

# FEASIBILITY STUDY TO MEASURE HDO/H<sub>2</sub>O ATMOSPHERIC PROFILES THROUGH A RAMAN LIDAR

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## ABSTRACT

The Raman lidar technique as currently applied for the retrieval of WV mixing ratio profiles allows, in theory, to estimate HDO/H<sub>2</sub>O atmospheric profiles. The objective of this study is to develop a lidar simulator and to study the feasibility of the ground-based Raman lidar HDO measurement. Thus, the characteristics of a realistic lidar system for the estimation of HDO/H<sub>2</sub>O atmospheric profiles are investigated through a set of numerical simulations.

## 1 INTRODUCTION

Improving the climate models' treatment of the complex water cycle is one of the key goals set by the Intergovernmental Panel on Climate Change. In this context, the isotopologue ratios of water (e.g. HD<sup>16</sup>O/H<sub>2</sub><sup>16</sup>O) contain valuable information about the history of the water content and are a powerful tool for investigating the water cycle (e.g. moistening and dehydration in the tropical troposphere [1], precipitation evaporation in the lower troposphere [2], etc...).

Remote-sensing measurements of HDO are a recent development and there are several satellites providing global HDO datasets [3]. However, absolute measurement calibration is dependent on accurate spectroscopy, while retrieval validation requires in situ profiling capability [3]. In principle, the Raman lidar technique allows estimating HDO/H<sub>2</sub>O atmospheric profiles. Expected advantages of this technique, that could be used to validate the existing measurements, are the temporal and vertical sampling (depending on the required integration) but, more important, independent retrieval for each layer.

The objective of this study is to present a set of numerical tools developed to investigate the feasibility of a Raman lidar system to estimate the HDO/H<sub>2</sub>O profiles. In particular:

- to define the optimal position and width of the filter for the measurement of the Raman backscattering from HDO.
- to define the signal integration over the vertical and over the time of the signal needed to reach a useful SNR
- to assess the capability to estimate variability of the HDO/H<sub>2</sub>O ratio.

The above objectives were obtained developing a radiative transfer model able to simulate a Raman lidar (Sect.2) and using a set of profiles of species of interests, as produced by a Climate Model (Sect.3), as input to produce a statistical dataset of estimated the measurements.

## 2 RAMAN LIDAR SIMULATOR

The model used to simulate the received lidar signal ( $P$ ) for a given wavelength can be schematized as follow:

$$P = INS \cdot EXT \cdot BKS + BKG + NOISE \quad (1)$$

**INS:** contains all terms describing the characteristics of the lidar system. The current version includes the following variables (in parenthesis the values adopted for the simulation discussed in this study):

- laser power (0.8 J), frequency (10 Hz) and wavelength (354.71333 nm) assumed as monochromatic.
  - lidar altitude (0 aslm)
  - overall transfer efficiency, accounting for transmission of optical components and quantum efficiency of the photomultiplier. Current version assumes a spectrally independent value (0.5).
  - telescope radius (1 m).
- Full overlap is assumed for the whole atmosphere.

**EXT:** contains all terms needed to account for the two wavelengths extinction. The following species have been included:

- **Atmospheric gases.** O<sub>3</sub>[4], NO<sub>2</sub> [5] and SO<sub>2</sub> [6] including temperature and pressure dependence

when available in the original data base of absorption cross sections.

- **Molecular scattering.** Molecular scattering extinction is estimated according to [7].
- **Aerosols extinction.** In the version used to produce the results discussed, the profile of aerosols spectral extinction for each wavelength is computed from a highly parametrized representation based on 3 parameters: AOT@380 nm (0.2), Angstrom exponent (1.5) and a scale height (2 km) having assumed the aerosols profile decreasing exponentially.

**BKG:** this term account for the contribution from background assumed as constant (0.1 photons/s) and spectrally independent.

**NOISE:** noise is assumed as the square root of the signal including the background.

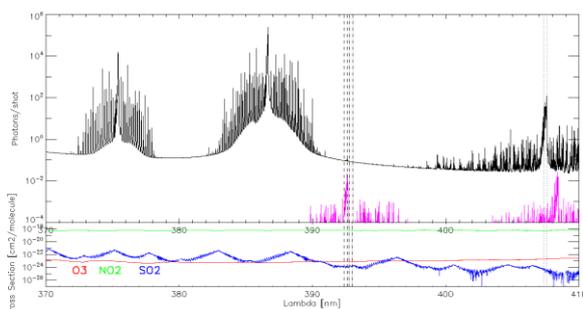


Figure 1. Example of spectrum of simulated numbers of photons per shot received by the system at  $z=75\text{m}$  for the modeled roto-vibrational Raman spectra of  $\text{N}_2$ ,  $\text{O}_2$ ,  $\text{H}_2\text{O}$  and  $\text{HDO}$  (black curve) and, as a reference, from  $\text{HDO}$  (purple curve). Black vertical dashed lines indicate the filter width of the  $\text{HDO}$ ,  $\text{N}_2+$  and  $\text{H}_2\text{O}$  channels selected through the lidar simulator. Absorbing cross section of  $\text{O}_3$ ,  $\text{NO}_2$  and  $\text{SO}_2$  are also depicted (lower panel).

