P1.7 MEASUREMENTS OF THE RADIATIVE SURFACE FORCING OF CLIMATE

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1. INTRODUCTION

One of the most pressing environmental issues encountered in this present century is that concerning the global warming of the earth's surface. This change may have a detrimental impact on society as a whole, and particularly on human health, ecology and, consequentially, on economical viability.

Over the past century the world has been getting warmer; an average 0.7°C increase has been observed in the global land-surface temperature records (IPCC, 2001) and the past decade has been the warmest yet since records were first kept. The potential impact that global warming has on society is extensive, particularly through the associated change in climate that has been predicted by many sophisticated climate models. The areas that may be affected the most by global warming include: water and coastal resources, health, agriculture, fisheries, forests and energy (Canada's Second National Report on Climate Change, 1997).

In order to investigate this global threat, an ongoing program of measurements of the downward atmospheric infrared radiation, otherwise known as the green-house radiation of the atmosphere, was undertaken at Trent University in Peterborough, Ontario (44°N, 78°W).

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The measurements have been obtained using commercial Fourier-transform infrared (FTIR) spectrometers. These measurements have been used to quantify the radiative flux associated with a number of greenhouse gases. It is this radiative flux that provides an additional source of warming for the planet's surface, and ultimately is responsible for any change in climate. We have provided the first direct measurements of the greenhouse effect for a number of trace gases in the atmosphere. These gases include trichlorofluoromethane (CFC-11), dichlorodifluoromethane (CFC-12), carbon tetrachloride (CCl_4), methane (CH_4), nitrous oxide (N_2O), carbon monoxide (CO), nitric acid (HNO_3), and tropospheric ozone (Evans and Puckrin, 1994-1997; Puckrin et al., 1996). Not only do these results prove that an increase in the greenhouse effect is real, and that trace gases in the atmosphere are adding a significant radiative burden to the energy budget of the atmosphere, but they also provide a means of validating the predictions that are made by global warming models (Ellingson et al., 1991). This last point is crucial since the temperature increases predicted by the various climate models can vary by several degrees; even a change of 0.7°C can have significant consequences on different parts of the globe. The cause of the large uncertainty in the models resides in the difficulty of accurately predicting the climate feedback mechanisms that are associated with the interaction of oceans, vegetation, and clouds and water vapour with the greenhouse effect.

In order to evaluate the contributions of the various gases to global warming, the concept of radiative forcing, or absorption of

the upward longwave radiation from the earth's surface by the atmosphere, has been formulated (Ramanathan, 1987; IPCC, 1994). The radiative forcing at the tropopause is then used as an input to drive climate models for the purpose of evaluating the global warming for various gases. Model calculations of this radiative forcing have been conducted by several authors (e.g., Dickinson and Cicerone, 1986; Hauglustaine et al., 1994). Earlier estimates of the greenhouse effect were in terms of the overall loss of radiation to space or of the downward radiation at the surface. It may be noted that, when convection just above the ground is small, the earth's surface actually responds to the downward greenhouse radiation from the atmosphere at the surface; over land, the temperature responds to the greenhouse radiation in a few hours. The greenhouse radiation is typically about 150 W/m^2; the modelled increases in the greenhouse radiation since pre-industrial times are about 3 W/m^2. The increase in downward surface greenhouse radiation for a particular gas is quite similar to the absorption of upward longwave radiation by the atmosphere (radiative forcing) as has been demonstrated by Sinha and Toumi (1996). An estimate of the radiative forcing (or net change in absorbed upward flux at the tropopause) for several gases is shown in Table 1.

Table 1: Radiative Forcing at the Tropopause

Greenhouse Gas	Radiative Forcing (W/m^2)
H_2O	~90
CO_2	~50
CH_4	1.7
N_2O	1.3
Tropospheric O_3	1.3
CFC-12	0.16
CFC-11	0.062

Note: The modelled forcing for the first five gases is based on the work of Dickinson and Cicerone (1986), and the forcing attributed to the CFCs is based on the IPCC report (1995).

The data is based on models by Dickinson and Cicerone (1986) and those which are included in the IPCC report (IPCC, 1990). Although there are extensive model calculations of radiative forcing in the literature, there are few experimental measurements of the greenhouse radiation which is responsible for global warming. In what follows, measurements of the surface greenhouse radiation are described and compared with the modelled estimates of radiative forcing.

