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RESEARCH COMMUNICATIONS

Effect of solar activity on the minor constituents in the mesosphere and lower thermosphere

T. S. N. Somayaji and T. Aruna Mani

Physics Department, Andhra University, Visakhapatnam 530 003, India

The effect of solar activity on the different minor species in the mesosphere and lower thermosphere has been studied using the one-dimensional model. The concentrations of all the species show significant increase with solar activity. The variations in the hydrogen and oxygen species are attributed to the variations in photodissociation coefficients of H_2O and O_2. The increase in the concentration of atomic nitrogen above 90 km is due to the ionic reactions in the lower thermosphere.

In the past decade studies of minor constituents in the middle atmosphere have acquired great importance, particularly with respect to the stratospheric ozone problem. Indeed, ozone is one of the many minor constituents produced by photochemical reactions in the middle atmosphere and its concentration is greatly affected through photochemical reactions involving other minor constituents. The UV region of the solar spectrum is known to vary over the 11 year solar cycle[1-3] and since the minor constituents in the middle atmosphere are produced mainly by the photochemical reactions that take place there, a solar cycle variability for the distributions of the minor constituents is expected. The solar cycle dependence of the distribution of minor species is important for investigations such as the composition, thermal structure and dynamics of the upper atmosphere and its state of ionization. Several sophisticated models of the mesosphere/lower thermosphere minor constituents have been developed and the variation of the distributions of the minor constituents arising from variations in solar illumination for different time scales have been studied earlier.[4,5]

As a part of the Indian Middle Atmosphere Programme, we have developed a time-dependent one-dimensional model for the minor constituents in the mesosphere and lower thermosphere[6].

In view of the aforementioned importance of the solar cycle dependence of the distribution of minor constituents, we have applied this model for studying the variation of the distributions of the minor constituents from solar minimum ($R_z = 20$) to solar maximum ($R_z = 200$). The results of this study are presented and discussed in this paper.

The details of the computation technique were described earlier[6]. The solar irradiance and absorption cross-section data for the two solar activity conditions were taken from Deshpande and Mitra[7]. The vertical distribution of $O(^3P)$, $O(^1D)$, O_3, O_2 ($^1\Delta g$), H, OH, HO_2, NO, NO_2, $N(^4S)$ and $N(^2D)$ appropriate for the two levels of solar activity conditions were determined from the one-dimensional model.

The solar cycle dependence of the minor constituents is studied by the altitude distribution of a sensitivity parameter (S, as percentage variation) computed as

$$S = (Y_{max} - Y_{min})/Y_{min} \times 100$$

where Y_{max} and Y_{min} are the concentrations of the particular minor species for solar maximum and solar minimum respectively.

The variation of photodissociation coefficients (J) for H_2O, O_2 are shown in Figure 1. The solar cycle dependence of hydrogen (H, OH and HO_2), oxygen ($O(^3P)$,O D,O_3 and O_2^1 g), nitrogen (NO, NO_2, N^4S and N^2D) species is shown in Figures 2–4 respectively.

