

RADIATION AND CHEMISTRY IN THE STRATOSPHERE: SENSITIVITY TO O<sub>2</sub> ABSORPTION CROSS SECTIONS IN THE HERZBERG CONTINUUM

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**Abstract.** We propose that a significant overestimate of the molecular oxygen absorption cross sections in the important spectral window from 200-220 nm is in large part responsible for the discrepancy between observed and modeled vertical profiles of some halocarbons (CFC<sub>13</sub> in particular), as well as for the long-standing problem of simultaneously fitting N<sub>2</sub>O, CH<sub>4</sub>, CF<sub>2</sub>Cl<sub>2</sub> and CFC<sub>13</sub> profiles with a single eddy diffusion model. Recent measurements of the direct solar flux in the stratosphere by J.R. Herman and co-workers seem to support this idea. Replacing our current O<sub>2</sub> cross sections in the 200-220 nm range by values in better agreement with the results of the above group leads to a reduction in N<sub>2</sub>O, CF<sub>2</sub>Cl<sub>2</sub> and CFC<sub>13</sub> concentrations (by factors of 0.70, 0.62 and 0.19, respectively, at 30 km), while CH<sub>4</sub>, H<sub>2</sub> and CO profiles are essentially unchanged. Moreover, the predicted concentration of HNO<sub>3</sub> above 30 km is reduced by ~50%, yielding better agreement with observations. The reduction in O<sub>2</sub> cross sections produces a 10-20% decrease in ozone above about 35 km, but a fairly large increase (~30%) near the peak around 20-25 km. The changes in other stratospheric species are also briefly discussed.

## Introduction

The present general understanding of stratospheric chemistry and comparisons between models and observations have recently been conveniently summarized by Hudson et al. (1982), in the latest WMO summary report. One persistent problem that still remains, both in 1-D and 2-D models, is the inability to produce a satisfactory fit to the altitude distributions of all long-lived source species that diffuse upwards from the troposphere and undergo relatively simple chemistry in the stratosphere. In particular, it has been difficult to produce good simultaneous fits to N<sub>2</sub>O, CH<sub>4</sub>, CF<sub>2</sub>Cl<sub>2</sub>(FC12) and CFC<sub>13</sub>(FC11) profiles above 20 km. The calculated mixing ratios near 30 km for FC12 and FC11 are generally overestimated by a factor of about two and five or more, respectively, given a model that is in reasonable agreement with N<sub>2</sub>O and CH<sub>4</sub>. This general discrepancy holds for 1-D and 2-D models alike and seems to be fairly independent of latitude or season (see Hudson et al., 1982; Miller et al., 1981). Possible solutions including either transport, unknown chlorofluorocarbon sinks or inaccurate solar radiation calculations in the Schumann-Runge bands have been suggested. In this paper, we discuss in detail the most plausible solution, which involves the uncertainty in the photodissociation rate of some stratospheric molecules (such as N<sub>2</sub>O, FC11 and FC12) due to uncertainties in molecular oxygen absorption cross sections near 200 nm. We briefly discuss the uncertainty in the O<sub>2</sub> cross sections and the importance of the spectral region from ~195 to 220 nm for the photolysis of certain stratospheric species. The sensitivity of N<sub>2</sub>O, CH<sub>4</sub>, FC11, FC12, HNO<sub>3</sub>, O<sub>3</sub> and other trace gases to the O<sub>2</sub> absorption near 200 nm is then presented and available mid-latitude observations are compared to theoretical profiles from a complete one-dimensional model.

