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HAL Id: hal-01310834
https://hal.sorbonne-universite.fr/hal-01310834

Submitted on 3 May 2016

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Keywords (up to 6)

Abstract

The quality of spectroscopic parameters that serve as input to forward radiative transfer models are essential to fully exploit remote sensing of Earth atmosphere. However, the process of updating spectroscopic databases in order to provide the users with a database that insures an optimal characterization of spectral properties of molecular absorption for radiative transfer modelling is challenging. The evaluation of the databases content and the underlying choices made by the managing team is thus a crucial step. Here, we introduce an original and powerful approach for evaluating spectroscopic parameters: the Spectroscopic Parameters And Radiative Transfer Evaluation (SPARTE) chain. The SPARTE chain relies on the comparison between forward radiative transfer simulations made by the 4A radiative transfer model and observations of spectra made from various observations collocated over several thousands of well-characterized atmospheric situations. Averaging the resulting 'calculated-observed spectral' residuals minimizes the random errors coming from both the radiometric noise of the instruments and the imperfect description of the atmospheric state. The SPARTE chain can be used to evaluate any spectroscopic databases, from the visible to the

microwave, using any type of remote sensing observations (ground-based, airborne or space-borne). We show that the comparison of the shape of the residuals enables: (i) identifying incorrect line parameters (line position, intensity, width, pressure shift, …), even for molecules for which interferences between the lines have to be taken into account; (ii) proposing revised values, in cooperation with contributing teams; (iii) validating the final updated parameters. In particular, we show that the simultaneous availability of two databases such as GEISA and HITRAN helps identifying remaining issues in each database. The SPARTE chain has been here applied to the validation of the update of GEISA-2015 in 2 spectral regions of particular interest for several currently exploited or planned Earth space missions: the thermal infrared domain and the short-wave infrared domain, for which observations from the space-borne IASI instrument and from the ground-based FTS instruments at the Parkfalls TCCON site are used respectively. Main results include: (i) the validation of the positions and intensities of line parameters, with overall significantly lower residuals for GEISA-2015 than for GEISA-2011; (iii) the validation of the choice made on the parameters (such as pressure shift and air-broadened width) which has not been given by the provider but completed by ourselves. For example, comparisons between residuals obtained with GEISA-2015 and HITRAN-2012 have highlighted a specific issue with some HWHM values in the latter that can be clearly identified on the 'calculated-observed' residuals.

1. Introduction

Remote sensing of Earth atmosphere from space heavily relies on the quality of spectroscopic parameters involved as input to the forward Radiative Transfer Models (RTMs) used to calculate the top of the atmosphere radiance spectra. The general principle of the inversion process, which aims at interpreting the observed spectra of radiances in terms of geophysical variables (such as vertical distribution of temperature, humidity, gas concentrations, clouds, surface characteristics, etc.), consists in minimising residuals between radiances spectra actually measured by the instrument at any given time and location and simulated spectra generated by RTMs from an initial guess of the atmospheric state vector. Over the years, studies relying on various inverse approaches, such as Bayesian or neural network (e.g. Chédin et al., 1985, Escobar et al., 1993) have highlighted that errors in the spectroscopic parameters used in the forward RTMs could introduce spatially, temporally, or even altitude-dependent biases in the retrievals. The accuracy of the retrieved geophysical fields thus ultimately depends on the accuracy of the forward model used and on the quality of the under-lying spectroscopy. Reducing the uncertainties in our knowledge of spectroscopic line parameters and continuum absorption is thus important to improve the

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application of satellite data to weather forecasting, atmospheric composition and climate studies. This also applies to remote sensing from airborne platforms and from the ground.

Spectroscopic parameters used as inputs to RTMs usually come from spectroscopic databases, which provide information on spectral properties of various molecular species: (i) parameters for the molecules having an absorption which can be described in terms of line spectrum (CO_2, H_2O, O_3, CH_4, N_2O, CO, etc.); (ii) far wing absorption and continuum data for H_2O, N2, O2, … ; (iii) far wing absorption and line interference effects data for CO_2 or CH_4; (iv) cross-section parameters for heavy molecules (such as CFCs). The present status of the spectroscopic databases is the result of numerous studies performed during the last 50 years in several dedicated spectroscopic laboratories all over the world. International cooperation contributed to the establishment of widely used spectroscopic databases for atmospheric applications, such as: HITRAN (http://hitran.org ; see Rothman et al., 2013 for its latest version HITRAN-2012) and GEISA (http://www.pole-ether.fr/geisa/ ; see Jacquinet-Husson et al., 2016, this issue for its latest version GEISA-2015). The evaluation of existing spectroscopic databases and their update with the latest spectroscopic data requires a continuing effort. In particular, the validation of the spectroscopic parameters in the most extended range of temperature, pressure and absorber amounts appropriate to the specifications of existing or forthcoming instruments is essential to fully exploit their observing capabilities.

