JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 92, NO. D1, PAGES 903-914, JANUARY 20, 1987

Response of Middle Atmosphere to Short-Term Solar Ultraviolet Variations: 2. Theory

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Ozone and temperature responses to solar variability, based on satellite data, have been reported in a companion paper (Keating et al., this issue). The present paper is intended to present a theoretical interpretation of this analysis with the purpose of better understanding the chemical behavior of the stratosphere and the coupling between temperature and ozone concentration, when a periodic forcing is applied to the solar ultraviolet (UV) flux. The response of the temperature and of the trace species concentrations, including ozone, to short-term variations in the solar UV irradiance is calculated by a one-dimensional chemical-radiative time-dependent model. The applied solar variability is assumed to be sinusoidal with a period of 27 days (in accordance with the rotation period of the sun) or 13.5 days (when two active regions are on opposite sides of the sun). The amplitude varies with wavelength, which is consistent with observations made by the Nimbus 7 solar backscattered ultraviolet (SBUV) experiment. The maximum ozone sensitivity in the stratosphere appears to be located near 3 mbar. The calculated amplitude and phase of the ozone response are significantly modified when the feedback between ozone and temperature is taken into account. The ozone/temperature coupling tends to modify the ozone phase lag such that, in the upper stratosphere and in the mesosphere, the ozone peak occurs a few days before the UV peak. Comparison of the model results with the observed ozone and temperature responses, based on satellite data, shows that the theory is consistent in many respects with observations. The calculated time lag of the temperature response, however, is approximately a factor of 2–4 smaller than the time lags derived from the measurements, suggesting evidence for some additional process not included in the model calculation. Large negative ozone sensitivities and positive temperature responses are predicted in the mesosphere as a result of the absorption by O_2 of the solar irradiance at the Lyman α wavelength. The model also shows that the expected variation in the stratospheric nitric acid mixing ratio is a factor 2 larger than the corresponding opposite variation in the ozone concentration.

1. INTRODUCTION

The possibility of a link between ozone and solar variability was raised early in this century by *Humphreys* [1910] and has been extensively discussed in the past (see, for example, *Willett* [1962]; *London and Oltmans* [1973]; *Angell and Korshover* [1973, 1976]; *Keating et al.* [1981]; *Keating* [1981]). The understanding of such a relationship is indeed a prerequisite for detecting possible changes in the atmospheric composition due to anthropogenic effects. After the first theoretical investigations by *Crutzen* [1973, 1974], the response of ozone to changes in the solar emission has been estimated by means of one- and two-dimensional photochemical models. In some of these models the spectral distribution of the ultraviolet variability was chosen to be representative of the 11-year cycle [*Callis and Nealy*, 1978; *Penner and Chang*, 1978, 1979; *Brasseur and Simon*, 1981; *Natarajan et al.*, 1981; *Garcia et al.*, 1984]. In other studies the effect of the 27-day rotation period of the sun was considered [*Frederick*, 1977; *Theobald et al.*, 1977; *Keating et al.*, 1985]. The variability in the solar output adopted by the earliest of these models was larger than the values which are now generally accepted, especially above 200 nm. In fact, the changes in the solar irradiance over the entire 11-year cycle are difficult to assess, since they have to be derived either from a number of individual measurements made by several instruments with different calibrations or from some continuous satellite observations which might be biased by instrument drift.

Observations of the solar variability over several rotation periods of the sun are by far more accurate and are now becoming available. These have been used, together with satellite observations of ozone and in some cases of temperature to detect a possible solar signal in the middle atmosphere. *Gille et al.* [1984], for example, using the ozone data obtained through the LIMS experiment and the solar irradiance provided by the solar backscattered ultraviolet (SBUV) instrument, both on board the Nimbus 7 satellite, estimated that the ratio between the relative ozone variation and the relative UV change at 205 nm (hereafter called the ozone "sensitivity") varies with increasing altitude from 0.17 at 31 km to 0.38 at 54 km. *Keating et al.* [1985], using the entire LIMS data set and correcting for the effect of temperature variations, obtained very high correlations between ozone and solar variations. Keating et al. found a sensitivity of the order of 0.3 between 5 and 0.5 mbar, in good agreement with a theoretical analysis. *Heath and Schlesinger* [1985], analyzing the Nimbus 7 SBUV data from May 14 to August 30, 1980, showed that the maximum correlation of the solar-induced ozone variation was located between 1.5 and 3 mbar, depending on the latitude. *Hood* [1984, 1986] used the ozone measurements provided by the Nimbus 4 backscattered ultraviolet (BUV) and Nimbus 7 SBUV experiments, respectively, to estimate the ozone sensitivity to UV variations. In his most recent study, *Hood* [1986] derives ozone sensitivities of about 0.14 at 10 mbar, 0.37 at 5 mbar, 0.48 at 2 mbar, and 0.37 at 1 mbar. The phase lag of the local O_3 mixing ratio relative to the 205-nm solar irradiance decreases with altitude and even becomes negative above the level of 3 mbar. The fact that the ozone mixing ratio peaks before the UV flux is interpreted by *Eckman* [1986*a*] and *Hood* [1986] as a consequence of temperature effects. *Chandra*

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Paper number 6D0518.
0148-0227/87/006D-0518$05.00

[1985], using similar ozone data to those of *Hood* [1984], indicates that a great deal of the ozone and temperature oscillations observed in the tropics are manifestations of high-latitude winter planetary waves, which are apparently not related to solar variability. Ozone measurements made by the ultraviolet spectrometer on board the Solar Mesosphere Explorer (SME) were used by *Eckman* [1986*b*] to study the sun-ozone relationship above the level of 1 mbar. From this study, a 1.3% variation is derived in the tropical ozone concentration near the stratosphere for a 2.5% change in the 205-nm solar irradiance. The fundamental importance of the coupling between chemistry and radiation to explain the observed amplitude and phase of the ozone response is emphasized in a related study by *Eckman* [1986*a*], who shows from theoretical considerations that the magnitude of the ozone variation is 25% smaller when the temperature feedback is included in this calculation than when the temperature is not allowed to vary. Also, in Eckman's study the ozone peak near the stratopause occurs about 1.5 day before the UV peak, as a consequence of the temperature/ozone coupling. However, the data analysis shows, during certain periods, a negative phase lag for ozone as high as 6–8 days, inconsistent with theory. The calculated temperature change over the solar period is found by *Eckman* [1986*a*] to be 0.6 K in the upper stratosphere, with a time lag of about 3 days. This lag is significantly shorter than the phase observed by *Keating et al.* [1986] and *Hood* [1986].

In all data analyses the ozone sensitivity is of the order of 0.1 at 10 mbar (~32 km) and increase with altitude to reach a value between 0.25 and 0.60 at 2 mbar (~44 km). A large part of the differences in the derived ozone sensitivities can probably be attributed to the analysis technique or to the statistical method applied to the data. For example, *Keating et al.* [1985] correct for ozone variations due to temperature fluctuations, while in most other treatments of the observations [*Gille et al.*, 1984; *Chandra*, 1985; *Hood*, 1986, etc.], this correction is not applied.

The purpose of this work is to identify, through a theoretical approach, the key processes that control the ozone and temperature response to solar variability. This study will be based on a one-dimensional chemical-radiative time-dependent model of the middle atmosphere, in which a periodic solar irradiance is applied as an external forcing. The resulting response (amplitude and phase) of the temperature and of the concentration of ozone and other trace species is determined and discussed. The results are intended to help in interpreting the recent data analyses by allowing a better understanding of the involved physical and photochemical processes in the middle atmosphere. A companion paper [*Keating et al.*, this issue] concentrates on the observed response of ozone and temperature in the stratosphere and mesosphere, based on the most recent satellite data.

