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RADIATIVE FORCING OF TROPOSPHERIC OZONE

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1. INTRODUCTION

The capability of GCMs to predict the radiative forcing of gases such as tropospheric ozone in the lower troposphere is limited. The confidence level reported in IPCC (2001) for the radiative forcing of tropospheric ozone is medium; whereas for other gases distributed throughout the atmosphere it is high. The low confidence level is mainly associated with the fact that ozone has large spatial and temporal variations that make it difficult to accurately model. Another contribution to the low confidence level is related to the presence of cloud cover. The radiative forcing of gases that are present beneath clouds is a model unsolvable problem with current GCMs due to the definition of radiative forcing that is used in these models. This definition involves modelling the radiative forcing at the top of the tropopause. Based on this definition, any changes in the radiative forcing of gases beneath clouds are completely obscured by the overlying cloud layer. Hence, the GCMs cannot calculate the radiative forcing of gases beneath cloud cover, and only an estimate of this quantity is provided in the IPCC reports (2001). Perhaps as a result of the poor state of knowledge of the radiative forcing associated with tropospheric ozone, its reduction has been omitted in the recent Kyoto protocol for the reduction of greenhouse gases.

In this paper we present the results of a number of measurements of the radiative forcing from tropospheric greenhouse gases that were obtained by a ground-based remote-sensing technique. The technique is based on earlier work (Puckrin et al., 1996) which uses Fourier-transform infrared (FTIR) spectroscopy to measure the atmospheric thermal emission from gases beneath uniform cloud cover. The cloud acts as a cold background emission source against which the emission from gases in the warmer atmosphere beneath the cloud may be detected. The region of the infrared spectrum near 2400 cm^{-1}, which has minimal atmospheric water vapour emission features, is used to infer the cloud base temperature. The FASCOD3 atmospheric transmission code (Clough et al., 1988) is used to simulate the background emission spectrum below the cloud, which is then subtracted from the measured spectrum to yield the thermal emission band of a particular gas in the lower troposphere. By matching the measured band intensity with the simulated one, the radiative forcing of the gas below the cloud can be determined. In order to have sufficient detection sensitivity for the gas column amount, the cloud base should exceed an altitude of about 1 km. The gases that have been successfully measured with this technique include tropospheric ozone, carbon monoxide, carbon dioxide, methane and nitrous oxide. However, since tropospheric ozone plays one of the more important roles in climate forcing, its investigation is the major focus of this paper.

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2. EXPERIMENTAL TECHNIQUE

Downwelling thermal emission spectra of the atmosphere beneath cloud cover were measured at a resolution of 0.25 cm^{-1} using a FTIR spectrometer (Bomem, model DA8) with a half-angle field-of-view of 0.2 degrees. The instrument incorporated either a liquid-nitrogen-cooled, narrow-band, MCT detector or an InSb detector.

The downward zenith sky radiation emitted by the cloud and the underlying atmosphere was collected by positioning a gold-coated mirror at a 45-degree angle relative to the emission port of the instrument. Immediately after the measurement of the sky radiation the mirror was pointed towards a blackened dewar of liquid nitrogen. The subsequent spectrum collected from this very low emittance source characterised the thermal emission background of the instrument. The subtraction of this background from the sky spectrum revealed an emission spectrum pertaining only to the cloud and the atmosphere below the cloud.

The calibration of the emission intensity scale was performed by placing a blackbody source beneath the gold mirror, filling the field-of-view of the spectrometer. The temperature of the blackbody was monitored by a chromel-alumel thermocouple. Each thermal emission measurement lasted for a period ranging from one to ten minutes, depending on the uniformity of the cloud cover. The resulting rms signal-to-noise ratio was usually better than 100:1.

3. RESULTS AND DISCUSSION

An example of a thermal emission spectrum measured beneath cloud on May 3, 2000 is shown in Figure 1. The contributions to the total thermal emission intensity include the cloud emission and the water continuum of the lower atmosphere beneath the cloud. Other influences include the intense emission from the carbon dioxide band at 600 cm^{-1}, methane at 1300 cm^{-1}, ozone at 1050 cm^{-1} and several minor contributions from other gases. By far, the largest contribution to the total radiance is from the blackbody emission from the cloud itself.

In order to extract useful information from the cloud emission measurement, it is imperative that the cloud exhibit blackbody behaviour; that is, the cloud must absorb all of the radiation incident upon it, and reflect and transmit none of it. Under these conditions the measured emission spectrum is solely representative of the emission from the cloud base and the gases beneath it. To determine the optical depth that a cloud must have in order for it to display blackbody characteristics, several emission spectra were simulated for clouds of varying thickness. The results of these simulations are shown in Figure 2. The simulations were carried out with the MODTRAN4 transmission code (Abreu et al., 1991), which correctly accounts for the multiple scattering in the cloud. From Figure 2, it

