Photochemical Production of $\text{H}_2\text{SO}_4$ Aerosols on Venus

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Evidence for SO₂ injection (by a volcano?) into the upper atmosphere of Venus

The quantum yields for producing O₂(a¹Δₐ) and O₂(b¹Σ⁺) for reaction (3) are summarized in Table I which also includes results for other simple reactions capable of producing the singlet oxygen states. In general, the quantum yields are very small, ranging from less than 10⁻⁵ to 4 x 10⁻².

According to Yung and DeMore (1982), the O₂(a¹Δₐ) emissions due to (1), (2), and (3) could be as high as 0.72, 0.37, and 0.72 MR, respectively, adding up to a total of 1.8 MR, if the quantum yields for forming O₂(a¹Δₐ) in the reactions were unity. However, using the current values for quantum yields, we obtain revised estimates of O₂(a¹Δₐ) emission (including cascading from O₂(b¹Σ⁺)) due to (1), (2), and (3) as 0.029, 0.007, and 0.014 MR, respectively. The total amounts to 0.05 MR, considerably less than the observed 1.2-1.5 MR.

We now face a fundamental dilemma. Reaction (1) turns oxygen atoms into O₂. Reactions (2) and (3) constitute a catalytic cycle for converting odd oxygen into O₂. The ultimate source of O atoms in the upper atmosphere of Venus in the present model is CO₂ photolysis, which proceeds at a mean rate of 8 x 10¹² cm⁻² s⁻¹. This implies that the total rate of O-O bond formation cannot exceed 4 x 10¹² cm⁻² s⁻¹. Therefore, if the quantum yield for producing the a¹Δₐ state is q in each O-O bond forming reaction, the airglow would be less than 4q MR. The recent laboratory results suggest that q is a few percent or less. Hence, the O₂(a¹Δₐ) emission of Venus cannot arise from O-O bond
forming reactions in which the O atoms are derived from CO$_2$ dissociation. The simplicity of the argument outlined here suggests that our conclusion is largely independent of the details of the photochemical model.

One obvious possibility is that there might have been a calibration error in the observations. A discussion with one of the authors (Traub, 1987, private communication) led to a critical re-examination of the data analysis procedure used in Connes et al.'s (1979) paper. It was concluded that calibration errors could have been at most a factor of 2, since two independent methods, based on the terrestrial airglow and the Venus continuum brightness were used, and both methods gave essentially the same result.

We can speculate on other explanations. The crucial clue is the source of O atoms. As was first pointed out by Winnick and Stewart (1980), photolysis of SO$_2$ is a major source of O atoms in the stratosphere of Venus. Indeed, according to the models of Winnick and Stewart (1980), and Yung and DeMore (1982), the photolysis rate of SO$_2$ on Venus is considerably higher than that of CO$_2$. However, most of the photolysis of SO$_2$ occurs in the lower part of the stratosphere, just above the cloud tops (~60 km). Any O$_2$(a' $\Delta_g$) produced in this region would be effectively quenched, and could not contribute to the observed emissions under "normal" circumstances.

Consider now an unusual event, an episodic injection of SO$_2$ into the upper atmosphere of Venus. We shall leave the cause of this event unspecified, although volcanism is a possibility (Esposito, 1984). The presence of large quantities of SO$_2$ in the upper stratosphere (above 80 km) would lead to rapid photolysis and production of oxygen atoms. Recombination of these atoms either by (1) or the catalytic cycle (2) and (3) would yield a source of O$_2$(a' $\Delta_g$).

Detailed modeling of this event appears difficult since the O$_2$(a' $\Delta_g$) airglow observations were carried out in 1975, before there were any quantitative data on SO$_2$ (Pioneer-Venus started collecting data in 1978).
Table I

List of the relevant reactions that produce \( \text{O}_2(a^1 \Delta_g) \) and \( \text{O}_2(b^1 \Sigma^+_g) \) in the atmosphere of Venus and their quantum yields.