The role of molecular spectroscopy in modern atmospheric research has indeed entered a new phase with the advent of highly sophisticated spectroscopic instruments and computers. The launch of high spectral resolution vertical infrared sounders like the Atmospheric Infrared Sounder (AIRS, Aumann et al., 2003; http://airs.jpl.nasa.gov/) on board NASA/Aqua satellite since May 2002, or the Infrared Atmospheric Sounding Interferometer (IASI, Chalon et al., 2001; https://iasi.cnes.fr/) on board the European polar satellites Metop since October 2006, have opened new perspectives for remote sensing applications, for numerical weather prediction, atmospheric composition and climate studies. The launches of the Thermal And Near infrared Sensor for carbon Observation (TANSO) onboard JAXA Greenhouse Gases Observing Satellite (GOSAT, Yokota et al., 2009; http://www.gosat.nies.go.jp/en/) in January 2009 and NASA Orbiting Carbon Observatory (OCO-2, Crisp et al., 2004; http://oco.jpl.nasa.gov/) in February 2014 have called for the need of improved spectroscopic parameters in the short-wave absorption bands of carbon dioxide (CO_2), methane (CH_4) and oxygen (O_2). Moreover, the need to improve and consolidate the spectroscopic parameters and the RTMs that use them will become a priority in order to exploit the increased spectral

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resolution and radiometric accuracy of new atmospheric instruments like IASI-New Generation (IASI-NG), which has been designed to improve by a factor of 2 the spectral resolution and a factor 2 to 4 the radiometric characteristics as compared to IASI (Crevoisier et al., 2014; https://iasi-ng.cnes.fr).

Several studies have focused on the evaluation of the quality of spectroscopic databases by studying the impact of updated spectroscopy on the estimation of various geophysical parameters from remote sensing observation. For instance, Frankenberg et al. (2008) identified an error in the methane column retrieved from near-infrared spectra recorded by the SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY) instrument onboard ENVISAT related to inaccuracies in water vapour spectroscopic parameters. This error resulted in a positive correlation of retrieved methane with water vapor abundances and thereby led to a systematic overestimation of tropical methane abundances. Updating the water spectroscopy largely eliminated this dependence. Scheepmaker et al. (2013) further evaluated an improved water spectroscopy in the range 4174–4300 cm^{-1}, a region where the spectroscopy of water lines remains a large source of uncertainty, in order to reduce systematic uncertainties observed in the SCIAMACHY retrievals of HDO/H_2O. Similarly, Alvarado et al. (2015) evaluated updated spectroscopic parameters for methane (CH_4), water vapor (H_2O) and nitrous oxide (N_2O) by performing thermal infrared retrievals of methane from the NASA Aura Tropospheric Emission Spectrometer (TES) and comparing the bias of the retrieved values when compared to in-situ measurements. Avarado et al. (2013) implemented a validation of spectroscopy by comparing a global dataset of 120 near-nadir, over-ocean, night-time spectra from IASI to calculations from two versions of the RTM (update or previous version of the spectroscopic database) to determine the impact of spectroscopic updates to the model on spectral residuals as well as retrieved temperature and H_2O profiles. Although efficient in detecting potential issues with spectroscopy of the targeted atmospheric variables, these studies were all based on an *a posteriori* evaluation consisting in evaluating the impact of updated spectroscopy on the retrieved geophysical variables. Several issues limit the use of this kind of approach for validating spectroscopic databases: (i) they rely on the existence of a retrieval or assimilation scheme used to interpret the measured radiances in terms of geophysical variables; (ii) they require the availability of a validation dataset for the targeted geophysical variables; (iii) they are limited to the specific spectral domains used in the retrieval schemes.