## Photolysis of Stratospheric Gases in the 190-220 nm Spectral Region

Photodissociation of various molecules plays a major role in stratospheric chemistry and the solar flux provides

the driving term for many chemical interactions. Nicolet (1980,1981) has published recent reviews on the subject, with some emphasis on molecular oxygen and ozone absorption; it certainly seems that the O<sub>2</sub> cross sections in the Herzberg continuum (200-242 nm) are uncertain by at least 25%. Moreover, the solar flux at the top of the atmosphere is not known to much better than 15% in this spectral region (see Hudson et al., 1982). However, molecules that dissociate near 200 nm, where O<sub>2</sub> is the main opacity source, will be more sensitive to the O<sub>2</sub> cross sections  $\sigma_{\lambda}(O_2)$  than to the solar flux, since  $\sigma_{\lambda}(O_2)$  enters as an exponential factor in the photodissociation rate ( $J_{\lambda}$ ) calculations. Indeed,  $\Delta J_{\lambda}/J_{\lambda} = -\Delta \tau_{\lambda} = -\tau_{\lambda}(\Delta \tau_{\lambda}/\tau_{\lambda})$ , which implies that a small percentage change in total optical depth  $\tau_{\lambda}$  can lead to a larger relative change in  $J_{\lambda}$ , if  $\tau_{\lambda}$  is larger than unity (below about 35 km in this case).

Figure 1 illustrates the importance of the 190-220 nm region for N<sub>2</sub>O, HNO<sub>3</sub>, CF<sub>2</sub>Cl<sub>2</sub> and CFC<sub>13</sub>, for which  $J_{\lambda}$  peaks in the center of this spectral range; however, the total photolysis rate of HNO<sub>3</sub> also depends on the flux longward of 300 nm and actually peaks at ~310 nm below about 20 km. Rates in Figure 1 were calculated with O<sub>2</sub> and O<sub>3</sub> profiles from the U.S. Standard Atmosphere 1976 and 24 hr diurnally-averaged transmission. The optical depths  $\tau_{\lambda}(O_2)$  and  $\tau_{\lambda}(O_3)$  in the 200-220 nm range are shown in Figure 2. O<sub>2</sub> and O<sub>3</sub> contribute roughly equally to the total opacity in this region, and it is clear that this model yields total  $\tau_{\lambda}$  values of order 1 to 10 between 20 and 30 km. The cross sections used in the above figures are generally averaged over 5 nm bins, as are the solar flux values, taken from Mount and Rottman (1981). The O<sub>2</sub> cross sections above 207.5 nm follow the recommendation of the 1979 NASA Report (Hudson and Reed, 1979) and the O<sub>3</sub> values are from Ackerman (1971). Below 207.5 nm, effective O<sub>2</sub> cross sections, depending on height and zenith angle, are calculated according to the work of Allen and Frederick (1982). Above 197.5 nm, the contribution from the bands should be about 5% or less, most of the absorption being due to the continuum (Hudson and Mahle, 1972). Sharanand and Prasad Rao (1977) have obtained the latest (and smallest) measurements of O<sub>2</sub> cross sections in the Herzberg continuum and have described the problems associated with the laboratory experiments. The reader is referred to their work for a summary of previous measurements (some of which differ by 50% or more).

If our model cross sections (average values of 14.5, 11.5, 8.85, 7.43 and 5.75, in units of 10<sup>-24</sup> cm<sup>2</sup>, for 200, 205, 210, 215 and 220 nm, respectively) are reduced by a factor of about 0.6, we find some fairly significant changes in relevant stratospheric profiles. Some of our preliminary sensitivity tests were presented by Y. Yung at the Chemical Manufacturers Association Meeting (Steed et al., 1982). During that month, related work from the NASA/Goddard Space Flight Center became available to us, with quite timely and direct implications for the Herzberg continuum absorption of O<sub>2</sub>. Frederick and Mentall (1982) discuss some of the direct solar flux measurements within the stratosphere (30-40 km) and conclude that the atmospheric transmission in the 200-210 nm range is larger than expected from laboratory data on O<sub>2</sub> and O<sub>3</sub> cross sections. Herman and Mentall (1982) give a more expanded analysis of the transmitted radiation from 190 to 320 nm, from which they derive some constraints on the absorption characteristics of O<sub>2</sub> and O<sub>3</sub>. They find that the O<sub>3</sub> cross sections seem to agree within a few percent with the laboratory data, whereas the O<sub>2</sub> cross sections seem to have been overestimated by 30% or more by all laboratory measurements, in agreement with our own suggestion based on more indirect modeling tests of N<sub>2</sub>O, HNO<sub>3</sub> and chlorofluorocarbon profiles.