2. Brief Description of the Model

The model which is considered in the present study has been described by *Brasseur et al.* [1982, 1985] and used, for example, in the work by *Keating et al.* [1985]. It extends from the earth's surface to the altitude of 100 km. The vertical distribution of about 40 species, belonging to the oxygen, hydrogen, carbon, nitrogen and chlorine families, is calculated, together with the vertical temperature profile.

The chemical code derives the production and destruction rate for each species, using the chemical and photochemical parameters (temperature-dependent rate constants, absorption cross sections) compiled by *DeMore et al.* [1985] and the spectral distribution of the solar irradiance provided by *Brasseur and Simon* [1981]. Diurnal average conditions are assumed for the solar illumination, corresponding to 30° latitude and to equinox. The solar penetration in the region of the Schumann-Runge bands of molecular oxygen and the O_2 photodissociation coefficient in this spectral interval are computed using the parameterization of *Kockarts* [1976]. The absorption cross sections of O_2 in the Herzberg continuum, on which the stratospheric ozone production rate is strongly dependent, are similar to the values derived from in situ observations by *Herman and Mentall* [1982]. The water vapor mixing ratio is determined above the tropopause, such that the total number of hydrogen atoms appearing in H_2O, CH_4, and H_2 is conserved. The short-lived species are assumed to be in immediate photochemical equilibrium, while the long-lived trace gases (including the families) and the potential temperature are transported in the vertical by an "eddy diffusion" type exchange.

The radiative code provides the net heating rate resulting (1) from the absorption of solar radiation by O_3 and O_2, (based on the formulation of *Schoeberl and Strobel* [1978]), and (2) from the emission and absorption of terrestrial radiation by CO_2, O_3, and H_2O. The code for long-wave radiation (J. J. Mocrette, private communication, 1983) solves the one-dimensional radiative transfer equation in which the transmission functions are parameterized for four broad wavelength intervals, namely the 15-μm CO_2 band, the 9.6-μm O_3 band, the rotational and 6.3-μm H_2O bands, and the atmospheric window (8–12 μm). This parameterization is based on a more detailed representation of the transmission over 116 spectral intervals, in which a random model [*Goody*, 1952] is used for H_2O and CO_2, and a Malkmus model [*Malkmus*, 1967] is adopted for CO_2 and O_3. Overlapping of several bands is taken into account, while temperature and pressure effects on the spectral parameters are treated according to the methods suggested by *Curtis* [1952], *Godson* [1954], and *Rodgers and Walshaw* [1966]. Calculations are performed for a global cloud cover of 50% with a top altitude of 5 km and a fixed relative humidity in the troposphere, following *Manabe and Wetherald* [1964].

The vertical transport of heat is parameterized using a first-order closure scheme with an eddy thermal diffusion coefficient [*Liou and Ou*, 1983]. This eddy thermal diffusion is largest in the troposphere where convective instability occurs and is several orders of magnitude smaller in the stratosphere where, except near the tropopause, the globally averaged temperature profile essentially results from radiative equilibrium conditions. The quantitative value of the eddy thermal diffusion coefficient is similar to the value used for the vertical transport of trace species.

In order to predict the ozone and temperature response to changes in the solar UV radiation, a sinusoidal variation in the solar irradiance is added to the average value as given by *Brasseur and Simon* [1981]. The period of this forcing has been chosen to be the rotation period of the sun (27 days) or half of this period (13.5 days). Indeed, the observed short-term UV variability exhibits these two types of behavior, depending on the number and distribution of active regions on the Sun. The amplitude of the UV variation is a function of wavelength and changes from cycle to cycle. However, in order to deal with mean situations, we have adopted for the 27-day cycle the spectral variability given in Table 1 [from *Keating et al.*,

TABLE 1. The 27-Day Spectral Variability of the Solar Irradiance Adopted in the Model

Wavelength, nm	I/I_{AV}, %	$\frac{I/I_{AV}}{I_{205}/I_{205,AV}}$
Lyman α(121.6)	10.00	3.90
170.0–188.7	3.34	1.30
188.7–198.0	2.80	1.09
198.0–208.3	2.57	1.00
208.3–250.0	1.09	0.42
250.0–270.3	0.58	0.23
270.3–317.5	0.22	0.09
>317.5	0.00	0.00

1985], which is in accord with the values measured by the Nimbus 7 SBUV instrument, averaged over 15 solar oscillations [*Heath et al.*, 1983]. The amplitude of the variation in solar irradiance at 205 nm is taken equal to 2.57% of the average irradiance. Because of smaller average variability during 13.5-day cycles, the solar irradiance variability, as it appears in Table 1, is reduced at all wavelengths by 10.5% for model runs simulating 13.5-day cycles, so that the variability at 205 nm becomes 2.30% instead of 2.57. The averaged variability in the irradiance responsible for solar heating by ozone absorption has been taken equal to 0.8% in the Hartley band and 0.15% in the Huggins bands. The time step for the numerical integration is chosen as 6 hours (but no diurnal variation of the solar irradiance is allowed). In general, the ozone and temperature response will be expressed in terms of sensitivity, referring to a 1% change in the solar irradiance at 205 nm [*Gille et al.*, 1984; *Keating et al.*, 1985].

3. Model Results and Discussion

3.1. *Simplified Model*

The calculated response of the ozone and of the temperature to an applied sinusoidal variation in the solar irradiance has an amplitude and phase varying with height. The relation between ozone and UV changes can be inferred, as a first approximation, from the continuity equation of ozone

$$\frac{dO_3}{dt} + \alpha O_3 + \beta {O_3}^2 = 2J_2O_2$$

in which it is assumed that the loss can be expressed by a linear ($L_1 = \alpha O_3$) and a quadratic ($L_2 = \beta {O_3}^2$) destruction term. Applying to the O_2 photodissociation coefficient J_2 a periodic variation with frequency ω_0, assuming that α, β, the concentration of molecular oxygen O_2, and the temperature T remain constant during the entire oscillation, and neglecting second-order effects, the relative amplitude $\Delta O_3/O_3$ and the phase Φ_{oz} of the O_3 variation can be expressed as

$$\frac{\Delta O_3}{O_3} = \frac{\Delta J_2}{J_2} \frac{X}{[1 + \omega_0 \tau_c]^{1/2}} \quad (1)$$

and

$$\Phi_{oz} = \tan^{-1} [\omega_0 \tau_c] \quad (2)$$

where $\tau_c = [\alpha + \beta \bar{O}_3]^{-1}$ is the relaxation time of ozone, $\bar{O}_3$ the average ozone concentration, and $X = (L_1 + L_2)/(L_1 + 2L_2)$ a parameter equal to 1, if the destruction rate is purely linear, and to 0.5, if it is entirely quadratic. An intermediate value is used in this work, based on an inspection of the ozone loss terms, as calculated by our one-dimensional model (0.6 at 0.5 and at 1 mbar, 0.8 at 3 mbar, and 0.9 at 5 and 10 mbar).