2. METHODOLOGY

The measurements of the downward atmospheric thermal emission were collected using a Magna 550 FTIR spectrometer or a high resolution Bomem DA8 system; the instruments were capable of resolutions of 0.25 cm^{-1} and 0.02 cm^{-1}, respectively. Both instruments incorporated a liquid-nitrogen-cooled, narrow-band, MCT detector with a 1 mm^2 element. The downward zenith sky radiation from the clear sky was collected by positioning a gold-coated mirror at the emission port along the optical axis of the instrument. A stored-phase correction was applied to the measured interferogram before conversion was made to the spectral domain in order to account for phase changes that were present at 750 and 2000 cm^{-1}. The thermal emission background of the instrument was characterized by measuring a negligible source of thermal radiation which consisted of a blackened dewar containing liquid nitrogen. The background measurement was taken immediately prior to and after the measurement of the sky radiation to ensure that the spectrometer was thermally stabilized.

The calibration of the atmospheric measurements was performed by placing an ambient 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. The atmospheric emission measurements required 15-30

minutes of observing time. This resulted in a typical root-mean-square noise value of about 5.0×10^{-9} W/(cm^2 sr cm^{-1}) in the mid-infrared region.

The greenhouse radiation from tropospheric ozone was measured by a technique in which the base of cold clouds was used as a target. The thermal emission from the warm atmosphere below the cloud was measured against the low background emission from the cold cloud base (Puckrin et al., 1996). The cloud also screened out the emission from the stratospheric ozone above it, effectively restricting the sampling area to the lower troposphere.

3. RESULTS AND DISCUSSION

A typical winter spectrum of the downward radiance in the 5-16 μm wavelength range is shown in Figure 1, with the emission from several greenhouse gases identified. The spectrum was measured at a resolution of 0.25 cm^{-1}.

In order to extract the greenhouse flux from individual gases, the background emission of the atmosphere was simulated using the radiative transfer code, FASCOD3 (Clough et al., 1988). The simulations incorporated the temperature, relative humidity and pressure profiles from radiosonde measurements obtained at Maniwaki, Quebec, a location 280 km distant from Peterborough. A constant mixing ratio profile of 360 ppmv was used for carbon dioxide (IPCC, 1995) and the concentrations of other background gases were taken from the AFGL Atmospheric Constituent profiles (Anderson et al., 1986) and scaled to current tropospheric concentrations (IPCC, 1995). The line transition parameters for the molecules were provided from the 1992 AFGL HITRAN database (Rothman et al., 1992). The model utilized an aerosol profile that was representative of the visibility conditions as monitored by the local weather office. An example using this procedure is illustrated for CFC-12.

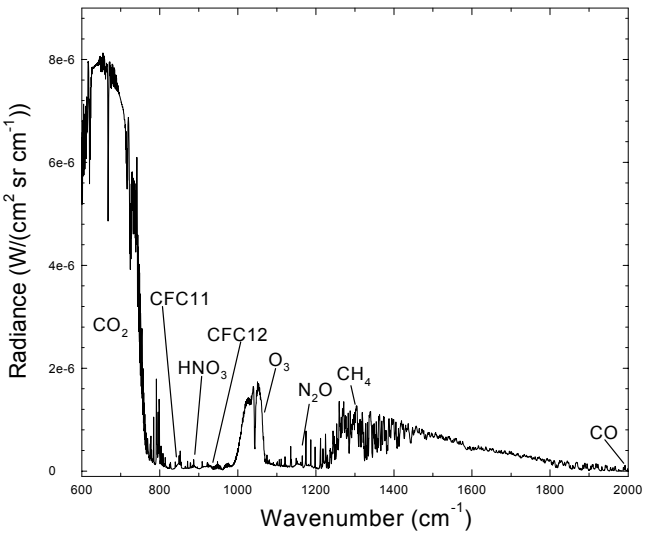

Figure 1. A spectrum of the greenhouse radiation at the surface measured for February, 1996, showing the contributions of several greenhouse gases.

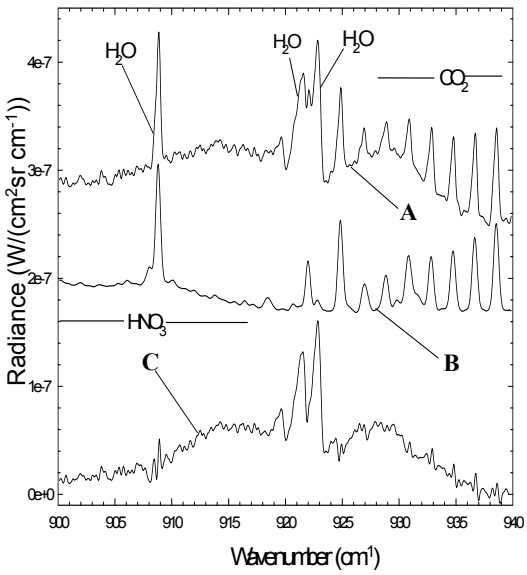

Figure 2. The extraction of the thermal emission band of CFC-12 from the measured atmospheric emission spectrum (curve A). Curve B represents the simulated background thermal emission in the

