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MODEL/DATA COMPARISONS OF OZONE IN THE UPPER STRATOSPHERE AND MESOSPHERE

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Abstract. We compare ground-based microwave observations of ozone in the upper stratosphere and mesosphere with daytime observations made from the SME satellite, with nighttime data from the LIMS instrument, and with a diurnal photochemical model. The results suggest that the data are all in reasonable agreement and that the model-data discrepancy is much less than previously thought, particularly in the mesosphere. This appears to be due to the fact that the latest data are lower than earlier reports and the updated model predicts more ozone than older versions. The model and the data agree to within a factor of 1.5 at all altitudes and typically are within 20%.

Introduction

A long standing problem in middle atmospheric science is the fact that photochemical models of ozone from 40 to 80 km have historically predicted significantly less ozone than is observed. In the review by Rusch and Clancy (1987), they state that the discrepancy is on the order of 30-50% in the upper stratosphere, increasing to a factor of 2-3 in the upper mesosphere. There have been a number of proposals advanced to try and reconcile the models and the observations. For example, it has been suggested that the catalytic cycles, such as that due to HO_x (Rusch and Eckman, 1985) which destroy ozone are less efficient than currently assumed. Others have suggested that larger O_2 photolysis cross sections are required (e.g Allen and Delitsky, 1991). This would then increase the production of odd oxygen. Finally, there has also been considerable effort to try and identify a missing ozone source that is not included in the current chemical scheme (e.g. Slanger et al., 1988).

Recently, Natarajan and Callis (1989) have pointed out that the discrepancy in the upper stratosphere can be reduced somewhat if updated rate coefficients and solar fluxes are used in their model along with the somewhat lower total chlorine estimates obtained from the ATMOS data. On the other hand, Allen and Delitsky (1991) argue that by combining the analysis of stratospheric and mesospheric ozone from ATMOS, a consistent, significant discrepancy still exists.

In this paper we pursue the question of the model-data comparison using a variety of datasets including that from the Solar Mesosphere Explorer (SME) for the daytime mesosphere, from the Limb Infrared Monitor of the Stratosphere (LIMS) for the nighttime upper stratosphere and mesosphere and from ground-based microwave data which span the altitude region from 40 to 70 km for both day and night conditions.

Comparison of Observational Data

Figure 1 presents an overview of the ozone data used in our study. The data are all mean profiles for the month of March, at a latitude of 35N, and include both day and night conditions. Although the measurement set spans 12 years (1979 for LIMS, 1982 for SME, and 1991 for the microwave) these data were all obtained during comparable solar activity conditions, near the maximum of either cycles 21 or 22. The figure shows a consistent pattern in that the nighttime data exceed the daytime data, by an amount which increases with decreasing pressure. This reflects the well known mesospheric diurnal variation of ozone whereby atomic oxygen recombines after sunset to increase the ozone density (Zommerfelds et al., 1989). Specific features of these profiles will be discussed below.

The LIMS data that we used were described by Remsberg et al. (1984) and consist of zonal mean ozone data up to 0.1 mb. We have exclusively used nighttime data in order to avoid the non-LTE effects known to be present above about 0.5 mb in the daytime data (Solomon et al., 1986).

SME measured ozone from 1982 to 1986 using an ultraviolet absorption (UVS) technique from 1.0 to 0.1 mb (Rusch et al., 1984) and a near infrared (NIR) emission technique from 0.75 to 0.002 mb (Thomas et al., 1984). Both techniques are valid during daytime conditions only. The SME dataset was reprocessed in 1988 (WMO, 1988) and we used this dataset for our study.

The microwave data come from an instrument at the Table Mountain Observatory (34 deg N, 118 deg W). The instrument was developed at the Millitech Corporation and the data is processed at NASA's Langley Research Center. The data cover the altitude range from 20 to 70 km (.05 mb) and record an observation every 20 minutes. A complete description of the instrument, the observing tech-

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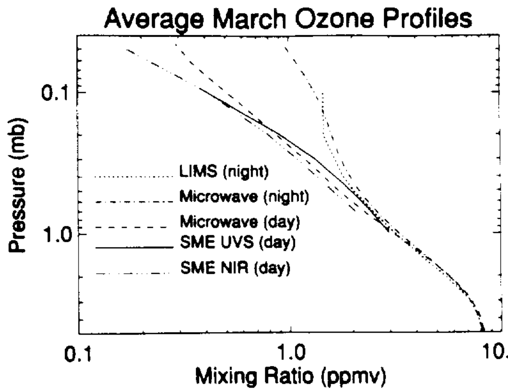