Since a large part of the radiation contributing to the dissociation of O_2 in the stratosphere and mesosphere is confined in a spectral region around 200 nm, it is expected that the 205-nm solar irradiance is a good index to monitor the ozone production rate. The ozone sensitivity to a change in the 205-nm solar irradiance,

$$S = \frac{\Delta O_3}{O_3} \Big/ \frac{\Delta I_{205}}{I_{205}}$$

can be expressed by the product of two factors: (1) the variation of the ozone concentration to a change in the O_2 photodissociation frequency J_2; and (2) the variation of the J_2 coefficient to a change in the solar flux (expressed as a change in the 205-nm irradiance). Factor 2 is a function of the chosen spectral dependence of the solar variability. As shown, for example, by column 4 in Table 2, the relative variation in the J_2 coefficient (and thus in the ozone formation rate) for a 1% change in the 205-nm solar flux is found to be 0.6 at 1 mbar, when adopting the spectral variability given in Table 1 and accounting for the effect of the change in O_3 column abundance resulting from UV radiation. This factor decreases with decreasing altitude (e.g., 0.24 at 10 mbar), since the UV radiation at longer wavelengths, which has smaller variability, penetrates deeper in the atmosphere than the radiation at shorter wavelengths, which has higher variability.

The ozone sensitivity to a change in the J_2 coefficient is, according to expression (1), equal to X when the ozone lifetime is short. The effect of the lifetime becomes apparent below the level of 3 mbar. In other words, the ozone sensitivity in the lower stratosphere becomes dependent on the period of the solar UV variation. Therefore calculated sensitivities for a 13.5-day period may not be extrapolated for 27-day or 11-year periods without introducing a correction for the "lifetime" effect. As will be shown below, the ozone/temperature coupling, which is not considered in (1) and (2), introduces a more complex relation between the ozone sensitivity and the period of the solar forcing.

Table 2 also compares the ozone response predicted by (1) and (2) (columns 5 and 6) to the sensitivity (column 7) and phase lag (column 8) obtained from a full one-dimensional calculation in which the temperature feedback is omitted, but the concentration of all atmospheric species is allowed to vary as a response to the change in solar UV. The general trends in the sensitivity and phase are similar (except in the upper stratosphere and mesosphere, where the odd hydrogen chemistry, whose effect is not included in expressions (1) and (2), plays a major role and tends to reduce the ozone amplitude).

The sensitivity and phase of ozone are not only sensitive to the chemical lifetime of O_3, but also to the radiative lifetime of the atmosphere, which affects the temperature response. The values of these quantities derived in the model are shown in Table 3. Also given in the Table 3 is the corrected radiative lifetime obtained when considering the adjustment of the ozone concentration to changes in the temperature (photochemical acceleration). The residence time for vertical transport (H^2/K, where H ($\sim$7 km) is the atmospheric scale height and K is the eddy diffusion coefficient used in the model) is given for comparison purposes.

3.2. *Temperature Response in Stratosphere and Lower Mesosphere*

The temperature sensitivity to changes in the solar flux increases with altitude, but the calculated amplitudes are significantly smaller than the current variability associated with all

TABLE 2. Sensitivity of the Photodissociation Rate of O_2 and of the O_3 Concentration to the Variation in the 205-nm Solar Irradiance: Time Lag of the Ozone Response

Level, mbar	Φ_{oz}, days	$\frac{\Delta O_3/O_3}{\Delta J_2/J_2}$ [a]	$\frac{\Delta J_2/J_2}{\Delta I_{205}/I_{205}}$ [b]	S[c]	Φ_{oz}, [d] days	S[e],	Φ_{oz}, [f] days (±0.25)
0.5	0.03	0.60	1.26	0.70	0.03	0.26 (0.36)[g]	0.125
1	0.1	0.60	0.60	0.36	0.1	0.31	0.250
3	0.5	0.78	0.54	0.42	0.5	0.41	0.750
5	1	0.86	0.37	0.31	1.0	0.36	1.625
10	15	0.66	0.24	0.16	5.5	0.16	4.125

[a]Calculated from expression (1) with $\omega_0 = 2\pi/27$ day^{-1}.
[b]Taken from the one-dimensional model calculation for a 24-hour averaged solar insolation (30° latitude, equinox).
[c]Ozone sensitivity derived from the two previous columns.
[d]Calculated from expression (2) with $\omega_0 = 2\pi/27$ day^{-1}.
[e]Sensitivity derived from the full one-dimensional model, neglecting temperature feedback.
[f]Phase lag derived from the full one-dimensional model, neglecting temperature feedback.
[g]Same as footnote *e*, but neglecting the effect of water vapor and odd hydrogen variability.

kinds of dynamical features in the atmosphere (see Figure 1 in the works by *Keating et al.* [1985] or *Chandra* [1985]). The calculated temperature variation in percent for a 1% change in the solar irradiance at 205 nm and the corresponding time lag versus the UV forcing are shown in Figures 1*a* and 1*b*. The model predicts, at 1 mbar, a corresponding temperature increase of 0.13 K, when the solar irradiance at 205 nm rises by 1% from its minimum to its maximum value. This value is in good agreement with the data analysis of *Hood* [1986] (0.17 K/percent) and of *Keating* [this issue] (0.14 K/percent) which was based on 22 months and 4 years of SAMS data, respectively. The calculated phase lag at 1 mbar is 1.5 days. This number is consistent with the radiation relaxation time predicted by the model in the upper stratosphere (3.6 days for the effect of infrared radiation on temperature variations (See Table 3)), provided the fact that the atmosphere response is slightly accelerated by photochemical relaxation, as shown, for example, by *Hartmann* [1981] and *Ghazi et al.* [1985]. The global radiative response time (infrared and solar contributions) provided by the model is indeed 4.8 days at 1 mbar. The calculated phase lag (1.5 days) is somewhat smaller than the value obtained by *Eckman* [1986*a*] but larger than the value derived by *Wuebbles* (private communication, 1986).

In all cases, the calculated phase lag is considerably smaller than the value provided by the observations (about 6 days in the studies of Keating et al. and of Hood). *Hood* [1986] concludes that an additional dynamically induced temperature component is required to explain the large temperature time lags. The dynamical response of the middle atmosphere to solar activity has been studied by *Geller and Alpert* (1980), *Bates* [1977, 1981], and more recently, by *Callis et al.* [1985]. *Ebel et al.* [1981] have found significant spectral coherences between the 10.7-cm solar flux and various harmonic components of planetary waves in the middle and lower stratosphere. *Schmidt* [1985] reports from a data analysis that the zonal and meridional circulations in mid-latitudes of the northern hemisphere are largely influenced by solar activity if, in addition to the sunspot number, the solar facula areas are also taken into account.

More recently, *Ebel et al.* [1986] have found significant coherence between the 27- and 13.5-day solar flux variation at 10.7-cm and atmospheric oscillations. They indicate that a large fraction of the temperature disturbance in response to solar variability is of dynamical nature, which represents an indirect effect of the short-term solar variation, in contrast with the direct response associated with the diabatic and chemical effects. Their study also suggests that planetary waves are an essential part of the components of this indirect temperature modulation. *Dameris et al.* [1986], using a three-dimensional mechanistic model similar to that of *Rose* [1983], show that a weak temperature disturbance applied near the stratopause (and assumed to result from a solar-induced ozone change) may propagate to lower altitudes under specific dynamical conditions (reduced inertial stability at low latitudes). Dameris et al. also show that the period of the oscillation found in the lower statosphere may be different from the solar period, as a result of the nonlinear behavior of the atmosphere. Dynamical processes may thus play an important role and introduce complex nonlinear feedback mechanisms. The nature of such dynamical coupling between direct and indirect effects of solar UV modulation is not yet understood and requires further investigation. Dynamical oscillations coherent with solar variability could be partly related to wave activity [*Chandra* [1985, 1986]. Other possible causes of the discrepancy between the temperature response derived from satellite data and the one predicted by one-dimensional chemical/radiative models have to be explored.