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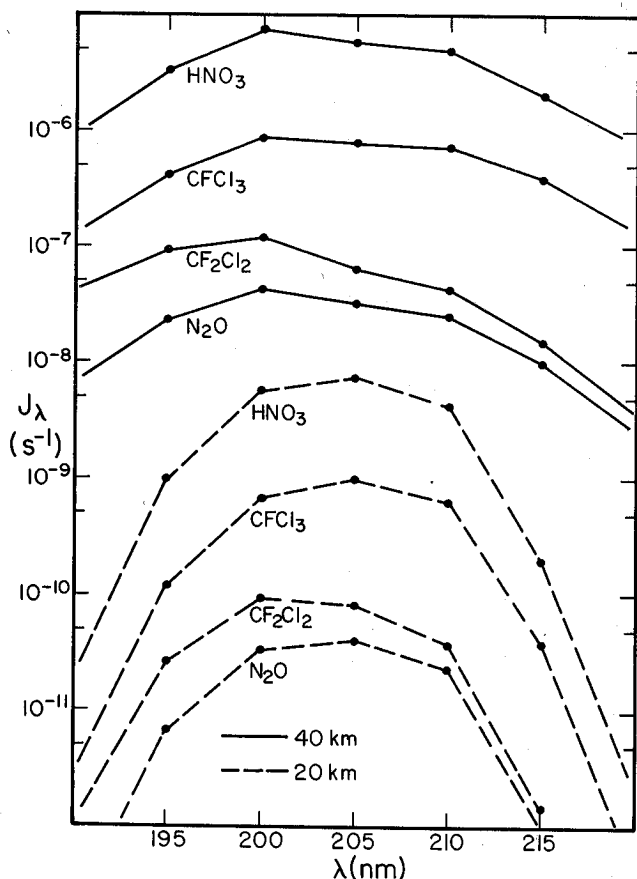


Fig. 1. Diurnally-averaged photodissociation rates ( $J_\lambda$ ) for  $\text{HNO}_3$ ,  $\text{CFCl}_3$ ,  $\text{CF}_2\text{Cl}_2$  and  $\text{N}_2\text{O}$  at 40 and 20 km, in the  $\text{O}_2$ - $\text{O}_3$  spectral window near 200 nm.

#### Modeling of Stratospheric Species and Sensitivity to O<sub>2</sub> Cross Sections

Since Herman and Mental (1982) have recently estimated that the O<sub>2</sub> cross sections in the Herzberg continuum region (up to 222 nm) should be even lower (by ~30%) than the lowest laboratory values, we adopt average cross sections in agreement with their results. We use reduced values of 5.6, 5.1 and 3.5 ( $10^{-24}$  cm<sup>2</sup>) at 210, 215 and 220 nm, respectively, keeping in mind that these values have associated error bars of 10-30%. Below 207.5 nm we multiply the effective O<sub>2</sub> cross sections by a factor of 0.55, down to 196.1 nm (spectral interval 5 in Allen and Frederick, 1982). This produces effective average cross sections of about  $8.0 \times 10^{-24}$  cm<sup>2</sup> (200 nm) and  $6.3 \times 10^{-24}$  cm<sup>2</sup> (205 nm) at an altitude where the O<sub>2</sub> absorption effect is maximized.

The 1-D Caltech photochemical model numerically solves the coupled set of continuity equations involving 47 species and over 130 reactions at 2 km intervals between 0 and 80 km. Allen et al. (1981) have described some of the model features, as applied to mesospheric trace species and transport and we have up-dated and expanded this model for stratospheric chemistry. A detailed discussion and comparison with other important species can unfortunately not be presented in this short paper, but a complete description of the model will be found elsewhere (Froidevaux, Allen and Yung, in preparation). Ground albedo and atmospheric Rayleigh scattering effects have been included and 24 hour diurnally-averaged species profiles are sufficient for the present sensitivity study. Transport is described by the standard vertical eddy-diffusion parameterization  $K(z)$ , which attempts to include all averaged dynamical effects affecting long-lived trace species in the earth's atmosphere. Massie and Hunten (1981) (hereafter MH) recently published a preferred