<table>
<thead>
<tr>
<th>Reactions</th>
<th>( \text{O}_2(a) )</th>
<th>( \text{O}_2(b) )</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 ( \text{O} + \text{O} + \text{M} \rightarrow \text{O}_2 + \text{M} )</td>
<td>4(-2)</td>
<td>2(-2)</td>
<td>Wraight</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Ogryzlo et al.</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3(-2)</td>
<td>Young and Black</td>
</tr>
<tr>
<td>2 ( \text{Cl} + \text{O}_3 \rightarrow \text{ClO} + \text{O}_2 )</td>
<td>&lt;2(-2)</td>
<td>&lt;5(-4)</td>
<td>Choo and Leu</td>
</tr>
<tr>
<td>3 ( \text{O} + \text{ClO} \rightarrow \text{Cl} + \text{O}_2 )</td>
<td>&lt;2.5(-2)</td>
<td>4.4(-4)</td>
<td>This work</td>
</tr>
<tr>
<td>4 ( \text{Cl} + \text{ClO}_2 \rightarrow \text{Cl}_2 + \text{O}_2 )</td>
<td>1.4(-2)</td>
<td>6(-3)</td>
<td>Choo and Leu</td>
</tr>
<tr>
<td>5 ( \text{Cl} + \text{HO}_2 \rightarrow \text{HCl} + \text{O}_2 )</td>
<td>&lt;5(-4)</td>
<td></td>
<td>Keyser et al.</td>
</tr>
<tr>
<td>6 ( \text{H} + \text{HO}_2 \rightarrow \text{H}_2 + \text{O}_2 )</td>
<td>1.5(-2)</td>
<td></td>
<td>Washida et al.</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.8(-4)</td>
<td>Hislop and Wayne</td>
</tr>
<tr>
<td></td>
<td></td>
<td>&lt;1(-2)</td>
<td>Keyser et al.</td>
</tr>
<tr>
<td>7 ( \text{O} + \text{HO}_2 \rightarrow \text{OH} + \text{O}_2 )</td>
<td>&lt;8(-3)</td>
<td></td>
<td>Keyser et al.</td>
</tr>
</tbody>
</table>

*Defined as the rate coefficient for the branch divided by the total rate coefficient.*
Determination of $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g^+)$ Yields in the Reaction $O + ClO \rightarrow Cl + O_2$: Implications for Photochemistry in the Atmosphere of Venus

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Abstract

A discharge flow apparatus with a chemiluminescence detector has been used to study the reaction $0 + \text{ClO} \rightarrow \text{Cl} + 0_2^*$, where $0_2^* = 0_2(a^1\Delta_g)$ or $0_2(b^1\Sigma_g^+)$, and the radiative decay of $0_2^*$. The measured quantum yields for producing $0_2(a^1\Delta_g)$ and $0_2(b^1\Sigma_g^+)$ in the above reaction are less than $2.5 \times 10^{-2}$ and $(4.4 \pm 1.1) \times 10^{-4}$, respectively. The observed $0_2(a^1\Delta_g)$ airglow of Venus cannot be explained in the context of standard photochemistry using our experimental results and those reported in recent literature. The possibility of an episodic injection of $\text{SO}_2$ into the mesosphere of Venus is speculated.
Introduction

The intense $O_2(a^1\Delta_g)$ emission of Venus at 1.27 $\mu$m was discovered by Connes et al. (1979), who estimated the dayglow and nightglow brightness to be 1.5 and 1.2 MR ($1$ Megarayleigh $= 1 \times 10^{12}$ photons cm$^{-2}$ s$^{-1}$), respectively. The aforecited authors also proposed that the most likely processes for producing $O_2(a^1\Delta_g)$ on Venus are:

1. $O + O + CO_2 \rightarrow O_2 + CO_2 + 498$ KJ/mol

2. $Cl + O_3 \rightarrow ClO + O_2 + 161$ KJ/mol

3. $O + ClO \rightarrow Cl + O_2 + 236$ KJ/mol

Detailed modeling by Yung and DeMore (1982) and Krasnopolsky and Parshev (1983) showed that, in order to explain the Venus observations, the quantum yields in reactions (1)-(3) must be close to unity.

Reaction (1) has been studied by a number of workers (see recent review by Wayne, 1985). The quantum yields for singlet oxygen (both a and b states), as summarized in Table III, are low. Reaction (2) was studied by Choo and Leu (1985), who found no evidence for the production of excited $O_2$ (see Table III). In this article we shall report new experimental results on the quantum yields of singlet oxygen produced by reaction (3). Using the known efficiencies for producing $O_2(a^1\Delta_g)$ in (1)-(3), we can make a quantitative comparison of the modeled and observed airglow brightness of Venus. The results will be briefly discussed.
The experimental technique used in this study has been described in detail in previous publications (Leu, 1984; Choo and Leu, 1985). The production and detection of the single oxygen molecules, $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma^+_g)$, were carried out in a discharge flow apparatus with a chemiluminescence detector.