This paper aims at introducing an original and powerful approach for evaluating spectroscopy: the Spectroscopic Parameters And Radiative Transfer Evaluation (SPARTE) chain. The SPARTE chain relies

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on the comparison, throughout the whole spectrum, between forward radiative transfer simulations and observations of spectra made from various instruments, both collocated over several thousands of well-characterized atmospheric situations. Considering that observed spectra come from instruments characterized by high radiometric and spectral stabilities, each individual residual can be affected by two kinds of errors: (i) a random error due to the imperfect description of the atmospheric state or to instrumental noise; (ii) systematic errors more related to the spectral line calculation (spectroscopic parameters, line shape, line mixing, continua, etc). Averaging the residuals leads to minimizing the random errors to highlight more systematic error due to spectroscopic parameters. This chain is built on the long-term involvement of Laboratoire de Météorologie Dynamique (LMD) in: (i) developing and maintaining the spectroscopic database GEISA; (ii) developing and maintaining forward RTMs; (iii) performing calibration/validation activities of several Earth Observation space missions. Although the SPARTE chain can be used to evaluate spectroscopy from any spectroscopic databases, from the visible to the microwave, taking into account or not the cross-sections or the diffusion by the aerosols, this paper will focus only on the validation of the line parameters sub-databases, for two spectral regions of particular interest for several currently exploited or planned space missions: the thermal infrared domain and the short-wave infrared domain, for which observations from the space-borne IASI instrument and from the ground-based Fourier Transform Spectrometer (FTS) instruments at the Parkfalls Total Carbon Column Observing Network (TCCON) site will be used respectively.

Section 2 describes the models, databases and instruments used in this study. Section 3 details the SPARTE chain. Section 4 presents the evaluation of various spectroscopic databases by focusing on the evaluation of methane and water vapour spectroscopic parameters, and of CO_2 line-mixing. Section 5 gives the conclusion.

2. Models and data

2.1. Spectroscopic databases GEISA and HITRAN

Several types of spectroscopic parameters, depending on the absorbing molecules, are needed by RTMs: (i) line parameters; (ii) far wing absorption and/or continuum for H_2O; (iii) far wing absorption and line interference effects for CO_2 or CH_4; (iv) cross-section parameters for heavy molecules (CFCs, etc.) or for the major constituents O_2 and N_2 presenting a collision-induced absorption spectrum for which a line-by-line approach is unpractical or improper.

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The line parameters (e.g. position, intensity, width, temperature dependence, lower-state energy, etc.) are usually obtained through experimental studies of pure gas and mixtures (with O_2, N_2 or synthetic air) using very high spectral resolution laboratory instruments (typically 0.002 cm^{-1}) and various absorption cells with controllable path length, temperature and pressure conditions. A theoretical analysis and modelling of the experimentally acquired spectra using vibration-rotation Hamiltonian and dipole moment operators is often necessary, in order to check the consistency of the measurements and/or to complete the parameters that could not be measured. For intensity and position, measured and theoretical parameters are complementary to cover the whole spectral range of GEISA or HITRAN. For parameters such as air-broadened HWHM and pressure shift, values mostly come from theoretical calculations.

Most of these spectroscopic parameters, especially line parameters and cross sections, are compiled in spectroscopic databases like GEISA and HITRAN. The GEISA database has been created in 1976 at LMD and updated versions have been regularly distributed since then, the two latest being the 2011 (Jacquinet et al., 2012) and 2015 (Jacquinet et al., 2016, this issue). The HITRAN database originates from the Air Force Geophysics Laboratory (AFGL) database, initially dedicated to military applications. The two latest versions of HITRAN were released in 2008 (Rothman et al., 2009) and 2012 (Rothman et al., 2013). Because of its origin, certain molecules, mainly related with planetary atmospheres (especially those of the giant planets) are specific to GEISA, such as: GeH_4, C_3H_8, C_2N_2, C_3H_4, HNC, and C_6H_6. Also specific to GEISA is the fact that CH_3D and C_2HD are considered as independent molecules, whereas in HITRAN, they continue to be one isotopologue of respectively methane and acetylene. The successive alternative updates between both databases insure to the users an access to the latest up-to-date spectroscopic parameters. For instance, The GEISA-2015 sub-database of line parameters archives, given at the reference temperature of 296K, contains the spectral properties of 52 molecular species (113 isotopologues) corresponding to a total of 4,800,000 entries in the spectral range from 10^{-6} to 35,877.031 cm^{-1} (1010 to 0.28 µm). Among them, 22 molecular species have been updated since the GEISA-2011 edition. In particular, HDO is now considered as an independent molecule (see Section 4.1) and SO_3 is a newly added molecular species. Although the cross-section and aerosol sub-databases of GEISA have also been updated, this paper focuses on the evaluation of the line spectroscopic sub-database only.