TABLE 3. Characteristic Times for Ozone, Temperature, and Vertical Transport

Level, mbar	Approximate Height, km	Ozone Chemical Lifetime, days	Radiative Lifetime, days	Residence Time for Vertical Transport, days
0.5	53	0.10	6.7[a], 5.0[b]	25
1	48	0.17	4.3[a], 3.6[b]	45
2	44	0.31	6.3[a], 5.3[b]	74
3	41	0.60	8.3[a], 8.0[b]	106
5	37	2.0	11.1[a], 11.0[b]	176
10	32	14.	16.7[a], 16.7[b]	326
20	27	91	25.0[a], 25.0[b]	629

[a]Due to infrared cooling only.
[b]Includes effect of photochemical acceleration.

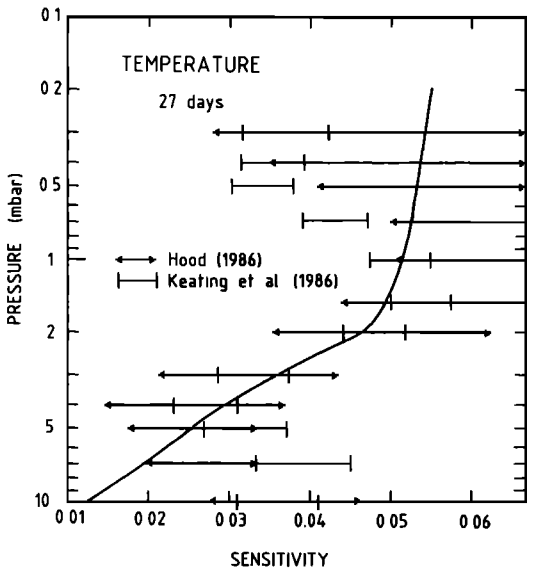

Fig. 1*a*. Amplitude of the temperature variation (expressed in percent of the background temperature) calculated in response to a variation in the solar irradiance of 1% at 205 nm, with a period of 27 days. The values provided by the model are compared to the observational data from *Keating et al.* [this issue] (labeled Keating et al. (1986)) and *Hood* [1986].

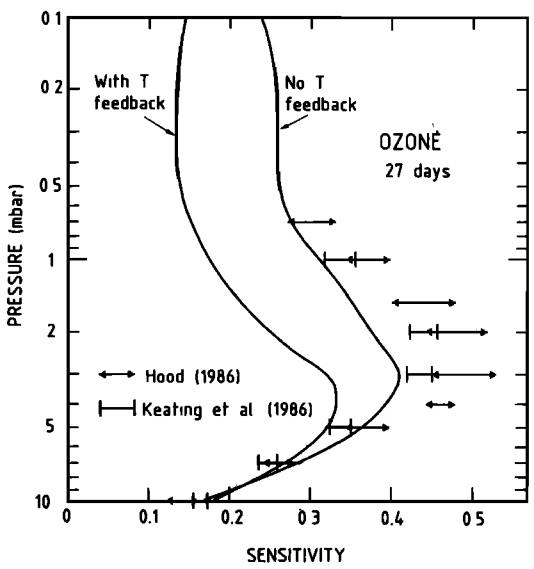

Fig. 2*a*. Amplitude of the ozone variation (expressed in percent of the background concentration) calculated in response to a variation in the solar irradiance of 1% at 205 nm, with a period of 27 days. The ozone sensitivity is calculated with and without temperature feedback and compared to the observational data of *Keating et al.* [this issue] (labeled Keating et al. (1986)) and *Hood* [1986].

3.3. *Ozone Response in the Stratosphere and Lower Mesosphere*

The ozone sensitivity and phase lag, calculated as a function of altitude with the full one-dimensional time-dependent chemical model, are shown in Figures 2*a* and 2*b*. In one of the model cases the temperature is allowed to vary as a result of changes in the heating and cooling rates along the 27-day solar rotation period. In the other case the temperature is kept fixed. Although the temperature variations calculated in the first case have different phases than the observed variations, a comparison between the two cases is nevertheless useful to understand the qualitative effect of temperature feedback on the UV/ozone relationship. It can be seen, for example, (Figure 2*a*) when the temperature phase lag is small (see Figure 1*b*) the ozone sensitivity above 10 mbar is reduced when the temperature coupling is included in the calculation. This is a manifestation of the negative feedback mechanism between ozone and temperature in the chemically controlled region of the stratosphere. For example, the ozone sensitivity calculated without temperature feedback is predicted to be 0.31 at 1 mbar, in good agreement with the value derived from (1), but is is reduced to 0.18 when the temperature feedback is

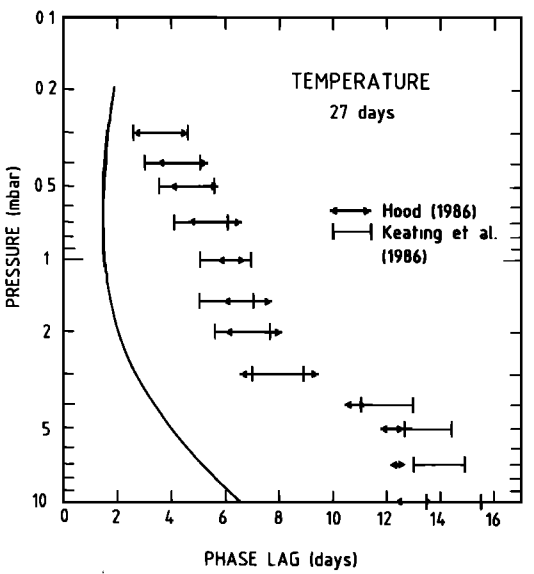

Fig. 1*b*. Same as Figure 1*a*, except for the temperature phase lag (expressed in days).

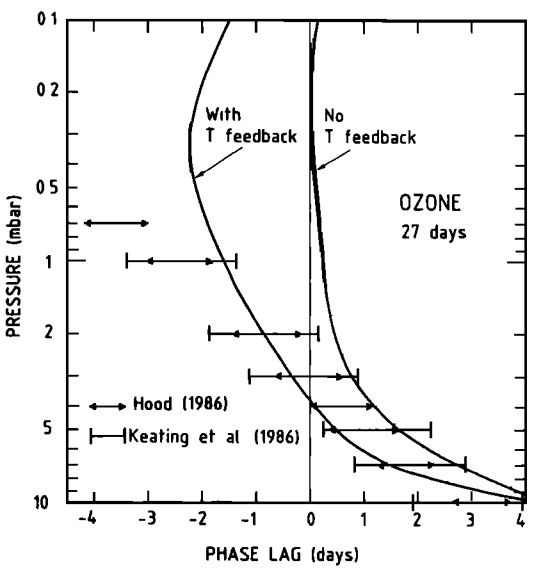

Fig. 2*b*. Same as Figure 2*a*, except for the ozone phase lag (expressed in days).