Figure 2 shows that, below 90 km, the hydrogen species show a structure in the vertical distribution of their sensitivity to solar activity changes with three dominant maxima of about 45% at around 78, 86 and 90 km. The altitude region of these maxima agrees nearly with the region (70–85 km), where the photodissociation of H_2O has a maximum (as shown in
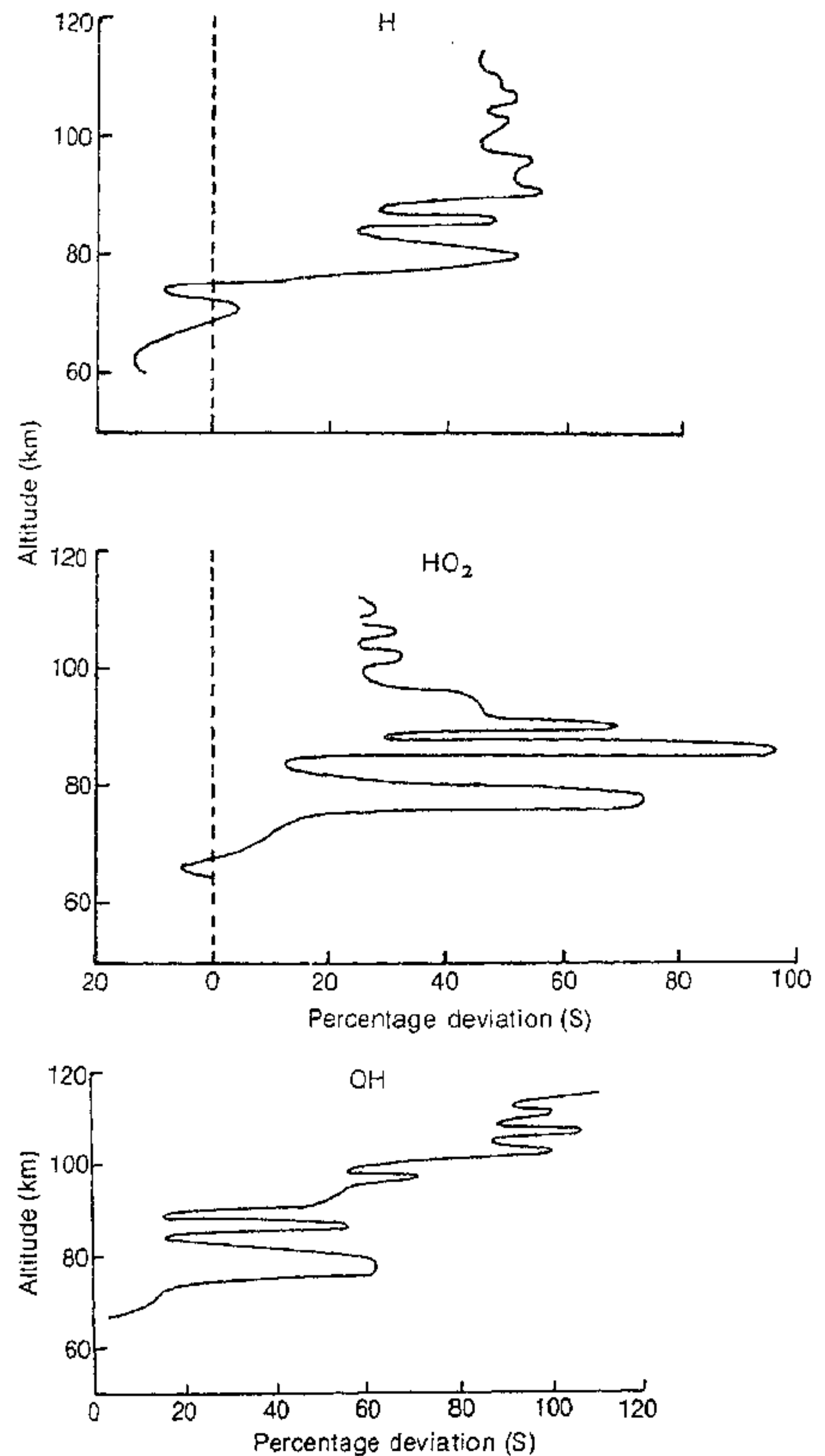
988
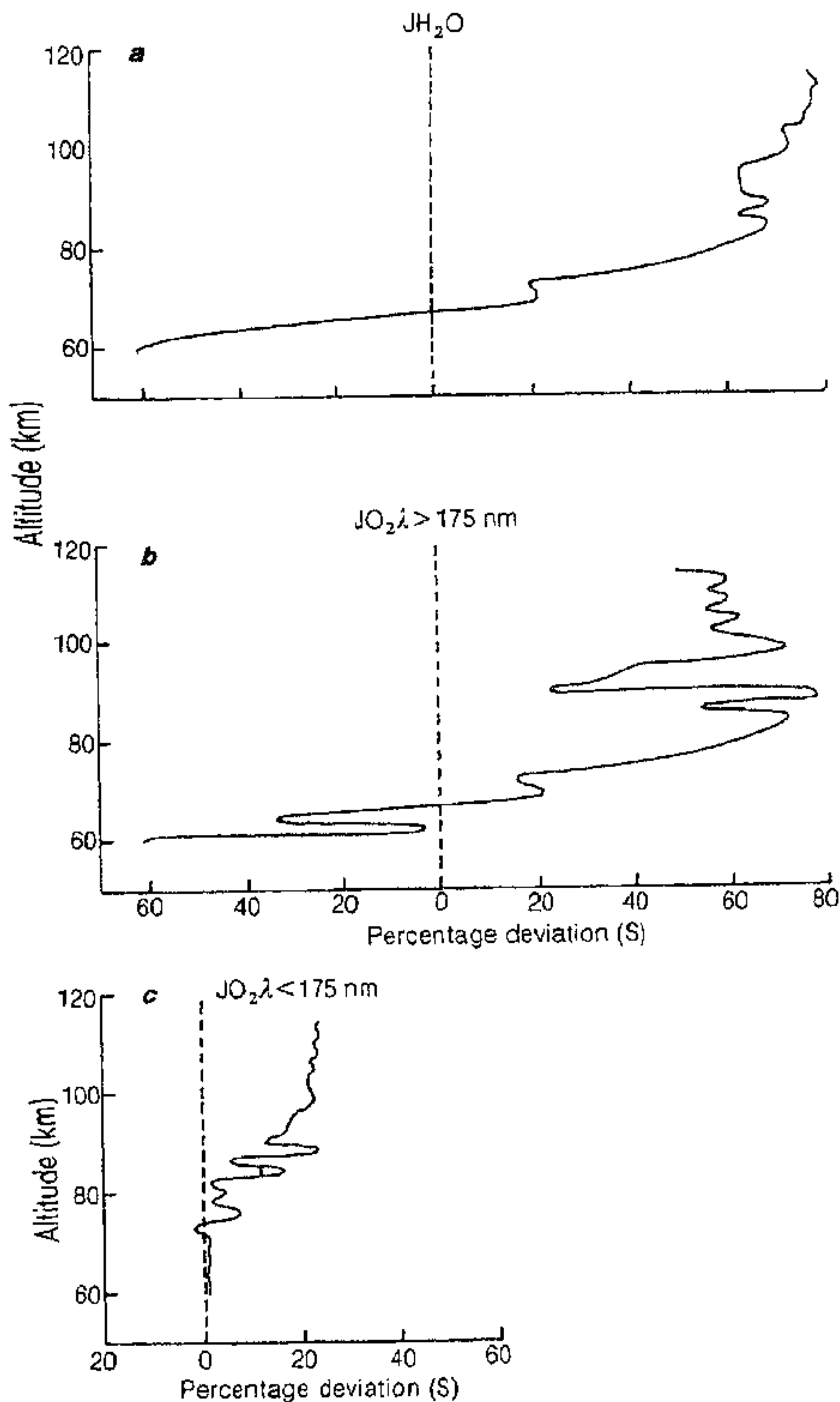
Figure 1. Altitude profile of percentage variation of photo dissociation coefficients (J) from solar minimum to solar maximum.

