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The Titan Haze Revisited: Magnetospheric Energy Sources and Quantitative Tholin Yields

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We present laboratory measurements of the radiation yields of complex organic solids produced from N_2/CH_4 gas mixtures containing 10 or 0.1% CH_4. These tholins are thought to resemble organic aerosols produced in the atmospheres of Titan, Pluto, and Triton. The tholin yields are large compared to the total yield of gaseous products: nominally, 13 (C + N)/100 eV for Titan tholin and 2.1 (C + N)/100 eV for Triton tholin. High-energy magnetospheric electrons responsible for tholin production represent a class distinct from the plasma electrons considered in models of Titan's airglow. Electrons with $E > 20$ keV provide an energy flux $\sim 1 \times 10^{-2}$ erg cm^{-2} sec^{-1}, implying from our measured tholin yields a mass flux of 0.5 to 4.0×10^{-14} g cm^{-2} sec^{-1} of tholin. (The corresponding thickness of the tholin sedimentary column accumulated over 4 Gyr on Titan's surface is 4 to 30 m.) This figure is in agreement with required mass fluxes computed from recent radiative transfer and sedimentation models. If, however, these results, derived from experiments at ~2 mb, are applied to lower pressure levels toward peak auroral electron energy deposition and scaled with pressure as the gas-phase organic yields, the derived tholin mass flux is at least an order of magnitude less. We attribute this difference to the fact that tholin synthesis occurs well below the level of maximum electron energy deposition and to possible contributions to tholins from UV-derived C_2-hydrocarbons. We conclude that Titan tholin, produced by magnetospheric electrons, is alone sufficient to supply at least a significant fraction of Titan's haze—a result consistent with the fact that the optical properties of Titan tholin, among all proposed materials, are best at reproducing Titan's geometric albedo spectrum from near UV to mid-IR in light-scattering models. © 1994 Academic Press, Inc.

OPTICAL PROPERTIES AND LIGHT-SCATTERING MODELS

This paper is a study of whether quantitative production rates of Titan haze, derived from the leakage of saturnian magnetospheric auroral electrons, is adequate to match modern values of the sedimentation rate of the haze particles out of the upper atmosphere.

Titan tholin is the term used for the complex organic solid produced from plasma discharge irradiation in an N_2/CH_4 atmosphere containing about 10% CH_4—approximately the abundance in Titan's upper stratosphere. Its production and optical properties were documented by Khare *et al.* (1984), who noted the similarity to the properties of the Titan haze. The first studies showing that Titan tholin could quantitatively reproduce Titan's geometric albedo spectrum in light-scattering models were by Sagan *et al.* (1983), Thompson (1984) and Sagan *et al.* (1985) (cf. figures in Sagan *et al.* 1992); Titan tholin particles of ~0.1–0.5 μm radius (with 0.1 μm preferred) very well reproduced Titan's reflection spectrum outside the CH_4 absorption bands. More recent sedimentation plus light-scattering models have provided many more details and show that a material with optical properties very similar to Titan tholin (but with the imaginary refractive index k multiplied by ~1.3) provides an excellent match to Titan's spectrum (cf. McKay and Toon 1992, McKay *et al.* 1989, Samuelson and Mayo 1991, Sagan *et al.* 1992). The 30% offset is within the probable errors of measurement (Sagan *et al.* 1992), but may also be radiation-dose related (Khare *et al.* (1993b). Poly-HCN badly matches Titan tholin (Khare *et al.* (1993a) and is unlikely to be a major constituent of the Titan haze. McKay (personal communication, 1993) also finds that alternative materials such as polyacetylene and poly-HCN are inconsistent with Titan's reflection spectrum.

The mass flux of haze required is $\sim 1 \times 10^{-14}$ g cm^{-2} sec^{-1} for the McKay *et al.* models and (insignificantly different) $\sim 0.8 \times 10^{-14}$ g cm^{-2} sec^{-1} for the model of Samuelson and Mayo (1991). Earlier models by Podolak *et al.* (1984), updated by Podolak (1984), inferred fluxes of $0.35–3.5 \times 10^{-14}$ g cm^{-2} sec^{-1}; Podolak (1984) noted that a more absorbing material than the polyethylene used by Podolak *et al.* (1984) (for the lower limit of the mass flux) was required for the model to be consistent with absorption properties inferred from polarization data.