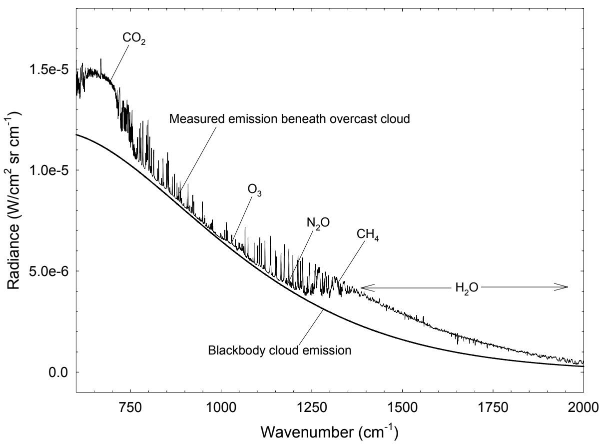

Figure 1: An example of the measurement of the thermal emission spectrum beneath overcast cloud. Several emission features are identified in the spectrum, including the large thermal contribution from the cloud itself, which behaves as a blackbody radiator.

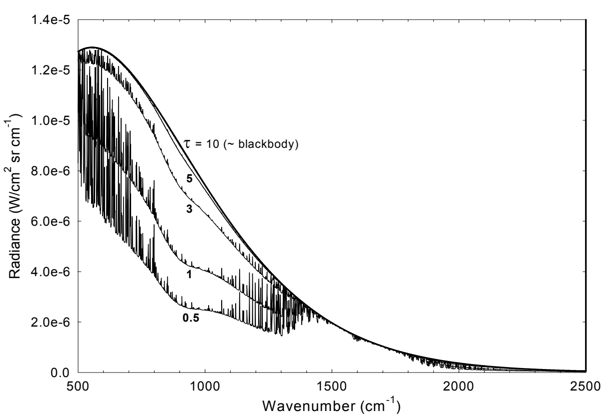

Figure 2: The thermal emission of clouds of varying optical depth, τ. The spectra were simulated with the MODTRAN4 transmission model for varying cloud thickness. When an optical thickness of about eight is achieved then the cloud takes on the characteristic shape of a blackbody radiator. An optical depth of 8 translates to a cloud thickness of about 100m.

can be seen that when a cloud reaches an optical depth of about eight it exhibits blackbody behaviour, as indicated by the good agreement with the emission spectrum of a true Planck blackbody. An optical depth of eight translates into a physical cloud thickness of about 100 m, depending on the cloud type. In practice, whenever the solar disk of the sun is completely obscured, the cloud is assumed to behave as a perfect blackbody. The next parameter that needs to be determined is the cloud base temperature. In order to determine this quantity a region of the thermal emission spectrum was chosen which exhibited a minimal influence from atmospheric water vapour, so as not to mask the cloud emission intensity. From the water line strengths listed in the 1996 HITRAN database (Rothman et al., 1998), it was observed that the best region of the spectrum with minimal water emission intensity was in the 2400 - 2500 cm^{-1} region. In this region the water vapour line strength is several orders of magnitude less than that of surrounding regions, and the atmosphere is nearly transparent in this region. Hence, the region from 2400 - 2500 cm^{-1} can be used to estimate the cloud base temperature. This figure shows the measured spectrum obtained beneath a cloud and two simulated spectra; one with water vapour present in the atmosphere beneath the cloud and the other without water vapour present. The two simulations show that no significant difference occurs in the intensity in the 2400 - 2500 cm^{-1} region, and we conclude that atmospheric water vapour, or any other gas for that matter, contributes insignificantly to the thermal emission spectrum in this region. By fitting a Planck blackbody curve to this portion of the thermal emission spectrum, we are able to estimate the cloud base temperature, as shown in Figure 3. For this cloud measurement, it was determined that the cloud base was at a temperature of about 285.3 K.

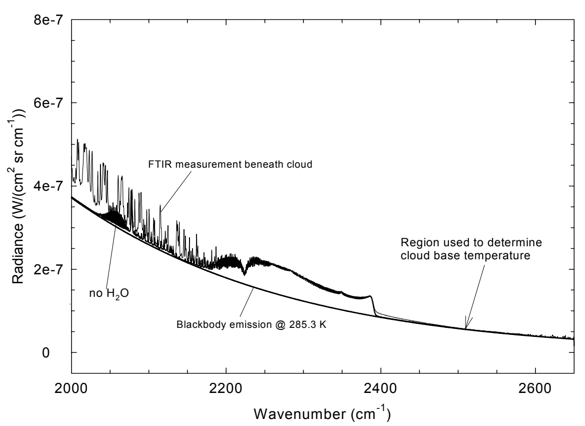

Figure 3: Use of the thermal emission spectrum to determine the cloud base temperature. Matching the emission intensity in the 2500 cm^{-1} region to an appropriate Planck blackbody emission curve yields the temperature of the cloud base.

With the cloud base and surface temperatures known, and the height of the cloud determined from the local weather office, the temperature profile between the Earth's surface and the cloud base can be determined by interpolation of the temperature end points. This information is input into the FASCOD3 transmission model to simulate the background radiance spectrum in the absence of ozone, as shown in Figure 4. The subtraction of the simulated background emission spectrum from the measured total spectrum yields the thermal emission band of ozone beneath cloud cover. Integrating this result over the full sky yields a greenhouse forcing flux of 0.43 W/m^2.

