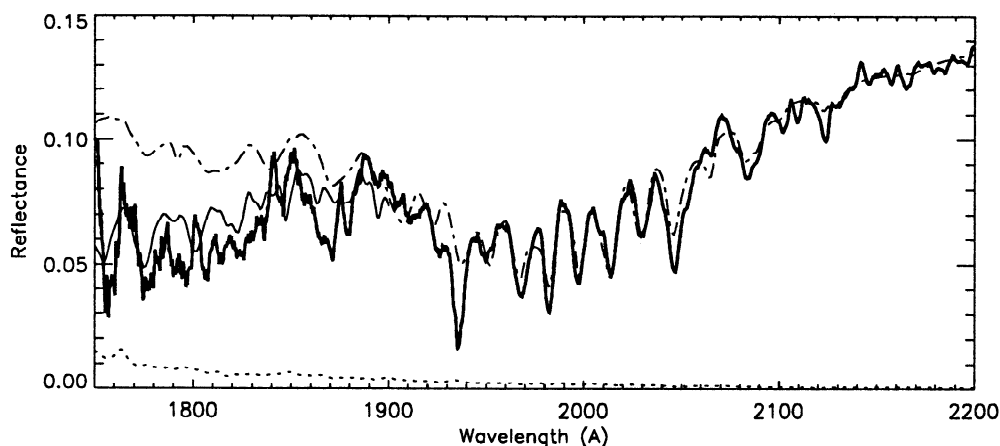
## Discussion and Conclusions

Our model fits give abundances above the level of unit optical depth in the atmosphere (Table 1). The NH<sub>3</sub> abundance above the aerosol unit optical depth level is found to be in the range  $(0.61 - 1.3) \times 10^{16} \text{ cm}^{-2}$  for  $0.001 < k < 0.03$ . The corresponding range for CS<sub>2</sub> is  $(0.56 - 1.1) \times 10^{15} \text{ cm}^{-2}$ . A value of 0.02-0.03 for  $k$  has been inferred for the impact debris at 2700Å, and is largely representative of silicates (West *et al.*, [1995]; similar values of  $k$  apply down to  $\sim 2000$ Å). Field *et al.* [1995] have also proposed silicates as a major component of the aerosol material. Nicholson *et al.* [1995] observed a feature near 10  $\mu\text{m}$  at the R-impact site that has been interpreted as silicate emission. West *et al.* [1995] also suggest HCN polymer ( $k = 0.5$  at 2000Å) as a likely candidate for some of the aerosol material. In that case the NH<sub>3</sub> and CS<sub>2</sub> abundances above the  $\tau = 1$  level would be in the range  $(2.5 - 6.0) \times 10^{15} \text{ cm}^{-2}$  and  $(4.3 - 5.6) \times 10^{14} \text{ cm}^{-2}$ , respectively (Table 1). It does not seem likely that the aerosols

**Table 1.** NH<sub>3</sub> and CS<sub>2</sub> Abundances Above the Aerosol Unit Optical Depth Level ( $\tau = 1$ )

N <sub>i</sub> (cm <sup>-2</sup> )	$\tau = 1$ @ 0.3 mb				$\tau = 1$ @ 10 mb			
	k=0.001	k=0.02	k=0.03	k=0.5	k=0.001	k=0.02	k=0.03	k=0.5
NH <sub>3</sub>	1.0(16)	8.2(15)	6.1(15)	2.5(15)	1.3(16)	1.1(16)	7.6(15)	6.0(15)
CS <sub>2</sub>	8.7(14)	6.7(14)	5.6(14)	4.3(14)	1.1(15)	9.6(14)	7.7(14)	5.6(14)

N<sub>i</sub> is column abundance above  $\tau = 1$  level; 1.0(16) =  $1.0 \times 10^{16}$ ;  $k$  is the imaginary index of refraction