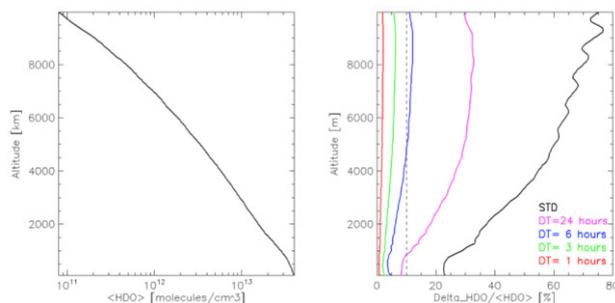


Figure 2. left: Mean profile of the  $\text{HDO}$  concentration in the outputs used; right: mean profiles of relative [%]  $\text{HDO}$  temporal variability for different time intervals.

**BKS:** Raman backscattering cross sections for:  $\text{N}_2$ ,  $\text{O}_2$ ,  $\text{H}_2\text{O}$  and  $\text{HDO}$ . Bi-atomic molecules, roto-vibrational Raman spectra are computed according to [8]. Cross sections for the  $\text{H}_2\text{O}$  and  $\text{HDO}$  Raman lines are computed according to [9] that provide tables for parallel and perpendicular polarizability coefficients and other relevant information for  $\text{H}_2\text{O}$  and  $\text{HDO}$ . Partition function is estimated with a 2<sup>nd</sup> order fit of the values tabled in [10] within the temperature range 175-325 K.

Single wavelength Raman cross sections contribution is computed summing contributions of each single line whose line broadening is estimated assuming collisional broadening as dominant process being the troposphere the region of interest.

The atmosphere is divided in 75 m thick layers from 0 to 10 km. Spectrally the model is limited by the availability of precomputed radiative properties: 370-410 nm with a resolution of 0.01 nm. Figure 1 shows an example of simulated spectrum.

### 3 LMDZ5A simulated data sets

The General Circulation Model (GCM) LMDZ5A (Laboratoire de Météorologie Dynamique- Zoom) is the atmospheric component of the Institut Pierre-Simon Laplace coupled model (IPSL-CM5A). Simulations were performed with forced sea surface conditions following the AMIP protocol [11] with the isotopic version of LMDZ [12] for the 1979-present period. Horizontal wind fields are nudged by ECMWF reanalyses [13] to ensure that simulated meteorological conditions are realistic on a day-to-day basis. Hourly outputs for the year 2007 at Rome Tor Vergata (41.8°N, 12.6°E) are used. Simulated profiles of  $\text{H}_2\text{O}$ ,  $\text{HDO}$ ,  $\text{O}_3$ , T as well as surface pressure are used as input for the lidar simulator. Profiles of  $\text{SO}_2$ ,  $\text{NO}_2$  and aerosols are assumed as constant.

Figure 2 shows the average  $\text{HDO}$  profile and its relative temporal variability for the outputs used in this study.

### 4 PRELIMINARY RESULTS

Once produced a set of simulated spectra from the whole time series of model derived profiles, the first step has been the estimation of the optimal position for a channel to measure the  $\text{HDO}$  Raman scattering. This has been done by searching, within

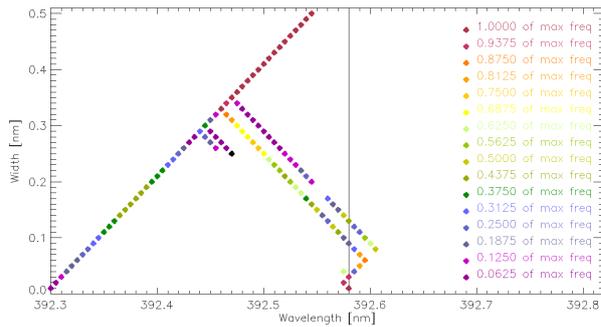


Figure 3 Central wavelength at which the maximum signal occurs (color=frequency of occurrence) as a function of the channel width. Only levels below 3 km are used to select the position of the HDO channel.