QUANTITATIVE YIELDS OF TITAN AND TRITON THOLINS

The yield for charged particle production of Titan tholin was estimated by Thompson *et al.* (1989a) to be at

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least equal to the total yield for all gas phase molecules and perhaps substantially greater. (For UV irradiation of N_2/CH_4 mixtures the ratio of solid phase to gas phase products may be $\ll 1$.) A mass flux of 0.3–3 × 10^{-14} g cm^{-2} sec^{-1} derived from this work was later used by McKay and Toon (1992), but our previous estimate was merely an educated guess, and a direct measurement of tholin production efficiency is reported here for the first time.

The tholin yield measurements were made in the same N_2/CH_4 production runs of gas phase organics reported in McDonald *et al.* (1994), by our usual method of exciting a high-frequency coronal discharge in a flow-through system. Such a system is described by Thompson *et al.* (1991), who measured the yields of gas-phase molecules for the 10% CH_4, Titan-type mixture. Thompson *et al.* (1989b) made similar gas-phase measurements for a 0.1% CH_4, Triton-type mixture. (This is too CH_4- rich by at least a factor of 10 for Triton, but serves as our best available simulation.) Pluto CH_4 mixing ratios have intermediate values (cf. Owen *et al.* 1993, Yelle 1994).

The yields were derived as follows: Glass discharge tubes were weighed before and after a run of known duration and gas flow rate, giving the total mass of tholin produced. The effective energy delivered to the gas was computed by ethene (ethylene) dosimetry (cf. Thompson *et al.* 1991) to be ~0.03 W at 2.8–2.0 mbar discharge pressure. Results are shown in Table I. The nominal tholin yields are large; for Titan tholin, 13 (C + N)/100 eV, or 1.7 × 10^{-12} g erg^{-1}. Comparison with gas-phase yields requires an extrapolation to the pressure of this experiment. From Thompson *et al.* (1991) we crudely estimate that the yield $Y \propto p^{0.4}$. Use of this relation produces a multiplicative factor of 2.7 for the 0.24-mbar results of Thompson *et al.* (1991) and 1.9 for the 0.56-mbar results of Thompson *et al.* (1989b), resulting in the entries for total gas-phase yields in Table I. (Other assumed functional forms of the pressure dependence of yield give similar results). The uncertainties in the relative yields of the two tholins should be somewhat less than that for the absolute yields, since every effort was made to duplicate conditions in the two experiments.

The range in tholin yields results from some uncertainty in the energy calibration. Using a similar experimental system, Thompson *et al.* (1989b, 1991) measured a somewhat higher delivered power. Using the two pressures in Thompson *et al.* (1991), one can estimate that power $P \propto p^{0.6}$ and predict from the measured power in Thompson *et al.* (1989b) (0.02 W at 0.56 mbar) a value of 0.09 W at 2.8 mbar; using Thompson *et al.* (1991) (0.04 W at 0.24 mbar) one would predict 0.17 W at 2.8 mbar. Although this argument is weak (since the experimental systems were not exactly the same), to be cautious we have listed tholin yields in Table I ranging from the preferred, higher values, to lower limits about one-quarter of the nominal values. (The corresponding range of productivity is 3.9 × 10^{-13} to 1.7 × 10^{-12} g erg^{-1}.)

TABLE I
Comparison of Plasma Discharge Yields of Titan and Triton Tholins

	Titan tholin (from 10% CH_4 in N_2)	Triton tholin (from 0.1% CH_4 in N_2)
Discharge pressure (torr)	2.1	2.2
Run time (hr)	24	72
Tholin produced (g)	0.0410	0.0148
Discharge power (eV/hr)	6.3 × 10^{20}	6.3 × 10^{20}
Tholin yields		
g/eV	2.7 × 10^{-24}	3.3 × 10^{-25}
g/g Reagent gas	1.8 × 10^{-3}	2.2 × 10^{-4}
C + N/100 eV (tholin)	3–13	0.5–2.1
C + N/100 eV low-molecular-weight products)	2.2 (Thompson *et al.* 1991)	0.30 (Thompson *et al.* (1989)
Productivity ratio, Tholin/organic gases	1.5–6.2	1.7–7.0
Titan tholin flux (from magnetospheric e^-)	0.5–4 × 10^{-14} gm cm^{-2} sec^{-1}	