globally-averaged  $K(z)$  profile, based on a combined analysis of N<sub>2</sub>O, CH<sub>4</sub>, O<sub>3</sub> and <sup>14</sup>C (time-dependent) tracer data. The diffusion rate of N<sub>2</sub>O, CH<sub>4</sub> and halocarbons from the troposphere up into the stratosphere will undoubtedly depend on  $K(z)$ . FC11 responds more strongly (at a given altitude) than FC12 or N<sub>2</sub>O to changes in either  $K(z)$  or O<sub>2</sub> opacity due to its faster photolysis rate and sharper fall-off in mixing ratio.

The mid-latitude models presented below are compared to N<sub>2</sub>O, CH<sub>4</sub>, CF<sub>2</sub>Cl<sub>2</sub>, CFCl<sub>3</sub> and HNO<sub>3</sub> observations graphically summarized in Hudson et al. (1982); the latter report describes in more detail the data base and the relevant references. Most of the observations were taken between 40°N and 50°N, during the summer, and the calculations refer to 45°N latitude and summer solstice solar illumination. Two eddy-diffusion profiles are used (see Table I). Profile  $K_1$  is very similar to the MH composite profile, but joins smoothly with the 70 km value ( $1.5 \times 10^6$  cm<sup>2</sup> s<sup>-1</sup>) of Allen et al. (1981). Profile  $K_2$  is everywhere lower than the MH composite profile, but is in better agreement with their <sup>14</sup>C tracer-inferred profile. Clearly, there is no "ideal" profile in this oversimplified representation of transport processes, but it will be seen that the slower  $K_2$  model will result in better fits with observations near 45°N; indeed there is strong evidence that vertical transport is latitude-dependent and increases towards the tropics. In model A, we use the standard O<sub>2</sub> cross sections and transport profile  $K_2$ . Model B differs from A simply by the reduction (factor of ~0.6) in O<sub>2</sub> cross sections described above and Model C is the same as case B, except that the  $K(z)$  profile is the faster  $K_1$  model.

The N<sub>2</sub>O profiles shown in Figure 3 illustrate the fact that both an increase in transport rates and an increase in O<sub>2</sub> cross sections can increase the mixing ratios above 20 km. Methane also shows an increase due to transport (from Model B to C), but is insensitive to the 200-220 nm spectral region and Models A and B yield similar profiles. Nevertheless, the N<sub>2</sub>O and CH<sub>4</sub> observations do not provide the most sensitive test of these three models. FC11 and

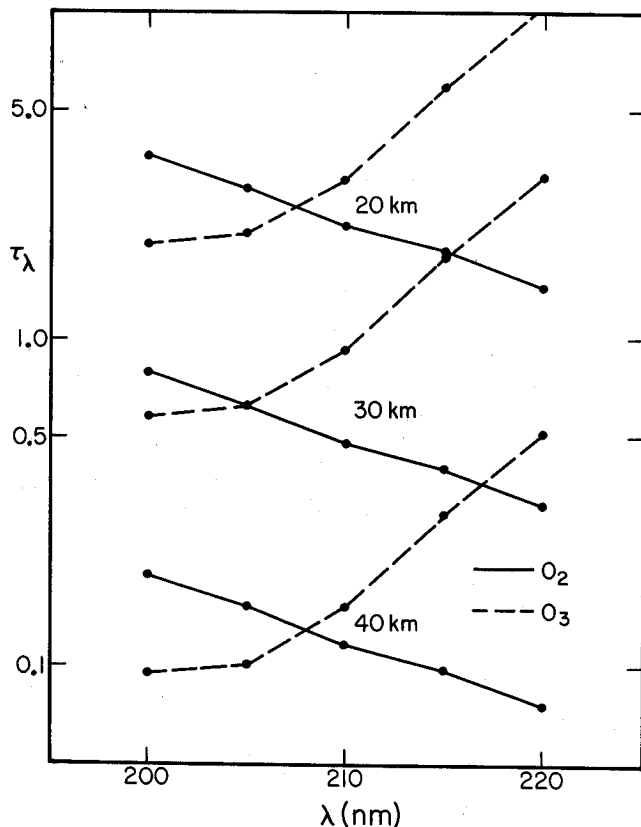


Fig. 2. Normal optical depths ( $\tau_\lambda$ ) for O<sub>2</sub> and O<sub>3</sub> at various altitudes; standard model O<sub>2</sub> cross sections are used (see text).