included. In fact, the temperature/ozone coupling can be described in terms of a θ parameter expressed in degrees Kelvin and defined by

$$\theta = \frac{\partial \ln [O_3]}{\partial T^{-1}}$$

where $[O_3]$ is the ozone concentration (in cubic centimeters), and T is the temperature. A similar parameter can be defined by using the mixing ratio of O_3 instead of the concentration; its value is smaller than θ by approximately the magnitude of the local temperature T. The regions with the highest negative feedback between ozone and temperature are characterized by the highest positive values of θ. Approximate analytical expressions for this parameter can be given [*Barnett et al.*, 1975, *Chandra et al.*, 1978, *Haigh and Pyle*, 1982], introducing possibly a correction for hydrostatic coupling. For an atmosphere with pure oxygen chemistry, in which the hydrostatic adjustment is neglected, the θ parameter is found to be close to 1400 K, indicating a strong negative coupling between temperature and ozone. If the loss of ozone was entirely due to HO_x, ClO_x, or NO_x, the theoretical value of θ would be 510, 260, or 1200 K, respectively [*Haigh and Pyle*, 1982]. In other words, in the mesosphere, where the HO_x chemistry dominates the ozone balance, the ozone/temperature relation is expected to be weak; in the stratosphere, where the NO_x chemistry plays the major role, this link should be stronger, with a maximum around 40–45 km [*Keating et al.*, 1983], where the Chapman reactions contribute the most to the ozone loss. Our model, which considers simultaneously the action of all chemical families and corrects for hydrostatic changes, provides θ values of 980 K at 35 km, 1120 K at 40 km, 1040 K at 45 km, 952 K at 50 km, 660 K at 60 km, and 609 K at 70 km. *Keating et al.* [1985] have reported for θ a value of 1128 K at 2 mbar (44 km), based on the analysis of LIMS data and in good agreement with our theoretical estimates. The strong anticorrelation observed between ozone and temperature does not necessarily imply that the dynamical terms in the ozone continuity equation are unimportant compared to photochemical terms. In fact, as shown by *Rood and Douglass* [1985], the dynamically induced phase between O_3 and temperature can mimic the phase expected from photochemical equilibrium conditions. This emphasizes again the importance of possible dynamical influence in the relationship between ozone, temperature and solar activity.

Because of this negative ozone/temperature feedback, a temperature disturbance which is in phase (or almost in phase) with the periodic variation of the solar UV irradiance tends to reduce the amplitude of the ozone response (as seen in Figure 2*a*). However, a temperature wave which is 180° out of phase with the sun tends to increase the amplitude of the ozone signal. Finally, if the phase of the temperature is intermediate, (say, 90°) the phase of the ozone response will be shifted so that the ozone signal will possibly peak before that of the solar irradiance. An accurate determination of the time lag in the temperature response, particularly in the upper stratosphere where the value of Θ is high, is thus crucial to determine the exact value of the ozone sensitivity and phase. These considerations have to be kept in mind when comparing the calculated ozone response with values derived from observations.

The ozone sensitivity predicted by the model is somewhat smaller than the values inferred from the satellite data (see Figure 2*a* and the companion paper by *Keating et al.* [this issue]. The agreement, however, is improved when the "no-temperature feedback" model case is compared to the data which have been corrected for temperature variability [*Keating et al.*, 1985].

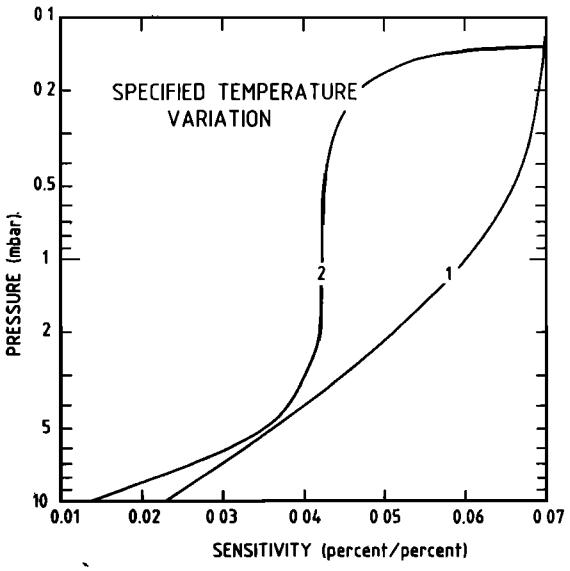

Fig. 3*a*. Amplitude of the temperature variation (expressed in percent of the background temperature for a 1% change in the solar irradiance at 205 nm), as specified in several numerical experiments discussed in the text. Two different vertical distributions (cases 1 and 2) of this temperature variation are considered.

The calculated time lag between ozone and UV variations decreased with increasing altitude, as a result of the variation with height of the ozone lifetime. In the upper stratosphere, however, where photochemical conditions are reached rapidly, the time lag approaches zero when the temperature is kept constant but becomes negative when the temperature feedback is included in the model. These negative time lags have been found in the Nimbus 7 SBUV observational data by *Hood* [1986], *Keating et al.*, [this issue], and *Eckman* [1986*a*], but with considerably larger negative values. A linearized analytical model used by *Hood* [1986] indicates that negative phase

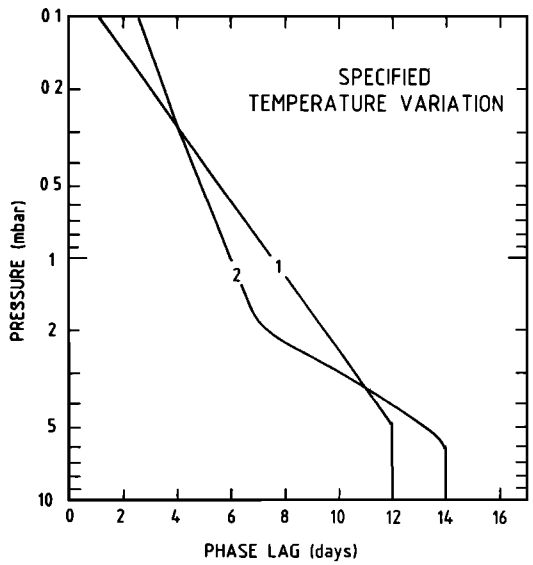

Fig. 3*b*. Same as Figure 3*a*, except for the temperature lag (expressed in days).

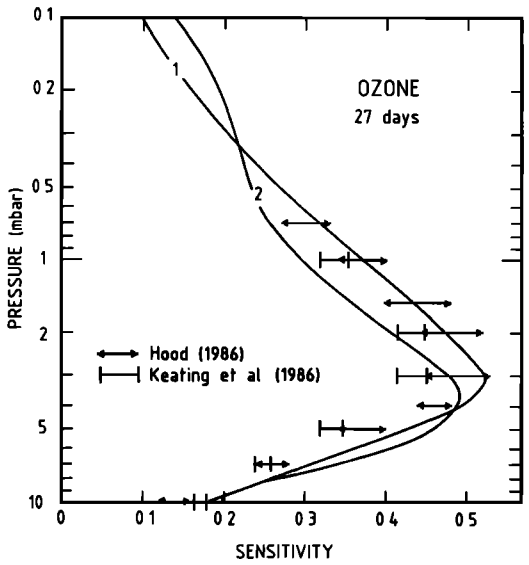

Fig. 4*a*. Amplitude of the ozone variation (expressed in percent of the background concentration) calculated in response to a 27-day variation in the solar irradiance of 1% at 205 nm and a simultaneous temperature change specified as in Figures 3*a* and 3*b* (cases 1 and 2).

lags can be found for small values of the ozone lifetime (upper stratosphere and the mesosphere) and for positive values of the temperature phase lag.

3.4. *Relation Between Temperature and Ozone Response*

Since the predicted negative phase associated with the ozone variation is significantly smaller than the observed phase (e.g., −1.4 days versus −2.4 days at 1 mbar), it is worthwhile to estimate if this discrepancy can be attributed to the incorrect prediction in the temperature phase lag, as discussed above. In order to test this hypothesis a new model run has been performed, in which the temperature response is no longer self-consistently calculated but is specified in close agreement with the data analysis. The amplitude and phase of this specified periodic variation in the temperature is shown in Figures 3*a* and 3*b*. Two different cases (labeled 1 and 2), both fairly consistent with data analyses, have been chosen to provide indications on the sensitivity of the ozone response to the amplitude and phase of the temperature variation. Case 1 is close to the values reported by *Hood* [1986], while case 2 is close to the data provided by *Keating et al.* [this issue]. The ozone response resulting from the simultaneous variation applied to the solar irradiance and to the temperature is displayed in Figures 4*a* and 4*b*. It can be seen that the agreement with observed O_3 sensitivity is significantly improved, indicating that theory and observations are consistent in regard to the temperature/ozone feedback. For example, in case 1 the amplitude of the ozone modulation is increased from 0.34 to 0.52 at 3 mbar and from 0.33 to 0.43 at 5 mbar, bringing the model prediction in closer agreement with the data analysis of *Hood* [1986] and *Keating et al.* [this issue] (see Figure 4*a*). The phases are also modified, particularly at high altitude. At 0.5 mbar, for example, the lag in case 1 becomes −2.75 days instead of −2.0 days, and at 0.2 mbar, −2.7 days instead of −1.75 days. Thus the ozone response, obtained by the model when the temperature amplitudes and lags are specified in accordance with the data analysis, is in better agreement with the satellite observations than if the temperature response is self-consistently calculated.