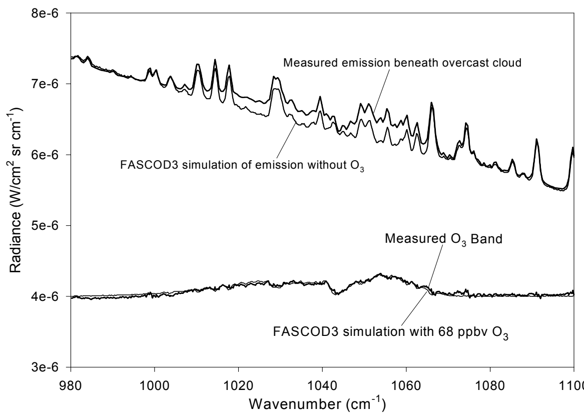

Figure 4: Determination of the ozone surface forcing beneath overcast cloud. A FASCOD3 simulation of the background emission in the absence of ozone was performed and subtracted from the measured total emission, which isolated the thermal emission band of tropospheric ozone. The radiative forcing was determined to be 0.43 W/m^2.

This analysis was repeated for several other measurements taken for summer days during the past two years. A summary of the surface radiative forcing fluxes for tropospheric ozone is shown in Figure 5. The average surface forcing associated with tropospheric ozone was measured to be 0.23 W/m^2. Along with the measured surface forcing values, the radiative trapping flux was also calculated in order to make a comparison with the current ozone forcing reported by IPCC (1995). The trapped fluxes were simulated with the FASCOD3

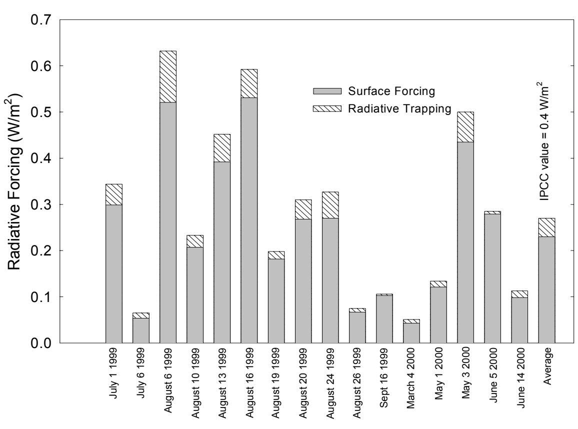

Figure 5: Summary of the surface forcing and radiative trapping of tropospheric ozone for the 2000-01 summers. The surface forcing fluxes are based on the FTIR measurements. The radiative trapping fluxes are based on simulations with the FASCOD3 model.

model by reversing the model geometry so as to calculate the change in the upward flux at the tropopause due to the measured tropospheric ozone layer, and with no cloud present. As shown in Figure 5, the trapped flux is always greater than the surface flux due to the interference role that water vapour plays in reducing the surface forcing flux (Evans and Puckrin, 2001). The average trapped flux of about 0.3 W/m^2 shown in Figure 5 for tropospheric ozone is about 25% less than the accepted value of 0.4 W/m^2 (IPCC, 1995). However, it is interesting to note that the latest IPCC2001 report has downgraded the radiative forcing estimate to a value of 0.3 W/m^2.

The results of more recent measurements made during the past year are summarized in Figure 6. This figure shows the seasonal variation in the forcing attributed to tropospheric ozone. As expected, there is only a small forcing in the winter months due to the lack of photochemical activity.

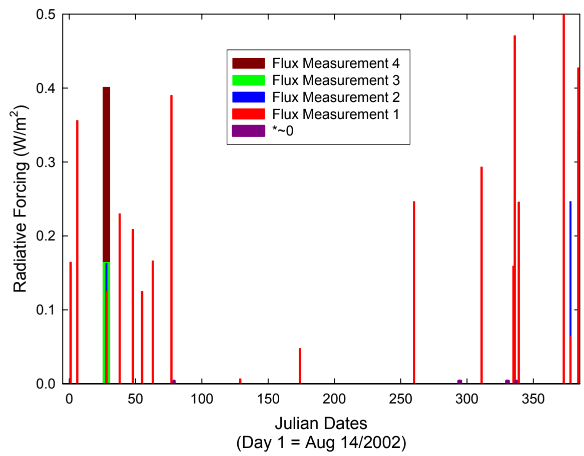

Figure 6: The seasonal variation in the radiative forcing of tropospheric ozone over a one-year period. Note: the measurements indicated by ~0 represent results that yielded negligible radiative forcing from ozone. This was attributed to relatively high cloud temperatures which gave minimal emission features and in some cases absorption features.

4. CONCLUSIONS

Significant progress has been made towards increasing the certainty of the climate forcing associated with tropospheric ozone. Measurements of the tropospheric ozone surface forcing made for a number of summer days during the past two years have shown that the average surface forcing is about 0.23 W/m^2. This translates to a radiative forcing of about 0.3 W/m^2, consistent with the value of 0.3 W/m^2, which is cited in the IPCC 2001 report. In order to decrease the uncertainty of the tropospheric ozone forcing measurements further, more measurements need to be made at different latitudes and climates.

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