Table I. Mid-latitude Model Eddy-diffusion Coefficient  $K(z)^*$ 

Altitude $z$ (km)	Model $K_1$	Model $K_2$
$z \leq 8$	As in Massie and Hunten (1981)	As in Massie and Hunten (1981)
$8 \leq z \leq 14$	As in Massie and Hunten (1981)	$K(z) = 1.0 \times 10^8 \exp\{-(z-8)/1.59\}$
$z = 16; z = 18$	As in Massie and Hunten (1981)	$K(z) = 2.1 \times 10^8$
$20 \leq z \leq 70$	$K(z) = 4.6 \times 10^8 \exp\{(z-20)/8.64\}$	$K(z) = 2.5 \times 10^8 \exp\{(z-20)/7.82\}$
$z \geq 70$	As in Allen et al. (1981)	As in Allen et al. (1981)

\*Units are  $cm^2 s^{-1}$ ; only 2 significant digits are used in model values.

FC12 show larger reductions in mixing ratios in the middle and upper stratospheres, if Model B is used instead of A (see Figure 4):  $CF_2Cl_2$  is reduced by factors of 0.62 and 0.41 at 30 and 40 km, respectively, whereas  $CFCl_3$  is decreased by factors of 0.19 and 0.06 at these altitudes. A much better fit is obtained with Model B; use of the faster transport profile (Model C) increases the mixing ratios back to values similar to Model A. An additional improvement due to Model B is shown in Figure 5. Nitric acid ( $HNO_3$ ) has always been in strong disagreement with observations above about 30 km. The increase in photolysis between models A and B is translated into a 50% decrease above 30 km, and much smaller changes in the lower stratosphere, where the total photolysis rate becomes insensitive to radiation in the  $O_2$  Herzberg continuum. While some of the  $HNO_3$  data points are in reasonable agreement with model B, there is still over a factor of two discrepancy with the data of Evans et al. (1981) at 35 km. We emphasize that the significant lower stratospheric increase in flux near 200 nm in Model B (flux higher than in Model A by a factor of 2-5) cannot be caused by a 10-20% change in the solar flux at the top of the atmosphere, since the latter uncertainty is not amplified by an exponential factor, as in the case for  $\sigma(O_2)$ .

Other stratospheric gases are also affected — directly or indirectly — by a reduction in  $O_3$  cross sections. The main direct effect is an increase in the photodissociation rates of other halocarbons in the middle and upper stratosphere, due to the larger fluxes in the 200-220 nm range.  $CH_3Cl$  is destroyed mainly by reaction with OH and is therefore not affected very much by an increase in photolysis (22% decrease in concentration at 30 km); we find reasonably good agreement with the few observations of this compound presented in Hudson et al. (1982). Large reductions in  $[CCl_4]$  and  $[C_2H_2Cl_2]$  are found, however, as for  $CFCl_3$  (factor of 0.16 at 30 km). No observations of  $CCl_4$  exist yet, and only tentative measurements (lower limit) of

$C_2H_2Cl_2$  are presented by Fabian et al. (1981); these authors find 1 pptv at 23.3 km, which is about an order of magnitude lower than in our model B. More observations of these compounds are needed.