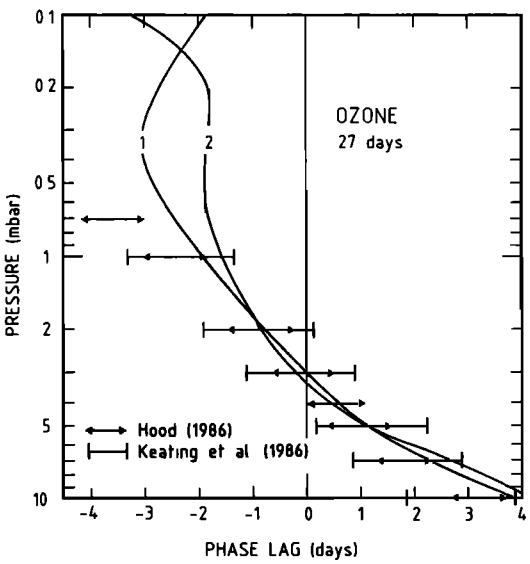

Fig. 4*b*. Same as in Figure 4*a*, except for the ozone phase lag (expressed in days).

3.5. *Sensitivity of the Ozone Response to the Adopted Chemical Scheme*

In order to investigate potential causes other than the temperature feedback effect to explain the discrepancy between the calculated and observed ozone responses (amplitude and phase) and, in particular, to estimate the effect of possible errors or omissions in the chemical scheme, additional model experiments have been performed. Such numerical studies, moreover, might help to explain the discrepancy between calculated and observed ozone concentrations in the upper stratosphere and in the mesosphere. The possible overestimation in the models of the ozone destruction rate, especially by HO_x, NO_x, and ClO_x, is one of the potential causes of this discrepancy [*Froidevaux et al.*, 1985; *Rusch and Eckman*, 1985; *World Meteorological Organization* (*WMO*), 1986]. The response of ozone has therefore been estimated for conditions in which the chemical action on ozone of HO_x, NO_x, or ClO_x is individually removed. The calculated amplitudes and phases of the ozone UV response are only slightly different from the standard case in which the effects of HO_x, NO_x, and ClO_x are simultaneously considered. Thus the numerical experiments do not clearly suggest an explanation for the ozone deficit found in the models near and above the stratopause. Only the high ozone sensitivity in the mesosphere derived from the LIMS data by *Keating et al.* [1985] and by *Gille et al.* [1984] seems to require lower concentrations of the HO_x radicals than generally predicted in the models, but those differences may be related to non-LTE (local thermodynamic equilibrium) effects on mesospheric ozone measurements by emission [*Solomon et al.*, 1986].

Another possible explanation of the discrepancy between theory and observation is the nonlinear behavior of the ozone response to the 205-nm solar variability. Indeed, the model assumes for the 27-day cycle a constant variation of 2.57% in the solar irradiance at 205 nm, while, as shown by the continuous monitoring of the solar emission, the maximum ampli-

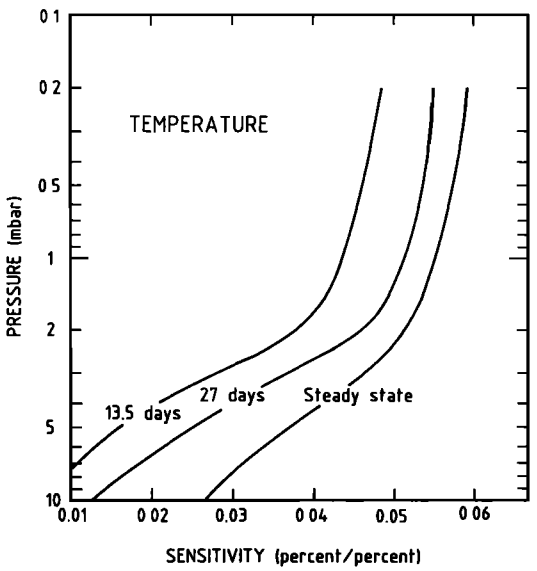

Fig. 5. Temperature sensitivity to a sinusoidal variation in the solar irradiance (1% at 205 nm) with different periods (13.5 days, 27 days, steady state).

tude of the ultraviolet signal varies significantly from one cycle to another (by about a factor of 2). The average strength of the ozone variation resulting from this irregular modulation of the solar flux is not necessarily equal to the amplitude of the ozone change calculated for an average amplitude of the solar variation. The importance of such nonlinear effects has been investigated by doubling at all wavelengths the applied solar forcing given in Table 1 (5.14 instead of 2.57 at 205 nm). The calculated ozone sensitivity (expressed relative to a 1% change at 205 nm) is almost unchanged except below about 20 mbar. At 25 mbar, for example, the departure from a perfect linear behavior is found, however, to be smaller than 5%.

3.6. *Effect of the Solar Period on Ozone Response*

Finally, the effect of a change in the period of the solar forcing is investigated. Indeed, on some occasions, when active regions appear on opposite sides of the sun, the variability in the solar irradiance is characterized by a period of 13.5 days instead of 27 days. Such behavior was observed, for example, in early 1979, when the LIMS instrument was in operation. Figures 5 and 6 show a comparison between the temperature and ozone sensitivities calculated for a 27-day and a 13.5-day period, as well as for steady state conditions. This latter case applies to a period in the solar variability much longer than the ozone lifetime, such as in the case of the 11-year cycle. As one can show from a simple analytical model (see expression (1) and *Hood* [1986]), the ozone and temperature sensitivities are dependent on the solar period if the length of this period is comparable to the lifetime of ozone and the radiative lifetime of the atmosphere, respectively. In the lower stratosphere, where the sensitivity of ozone to photochemical conditions is weak, the $(\omega_0\tau_c)^2$ term in (1) cannot be neglected compared to 1, so the amplitude and phase of the ozone response are obviously increasing with the period of the solar variation. The same type of behavior appears for the temperature response. At 1.5 mbar, for example, where the radiative lifetime τ_R is of the order of 5 days [*Schoeberl and Strobel*, 1978], the term $(\omega_0\tau_R)^2$, which is equal to 1.35 and 5.4 for the 27-and 13.5-day periods, respectively, cannot be neglected compared to 1. (See also expression (25) from *Hood* [1986]). The relative temperature sensitivity is therefore increasing with the period of the solar forcing at all altitudes between 10 and 0.1 mbar. The fact that the ozone sensitivity decreases with the solar period in the upper stratosphere, where $(\omega_0\tau_c)^2 < 1$, is explained by the ozone/temperature feedback mechanism, which at these altitudes is characterized by a clear anticorrelation between the ozone density and the temperature.

