Geophysical Research Abstracts
Vol. 17, EGU2015-2130, 2015
EGU General Assembly 2015
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Non-equilibrium radiation of water vapor and retrieval of H_2O in the mesosphere and lower thermosphere.

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In the study the modern investigations of vibrational kinetics of H_2O molecule in the middle atmosphere and development of the models of non-equilibrium radiation of water vapor in the IR ro-vibrational bands are discussed. Our model accounts for 13 excited vibrational states up to energies 7445 1/cm. In this model, 54 vibrational-translational (V-T) and vibrational-vibrational (V-V) processes of energy exchange at collisions of H_2O with N2, O_2 and O, which are important at the atmospheric conditions, were taken into account. Different variants of possible values of the rate constants of non-elastic collisional processes were analyzed considering the experimental data.

Our consideration of the source of excitation of the first vibrational level $H_2O(010)$ due to quasiresonance energy transfer between the first vibrational levels of O_2 and H_2O molecules is based on the YM-2011 model of electronic-vibrational kinetics of excited products of ozone and oxygen photolysis in the mesosphere and lower thermosphere (MLT). In the model we solved the system of 45 kinetic equations for populations of electronically-vibrationally excited levels of oxygen molecule and excited oxygen atom O(1D). Using the YM-2011 model of electronic-vibrational kinetics of excited products of ozone and oxygen photolysis in the MLT and the model of vibrational kinetics of H_2O molecule which has been developed enables us to retrieve altitude profiles of H_2O concentrations from the measurements of 6.3 mkm H_2O radiance in SABER/TIMED experiment.

Currently, the experimental data for the processes of (V-T) and (V–V) energy exchanges occurring at the collisions with atmospheric molecules and atoms are available only for the transitions involving the lowest vibrational levels. In our 14-level model (the upper levels are 002, 101, 200) and in the latest 25-level model, which utilizes 21 levels of the main isotope molecule up to 050, 031, 130, 210 and 012 at 9000 1/cm, and also in all former models, it was proposed that for all collisional transitions, at which the bending mode quantum number, v2, decreases by 1, the rate constants are equal to that of the transition between levels $H_2O(010)$ and $H_2O(000)$, and at which the bending mode quantum number, v2, increases by 2, the rate constants are equal to that of the transition between levels $H_2O(001, 100)$ and $H_2O(020)$. However, the higher the energy of the vibrational level, the more speculative assumptions are used for estimating the rate constants of collisional transitions from these levels. Based on the analysis of currently available experimental and theoretical data, we discuss the effect of uncertainty of the rate constants values on the H_2O vibrational levels populations, limb radiation spectra and on uncertainties of H_2O retrieval from the measurements of intensities in IR ro-vibrational bands.