Fig. 1. Summary of monthly mean ozone datasets for 35N latitude. The LIMS data are from March 1979, the SME data from March 1982, and the microwave data from March 1991.

nique and calibration method are described in Parrish et al., (1992). The retrieval method is described by Connor et al., (1991). Comparisons of these data with other ground and satellite obsevations are given by Parrish et al., (1992, these proceedings). It is important to note that the microwave experiment has much lower vertical resolution than the limb scanning satellites (12-16 km for the microwave, 3-4 km for LIMS and SME). Thus, in the discussion which follows we will convolve the high resolution satellite data (as well as the photochemical model) with the microwave averaging kernels as described by Connor et al. (1991).

Figure 2 shows March mean daytime ozone profiles from the SME NIR instrument from 1982 and the ground based microwave from 1991. Both the SME and the microwave data are for 1500 hours local time. Also shown in the figure is the SME data after convolution with the microwave averaging kernel. The effect of the convolution process is most noticeable at the lowest pressure (.05 mb). The figure shows that the convolved SME agrees with the microwave to within the estimated errors of the two experiments (25% for the microwave at .05 mb, 15-20% for SME). It should be noted that the narrower pressure range covered by the UVS instrument (1.0 to 0.1 mb) precludes a direct intercomparison with the low resolution microwave. On the other hand, in the next section we will compare all three daytime ozone profiles to a photochemical model.

Figure 3 shows the ratio of the nighttime LIMS to the nighttime microwave. Because the LIMS data only extend up to 0.1 mb, it is necessary to extrapolate the data up to 0.01 mb in order to convolve with the microwave averaging kernels. The three curves in Figure 3 show the results for three different extrapolations. The solid line uses a profile taken from the photochemical model (discussed below), normalizes it to the LIMS data at 0.1 mb and then is used to extend the data up to 0.01 mb. Using this extrapolation, we find that the LIMS and the microwave data agree to within 5% at all altitudes. The left dotted line simply assumes a constant mixing ratio from 0.1 to 0.01 mb, while the right dotted line uses the microwave a priori profile above 0.1 mb. The purpose of the two dotted curves

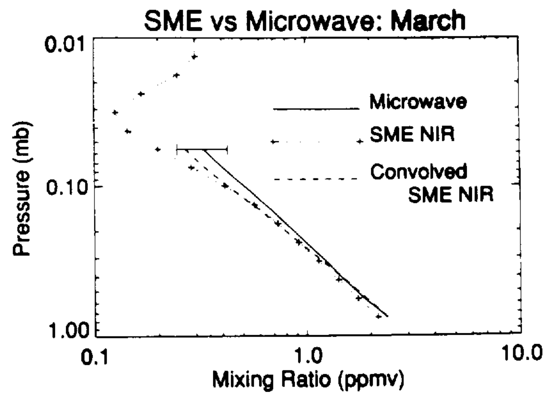

Fig. 2. Daytime ozone data. The dotted line is the SME NIR mean data for March 1982, the solid line is the mean microwave data for March 1991. The dashed curve is the SME data after convolving with the microwave averaging kernels.

is to show that although the agreement between LIMS and microwave is excellent everywhere, there is a mathematical uncertainty of +/- 10% at the top of the profile which results from the need to extrapolate the LIMS data.

Photochemical Model Calculations

The model we use is a one dimensional photochemical model of the middle atmosphere from 40 to 80 km. It has evolved from the model used by Rusch and Eckman (1985) to study daytime mesospheric ozone. For the present application we have expanded its capabilities in three ways. First, we now use complete spherical geometry to calculate the attenuation of the solar UV radiation by ozone and thus improve the O_2 and O_3 photolysis calculations during twilight. Second, while the previous model used the family method to combine O and O_3 as O_x, we now calculate the O and O_3 densities separately for altitudes greater than 60 km and for solar zenith angles greater than 92°. Third, we now account for account the enhanced photolysis of O_3 and NO_2 at near UV and visible wavelengths by inputting a ta-