The process of updating such databases is particularly challenging and requires several steps: (i) collecting the most recent measurements from every concerned laboratory; (ii) reconciling the measurements made by different teams; (iii) evaluating different line lists received to choose the

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updated parameters/lines as compared to the previous version; (iv) filling the absence of missing parameters (e.g. when only intensities are provided). In order to provide the users with a database that insures both an optimal characterization of spectral properties of molecular absorption, as well as an access to all parameters required for radiative transfer computations, the evaluation of the databases content and the underlying choices made by the managing team is a crucial step. In that sense, we will show that the availability in parallel of the two databases GEISA and HITRAN enables identifying remaining issues in each database by comparing the impact of their respective line parameters on radiative transfer simulations of same sets of observed spectra.

2.2. Radiative transfer model 4A in the thermal and shortwave infrared

4A (Automatized Atmospheric Absorption Atlas) is a fast and accurate line-by-line radiative transfer model (Scott and Chédin, 1981, Chéruy et al, 1995). It is an advanced version of the nominal line-by-line STRANSAC model (Scott, 1974, Tournier et al., 1995) and is basically a compressed look-up-table of optical depths calculated once and for all. It can be coupled to any spectroscopic databases and can simulate any instruments and observed configurations (ground, airborne, satellite). Since the beginning of 2001, an operational version 4A/OP has been developed by Noveltis (http://www.noveltis.com/4AOP/) in collaboration with the French national space agency CNES and LMD. Originally developed for the thermal infrared region, the model has recently been extended to the short-wave infrared region (4A/OP-SWIR) and is currently extended to the Ultra Violet and Visible domain. 4A is the official code chosen by CNES for calibration/validation activities of IASI, Merlin (https://merlin.cnes.fr) and MicroCarb (https://microcarb.cnes.fr) missions. Although not used in this study, 4A/OP has been coupled to DISORT to take into account scattering effects.

4A has a long history of validation within the frame of the international radiative transfer community. Most of the validation results have been extensively discussed in a number of intercomparison exercises and in particular during the ITRA (Intercomparison of Transmittance and Radiance Algorithms) working groups - 1983, 1985, 1988, 1991 of the International Radiation Commission (see Chédin et al., 1988) and during the ICRCCM (InterComparison of Radiation Codes in Climate Models) campaigns (Luther et al. 1988). More recently, launch of hyperspectral sounders such as AIRS and IASI have led to more extensive validations, within the frame of international campaigns or working groups.

Thanks to its precision, speed and spectral coverage, 4A is well suited to evaluate the line parameters archived in spectroscopic databases over a large spectral range. By default, these parameters assume a Voigt line shape, combining the Doppler and the Lorentzian effects. In general, when a molecule does not follow this rule, the absorption has to be corrected by adding parameterizations representing physical phenomena such as, for instance, line mixing (interference between transitions of a same molecule), speed dependency (effects due to the speed of the molecule), or continua (aiming at correcting other defaults of absorption in the Voigt modelling). In 4A, line mixing effects have been included for CO_2 (Niro et al, 2005; Lamouroux et al, 2015), CH_4 (Pieroni et al, 1999; Tran et al, 2006; Tran et al, 2010) and O_2 (Tran et al, 2008). In the case of water vapour, the Voigt line shape is inconsistent, and a lack of absorption has been observed for a long time. To correct this effect, a continuum, derived from the widely-used 'Clough modelling' MT CKD 2.5 (Mlawer et al, 2012) which has been adjusted to the specific 4A line cut-off, is used.

2.3. Atmospheric inputs: the ARSA database

Radiosonde reports are critical for a wide range of applications as forward and inverse models validation, verification of satellite measurements, and any other application dealing with Earth observations from nadir or limb. In order to fulfil the needs of these applications, the ARSA (Analyzed RadioSoundings Archive) database (http://ara.abct.lmd.polytechnique.fr/index.php?page=arsa) has been elaborated, starting from observations by worldwide distributed radiosonde stations and combining them with surface and other auxiliary observations (Scott et al. in prep.). Physically coherent quality control tests have been developed to detect and eliminate gross errors: format problems, redundant radiosoundings and levels, unrealistic jumps, physically implausible values, temporal and vertical inconsistencies in temperature and dew point temperatures. Furthermore, relying upon still more elaborate quality control, we have made sure that every radiosonde report kept was fully compatible with the forward radiative transfer simulations of the high spectral resolution observations. The current ARSA database (about 6 million elements) starts in January 1979, and is extended onwards, on a monthly basis.