ELECTRON ENERGY INPUTS TO TITAN'S STRATOSPHERE

The total energy delivered by various charged particle and solar electromagnetic radiation sources to Titan's atmosphere was calculated by Sagan and Thompson (1984). (Diagrams of UV, magnetospheric electron, and cosmic ray deposition altitudes are given in that paper and in Sagan *et al.* 1992.) In particular, for magnetospheric electrons power law models of the high-energy electron fluxes (from the LECP and CRS experiments) measured by the Voyager spacecraft were employed. A relativistic energy loss equation was used to compute the energy deposited by a population of electrons having this measured power law spectrum, for an assumed uniform distribution of incidence angles over 2π sterad. Since these values are based on real Voyager data, the only assumption involved was the low-energy cutoff for the validity of the power law. This is of some importance, since the differential flux power index is −3.1 for the high-energy electrons (see Sagan and Thompson 1984), and for indices <-2 most of the power is delivered by the lowest energy component of the population.

Sagan and Thompson used 10 keV as the (unstated) low-energy cutoff, yielding a flux estimate of 2.4 × 10^{-2}

erg cm^{-2} sec^{-1}. The lowest actual Voyager energy bin used in the fit was 37 keV; truncation here yields 5×10^{-3} erg cm^{-2} sec^{-1}, while an intermediate cutoff at 20 keV yields 9×10^{-3} erg cm^{-2} sec^{-1}. The nature of these high-energy electrons is discussed by Van Allen (1984), who concludes that for 40 keV $< E <$ a few MeV, the electrons have an external source, and characterizes them as in loss-free and source-free diffusion inward to 10 Saturn radii, R_S. The spin-averaged overall flux is given as 10^4–10^5 cm^{-2} sec^{-1}, yielding a flux over 2π of 0.4–4×10^{-2} erg cm^{-2} sec^{-1} (for a 40-keV cutoff), consistent with the Sagan and Thompson estimates. Van Allen also cites a value of 10^5 cm^{-2} sec^{-1} sr^{-1} at $E > 15$ keV for Pioneer 11, yielding a flux of 1.5×10^{-2} erg cm^{-2} sec^{-1}. There seems little doubt, then, that a high-energy electron flux close to the values used by Sagan and Thompson (1984) is relevant for Titan.

The inbound *Voyager* LECP data show a broad gouge in the magnetospheric ion and electron distributions at approximately the orbit of Titan (Maclennan *et al.* 1982). In part because of the width of the gouge and in part because of magnetic field models predicting that the Titan L-shell should be elsewhere, the possible causal association of the gouge with Titan was not firmly drawn. However, in the comprehensive LECP paper on Saturn (Krimigis *et al.* 1983) the authors note that the gouge was present in the outbound as well as the inbound passes (their Fig. 6) and allow that it may be due to Titan. Krimigis' current view (private communication, 1993) is that magnetic field model predictions so far from the planet are unreliable and that Titan and/or its torus may in fact be responsible, both in electrons and in ions, for the gouge. Lanzerotti (private communication, 1994) agrees. The difference between the interpolated flux near 20 R_S and the residual flux in the gouge is here attributed to absorption by Titan.

Most of the theoretical modeling of the delivery of electron energy to Titan's atmosphere has concerned the low-energy plasma and the amount of power from this source required to sustain the observed EUV airglow. Strobel *et al.* (1992) infer that 2×10^8 W of total power (about 2×10^{-3} erg cm^{-2} sec^{-1}) are delivered by plasma electrons and model the delivery of low-energy plasma by curvature drift. They use the plasma properties of Neubauer *et al.* (1984), who characterized the plasma in terms of a 200-eV mean energy Maxwellian core with density 0.3 cm^{-3}, and a 1-keV superthermal tail with density 0.01 cm^{-3}. Clearly, this is a very different electron population from the high-energy component discussed previously (although one can estimate from the density and mean velocity that, if the plasma had free access to Titan, the total power would be within an order of magnitude, $\sim 2 \times 10^{-2}$ erg cm^{-2} sec^{-1}). First, as Strobel *et al.* model in detail, such low-energy electrons are impeded from free interaction with Titan's upper stratosphere, and second, the energy that they deliver is at very high altitudes ~1000 km above the surface. In contrast, the high-energy electrons penetrate freely into the stratosphere: Strobel *et al.* state that for $E > 20$ keV the electrons "would penetrate deep into the UV-absorbing hydrocarbon region." Sagan and Thompson show that this high-energy component deposits most of its energy below 650 km; airglow produced by this component will be unobservable due to UV opacity. While the total energy flux from high-energy plasma electrons is negligible, as Strobel *et al.* argue, the external source high-energy electrons are comparable in total energy to the plasma.

