

# Atmospheric Ozone as A Climate Gas

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## Project Objectives--

To develop and apply climate-chemistry models for:

- Understanding the physical, chemical and dynamic processes that control mid-latitude O<sub>3</sub> in the lower stratosphere and free troposphere;
- Developing improved predictions of future O<sub>3</sub> changes in these regions and their influence on (and response to) future climate changes due to increasing greenhouse gases CO<sub>2</sub>, N<sub>2</sub>O, CH<sub>4</sub> and the CFCs, and changes of O<sub>3</sub> precursor gases.

Two specific studies are reported :

- I. The effect of cirrus cloud on photodissociation rate coefficients J(O<sup>1</sup>D) and J(NO<sub>2</sub>)
- II. The Impact on radiative forcing from aircraft emissions

## I. The Effect of Cirrus Clouds on Photodissociation Rate Coefficients J(O<sup>1</sup>D) and J(NO<sub>2</sub>)

### Rationale

- Photodissociation rate coefficient, usually referred as *J* value, is calculated by

$$J = \int \sigma(\lambda) \phi(\lambda) F(\lambda) d\lambda ,$$

where  $\sigma(\lambda)$  is absorption cross section,  $\phi(\lambda)$  is quantum yield, and  $F(\lambda)$  is actinic flux. The effect of clouds is particularly important because of the great impact on actinic flux induced by the scattering of cloud drops. Such impact has been demonstrated by observational and theoretical modeling studies.

- The presence of cirrus cloud can alter O<sub>3</sub> photochemistry near the tropopause and consequently lead to changes in O<sub>3</sub> concentration near that region. Studies show that radiative forcing induced by O<sub>3</sub> changes near the tropopause has important climate implication. Therefore, this study will focus on the effect of cirrus clouds on *J* values.
- The production of O(<sup>1</sup>D) from O<sub>3</sub> photodissociation is a major source of hydroxyl radical OH, the most important oxidant in the tropospheric chemistry. NO<sub>2</sub> photodissociation is also important as it plays a key role in tropospheric O<sub>3</sub> formation. J(O<sup>1</sup>D) and J(NO<sub>2</sub>), therefore, are calculated to examine the impact of the presence of cirrus clouds.

## **Approach**

Detailed calculations are carried out to study the sensitivity of photodissociation rate coefficients  $J(^1D)$  and  $J(NO_2)$  to several important factors, *e.g.* cloud optical properties, solar zenith angle and surface albedo.

### 1. Model and parameterization

The model is PHODIS by Kylling (1994) customized for this study. The parameterization of cirrus clouds in Ebert and Curry (1992) is adopted. Optical depth ( $\tau$ ), single scattering albedo ( $\omega$ ) and asymmetry factor ( $g$ ) are calculated by the following formula using ice water path (IWP) and effective radius ( $r_e$ ):

$$\tau_i = IWP(a_i + b_i/r_e)$$

$$1 - \omega_i = c_i + d_i r_e$$

$$g_i = e_i + f_i r_e$$

over two bands: 0.25-0.7  $\mu\text{m}$ , and 0.7-0.85  $\mu\text{m}$ . The physical properties of cirrus clouds are derived from the observations of tropical cirrus clouds by McFarquhar et al. (1996, see Table 1).

Table 1: The cirrus cloud properties in the tropics (McFarquhar et al., 1996).

|  |               |
|--|---------------|
| Ice water content ( $\text{gm}^{-3}$ ) | 0.00102~1.022 |
| effective radius ( $\mu\text{m}$ )     | 16.2~105.5    |
| optical depth                          | 0.16~27.07    |
| single scattering albedo               | ~1.0          |
| asymmetric factor                      | ~0.8          |
| cloud base height (km)                 | 11-12         |

### 2. Input data

In this study, the temperature, moisture and ozone density profiles of the tropical atmosphere (Anderson, 1987) are adopted. Figure 1 shows the temperature and ozone vertical distribution with a total column of 283DU. A surface albedo 0.1 is used, corresponding to an average albedo of the tropics, and solar zenith angle is set for  $0^\circ$ .

