

Determination of the quantum yield for N(4S) atom production from N₂O photolysis at 193 nm

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Direct detection of N(4S) atom formation in the 193 nm photolysis of N₂O by a technique of vacuum ultraviolet (vuv) laser-induced fluorescence spectroscopy has been reported. Tunable vuv laser radiation around 120.071 nm that is resonant to the one-photon N(2p²3s 4P¹/₂ - 2p³ 4S³/₂) transition has been generated by two-photon resonant four-wave sum frequency mixing in Hg vapor. The quantum yield value for N(4S) formation in the N₂O photolysis at 193 nm has been determined to be 2.1 (+0.9) × 10⁻³. The N(4S) detection technique, which is developed in this study, is very sensitive and the minimum detection limit is estimated to be 2 × 10⁹ atoms cm⁻³. Impact of the photolytic N(4S) and NO(X²Pi) production from N₂O photolysis on stratospheric chemistry has been explored using a one-dimensional photochemical model while the fragmentation was not considered in former model calculations. When the N(4S)+NO dissociation channel is considered in the photochemical model, an enhancement of the NO_x production rate (up to 3%) is observed, which is followed by a decrease of the steady-state O₃ concentration throughout the stratosphere.