We illustrate this in Figs. 1a and 1b. The differential flux for the 200 eV plus 1 keV Maxwellian plasma of Neubauer *et al.* (1984) is plotted along with the Voyager-derived high-energy electron flux. The high-energy population has a shallow -3.1 power law slope compared to the sharp drop-off of the superthermal Maxwellian component. A possible interpolation between the populations is indicated. Figure 1b shows instead the energy deposited per δE bin for steps of 0.1 in log E. This illustrates the substantial amount of total energy in the external high-energy electron component and implies that a secondary maximum in deposited energy between the domains of the plasma and high-energy populations is likely.

THOLIN FLUXES FROM HIGH-ENERGY ELECTRONS

Using a 20-keV cutoff, the flux computed from the Sagan and Thompson parameterization is $\sim 1 \times 10^{-2}$ erg cm^{-2} sec^{-1}. Using the experimental yield of 1.7×10^{-2} g erg^{-1} for Titan tholin, the nominal production rate of tholin from this source is then 1.7×10^{-14} g cm^{-2} sec^{-1}, consistent with the mass fluxes estimated from recent radiative transfer and particle sedimentation models discussed above. (This agreement implies that $\lambda > 2000$ Å solar photons are ineffective in generating Titan tholin; cf. Sagan and Thompson 1984.) Allowing for an increase of a factor of two if the cutoff is taken to be 10 keV and a decrease of a factor of four if the lower estimate of tholin yield is employed, we obtain a probable range of 0.5–4.0×10^{-14} g cm^{-2} sec^{-1} for the mass flux due to tholin production by high-energy magnetospheric electrons. Over the lifetime of Titan, and for a density of tholin $\simeq 1.5$ g cm^{-3}, this is the equivalent of a layer of tholins 4 to 30 m thick accumulated on the surface of Titan.

However, Sagan and Thompson (1984) (see also Sagan *et al.* 1992) show electron deposition rates within a factor of 10 of maximum deposition between 850 and 425 km altitude, corresponding, respectively, to pressures of 5.6×10^{-6} and 6.3×10^{-3} mb, with a peak rate at 650 km altitude. The pressures of tholin production on Titan are