Fig. 1 The vertical profiles of temperature (solid) and ozone (dash) in the tropical atmosphere (Anderson, 1987). The upper x-axis is for temperature in K, and the lower x-axis is for ozone in molecules/ $\text{cm}^3$ .

### 3. Multiple cloud layer case

In reality not often there appears single layer cirrus cloud. Cirrus clouds usually overlap lower clouds half of the time over the land while such overlapping always occurs over the ocean. Therefore, we also have conducted some experiments with two layers clouds overlapping each other. The optical properties and heights of two cloud layers are given in Table 2.

Table 2. The optical properties and heights of two cloud layers.

|                                   | Water cloud | Cirrus |
|-----------------------------------|-------------|--------|
| lwc/iwc ( $\text{g/m}^3$ )        | 0.1         | 0.0409 |
| effective radius( $\mu\text{m}$ ) | 10          | 43.5   |
| optical depth                     | 15          | 2.4    |
| cloud height (km)                 | 4~5         | 11~12  |

## **Conclusions**

### 1. Single cloud layer

- a)  $J(O^1D)$  and  $J(NO_2)$  increase above and below cirrus cloud ( $\tau < 8$ ) vs. the values under clear sky condition (Fig. 2). Above-cloud  $J(O^1D)$  is increased more as the cloud layer is optically thicker; below-cloud  $J(O^1D)$  increases by nearly a factor of 1.2 at optical thickness around 2.5, which is the typical optical thickness for cirrus clouds, and then decreases as  $\tau$  increases.
- b)  $J(O^1D)$  and  $J(NO_2)$  decrease as solar zenith angle increases (Fig. 3) under both clear and cloudy sky conditions, above and below clouds. When solar zenith angle is greater than  $40^\circ$ ,  $J$  values below cloud are reduced no matter how thin the cloud layer is.
- c)  $J$  values near the surface are sensitive to surface albedo (Fig. 4). Higher surface albedo induce larger  $J$  values near the surface. With the presence of clouds, the multiple reflectivity between cloud base and surface can increase below-cloud  $J$  values, especially  $J(NO_2)$ .

### 2. Multiple cloud layers

- a) The presence of water cloud layer enhances the increase of the  $J$  values above and below the cirrus cloud layer. As solar zenith angle increases, the  $J$  values are attenuated significantly between and below two cloud layers (Fig. 5).
- b) The change in surface albedo only affects the  $J$  values near the surface, not those between and above the two cloud layers (Fig. 6).

## **II. The Impact on Radiative Forcing from Aircraft Emission**

- Increases in  $NO_x$  emission due to subsonic aircraft activities can affect the hydroxyl and hydrogen peroxy radical distributions with subsequent effects on the production of  $O_3$  and  $CH_4$  lifetime. Here, we present a model study of the potential impact on radiative forcing from aircraft emission in the next few decades with focus on three aspects: the seasonal and geographical characteristics of the impact; the relative contribution of the impact to the total radiative forcing, in particular the  $CO_2$  forcing; and the relative effect of  $O_3$  changes versus  $CH_4$  changes.
- The University of Oslo's 3-D CTM was used to study the impact of aircraft emission at 1992 and future (2015 and 2050) on the distribution of gases like  $NO$ ,  $NO_2$ ,  $OH$ ,  $O_3$  and  $CH_4$ . Changes in the background distribution due to changes in surface emission of pollutants like  $NO_x$ ,  $CO$ ,  $CH_4$  and NMHC (based on IPCC IS92a) are also included in the calculations. Ozone perturbations from aircraft emission at the three time frames show a similar pattern: enhancement at middle and high northern latitudes with maximum values in the upper troposphere (around 12 km) at  $60^\circ N$ , and a substantial part of the increase occurring in the lower stratosphere. The maximum  $O_3$  increases are 6, 11, and 17 ppbv for the years 1992, 2015, and 2050, respectively. In contrast to the inhomogeneous  $O_3$  increases, changes in  $CH_4$  is uniform globally and is determined by changes in its global average lifetime, which is calculated to be -1.82, -3.64, and -5.46% for the respective 1992, 2015, and 2050 values.
- These calculated changes in  $O_3$  and  $CH_4$  were then used in NCAR-CCM3 to calculate changes in radiative forcing. In addition to the greenhouse gases (Table 3), the climate parameters used in the radiative forcing calculations are taken to be the 1979-92 mean