Figure 2. Altitude profile of percentage variation of hydrogen species from solar minimum to solar maximum.

Figure 1a). Above 90 km the sensitivity of H and HO_2 remains nearly constant at about 50% and 30% respectively, while that for OH increases from 50% at 90 km to 90% at 100 km. These variations can be attributed to variations in JH_2O.

In Figure 3, below 90 km, the sensitivity of oxygen species shows a structure with positive maxima of about 20% or more at 70 km and 80 km and minima at 75 and 85 km and at some of the minima, the sensitivity has even reversed its sign. Above 90 km, the sensitivity of all the oxygen species shows an increasing trend with altitude up to about 100 km.

The main source for atomic oxygen (O), in its ground state and excited state, is photolysis of O_2 and the changes in O can be seen to be similar to changes in $J O_2$ (shown in Figure 1b and c). In the case of O_3 the important source is the three-body reaction of O with

atomic oxygen and the main sink is photodissociation of O_3, the $J O_3$ does not show any changes with solar activity, hence the changes in O_3 will follow the changes in O.

Variations in concentrations of all nitrogen species (N^2D, N^4S, NO and NO_2) are shown in Figure 4. Below 90 km, all the nitrogen species, except N^4S, show significant sensitivity with a maximum of 40% or higher at about 75 km and a negative maximum of about 20% at around 88 km. N^4S was relatively less sensitive, below 90 km, with the sensitivity values ranging between +10% and −20%. Above 90 km, while NO and NO_2 are relatively insensitive, atomic nitrogen, both in its ground state (^4S) and excited state (^2D) show a steep rise in sensitivity with altitude.

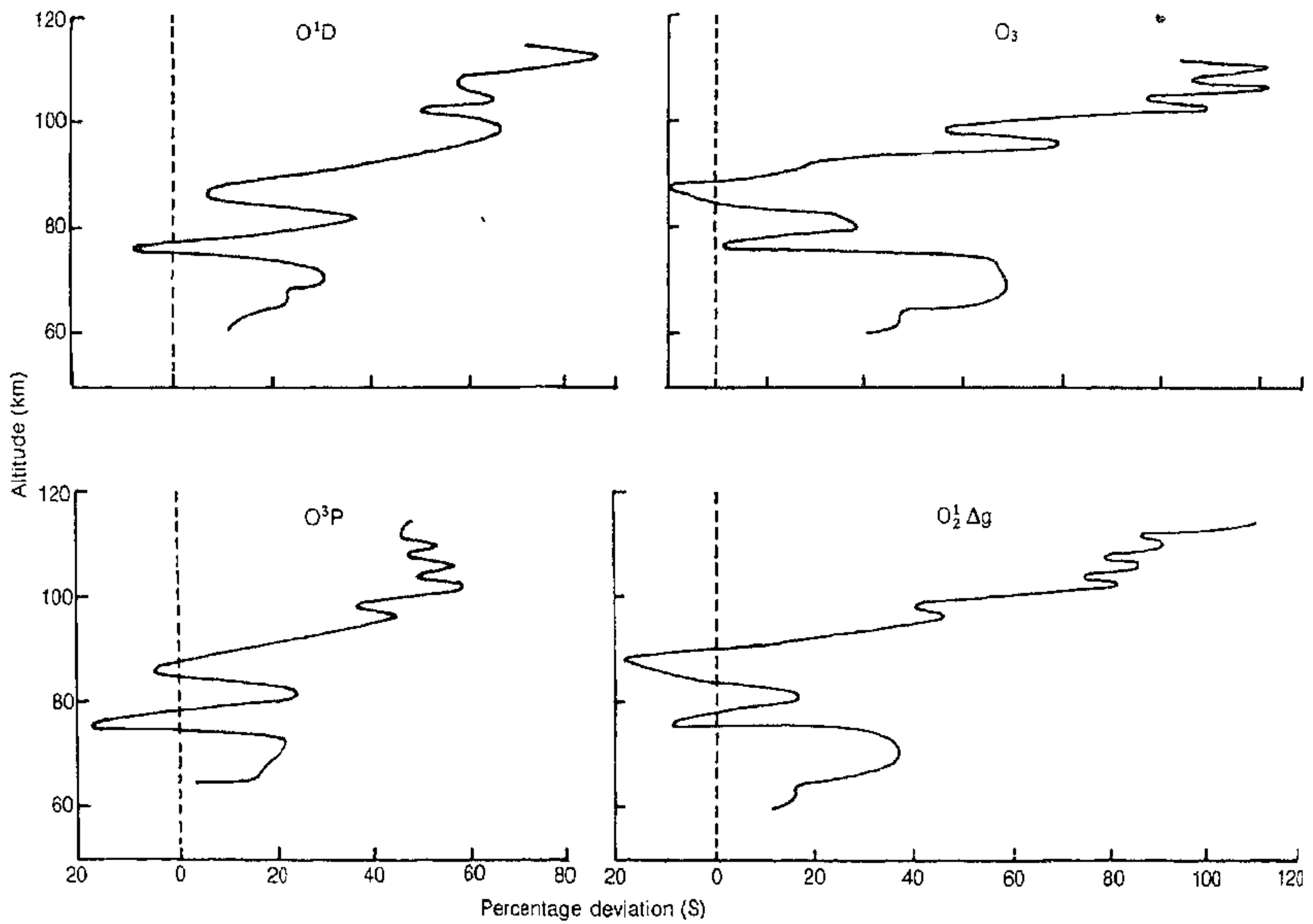The main production of atomic nitrogen above