Oxygen atoms were produced in the side arm of the flow tube via the fast reaction

$$\text{N} + \text{NO} \rightarrow \text{O} + \text{N}_2,$$

where the nitrogen atoms were generated in a microwave discharge by passing nitrogen gas through a quartz tube. Since the N atoms were slightly in excess, the final O atom concentration would be equal to that of NO, which was measured with a mass flowmeter.

ClO radicals were generated as follows: First, Cl atoms in the concentration range $(5-10) \times 10^{13} \text{ cm}^{-3}$ were produced by passing a small amount of Cl$_2$ in a helium carrier through a microwave discharge tube. Then, Cl atoms, in slight excess, were allowed to react with Cl$_2$O in a fast reaction leading to the production of ClO

$$\text{Cl} + \text{Cl}_2\text{O} \rightarrow \text{Cl}_2 + \text{ClO}.$$

The Cl$_2$O molecules used in this work were prepared as described in Leu (1984). The absolute concentration of Cl$_2$O was monitored by an optical absorption technique. The light source was a low pressure mercury lamp, which emits a strong line at 253.7 nm, and the adopted cross section for Cl$_2$O at this wavelength is $2.01 \times 10^{-18} \text{ cm}^2$. The detector was a photomultiplier equipped
with an interference filter. By virtue of the stoichiometry of reaction (5),
the final concentration of C10 must equal the initial concentration Cl2O.
Having prepared the C10 molecules in this manner, they were then introduced
into the reaction cell via a movable Pyrex injector.

The observation of the singlet oxygen emissions was carried out using the
chemiluminescence detection system. Radiation in the wavelength range 600-900
nm was resolved with a 1/4 m McPherson monochromator (Model 275) and registered
by a Hamamatsu cooled photomultiplier tube (Model R943-02) in conjunction with
a Keithley picoammeter (Model 480). Radiation in the wavelength range 900-1700
nm was resolved with an Oriel grating monochromator (Model 7240) and detected
with a liquid nitrogen cooled germanium detector (Applied Detector Corp. Model
403L). The signals were modulated with a 100 Hz chopper (Bulova) and a lock-in
amplifier (EG & G Model 5804) was used for the phase sensitive detection.
Detection of emissions from both singlet oxygen states were simultaneously
recorded on a two-channel recorder.

Figure 1 shows the chemiluminescence data taken under the following
conditions: \([0] = [C10] = 1.7 \times 10^{13} \text{ cm}^{-3} ([x] \text{ denotes the concentration of}
\text{ species } x)\); total pressure = 1.25 Torr; flow velocity = 3620 cm s\(^{-1}\); and
reaction time = 4.8 ms. \(O_2(b^1\Sigma^+_g)\) emission at 762 nm with a bandwidth of 10
nm was clearly detected. This result suggests that there were 3.8 \times 10^8 cm\(^{-3}\)
of \(O_2\) in the \(b^1\Sigma^+_g\) state, derived from reaction (3). \(O_2(a^1\Delta_g)\) emission at
1270 nm was not detectable in the same experiment. The experimental data are
summarized in Table I. In order to derive the quantum yields of \(O_2(a^1\Delta_g)\) and
\(O_2(b^1\Sigma^+_g)\), it is necessary to employ a computer simulation. The set of
relevant chemical reactions is listed in Table II. The rate of loss of
\(O_2(b^1\Sigma^+_g)\) on the walls of the reaction cell and quenching by \(N_2\) has been
measured using the same technique as reported here. The rate of loss of $O_2(a^1\Delta_g)$ by quenching on the walls and by collisions with $N_2$, Cl, O, and ClO can be shown to be negligible. The experimental conditions have been chosen so as to minimize interference from the reactions (1) and $Cl + ClO_2 \rightarrow Cl_2 + O_2$.