Above 30 km, the net effect of an increase in flux near 200 nm and a (larger) decrease in  $\sigma(O_2)$  is a slight (up to 20%) decrease in  $O_3$  photolysis rate. This leads to less ozone production and, along with the slight increase in ozone photolysis, to a 10-20% reduction in  $[O_3]$  above 35 km. Our originally somewhat low  $[O_3]$  values in the upper stratosphere thus become 20-30% smaller than the lower limits in the *U.S. Standard Atmosphere 1976*, a discrepancy that we cannot explain at this time. Below 30 km, the decrease in the large total opacity leads to a significant increase in flux between 200 and 220 nm, with a net result of ~30% larger  $O_3$  concentrations at the 20-25 km level. This change near the  $[O_3]$  peak leads to a 15% increase in total column ozone and brings our model into closer agreement with the *U.S. Standard Atmosphere 1976*. The 70% change in  $\sigma(O_2)$  is more important than the 10-20%  $O_3$  reduction above 30 km, which also leads to an increase in flux. Furthermore, the increase in  $O_3$  below 30 km produces a decrease in flux in the lower stratosphere which counteracts the upper stratospheric  $O_3$  reduction effect. To isolate the effect of a change in  $\sigma(O_2)$ , we have run a case identical to model B, but with the  $O_3$  profile fixed as in the Model A case. We find that the largest part (80-90%) of the reductions in the trace species discussed above is due to the change in  $\sigma(O_2)$ , not to the subsequent change in the ozone vertical profile. Moreover, if we fix the  $O_3$  profile as in the *U.S. Standard Atmosphere 1976*, we obtain chlorofluorocarbon concentrations close to the Model B values and actually smaller by up to 30% below 30 km. The Standard ozone concentrations are significantly higher than the model values above 35 km, but again, the chlorofluorocarbon profiles are more sensitive to the ozone profile in the lower stratosphere, where

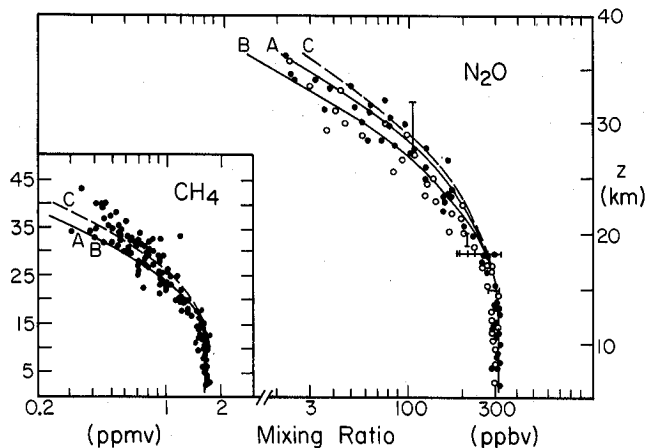


Fig. 3. One-dimensional model fits to  $N_2O$  and  $CH_4$  data.  $N_2O$  data is from 40-45°N, in the summer.  $CH_4$  data is from 40-60°N at various seasons (see Hudson et al., 1982). Models apply to 45°N, summer (see text for details).

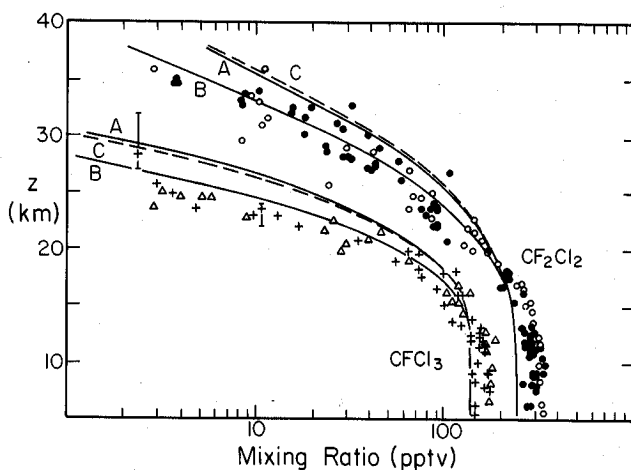


Fig. 4. Sensitivity of  $CF_2Cl_2$  and  $CFCl_3$  to  $O_2$  cross sections near 200 nm and transport (same models as in Fig. 3). Data are from 40-45°N, in the summer (see Hudson et al., 1982).