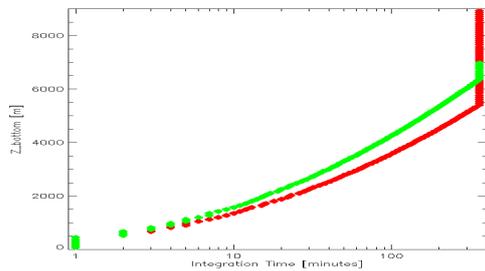


Figure 4 Integration time required to obtain for 95% of the simulated profiles  $SNR \geq 10$  as a function of height. Signal is integrated over 1/3 (Red/Green) km thick layers estimated every 75 m

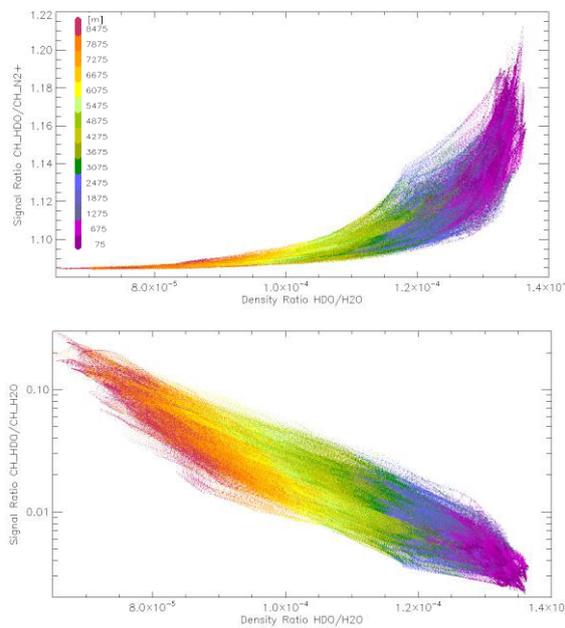


Figure 5. Ratio of the modeled Raman channel signals as a function of the ratio of the HDO/H<sub>2</sub>O densities (color scale indicates the signal height). Upper: HDO/H<sub>2</sub>O. Lower: HDO/N<sub>2</sub>+

a spectral interval including the largest HDO Raman signal, the position of an ideal car-box like spectral response channel maximizing the SNR as estimated from the whole set of simulated profiles. The search was done varying the channel width from 0.01-0.5 nm. Figure 3 shows the results of the channel definition statistics. The following results are relative to a 0.25 nm width channel centered at 392.5 nm.

The second step has been the estimation of expected temporal and vertical resolution. In order to do this, for the HDO channel, as defined in the previous step, the SNR was estimated by integrating over time from 1' to 360' a 1' step for different vertical integration layers. Fig.4 shows the profiles of integration time required to reach a single channel  $SNR \geq 10$  for 95% of the analyzed cases for 1 and 3 km layer thickness.

A third step consisted in investigating the sensitivity to the variability of HDO/H<sub>2</sub>O ratio of simulated signal ratios for different combinations of acquisition channels. Fig.5 shows the scatterplot of signal ratio for two combinations of Raman channels (see Fig.1) as a function of the HDO/H<sub>2</sub>O model derived density ratio.

## 5 CONCLUSIONS

Preliminary results show that, as expected, retrieving HDO/H<sub>2</sub>O profiles with Raman technique is a challenging issue. With a highly performing and somehow ideal system, useful information can be obtained from nighttime measurements on variability day-to-day variability of HDO vertical distribution in the boundary layer and lower free troposphere. However, in the remaining part of the troposphere, the uncertainty of the estimated ratio is of the order of magnitude of the expected dynamic of the variable of interest. In particular:

- from the simulation analyses it appears a strong contamination of the N<sub>2</sub> wings Raman signal in the selected HDO spectral window (Fig.1). This may be partially overestimated due to the assumption on the functional form of the line broadening as uniquely due to pressure broadening as well as by the number of rotational levels (60) considered to produce the N<sub>2</sub> Raman cross section spectra.
- The ratio of the HDO and H<sub>2</sub>O channels (Fig.5 upper panel) shows an inverse dependence from

the ratio of density. In fact, the HDO channel is sensitive to the air density through the contamination of N<sub>2</sub> wings and a moister atmosphere corresponds to lower pressure. A possible solution to remove this dependence is measuring the residual N<sub>2</sub> contribution with a channel (N<sub>2</sub><sup>+</sup>) close to the HDO one (see Fig.1 and Fig.5 lower panel)

- The results of the simulations indicate that, ideally, integrating the measurements to obtain a signal to noise ratio higher than 10 allows the estimation of the HDO day-by-day variability in the boundary layer.

Further activities will include:

- 1) simulation of more favorable scenarios: more powerful system (doubling the laser emission), site located at higher altitude
- 2) simulation for different regimes as for example sub-tropical regions;
- 3) inclusion of more realistic features in the simulator: spectral response of the system, spectrally dependent background, Raman backscattering from other potentially interfering species (e.g. CO<sub>2</sub>);
- 4) estimation of uncertainties due to the effects of variability of other atmospheric components (gas, aerosols) responsible for the extinction.
- 5) definition of a processing algorithm to retrieve HDO/H<sub>2</sub>O profiles and estimation of associated final uncertainty budget.

Finally, the developed code can be adapted to simulate other lidar systems including instrument proposed for candidate satellite missions.

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