Figure 3. Altitude profile of percentage variation of oxygen species from solar minimum to solar maximum.
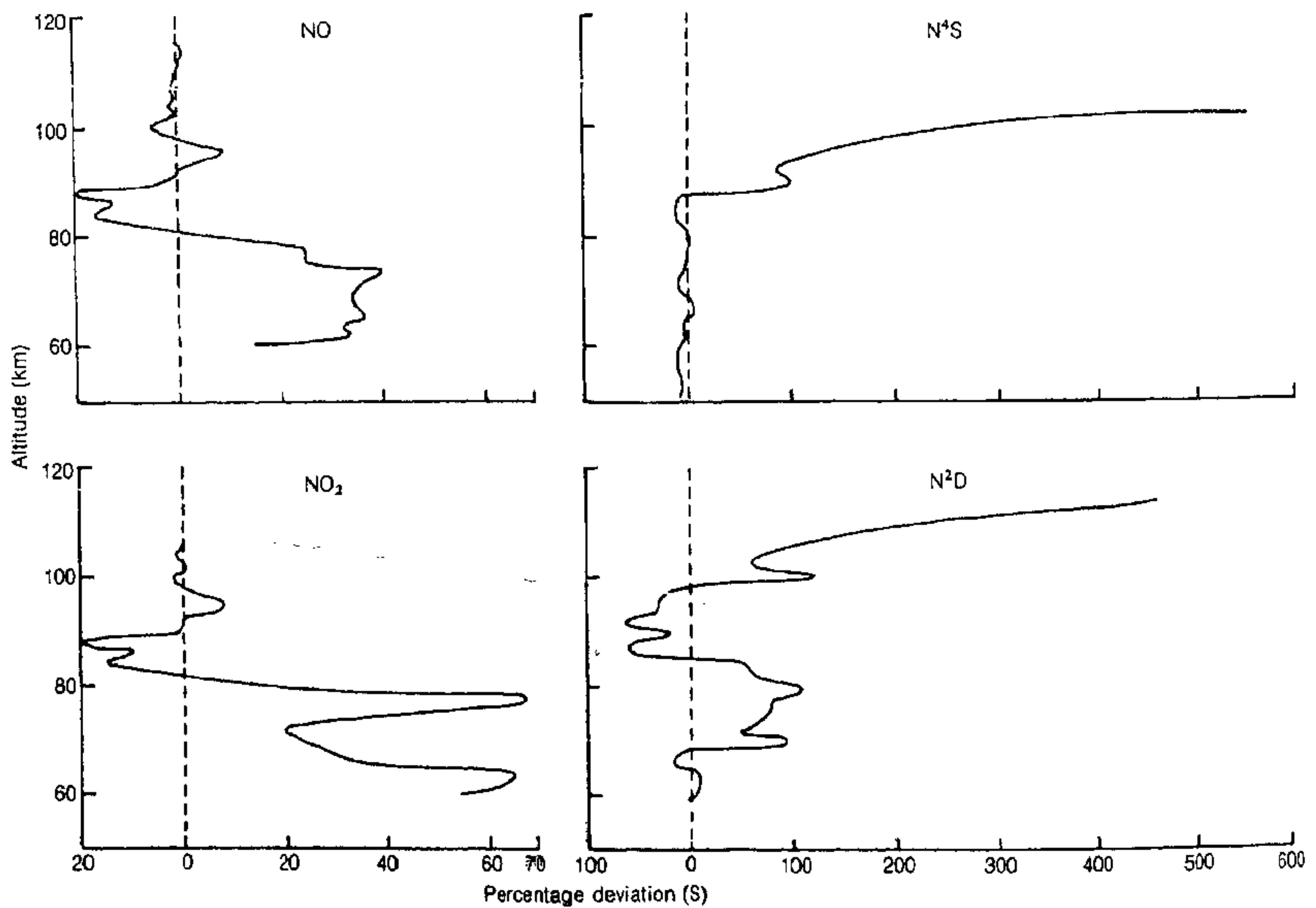
Figure 4. Altitude profile of percentage variation of nitrogen species from solar minimum to solar maximum.