The results of our analysis can be summarized as: $k_a < 9 \times 10^{-13}$ cm$^3$ s$^{-1}$, and $k_b = (1.6 \pm 0.4) \times 10^{-14}$ cm$^3$ s$^{-1}$ for the absolute rate constants for (3) to produce $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma^+_g)$, respectively. The uncertainty represents one standard deviation error.
Discussion

The quantum yields for producing $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma^+_g)$ for reaction (3) are summarized in Table III, which also includes results for other simple reactions capable of producing the singlet oxygen states. In general, the quantum yields are very small, ranging from less than $10^{-4}$ to $4 \times 10^{-2}$.

According to Yung and DeMore (1982), the $O_2(a^1\Delta_g)$ emissions due to (1), (2), and (3) could be as high as 0.72, 0.37, and 0.72 MR, respectively, adding up to a total of 1.8 MR, if the quantum yields for forming $O_2(a^1\Delta_g)$ in the reactions were unity. However, using the current values for quantum yields, we obtain revised estimates of $O_2(a^1\Delta_g)$ emission (including cascading from $O_2(b^1\Sigma^+_g)$) due to (1), (2), and (3) as 0.029, 0.007, and 0.014 MR, respectively. The total amounts to 0.05 MR, considerably less than the observed 1.2-1.5 MR.

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Conclusion

A discharge flow apparatus with a chemiluminescence detector has been used to determine the quantum yields of $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma^+_g)$ in the reaction $O + ClO \rightarrow Cl + O_2$. The results are summarized in Tables II and III. Application of these results to explain the airglow brightness of Venus reveals a fundamental difficulty with a "normal" photochemical model. The upper atmosphere of Venus might have been strongly perturbed by a massive injection of $SO_2$ in 1975.

It is important that our laboratory kinetics-chemiluminescence results be confirmed using an independent experimental technique. The $O_2(a^1\Delta_g)$ emission from Venus have been observed only once. These observations should be repeated to establish the magnitude and nature of possible variations, along with the simultaneous observation of other chemical species such as CO, H$_2$O, HCl, HF, $O_2$, and $SO_2$. 
Acknowledgements

We thank W.A. Traub, W.B. DeMore, M. Allen, J.P. Pinto, L.W. Esposito, and L.D. Travis for helpful discussions. This research was performed at the Jet Propulsion Laboratory, California Institute of Technology, under contract with NASA. YLY was supported as a guest investigator on the Pioneer-Venus Project under contract NAG 2267 to the California Institute of Technology.
Table I.

Summary of $O_2(b^1\Sigma^+_g)$ Measurements

<table>
<thead>
<tr>
<th>[O]</th>
<th>[ClO]</th>
<th>Reaction Time</th>
<th>$[O_2(b)]$</th>
<th>$k_b$</th>
<th>Number of Experiments</th>
</tr>
</thead>
<tbody>
<tr>
<td>(cm$^{-3}$)</td>
<td>(cm$^{-3}$)</td>
<td>(ms)</td>
<td>(cm$^{-3}$)</td>
<td>(cm$^3$ s$^{-1}$)</td>
<td></td>
</tr>
<tr>
<td>1.5(13)</td>
<td>4.9(12)</td>
<td>3.0</td>
<td>1.0(9)</td>
<td>1.2(-14)</td>
<td>1</td>
</tr>
<tr>
<td>1.5(13)</td>
<td>1.6(13)</td>
<td>3.0</td>
<td>2.8(9)</td>
<td>1.3(-14)</td>
<td>1</td>
</tr>
<tr>
<td>1.6(13)</td>
<td>1.1(13)</td>
<td>4.8</td>
<td>3.4(9)</td>
<td>1.9(-14)</td>
<td>3</td>
</tr>
<tr>
<td>1.6(13)</td>
<td>1.6(13)</td>
<td>4.8</td>
<td>3.6(9)</td>
<td>1.6(-14)</td>
<td>2</td>
</tr>
<tr>
<td>1.6(13)</td>
<td>1.7(13)</td>
<td>4.8</td>
<td>3.8(9)</td>
<td>1.4(-14)</td>
<td>6</td>
</tr>
<tr>
<td>1.6(13)</td>
<td>1.9(13)</td>
<td>4.8</td>
<td>5.2(9)</td>
<td>2.1(-14)</td>
<td>1</td>
</tr>
</tbody>
</table>

Average Value $(1.6 \pm 0.4) \times (-14)$

$^+$The numbers a(b) should be read as a x $10^{-b}$ in this table and Tables II and III.

$^*$Best fit of the $O_2(b^1\Sigma^+_g)$ measurement.