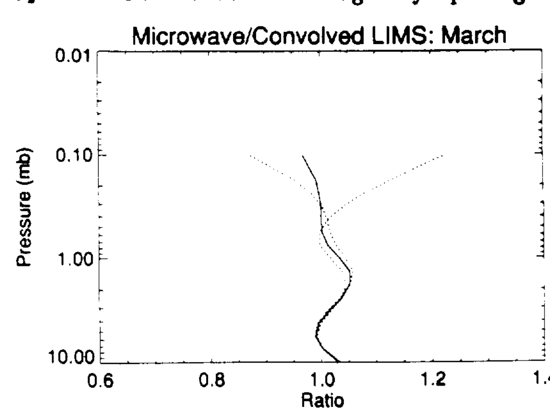

Fig. 3. Ratio of microwave to convolved LIMS. Three different assumptions were used to extrapolate the LIMS data above 0.1 mb when performing the convolution (see text).

ble of scattering enhancement factors which depend upon altitude and solar zenith angle.

The model uses fixed chlorine ($Cl_y = HCl + HOCl + ClO + Cl$), nitrogen ($NO_x = NO + NO_2$), and water vapor inputs. For Cl_y, we used a constant value of 2.2 ppbv for the comparison with the LIMS data, 2.4 ppbv for the comparison with SME, and 3.0 ppbv for the comparison with the microwave data. These chlorine quantities are consistent with that used by Natarajan and Callis (1989) and are lower than those used in earlier studies. For NO_x, we used a fixed value of 15 ppbv, except for the LIMS comparison where we used the LIMS nighttime NO_2 measurement as an indicator of the total NO_x. Our assumed water vapor profile is taken from the LIMS climatology (Remsberg et al., 1990) for the stratosphere and the microwave data of Bevilacqua et al. (1989) for the mesosphere. Finally, the model uses different temperatures when comparing with different datasets: for the LIMS comparison, simultaneously measured LIMS temperatures are used, for the microwave comparison, the temperatures are those used in the data processing, and for SME, the reference climatology of Cole and Kantor (1978) was adopted.

One limitation of the model is its neglect of vertical transport. While this will not be important for altitudes where the lifetime of ozone is short (< 75 km), it could introduce an uncertainty when comparing with the low resolution microwave data at 70 km. We therefore have limited our comparison of the microwave data and the convolved model to altitudes below 66 km (0.1 mb).

Figure 4a compares the ratio of the two nighttime datasets to either the model (in the case of LIMS) or the convolved model (for the microwave). A similar comparison with the March LIMS data was presented by Natarajan and Callis (1989); however, their analysis only went up to 0.5 mb (52 km). Here, we extend the comparison up to 0.1 mb. The figure shows that the data exceeds the model at all pressures, consistent with all previous studies of the problem. The detailed shape of the difference agrees very well with that seen by Natarajan and Callis in the pressure range where our two studies overlap, a minimum from 1.0 to 0.5 mb with a maximum at 3 mb. In addition, our analysis suggests that the discrepancy increases above 0.5 mb to reach a maximum of a factor of 1.3 at 0.1 mb. Also, the figure shows remarkable agreement between the LIMS and microwave comparisons, despite the different inputs to the model (described above) and the 12 year separation in the observations. Given the lack of other nighttime mesospheric ozone measurements, the combination of this comparison and that shown in Figure 3 serves as the first reliable validation of the LIMS nighttime ozone in the mesosphere.

Figure 4b presents a similar model-data comparison for the three daytime data sets. It shows the ratio of the three daytime datasets to either the model (in the case of SME) or the convolved model (for the microwave). At 1.0 mb, the UVS and the microwave data exceed the respective model by a factor of 1.15-1.20. At pressures below 1.0 mb, the discrepancy between the model and the UVS worsens to reach a factor of 1.35 at 0.3 mb. The discrepancy between the microwave and the SME NIR show somewhat different behavior and seem to agree slightly better. Above 0.1 mb the model and the NIR data come into closer agreement. The pattern here is not as consistent as that seen in Figure 4a. Part of this undoubtedly reflects the 20% disagreement which exists between the two simultaneous SME measurements at certain pressures (e.g 0.4 mb). In addition the temperatures used in the comparison between the model and the SME datasets, which are from the Cole and Kantor (1978) climatology, may not be as realistic as those used in analysing the microwave data (from the National Meteorological Center analysis and the MAP climatology).