90 km, is through reactions of ionic species and hence the increase in solar activity results in an increase in atomic nitrogen concentration as expected.

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Solvent effect on fluorescence lifetime of 8-anilino-naphthalene-1-sulphonate

T. Bhatt, H. B. Tripathi and D. D. Pant
Department of Physics, D. S. B. Campus, Kumaun University, Naini Tal 263 002, India

We report the fluorescence lifetime of 8,1-ANS in different solvents and compare them with the previously reported values. Effects of solvent viscosity and polarity on fluorescence lifetime have also been studied. A simple empirical relationship between fluorescence lifetime and solvent viscosity/polarity shows that the fluorescence lifetime is more sensitive to solvent polarity rather than viscosity.

THE extreme sensitivity of the fluorescence parameters to the environment of 8-anilino-naphthalene-1-sulphonate (8,1-ANS) was first recognized by Weber and Laurence[1]. In water 8,1-ANS exhibits weak green fluorescence whereas an intense blue fluorescence is observed when bound to serum albumin or proteins[2-6]. The fluorescence decay time, quantum yield and fluorescence maximum of this molecule in solvents of varying polarity have been investigated by several workers[7-10]. The picosecond time resolved fluorescence of 8,1-ANS in a mixed solvent system[11], its photophysical decay pathways[12], the edge excitation red shift at liquid air temperature[13], time-dependent spectral shift at two different temperatures[7] have also been studied. In spite of the extensive work already reported on the spectroscopy and photophysics of 8,1-ANS, a

large inconsistency is found in the fluorescence decay times as reported by different authors. In this connection we report the fluorescence lifetime of 8,1-ANS in different solvents and compare them with previously reported values. Effects of solvent viscosity and polarity have also been studied.

Spectroscopic grade solvents (Aldrich Chemicals) were used after distillation. Aqueous solutions were prepared from triple distilled water. 8,1-ANS (Aldrich Chemicals) obtained as ammonium salt was purified by crystallizing it in $MgCl_2$ solution as described by Zadkowski and Fleming[8]. Freshly prepared solutions (ANS concentration 5×10^{-5} M) were studied after bubbling nitrogen for 10 min. Lifetime was measured by degassing using thaw-freeze and pump method in several cycles (in n-propanol only). For fluorescence lifetime measurements a time correlated single photon counting[14] spectrometer was used with a coaxial flash lamp driven set-up with fwhm = 1 hs (Edinburgh Instruments 199 Spectrometer). Some measurements were carried out with a frequency doubled, cavity dumped dye laser synchronously pumped by a mode locked and frequency doubled Nd-YAG Laser (Spectra Physics) as the excitation source with fwhm 450 ps. Data were analysed with a PDP 11/2 micro computer by reconvolution method using a least-square fitting method. The goodness of the fit was estimated by reduced χ^2, distribution of residuals and standard deviation.

The steady state spectra of 8,1-ANS recorded by us are similar to those reported earlier[7,8]. The fluorescence decay times of 8,1-ANS in various solvents at 25°C are given in Table 1 along with their viscosities (η) and solvent polarity parameters[15] $[E_T(30)]$. The fluorescence lifetime values (at 25°C) reported in the literature[7-10] have also been tabulated for comparison. The lifetimes reported by Chakrabarti and Ware[7] are exceptionally high, probably due to the inaccuracies in measurements. However, our values of under-gassed solutions resemble close to those obtained by other workers[8,9]. In an aerated solution the fluorescence lifetime is quenched by the presence of dissolved oxygen. Nakamura and Tanaka[10] have shown that if the solution is degassed by thaw-freeze method, the lifetime increases. However, our values of lifetime (τ_f) (11.8 ns) after thaw-freeze and pump procedure (in n-propanol) did not match with those (14.5 ns) of Nakamura and Tanaka[10]. Our measurements for all other solutions have been done using the method of nitrogen bubbling for degassing.

From Table 1, it is evident that there is a large variation in lifetime in different solvents. In all the solvents, the fluorescence decay of 8,1-ANS exhibits a single exponential behaviour which remains invariant throughout the emission wavelength except in octanol where a double exponential behaviour[16] is observed