$^{**}$One standard deviation.
Table II.
Chemical scheme for computer simulation of the production of $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma^+_g)$ in the reaction $O + ClO \rightarrow Cl + O_2$.

<table>
<thead>
<tr>
<th>Reactions</th>
<th>Rate Constant (cm$^3$ s$^{-1}$)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>$O + ClO \rightarrow Cl + O_2$</td>
<td>3.6(-11)</td>
<td>Leu</td>
</tr>
<tr>
<td>$Cl + O_2(a)$</td>
<td>&lt;9.0(-13)</td>
<td>This work</td>
</tr>
<tr>
<td>$Cl + O_2(b)$</td>
<td>1.6(-14)</td>
<td>&quot;</td>
</tr>
<tr>
<td>$O_2(a) + wall \rightarrow O_2$</td>
<td>0</td>
<td>Choo and Leu</td>
</tr>
<tr>
<td>$O_2(a) + N_2 \rightarrow O_2 + N_2$</td>
<td>1.0(-19)</td>
<td>&quot;</td>
</tr>
<tr>
<td>$O_2(a) + Cl \rightarrow O_2 + Cl$</td>
<td>1.0(-16)</td>
<td>*</td>
</tr>
<tr>
<td>$O_2(a) + O \rightarrow O_2 + O$</td>
<td>1.0(-16)</td>
<td>*</td>
</tr>
<tr>
<td>$O_2(a) + ClO \rightarrow O_2 + ClO$</td>
<td>1.0(-16)</td>
<td>*</td>
</tr>
<tr>
<td>$O_2(b) + wall \rightarrow O_2$</td>
<td>80.0</td>
<td>Choo and Leu</td>
</tr>
<tr>
<td>$O_2(b) + N_2 \rightarrow O_2 + N_2$</td>
<td>1.7(-15)</td>
<td>&quot;</td>
</tr>
<tr>
<td>$O_2(b) + Cl \rightarrow O_2 + Cl$</td>
<td>1.0(-13)</td>
<td>*</td>
</tr>
<tr>
<td>$O_2(b) + O \rightarrow O_2 + O$</td>
<td>1.0(-13)</td>
<td>*</td>
</tr>
<tr>
<td>$O_2(b) + ClO \rightarrow O_2 + ClO$</td>
<td>1.0(-13)</td>
<td>*</td>
</tr>
</tbody>
</table>

*Estimated based on analogous reactions; see the review by Wayne (1985).
List of the relevant reactions that produce $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g^+)$ in the atmosphere of Venus and their quantum yields.

<table>
<thead>
<tr>
<th>Reactions</th>
<th>$O_2(a)$</th>
<th>$O_2(b)$</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>$O + O + M \rightarrow O_2 + M$</td>
<td>4(-2)</td>
<td>2(-2)</td>
<td>Wraight</td>
</tr>
<tr>
<td>Cl + $O_3 \rightarrow ClO + O_2$</td>
<td>&lt;2(-2)</td>
<td>&lt;5(-4)</td>
<td>Choo and Leu</td>
</tr>
<tr>
<td>Cl + ClO</td>
<td>2.5(-2)</td>
<td>4.4(-4)</td>
<td>This work</td>
</tr>
<tr>
<td>Cl + ClO$_2$</td>
<td>1.4(-2)</td>
<td>6(-3)</td>
<td>Choo and Leu</td>
</tr>
<tr>
<td>Cl + HO$_2$</td>
<td>&lt;5(-4)</td>
<td>Keyser et al.</td>
<td></td>
</tr>
<tr>
<td>H + HO$_2$</td>
<td>1.5(-2)</td>
<td>Washida et al.</td>
<td></td>
</tr>
<tr>
<td>O + HO$_2$</td>
<td>&lt;8(-3)</td>
<td>Keyser et al.</td>
<td></td>
</tr>
</tbody>
</table>

*Defined as the rate coefficient for the branch divided by the total rate coefficient.*
References


Choo, K.Y. and Leu, M.T. (1985). Determination of $O_2(1\Sigma_g^+)$ and $O_2(1\Delta_g)$ yields in $Cl + O_2$ and $Cl + O_3$ reactions. J. Phys. Chem. 89, 4832-4837.


Figure Caption

Figure 1. Chemiluminescence spectra taken from the reaction of atomic oxygen with C10 radical (see text for detail).