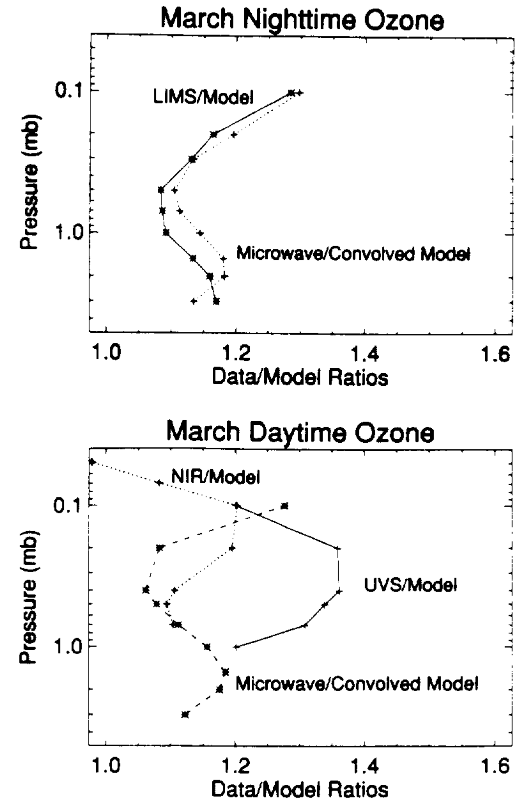

Fig. 4. (a) Ratio of March nighttime ozone to photochemical model (in the case of LIMS data- solid line) and convolved model (for the microwave- dotted line). (b) Ratio of March daytime ozone to photochemical model (solid line is for the SME UVS data, the dotted line is for the SME NIR data) and with convolved model (dashed line- for the microwave data).

Discussion

In both Figures 4a and 4b, the data-model ratios are lower than previously reported. For example, using SME UVS data from 1983, Rusch and Eckman (1985) reported a discrepancy of 1.8 during March at 40 N, with a larger discrepancy during winter. Here, however, the peak discrepancy from all 5 ozone observations lies in the range 1.2 - 1.35. One reason for this change is that the model now uses a faster $OH + HO_2$ rate coefficient (see JPL90 for a disussion) and thus predicts less HO_x and about 1.15 times more ozone at 0.1 mb. Second, the reprocessed SME data is generally lower than the original version used by Rusch and Eckman (1985). Some of this is discussed in the WMO (1988) report and is attributed to a removal of a calibration drift, corrections to the field of view and the determination of tangent point altitude as well as a 12% change in the absolute calibration of the NIR instrument (R. Thomas, private communication, 1992). A example of the difference is shown in Figure 5 which compares the new SME NIR data for March 1982 with the version 1 data as given the MAP Handbook (1985). It can be seen that at 0.1 mb, the old data is more than 1.3 times greater than the new. The combination of the newer, lower data (factor of 1.2 - 1.4) and the higher model (factor of 1.10 - 1.15) leads to a significantly reduced discrepancy between model and data.

Summary

We have compared ground based microwave data with satellite observations from LIMS and SME. For both day and night, the observations agree to within 25%. When compared to photochemical model calculations, the data exceed the model at all altitudes, but by an amount which is much smaller than earlier reports. This is because the revised model predicts more ozone while the newer observations report less.

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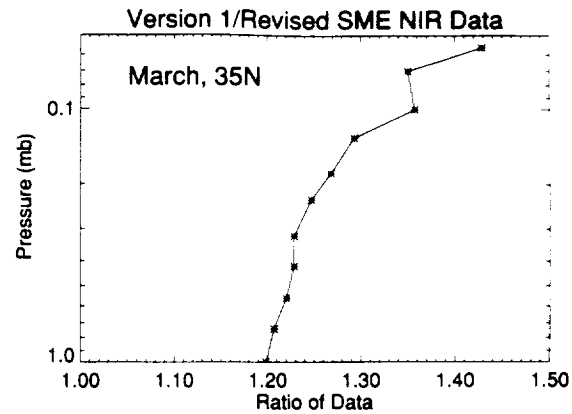

Fig. 5. A comparison of version 1 and the revised SME NIR for March 1982. The older dataset (version 1) contains larger values.

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