Possibility to monitor the HDO/H_2O content ratio in the atmosphere from space observations of the outgoing thermal radiation

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Rather intense spectral lines of HDO molecule have been detected in the emission spectra of the atmosphere observed from space by the IMG sensor in the region $650-2000 \text{ cm}^{-1}$ with the resolution of 0.1 cm^{-1} . The analysis of signals from the isotopic species of the water molecule suggests the possibility of their use for determination of the HDO/H₂O concentration ratio in the atmosphere. As an example of application of the proposed method, the results of IMG/ADEOS data processing at some observation points of the atmosphere over the ocean and land in tropical and middle latitudes under clear sky conditions are presented.

The aggravation of the greenhouse effect puts forward the problem of observation over the parameters of the hydrological cycle. Different methods for determining the parameters of the water cycle from space are being developed now.¹ The advent of the new generation of satellite sensors operating in the far IR with the spectral resolution higher than 1 cm^{-1} , such as IMG/ADEOS,² has allowed an observation of the emission molecular spectra with well resolved rotational structure, including spectral lines of different isotopic species of greenhouse gases.³ One of the promising ways to monitor the hydrological cycle is a measurement of HDO/H₂O concentration ratio the in the atmosphere.4,5 This circumstance stimulates analyzing the IMG spectra of outgoing radiation and developing the adequate methods for determination of the HDO/H₂O concentration ratio.



Fig. 1. Characteristic view of the HDO signal in the IMG/ADEOS atmospheric emission spectra.

In this paper we propose the method to determine the HDO/H₂O concentration ratio from the spectra of the HDO and H₂O molecules measured in the

atmospheric window. It is assumed that the vertical profiles of the HDO and H_2O concentrations coincide accurate to the sought factor.

With the use of the FIRE–ARMS software⁶ and the HITRAN–96 database,⁷ spectral lines of the HDO molecule nearby 1206 cm⁻¹ were reliably assigned in the spectra recorded by the IMG sensor (Fig. 1). The signal is sufficiently strong and is exclusively due to the HDO molecule. Contributions from other atmospheric gases, including $H_2^{16}O$, are practically absent in this spectral region.

The atmospheric spectrum of thermal radiation for the case of observation in the nadir direction is described by the following equation⁸:

$$W(\mathbf{v}) = \varepsilon(\mathbf{v})B(\mathbf{v}, T_0)\exp\left(-\int_0^H K_{\rm atm} dh\right) + \int_0^H K_{\rm atm}B(\mathbf{v}, T(h))\exp\left(-\int_h^H K_{\rm atm} dh'\right) dh.$$
(1)

Here W is the spectral density of the energy brightness of radiation, in $W/(m^2 \cdot cm^{-1} \cdot sr)$; h is the altitude; K_{atm} is the absorption coefficient of atmospheric gases; B(v, T(h)) is the black body emission function; $\varepsilon(v)$ is the emission coefficient of the Earth's surface; T_0 is the temperature of the Earth's surface; H is the altitude of the atmospheric top. The absorption coefficient can be expressed as follows:

$$K_{\rm atm}(\nu, h) = N_0(h) \times \sum_{i=1}^{N_g} n_i(h) \sum_{j=1}^{N_i} \alpha_{ij} \sigma_{ij}(\nu, T(h)), \quad (2)$$

where v is the wavenumber, in cm⁻¹; $N_0(h)$ is the total concentration of molecules at the altitude h, in mol/cm⁻³; T(h) is the altitude profile of the temperature; $n_i(h)$ is the relative concentration of molecules of all isotopic species of the *i*th gas; α_{ij} is the natural abundance of the *j*th isotopic specie of the *i*th gas (these constants can be found in the HITRAN–96 database); σ_{ij} is the absorption cross section, in cm², calculated by the line-by-line summation using spectral parameters from the HITRAN–96 database; N_g is the number of gases taken into account; N_i is the number of isotopic species of the *i*th gas.

To reconstruct altitude profiles, the FIRE–ARMS algorithm minimizes the following goal function:

$$F = \sum_{i=1}^{M} \left[\frac{W^{\text{obs}}(\mathbf{v}_i)}{W^{\text{calc}}(\mathbf{v}_i)} - \frac{W^{\text{calc}}(\mathbf{v}_i)}{W^{\text{obs}}(\mathbf{v}_i)} \right]^2, \quad (3)$$

where W^{obs} and W^{calc} are, respectively, the observed and calculated spectra.

HDO/H₂O concentration The ratio was reconstructed with the use of the FIRE-ARMS software in three steps. The first step involved reconstruction of the altitude profile of the temperature T(h). Toward this end, two spectral regions were used: 665-695 and 817-821 cm⁻¹. The latter was used exclusively to determine the surface temperature at the observation point. At the second stage, the altitude profile of H₂O [$n_1(h)$ in Eq. (2)] was reconstructed in the spectral region 1170-1200 cm⁻¹. Then it was assumed that the altitude profile of HDO coincides with the H₂O profile accurate to a factor, and this factor was fitted to minimize Eq. (3) in the spectral region 1205- 1207 cm^{-1} , where the signal is only due to HDO. Figure 2 shows an example of the calculated spectrum fitting to the measured one as a result of reconstruction of the temperature, H_2O , and α profiles.





Fig. 2. An example of fitting of the calculated spectrum (2) to the measured one (1) after reconstruction of the temperature, H_2O , and α profiles.

Table 1

Tuble 1	
Coordinates of the measurement point	α, in %
14.589 N/7.103 W	0.044
13.602 N/14.824 E	0.028
29.742 N/1.003 E	0.031
69.801 N/151.975 W.	0.019
49.873 N/1.422 W	0.068
$50.849 \ {\rm S}/66.402 \ {\rm W}$	0.031
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The results of reconstruction of the current values of α in the atmosphere at some observation points over the ocean and land are presented in the Table 1.

The obtained values of the HDO/H₂O concentration ratio presented in the Table for low and mid-latitudes demonstrate high variability of the observed α which makes up about 300%. The typical error of the H₂O concentration profile reconstruction from the IMG data varies within 10–20% at different altitudes.⁹ Keeping in mind the above-mentioned variability of α , this accuracy seems quite acceptable for the proposed method of determination of the HDO/H₂O concentration ratio in the atmosphere.

According to the literature data,^{4,5} the difference between the observed HDO/H₂O ratio and the equilibrium one (equilibrium α is about 0.031% according to the HITRAN-96 data) in different regions of the Earth, affords an important information on the water cycle. The global monitoring of long-term variations of the parameter α allows an accumulation of data needed to study characteristic trends in the hydrological cycle due to the growing greenhouse effect.

In conclusion it should be noted that the proposed method is suited to any ways of reconstruction of the altitude profiles and can be used efficiently for zoning of horizontal seasonal distributions of the HDO/H₂O concentration ratio from the data of space observations with such sensors as IMG/ADEOS, IASI/METOP, and others.

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