In order to perform a more detailed interpretation of the satellite data, an estimation of the ozone response to a 13.5-day variation, applied simultaneously to the solar irradiance and to the temperature, is compared with a similar case where the period of the forcing is 27 days. The specified amplitude and phase describing the temperature variation are the same as case 2 of Figures 3*a* and 3*b*. The ozone response is significantly different for the two cases (Figures 7*a* and 7*b*). Indeed, as the period of the solar and temperature forcing decreases, the maximum of the ozone amplitude is displaced towards higher altitudes (see Figure 7*a*). This behavior is the consequence of two different effects: (1) the ozone sensitivity decreases with the decreasing solar period in the lower and middle stratosphere; and (2) the phase of the specified temperature variation, although the same in both cases when expressed in days, is a factor of 2 different when expressed in radians or degrees. At 2 mbar, for example, the adopted time lag for the temperature is chosen to be 6.5 days, corresponding to 90° and 180° for the 27- and 13.5-day cases, respectively. In other words, when the solar UV irradiance reaches its maximum, the amplitude of the temperature variation at this height equals zero when the period is 27 days, but it reaches a negative maximum when the period is 13.5 days. Obviously, since the feedback between ozone and temperature is clearly negative in the upper stratosphere, the ozone sensitivity at 2 mbar should be larger in the 13.5-day case than in the 27-day case.

A comparison between the ozone lags (Figure 7*b*) obtained for both solar periods suggests that, for a given temperature

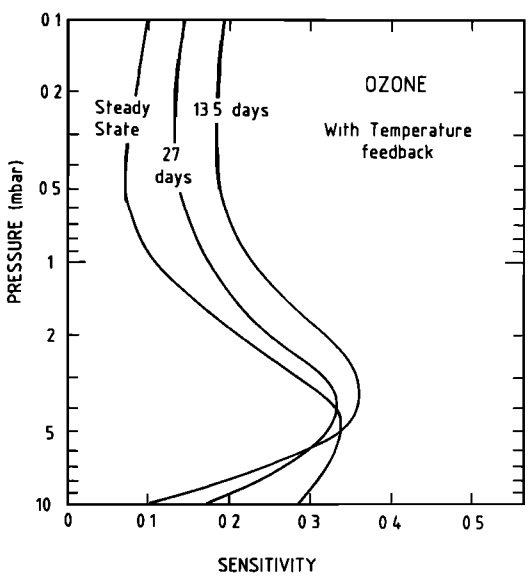

Fig. 6. Same as Figure 5, except for the ozone concentration. These three curves refer to cases in which the temperature variations are self-consistently calculated with the ozone variation.

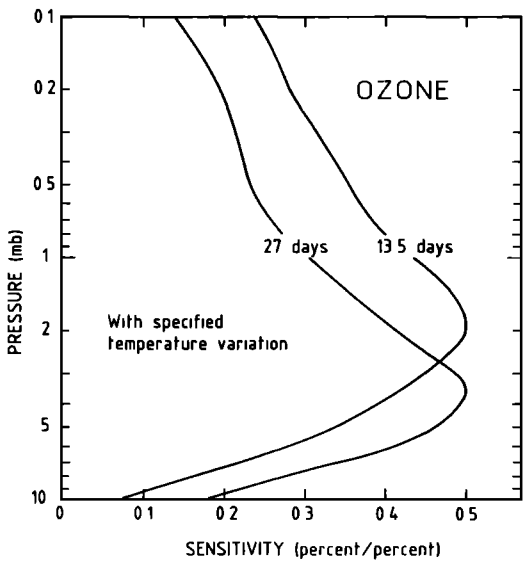

Fig. 7*a*. Effect of a change in the solar period (13.5 versus 27 days) on the amplitude of the ozone variation when the solar irradiance varies by 1% at 205 nm, and the change in the temperature is specified as in Figures 3*a*. and 3*b* (case 2).

forcing, larger time lags should be expected in the ozone signal detected in the upper stratosphere and mesosphere when the period of the solar irradiance is increased from 13.5 days to 27 days. Again, such differences can be understood when considering, for example, the temperature conditions at 2 mbar. As indicated previously, the 90° differences of phase between temperature and solar irradiance, which appears in the 27-day case, produce a negative phase in the ozone response (see above), whereas the 180° difference of phase appearing in the 13.5-day case gives smaller phase shifts in the ozone signal. Such behavior is clearly seen when comparing the time lags derived from the SBUV data over time intervals rich in 27-day signals and over time intervals rich in 13.5-day signals [*Keating et al.*, [this issue]

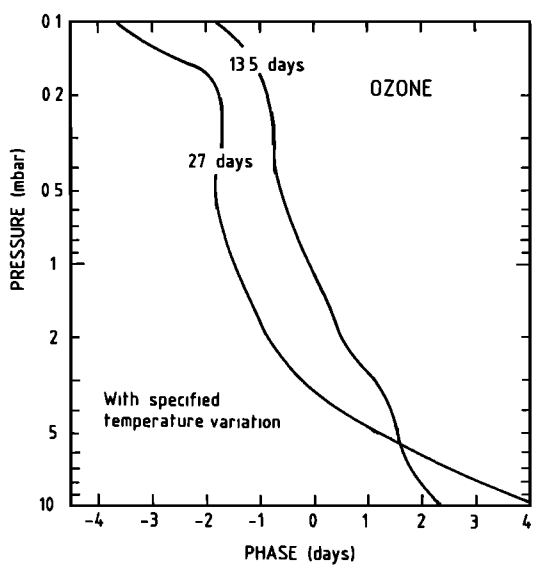

Fig. 7*b*. Same as Figure 7*a*, except for the ozone phase lag.

TABLE 4. Heating Rate Between 65 and 95 km Altitude

Altitude, km	Absorption by Ozone	Absorption by Molecular Oxygen		Total
		Schumann-Runge	Lyman α	
65	3.8	0.3	3×10^{-4}	4.2
70	1.9	0.4	2×10^{-2}	2.3
75	0.7	0.6	0.1	1.4
80	0.3	0.9	0.3	1.5
85	2.3	1.2	0.5	4.0
90	1.8	2.3	0.4	4.5
95	0.7	5.5	0.4	6.6

Heating rate is stated in degrees Kelvin per day.

3.7. *Ozone and Temperature Responses in the Upper Mesosphere*

Since radiative transfer in the upper mesosphere involves complex non-LTE processes, which are not considered in our radiative code, no detailed calculation of the ozone and temperature variations will be reported over 0.1 mbar (62 km). The key processes involved in the atmospheric response to solar variability can, however, be understood from simplified calculations. For example, the model indicates, when ignoring temperature feedback, that the ozone sensitivity changes sign sharply above 68 km. Such abrupt change occurs only if the variability in the solar radiation at Lyman α is included in the calculation. As a matter of fact, the ozone loss in the mesosphere is due mainly to the action of hydroxyl radicals, which are produced principally above ~68 km, essentially by the photodissociation of water vapor by solar radiation at Lyman α. The mixing ratio of mesospheric H_2O decreases with altitude so that the production rate of OH and the related negative ozone sensitivity to solar variability peaks near 75 km. This behavior has already been noted by *Frederick* [1977]. The model predicts a sensitivity of 1.4% per percent of 205-nm radiation for OH and a sensitivity of −0.32% per percent for ozone at 75 km.