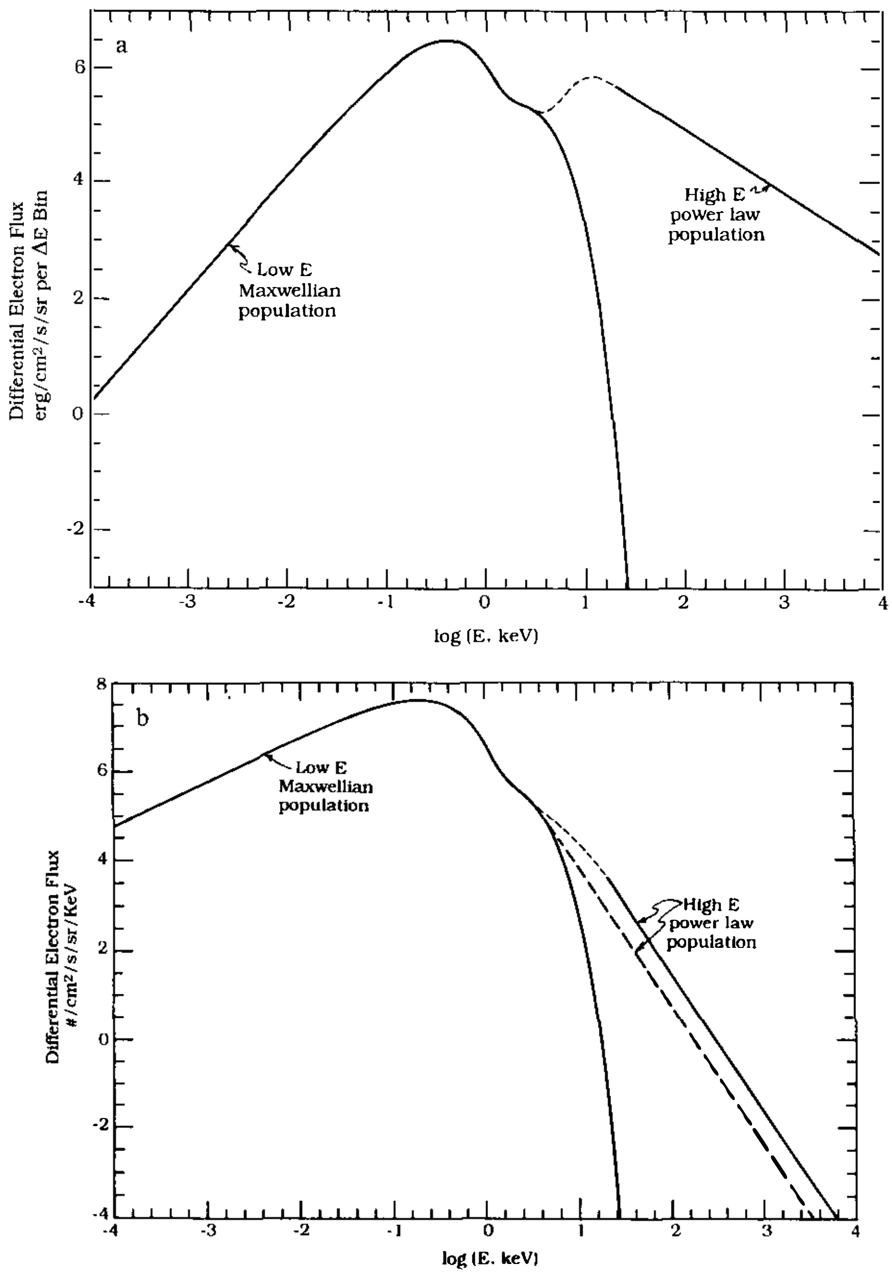

FIG. 1. (a, b) Differential flux, respectively, per keV and per unit energy bin (steps of 0.1 in log E) of the Saturn magnetospheric electrons at Titan, based on Voyager data. The light dashed line in both figures represents the inferred join between the high-energy power law and the low-energy Maxwellian electron populations.

then approximately 300 (at the 425 km, 10% of maximum level) to 2000 (at the 650 km maximum) times less than the 2-mb experimental pressure. Unfortunately, there are no experimental data that bear directly on the variation of tholin production efficiency with pressure. If one assumed that the efficiency varied as $p^{0.4}$ as it does for gases produced under similar experimental conditions, then tholin yields would be reduced by a factor of 10 at the 425 km, 10% level and by a factor of 50 at the 650 km, maximum input level.

Since the nominal production rate so well matches that predicted for Titan tholin, and the tholin optical constants so well match the Titan haze, we are faced with an interesting problem. The most obvious, but very uncertain, possibility is that the production of tholin aerosols does not decline with pressure as quickly as does the production of gas-phase organics. However, it is much more likely—in fact it is required (see below)—that tholin production does not mainly occur near the altitudes of maximum energy input. Reactions between unsaturated nitriles and hydrocarbons are well known (cf. Lichtin and Lin 1985, 1986, 1987) to have near-zero activation energies, so that gases transported downward from the altitude of maximum production can produce tholin at increasingly higher rates as atmospheric pressure increases. In essence, the total gas production rates (which have been shown to match Voyager infrared measurements closely) should strongly contribute to tholin production, but in a decoupled rather than an *in situ* process.

A mechanism of this type would seem to be required, since the main haze begins at about 300 km altitude, and since physical models for Titan's aerosols require all production to occur below 300 km, increasing to 50% of the total column density at 260 km and to 100% at 220 km (Toon *et al.* 1992). (Toon *et al.* note that this profile is also in disagreement with the CH_4 destruction profile of Yung *et al.* 1984.) Titan tholin must then be produced primarily at pressures between 0.1 and 0.3 mb, much nearer to our 2-mb experimental pressure and therefore with efficiencies much closer to those of our experimental values: conceivably ranging from no difference for an approximately pressure-independent rate to about 40% of our nominal value of the $p^{0.4}$ gas-phase production power law applies (merely a guess since the production processes clearly differ).