**Figure 2:** Reflectance spectra (heavy continuous curve), error (dotted curve) and the result of the model fit (dot-dashed curve) including NH<sub>3</sub>, CS<sub>2</sub>, H<sub>2</sub>S, PH<sub>3</sub>, and C<sub>6</sub>H<sub>6</sub>, but not C<sub>2</sub>H<sub>2</sub>. C<sub>2</sub>H<sub>2</sub> could have an impact on the reflectances below 1900Å. This is shown by the light continuous curve below 1900Å, assuming a C<sub>2</sub>H<sub>2</sub> column abundance of  $10^{17} \text{ cm}^{-2}$  at the  $\tau = 1$  level.

would be composed of pure HCN polymer as the measured amount of HCN gas is small,  $6 \times 10^{11}$  g [Marten *et al.*, 1995]. From Table 1, the NH<sub>3</sub> mixing ratio above  $\tau = 1$  level is found to be in the 5-10 ppb range for the case where  $\tau = 1$  is at 10 mb, and in the 60-250 ppb range for the case where  $\tau = 1$  is at 0.3 mb. It is interesting to note that the latter range is similar to the 80 ppb value derived by Orton *et al.* [1995] in the stratosphere above the K impact site, which seems to imply that K and G fragments and their impacts were similar and the aerosol scattering was taking place at high altitudes where pressures were about 0.3-1 mb. For the case with the unit aerosol optical depth at 0.3 mb, we also find the following upper limits for various constituents above this level: H<sub>2</sub>S  $< 1.2 \times 10^{16}$  cm<sup>-2</sup>, PH<sub>3</sub>  $< 3.3 \times 10^{14}$  cm<sup>-2</sup>, C<sub>2</sub>H<sub>2</sub>  $< 1.0 \times 10^{17}$  cm<sup>-2</sup>, and C<sub>6</sub>H<sub>6</sub>  $< 3.3 \times 10^{14}$  cm<sup>-2</sup>; the latter two should be considered tentative because of grating scatter. Noll *et al.* [1995] reported a detection of H<sub>2</sub>S, which we no longer find to be valid.

We find that the abundance of ammonia and the upper end of the range for carbon disulfide abundance are each approximately a factor of two lower than was reported previously [Noll *et al.*, 1995]. This we attribute to the more rigorous analysis of the data here which includes the possible range in aerosol material. The ammonia we measure in the stratosphere far exceeds 2 ppb just above the troposphere calculated on the basis of photochemical models [Atreya *et al.*, 1977; Atreya and Romani, 1985]. CS<sub>2</sub> was not previously detected in Jupiter's atmosphere. Ammonia could have been transported to the stratosphere of Jupiter from below if the fragments exploded in or below the NH<sub>3</sub> ice cloud, contributed by the comet, and/or formed in the shock induced chemistry following the impact from either cometary or the Jovian sources of N. Even if the fragments exploded above Jupiter's tropopause, there is a likelihood of the warming of the upper troposphere/lower stratosphere which could result in more of the ammonia existing in the vapor phase in the stratosphere. CS<sub>2</sub> is expected to have formed post-impact in a synthesis between carbon (such as Jupiter's CH<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>, or C, or carbon derived from the comet) and sulfur (such as Jupiter's H<sub>2</sub>S or elemental sulfur, or the comet's sulfur derived from CS, CS<sub>2</sub>, the parent of CS, or H<sub>2</sub>S). The question of the origin of the detected species is complex, and its resolution requires careful investigation of many other aspects of the SL9 observations which is beyond the scope of this short paper.