The change in the heating rate associated with the absorption of UV radiation by ozone in the Hartley band is essentially proportional to the change in the ozone concentration and is thus expected to be negative (cooling) at 75 km, as the solar irradiance increases. In the upper mesosphere and in the thermosphere, however, a significant part of the heating results from the absorption of solar energy by molecular oxygen. The contribution to the heating rate of the highly variable Lyman α radiation is of the order of 20% of the total heating near 80 km (see Table 4). At this height, for a solar zenith angle of 60°, the energy deposition by Lyman α radiation is maximum. For lower solar zenith angles the altitude of this maximum decreases. Adopting, for example, an ozone sensitivity of −0.60 at 80 km, a variation in the solar irradiance of 1.2% for the spectral region of the Schumann-Runge systems and 4% for Lyman α (assumed to correspond to a 1% change at 205 nm), the resulting change in the total heating rate is estimated to be 2.4×10^{-2} K/day, when the values quoted in Table 4 are used. If a Newtonian cooling coefficient of 0.1/day is adopted for illustrative purposes, this corresponds to a temperature variation of 0.24 K.

In conclusion, a negative sensitivity for ozone, together with a high positive sensitivity for the temperature, are predicted by

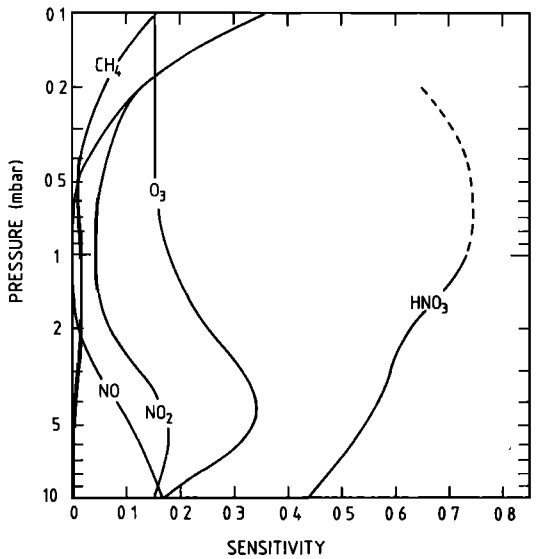

Fig. 8. Amplitude of the O_3, HNO_3, NO_2, NO, and CH_4 response (expressed in percent of the background concentration) to a change in the solar irradiance corresponding to 1% at 205 nm with a period of 27 days. These curves refer to the case where the temperature response is self-consistently calculated together with the changes in chemical composition.

theory in the upper mesosphere and are fairly consistent with the data analysis by *Keating et al.* [this issue]. The peak isolated in the observational study is, however, 5–10 km lower in the atmosphere than predicted by the model. Part of the difference may be attributed to the fact that the data used for the temperature analysis proceed from tropical regions (±20° latitude), whereas the model deals with globally averaged conditions. Moreover, the model in its present development assumes a constant water vapor mixing ratio with height up to the mesopause and no variation with solar irradiance. A self-consistent calculation of the H_2O vapor would reduce the ozone sensitivity near and above the mesopause and therefore sharpen and lower the peak in ozone/UV sensitivity.

3.8. *Response of Other Trace Species*

Finally, the sensitivity of selected species is compared in Figure 8 to the calculated sensitivity of ozone obtained when performing a fully interactive chemical-radiative model run. Clearly, the sensitivity of nitric acid to solar variability should be larger than that of ozone; it should therefore be detectable, at least in the atmospheric regions where the dynamical perturbations are the weakest. A negative correlation with the 205-nm index is expected because a large fraction of the photodissociation of nitric acid in the stratosphere takes place near 200 nm. *Keating et al.* [1986] have recently reported the solar signal in the LIMS HNO_3 data and derived at 10 mbar a sensitivity of about 0.4, in good agreement with model calculations. Further work is required to fully understand the response of other minor species to solar variability.

4. Ozone Response to 11-Year Solar Variability

The ozone response to solar variation associated with the 11-year cycle of the sun can be calculated theoretically using the steady state solution discussed in section 3.6. The amplitude of the solar emission variation over the 11-year cycle is poorly known, so that the model prediction is quite uncertain. If it is assumed that the spectral distribution of the long-term irradiance ratio is similar to the variability adopted for the 27-day period, only the 11-year change in the irradiance at 205 nm must be specified. This latter parameter was determined by a regression coefficient between the (Nimbus 7 SBUV) 205-nm "ratio" and the (Ottawa World Data Center) 10.7-cm "ratio" for the period between November 1978 and September 1981. The long-term variations of the 10.7-cm flux are known, and thus using the regression coefficient, an estimate can be made of the 205-nm variability if it is assumed that the regression coefficient for short-term variations is approximately the same as that for long-term variations. This study of the relationship between the 10.7-cm and 205-nm flux yields an estimated 10% variation over the solar cycle for the 205-nm solar flux. On the other hand, if the variations at 180 nm noted by *Hinteregger* [1981] are assumed, and the relative variations between 180 and 205 nm [*Keating et al.*, 1985] are taken into account, the 205-nm variation over the solar cycle could be as high as 15%. In contrast, recent studies with solar radiances measured by the SME satellite [*Rottman*, 1985] yield variations smaller than 10% and probably as low as 2–6% (G. Rottman, personal communication, 1986). We assume here that the 205-nm radiation variability ranges between 2 and 15% over the solar cycle. The corresponding variations in ozone mixing ratio, total column ozone, and temperature are shown in Table 5. Thus changes in ozone may reach 5% and changes in temperature 2 K in the stratosphere but are more probably 2.8–3.5% and 1.3–1.6 K, respectively, (assuming 8–10% 205-nm variability). It is interesting to note that the estimated long-term change in total column ozone (0.3–2.6%) is in accord with the observed change detected from analysis of Nimbus 4 BUV ozone after correcting for instrument drift [*Keating et al.*, 1981].

5. Conclusions

The relation between short-term periodic variations in the solar ultraviolet radiation and stratospheric ozone, which has been a subject of controversy in the past, seems now to be established through the analysis of satellite data and partly explained by model calculations. The coupling between ozone and temperature seems to play a fundamental role in this problem. Sensitivities predicted by fully coupled chemical-radiative time-dependent models are about 30% smaller than the amplitudes derived from satellite data. The discrepancy, however, seems to be resolved if the observed rather than the predicted phase lag of the temperature is used in the model calculation. *Hood* [1986] suggests that an additional dynamical forcing, fully coherent with the solar variation, is required to understand the phase lags of the temperature/UV relation. Such dynamical coupling is not considered in the present one-

TABLE 5. Estimated 11-Year Variations

Pressure, mbar	Approximate Altitude, km	O_3, %	T, °K
0.5	54	0.16–0.9	0.32–2.4
1.0	48	0.20–1.7	0.32–2.1
2.0	43	0.40–3.2	0.32–2.1
5.0	36	0.7–5.1	0.20–1.3
10.0	31	0.6–4.2	0.16–1.0

Assuming 8 ± 6% variation of 205-nm radiation. Steady state calculation with temperature feedback. Total column variation is 0.32–2.6%.

dimensional model, as it requires more sophisticated dynamical simulations, including the formation and the propagation of a large spectrum of atmospheric waves. However, other potential causes, related to radiative and chemical processes, should not necessarily be ruled out. Further work should consider the latitudinal and seasonal dependence of the ozone and temperature response to solar variablity.

Acknowledgments. Helpful discussions with S. W. Bougher, R. Dickinson, J. C. Gille, V. Ramanathan, R. J. Roble, and S. Solomon are gratefully acknowledged. We want to acknowledge the effort of R. F. Donnelly in organizing the special session at the 1985 Fall American Geophysical Union meeting entitled "Stratospheric, Mesospheric and Thermospheric Effects of Solar Variability." This work was partly supported by the Chemical Manufacturers Association under contract FC-84-534 and by NASA under contract NAS1-15785. G.B. is partially supported by the Belgian National Fund for Scientific Research (FNRS).

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(Received April 24, 1986;
revised September 3, 1986;
accepted September 8, 1986.)