In this decoupled model of the production of source molecules and of the tholin product, the photochemically produced gases (primarily, the C_2 hydrocarbons) will contribute, along with the radiation–chemically produced gases, to the total tholin production; this could easily make up for a possible shortfall of the charged particle-only production rate, provided that the admixture of larger quantities of photochemical products to the tholin production process does not significantly alter the optical constants. Note that while radiation–chemical production alone is sufficient to produce the quantities of more complex molecules required to match Voyager infrared observations, a photochemical input is required for C_2 hydrocarbons (especially C_2H_6 and C_2H_4), whose predicted radiation–chemical abundances are too low (Thompson *et al.* 1991). A model in which the photochemical products play at least a minor role in tholin production then seems reasonable. But we note again that hydrocarbon polymers such as polyacetylenes do not match the observed optical constants of the Titan haze.

SUMMARY AND DISCUSSION

Titan tholin has been shown in increasingly sophisticated radiative transfer plus particle sedimentation models to have optical properties consistent, within a small multiplicative factor, with those required to match Titan's geometric albedo spectrum. In this work we have shown that the high-energy electron population, distinct from the plasma electron population, contributes substantial energy to the mid-stratosphere. Combined with quantitative yields of tholin production measured in the laboratory and reported here, the derived mass flux of tholin is in agreement, within a factor of 2–4, with the requirements of haze coagulation–sedimentation models such as those summarized by McKay and Toon (1992)—provided that tholin production mainly occurs well below the level of maximum auroral electron energy deposition.

Certainly, photochemistry also contributes large amounts of material to Titan's stratosphere, some of which might be constituents of Titan tholin; apart from the simplest C_2-hydrocarbons, all the remaining minor constituents in the Titanian atmosphere, both nitriles and hydrocarbons, seem to be mainly produced by magnetospheric electrons (Thompson *et al.* 1989, Sagan *et al.* 1992). To the extent that these nitriles and hydrocarbons with more than two heavy atoms are responsible for the aerosols, charged particles dominate ultraviolet light.

The total production rate from CH_4 photolysis is estimated by Yung *et al.* (1984) as 4×10^{-13} g cm^{-2} sec^{-1}, at least 10 times the tholin production flux above. Of this, most is in the form of small molecules (hydrocarbons and nitriles), and does not contribute to the haze, but instead freezes out deep in the stratosphere, as illustrated by Sagan and Thompson (1984). Photochemical models predict that some fraction will be in the form of polyacetylenes with about six or more carbons; McKay *et al.* infer a mass production rate of 2×10^{-14} g cm^{-2} sec^{-1} from this source, comparable to the tholin production rate. Yet the optical properties of laboratory-produced polyacetylene are seriously at variance with Titan's spectrum.

There are several possible explanations, which we state for discussion. Perhaps this less absorbing material is intimately mixed with tholin that is actually even more absorbing than our nominal Titan tholin (conceivably because of the combined plasma/UV radiation dose), resulting in a material whose bulk properties result in the observed reflection spectrum. Or the actual situation could be more complicated, with poorly absorbing UV-derived aerosols (produced at higher altitudes, ~800–1000 km) generating a different grain population incident on the lower altitude level of primary tholin production. Radiative transfer and sedimentation models with more than one kind of grain input may be able to distinguish these possibilities, especially when orbital and *in situ* measurements from the Cassini/Huygens mission become available. Or when charged-particle and short-UV-derived N atoms are fully included in absolute reaction rate kinetics photochemical models, perhaps more of the Titan tholin will turn out to have been driven by UV. For example, polyacetylene and Titan tholin pathways may intertwine when both are being produced in the same volume.

However, recent recalculation of photochemical yields by Toublanc *et al.* (1994) give a production rate of C_2H_4 about one-sixth that of Yung *et al.* (1984). Photochemical organic solids may therefore be produced at no higher rate than those generated by auroral electrons. At the very least, charged particle radiation chemistry alone seems able to account for a significant fraction of the Titan haze production rate.

The laboratory data displayed in Table I show that yields of N_2/CH_4 tholins decrease by only about one order of magnitude over a two order of magnitude decrease in CH_4 mixing ratio. It should therefore not be routinely assumed that yields per unit energy of organic hazes in N_2/CH_4 atmospheres are similar unless the CH_4 mixing ratios are also similar.

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