distributions of temperature, moisture, clouds, and surface albedo simulated in the AMIP runs in which the observed monthly SSTs were used. Although the change in global and annual mean radiative forcing due to the combined changes in O<sub>3</sub> and CH<sub>4</sub> is calculated to be small (0.019 Wm<sup>-2</sup> for 1992, 0.043 Wm<sup>-2</sup> for 2015, and 0.058 Wm<sup>-2</sup> for 2050), the seasonal and geographical changes are much larger.

Table 3. Trace gas concentrations used in the calculations. IPCC IS92a (Houghton et al., 1995) was used for calculating the concentrations of the trace gases.

| <b>Gases</b>                  | <b>1992</b>          | <b>2015</b>          | <b>2050</b>          |
|-------------------------------|----------------------|----------------------|----------------------|
| CH <sub>4</sub> (ppmv):       |                      |                      |                      |
| A. Fixed CH <sub>4</sub> sink | 1.714                | 2.029                | 2.412                |
| B. IPCC                       | 1.714 (0%; vs A)     | 2.052 (1.15%; vs A)  | 2.793 (15.8%; vs A)  |
| C. IPCC+Aircraft              | 1.683 (-1.82%; vs B) | 1.977 (-3.64%; vs B) | 2.641 (-5.46%; vs B) |
| CO <sub>2</sub> (ppmv)        | 356                  | 405                  | 509                  |
| N <sub>2</sub> O (ppbv)       | 311                  | 333                  | 371                  |
| CFC-11 (pptv)                 | 268                  | 220                  | 120                  |
| CFC-12 (pptv)                 | 503                  | 470                  | 350                  |

Fig. 1 The vertical profiles of temperature (solid) and ozone (dash) in the tropical atmosphere (Anderson, 1987). The upper x-axis is for temperature in K, and the lower x-axis is for ozone in molecules/cm<sup>3</sup>.

Figure 2.  $J(^1D)$  at 7 heights vs. different optical thickness of the cloud layer. The single cirrus cloud layer is located between 11-12km.

Figure 3. In the upper panel  $J(O^1D)$  with six cirrus cloud optical depths vs. clear sky at solar zenith angles: a) 0°, b) 20°, c) 40°, and d) 60° in a standard tropical atmosphere. The surface albedo is 0.1. In the lower panel the same but for  $J(NO_2)$ .

Figure 4. In the upper panel  $J(O^1D)$  with six cirrus cloud optical depths vs. clear sky at surface albedos: a) 0.0, b) 0.1, and c) 0.2. In the lower panel the same but for  $J(NO_2)$ .

Figure 5. In the upper panel  $J(O^1D)$  under four conditions: 1). one cirrus cloud layer at 11-12km, 2). one water cloud layer at 4-5km, 3). those two cloud layers overlapping, and 4). clear sky at four solar zenith angles: a) 0°, b) 20°, c) 40°, and d) 60°. In the lower panel the same but for  $J(NO_2)$ .

Figure 6. In the upper panel  $J(O^1D)$  under four conditions: one cirrus cloud layer at 11-12km, one water cloud layer at 4-5km, those two cloud layers overlapping, and clear sky, at 3 surface albedos: a) 0.0, b) 0.1, and c) 0.2. In the lower panel the same but for  $J(NO_2)$ .