absence of CFC-12. The subtraction of curve B from curve A shows the thermal radiation associated with CFC-12 (curve C). Curve A in Figure 2 shows the measured downward thermal emission for January, 1994. Besides the emission from CFC-12 in the 900-940 cm^{-1} region, other gases including nitric acid, water vapour and carbon dioxide have emission bands that overlap with those of CFC-12, as noted in the figure. The simulated emission of these other gases is represented by curve B in Figure 2. Subtracting the simulated background thermal emission from the measured emission yields the measured downward greenhouse radiation at the surface that is associated with atmospheric CFC-12 (curve C). The downward radiation fluxes at the surface for all greenhouse gases measured up to 1996 are summarized in Table 2.

Table 2: Measured Greenhouse Fluxes at the Earth's Surface

Greenhouse Gas	Emission Band (cm^{-1})	Measured Flux (W/m^2)	Simulated Flux (W/m^2)
CFC11	830 – 860	0.14	0.12
CFC12	900 – 940	0.12	0.11
CFC12	all bands	0.28	0.26
CFC11 & 12	all bands	0.42	0.38
CCl_4	786 – 806	0.046	0.039
CFC113	800 – 830	NA	0.033
HCFC22	780 – 830	NA	0.031
HNO_3	850 – 920	0.085	0.060
N_2O	all bands	1.06	0.99
CH_4	1200 – 1400	0.85	0.80
CO	2000 – 2200	0.032	0.033
CO_2	all bands	26.0	24.8
O_3	950 – 1100	3.26	3.20
Trop. O_3	950 – 1100	0.61	0.58

These are relevant for a mid-latitude location in the wintertime. The measurement pertaining to tropospheric ozone was made in moderately polluted springtime air for an ozone concentration of 60 ppbv. A comparison with simulated fluxes predicted by the FASCOD3 atmospheric transmission code (Clough et al., 1988) for the actual atmospheric conditions and recent atmospheric gas concentrations (Kaye et al., 1994; IPCC, 1995) is shown in the last column of Table 2. In general, there is a good correlation between the measured and simulated radiative fluxes implying that the FASCOD3 radiation code is very reliable. The flux measurements presented in Table 2 provide important experimental verification of the driving radiation that is responsible for global warming. The effect of this radiative perturbation on the longwave radiation budget of the earth may introduce significant changes in the climate system. Continued measurements of the downward greenhouse flux made over a range of locations and seasons would provide useful information for validating global climate models and for resolving the discrepancies that currently exist between them. For example, a comparison of surface fluxes from 20 models has been reported by Ellingson et al. (1991). In most of these, only the CO_2 fluxes are reported although the combined CH_4 and N_2O flux were reported for one model; the reported modelled value of 1.7 W/m^2 was not inconsistent with our combined experimental value of 1.91 W/m^2.

Figure 3 shows a spectrum of greenhouse radiation forcing measured in summer at Peterborough (44N). In comparison with Figure 1, the spectrum has been altered by the appearance of many large water vapour lines in the emission spectrum. The water vapour provides an interference effect on the emission from the other greenhouse gases which is large.

Table 3a shows the measured downward surface radiation forcing for three winter cases. CO_2 is around 33 W/m^2, CH_4 is 1.25 W/m^2, N_2O is 1.25 W/m^2 and CFCs contribute around 0.25 W/m^2.

Table 3b shows the measured downward surface radiation forcing for three summer cases. The H_2O flux has increased from about 100 W/m^2 to 200 W/m^2 . CO_2 is reduced from 33 W/m^2 to 11 W/m^2 CH_4 is reduced from 1.25 W/m^2 to 0.8 W/m^2. N_2O is reduced from 1.25 W/m^2 to 0.8 W/m^2. O_3 is reduced from 3.2 W/m^2 to 2.6 W/m^2 in summer. The CFCs contribution is about the same around 0.25 W/m^2 and hence the relative contribution as compared to CO_2 has increased.

Table 4 shows the contribution of greenhouse gases to the increase in the downward greenhouse flux since the pre-industrial period. Simulations using FASCOD3 were performed to estimate the greenhouse flux from the various gases using their respective tropospheric concentrations from two centuries ago (IPCC, 1995; Dickinson and Cicerone, 1986). Most of these simulated fluxes have been verified with the experimental measurements in Table 4, so that there is some confidence that these modelled flux increases are representative of northern middle latitudes. In these calculations, the amount of atmospheric water vapour has been assumed to be invariant. The pre-industrial and current concentrations of gases used in the flux calculations were taken from IPCC, 1990. The current tropospheric ozone amount of 60 ppbv over the first two kilometers of the atmosphere is typical of a moderately polluted environment. From Table 4 it is apparent that the increase in the carbon dioxide concentration since pre-industrial times has resulted in the largest increase in the radiative forcing at the surface. After CO_2, the chlorofluorocarbons contribute the second largest amount to the flux (20.8%); however, carbon dioxide has increased about 100,000 times more than the combined concentrations of the chlorofluorocarbons since the pre-industrial period.