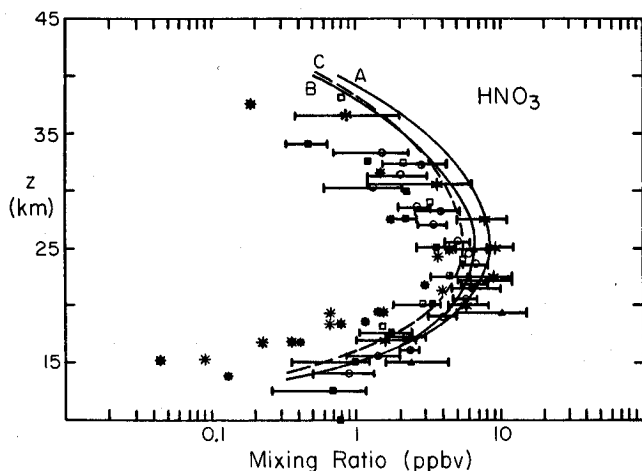


Fig. 5. Same models as for Figs. 3 and 4, but for HNO<sub>3</sub> at mid-latitudes. Data are reproduced from Hudson et al. (1982); see references therein.

the Standard concentrations are somewhat (up to 25%) lower than the Model B values. Uncertainties and variability in [O<sub>3</sub>] in the lower stratosphere can therefore also affect halocarbon and other species concentrations and accurate measurements of O<sub>3</sub> should be performed in conjunction with other observations whenever possible. In the upper stratosphere, we also find a reduction in NO<sub>y</sub> species (NO, NO<sub>2</sub>, NO<sub>3</sub>, N<sub>2</sub>O<sub>5</sub>, HNO<sub>2</sub>, HO<sub>2</sub>NO<sub>2</sub>) by 20-30%, due to the decrease in N<sub>2</sub>O (and a small decrease in O(<sup>1</sup>D)). In the 20-30 km region, the increase in [O<sub>3</sub>] and decrease in [NO] lead to a significant shift in the [ClO]/[Cl] and [HO<sub>2</sub>]/[OH] ratios, which are both almost linearly related to the [O<sub>3</sub>]/[NO] ratio there. [ClO] increases by a factor of 2.3 and [HO<sub>2</sub>] by 1.7 at 20 km; [H<sub>2</sub>O<sub>2</sub>] is increased by a factor of 3 at 20 km, since it depends quadratically on [HO<sub>2</sub>]. HCl, Cl, and OH show little change (~10%) at all altitudes.

#### Conclusions

It is argued that a significant overestimate in the O<sub>2</sub> Herzberg continuum cross sections may in large part be responsible for the overestimate of model source species concentrations (N<sub>2</sub>O, CF<sub>2</sub>Cl<sub>2</sub>, CFC<sub>12</sub> and other halocarbons) in the middle and upper stratosphere. Accurate simultaneous determinations of source species profiles (as presented by Fabian, 1981) provide constraints on both transport and solar flux penetration; we find better agreement with Fabian's data if the model O<sub>2</sub> cross sections are reduced. Recent measurements of solar fluxes near 200 nm in the middle stratosphere have directly revealed a discrepancy between laboratory experiments and atmospheric observations. It is certainly interesting that over 17 years ago, Brewer and Wilson (1965) had measured the direct solar flux in the lower stratosphere and that these somewhat crude observations had already indicated that the O<sub>2</sub> cross sections were probably overestimated by at least 30% near 210 nm. The present paper demonstrates the sensitivity of many stratospheric species to the radiation field in this spectral range, a feature that should be common to 2-D models as well. Similar conclusions have been arrived at independently by the Goddard group (J.R. Herman, private communication, 1982). Accurate knowledge of the lower stratospheric O<sub>3</sub> profile is also important in determining the attenuated solar flux, but current uncertainties in the solar flux outside the Earth's

atmosphere have a much smaller effect on model chlorofluorocarbon profiles than the uncertainties in  $\sigma(\text{O}_2)$  or [O<sub>3</sub>]. These results should motivate further refinement of both laboratory and solar flux measurements related to these small, but important, molecular oxygen cross sections.

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