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## References

- Allen, C.W., *Astrophysical Quantities*, The Athlone Press, 1973 (pp. 92-93)
- Atreya, S.K. and P.N. Romani, Photochemistry and clouds of Jupiter, Saturn, and Uranus. In *Planetary Meteorology* (G.E. Hunt, ed.), Cambridge Univ. Press, pp. 17-68, 1985
- Atreya, S.K., et al., The distribution of ammonia and its photochemical products on Jupiter, *Icarus*, 31,348,1977
- Atreya, S.K., et al., Jupiter: structure and composition of the upper atmosphere, *Ap.J.*, 247, L43

- Chandrasekhar, S., *Radiative Transfer*, Dover Publications, Inc., 1960
- Chen, F., et al., High-resolution, low temperature photoabsorption cross sections of C<sub>2</sub>H<sub>2</sub>, PH<sub>3</sub>, AsH<sub>3</sub>, and GeH<sub>4</sub>, with applications to Saturn's atmosphere, *J. Geophys. Res.*, 96, 17519, 1991
- Chen, F., et al., Low - and room temperature photoabsorption cross sections of NH<sub>3</sub> in the UV region, *J. Geophys. Res.*, submitted, 1995
- Dalgarno A. and D.A. Williams, Rayleigh scattering by molecular hydrogen, *Ap. J.*, 136, 690, 1962
- Field, G.B. et al., Dust as the cause of spots on Jupiter, *Astron. Astrophys.*, 1995, in press.
- Hansen, J.E. and L.D. Travis, Light scattering in planetary atmospheres, *Sp. Sci. Rev.*, 16, 527, 1974
- Huber, K.P. and G. Herzberg, *Constants of Diatomic Molecules*, Van Nostrand Reinhold, 1979
- Lee, L.C., et al., Quantitative photoabsorption and fluorescence spectroscopy of H<sub>2</sub>S and D<sub>2</sub>S at 49-240 nm., *J. Chem. Phys.*, 86, 4353, 1987
- Lindal, G.F., et al., The atmosphere of Jupiter: an analysis of Voyager radio occultation measurements, *J. Geophys. Res.*, 86, 8721, 1981
- Marten A. et al., The collision of Comet Shoemaker-Levy 9 with Jupiter: Detection and Evolution of HCN in the Stratosphere of the Planet, *Geophys. Res. Lett.*, 1995, in press.
- Nicholson, et al., Palomar observations of the R-impact of Comet SL9 Spectra II, *Geophys. Res. Lett.*, 1995, in press.
- Noll, K.S., et al., Hubble Space Telescope observations of Jupiter after the collision of Comet P/Shoemaker-Levy 9, *Science*, 267, 1307, 1995
- Orton G. et al., The NASA IRTF investigation of Comet Shoemaker-Levy 9 and its collision with Jupiter, *Science*, 267, 1277, 1995
- Rabalais, J.W., et al., Electronic spectroscopy of isoelectronic molecules. II. Linear triatomic groupings containing sixteen valence electrons., *Chem. Rev.*, 71, 73, 1971
- Rosa, M.R., Correcting FOS background, STSci/ECS Newsletter 20, vol.16, 1993.
- Toon, O.B., et al., Rapid calculation of radiative heating rates and photodissociation rates in inhomogeneous multiple scattering atmospheres, *J. Geophys. Res.*, 94, 16287, 1989
- Schmitt, R.G. and R.K. Brehm, Double beam spectroscopy in the far ultraviolet. 1: 1150Å to 3600Å, *Appl. Opt.*, 5, 1111, 1966
- West, R.A., et al., Impact debris particles in Jupiter's stratosphere, *Science*, 267, 1296, 1995
- Wu, C.Y.R., et al., Photoabsorption of C<sub>2</sub>H<sub>2</sub> in the 1530-1930Å region: a temperature dependence study, *J. Chem. Phys.*, 91, 272, 1989
- VanHoosier, M.E., et al., Absolute solar spectral irradiance 120nm-400nm (Results from the Solar Ultraviolet Spectral Irradiance Monitor- SUSIM- Experiment on board Spacelab 2). *Astro. Lett. Com.*, 27, 163, 1988
- Xu, H. and J.A. Joens, CS<sub>2</sub> Absorption cross section measurements from 187 nm to 230 nm., *Geophys. Res. Lett.*, 20, 1035, 1993

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