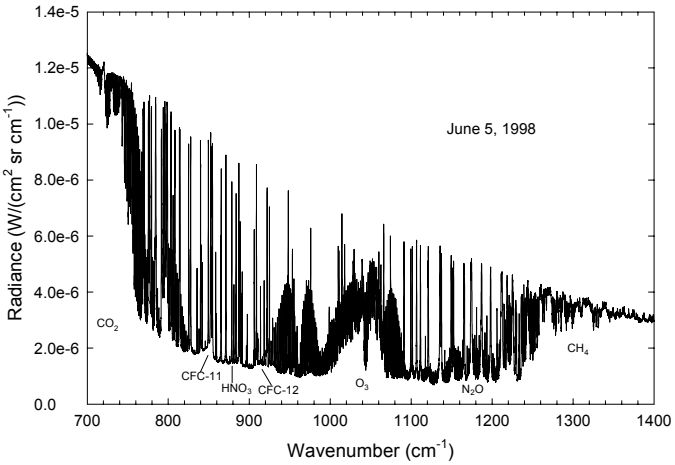

Figure 3. A spectrum of greenhouse radiation forcing measured in summer showing enhanced water lines.

Table 3a: Measured Winter Downward Surface Fluxes

Greenhouse Gas	Winter Fluxes (W/m^2) 1999	2000	Past
CFC11	0.13	0.10	0.14
CFC12	0.24	0.22	0.28
HNO_3	0.061	0.065	0.052
CH_4	1.30	1.29	0.96
N_2O	1.34	1.41	1.04
O_3	3.03	3.17	3.27
H_2O	94.1	105.4	125
CO_2		34.7	30.9
Total			

Table 3b: Measured Summer Downward Surface Fluxes

Greenhouse Gas	Summer Fluxes (W/m^2) 1998	1999	Past
CFC11 all	0.15	0.11	0.15
CFC12 all	0.29	0.24	0.27
HNO_3	0.075	0.063	0.066
CH_4	1.16	0.60	1.08
N_2O	1.14	0.64	0.89
O_3	2.57	2.47	2.61
H_2O	178	256	251
CO_2		10.5	10.5
Total			

Table 4: Comparison of Measured and Model Increases in Downward Surface Flux

Greenhouse Gas	Measured Flux Increase (W/m^2)	Model Flux Increase (W/m^2)
CO_2	2.10	1.30
CH_4	0.38	0.33
N_2O	0.15	0.13
Trop. O_3	0.40	0.40
CFC11	0.14	0.14
CFC12	0.28	0.28
CFC113	0.00	0.033
HCFC22	0.031	0.031
CCl_4	0.046	0.046
Total	3.52	2.55 (-3.1)

In Table 4, the contribution of greenhouse gases to the downward greenhouse flux is shown since the pre-industrial period. The increase in radiative forcing at the tropopause as taken from IPCC, 1995 is also shown in brackets for comparison in Table 4; the total increase in the modelled radiative trapping was 3.1 ± 0.5 W/m^2. The contribution of water vapour to the increase in greenhouse radiation has not been included since it is a part of the natural climate feedback. There is some argument to suggest that tropospheric water vapour has already increased by several percent; hence, the corresponding flux contribution may need to be included, but this effect is beyond the scope of current models. From Table 4 it is evident that the actual greenhouse radiation has increased by over 3.5 W/m^2 since pre-industrial times, or by about 2.3% of the total greenhouse radiation. Of this 3.5 W/m^2, about 0.5 W/m^2 has been measured due to the chlorofluorocarbons.

4. **CONCLUSIONS**

Measurements of the downward radiative flux have been made for several important greenhouse gases. At mid-latitudes in summer as compared to winter, our measurements show that the downward surface flux from H_2O has doubled to 200 W/m^2. The water increase causes a reduction of the fluxes from the other greenhouse gases. These measurements show that the greenhouse effect from trace gases in the atmosphere is real and adds significantly to the radiative burden of the atmosphere. The greenhouse radiation has increased by approximately 3.52 W/m^2 since pre-industrial times.

This compares favorably with a modeled prediction of 2.55 W/m^2. Measurements such as these can provide a means by which to verify the predictions made by global warming models (Puckrin et al; 2004).

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