Experimental determination of the formation rate of O (1D) and OH in the troposphere from the ozone photolysis by solar radiation (UV-B)

Fact Sheet

Project Information

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FP3-ENV 1C

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30 June 1994

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€ 0

EU contribution
€ 0

Coordinated by
Forschungszentrum Jülich GmbH

Germany

Objective

This project is aimed at improving the understanding of the primary OH production rate in the sunlit troposphere.

1996-10-24ve quantum yield of oxygen (a1 Delta g) from the photolysis of ozone between 270 and 329nm has been measured and placed on an absolute scale by comparison with previous studies. The yield follows that of oxygen (1D2) from 270 to 315nm, but at longer wavelengths deviates from the National Aeronautical and Space Administration (NASA) recommendation for the singlet atomic fragment, with a persistent tail of 10 to 20% out to at least 329nm. Spin forbidden production of oxygen (a1 Delta g) appears to occur, and may affect the recovery of global ozone.
concentrations from atmospheric infrared band emission data.

A spectroradiometer has been used for absolute measurements of solar actinic flux spectra in the wavelength range from 280 nm to 330 nm. The instrument, based on a scanning double monochromator, was equipped with a special receptor optic that has an almost uniform response to radiation for all angles of incidence. From the recorded spectra absolute values for \( J(O_{1D}) \), the rate coefficient for the photolysis of atmospheric ozone yielding \( O(1D) \) atoms, were calculated using published data for the ozone photodissociation cross section. In the autumn of 1993 this approach was compared with 2 other techniques, namely chemical actinometry and filter radiometry, in a series of field measurements in Juelich. In general, a good correlation was found between the \( J(O_{1D}) \) values obtained from the 3 methods. In particular, the good agreement between the 2 absolute and independent techniques, spectroradiometry and chemical actinometry, indicates that spectroradiometry could be used as a versatile method to obtain photolysis frequencies for different trace gases (ozone, nitrogen dioxide, hydrogen peroxide, \( CH_2O \), etc) from 1 actinic spectrum measured with a single instrument.

The oxidizing capacity of the troposphere is mainly determined by the concentration of hydroxyl radicals (OH). These are primarily formed by the reaction of \( O(1D) \) atoms with water vapour. The \( O(1D) \) atoms are produced by the photolysis of ambient ozone with solar radiation in the spectral range of UV-B. Thus, any change of solar UV-radiation has a major impact on the global oxidation potential of the atmosphere. At present, there is a great deficiency of experimental field data on the rate coefficient \( J(O_{1D}) \) for the \( O_3 \) photolysis forming \( O(1D) \) atoms. Furthermore, there is a lack of accurate \( J(O_{1D}) \) measurement techniques for field experiments. Laboratory work will be carried out to reinvestigate the parameters of two crucial photochemical gas phase reactions which limit the accuracy of experimental and theoretical \( J(O_{1D}) \) determinations.

First, the \( O(1D) \) quantum yield will be determined as a function of wavelength with high spectral resolution and for various temperatures over the pertinent range of tropospheric and stratospheric conditions. Second, the branching ratio of the reaction of \( O(1D) \) with \( N_2O \) will be reinvestigated in laser experiments. Field measurements will be carried out in order to intercompare three different techniques to measure \( J(O_{1D}) \) in the field: the chemical actinometry (direct chemical determination of the \( O(1D) \) formation, based on the reaction of \( O(1D) \) with \( N_2O \)), actinic filter radiometer (measurement of the spectrally integrated solar actinic flux) and actinic spectroradiometer (measurement of the spectrally resolved solar actinic flux).

Furthermore, \( J(O_{1D}) \), \( O_3 \) and \( H_2O \) will be measured continuously in the field in order to establish a dataset on the diurnal and seasonal variations of the \( O(1D) \) and OH production rates at 50°N. Its relationship to concurrently observed parameters, such as
as atmospheric aerosol or clouds, will be investigated and compared with theoretical J(OID) predictions from radiation flux models.

Programme(s)

Topic(s)

Funding Scheme

CSC - Cost-sharing contracts

Coordinator

Forschungszentrum Jülich GmbH

Address

Wilhelm-johnen-strasse
52405 Jülich
Germany

Participants (1)

The Chancellor, Masters and Scholars of the University of Oxford

Address

South Parks Road
OX1 3QZ Oxford

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