2.4. Observations: IASI and TCCON

2.4.1. The Infrared Atmospheric Sounding Interferometer

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IASI, developed by CNES in collaboration with EUMETSAT, is a Fourier Transform Spectrometer based on a Michelson Interferometer coupled to an integrated imaging system that measures infrared radiation emitted from the Earth. IASI provides 8461 spectral samples, aligned in three bands yielding a continuous spectral scanning from 645 to 2760 cm^{-1} (15.5μm and 3.63μm), with a spectral resolution of 0.50 cm^{-1} after apodisation and a spectral sampling interval of 0.25 cm^{-1}. IASI was launched on October 2006 onboard the European Metop-A platform and on September 2012 onboard Metop-B. Since then, it has been providing water vapour and temperature soundings for operational meteorology, while observing simultaneously a whole suite of trace gases, surface properties and aerosol properties (Hilton et al., 2012). Owing to its spectral and radiometric stability, IASI has become the reference of the WMO GSICS (Global Space-Based Inter-Calibration System, http://gsics.wmo.int/) program for the thermal infrared. Figure 1 shows a typical IASI spectrum (tropical airmass for January 2008) where the main absorption bands of various gases are indicated.
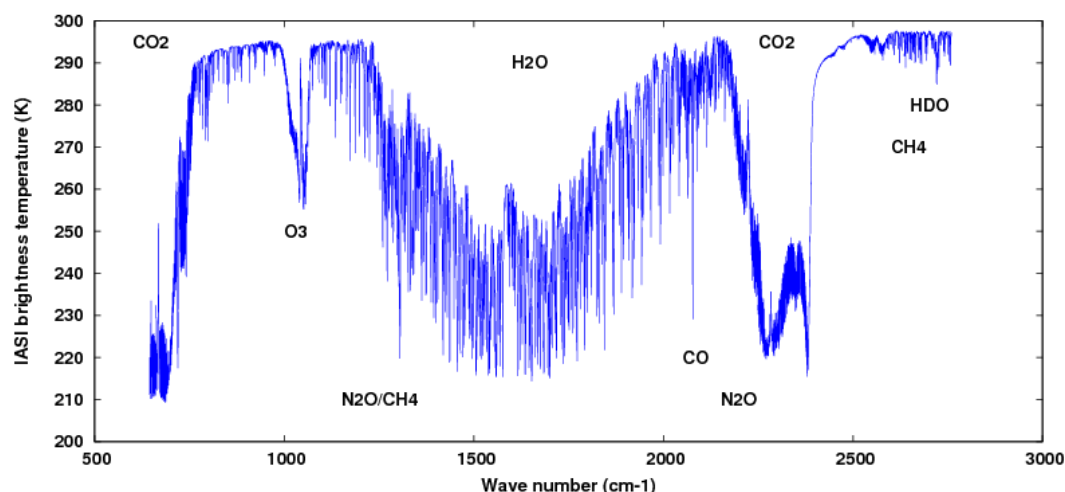

FIG. 1. Spectrum expressed in brightness temperature observed by IASI onboard Metop-A in the tropics (average over January 2008).

2.4.2. The Total Carbon Column Observing Network

The Total Carbon Column Observing Network (TCCON) network uses ground-based Fourier transform spectrometers (FTS) looking at the sun for the measurement of column abundances of CO_2, CO, CH_4, N_2O and other molecules that absorb in the near-infrared (Wunch et al., 2011). TCCON interferograms are recorded with a maximum OPD of 45 cm, giving a spectral resolution of approximately 0.02 cm^{-1}, which is the optimum resolution for a source-photon-noise-limited detector in the 6000–8000 cm^{-1} region. Two detectors are used: (i) InGaAs (Indium-Gallium-Arsenic) between 4000 and 11000 cm^{-1}; (ii) Silicium between 11000 and 13000 cm^{-1}. The thermodynamic

description mostly comes from NCEP profiles, and for each spectrum, total column abundances (H_2O, CO_2, N_2O, CO and CH_4) have been retrieved using version 2012 of GFIT algorithms in the spectral band 6220 cm^{-1} ± 40 cm^{-1} (Wunch et al. 2011).

Founded in 2004, the network has been established as a powerful tool for the validation of different satellites dedicated to the measurement of greenhouse gases in the short wave infrared, like SCIAMACHY, GOSAT and OCO-2. A key feature of this network is its stringent requirements with respect to instrumentation, acquisition procedures, data processing and calibration. So far, only the BRUKER high resolution interferometer IFS 125HR meets the requirements for sensitivity and long-term stability. Measurements can be performed throughout the whole day during clear sky conditions, with the sun typically 10° above the horizon. Spectra are recorded individually within 2-3 min.

In this study, use is made of 465 spectra acquired between May 2008 and April 2011 at Parkfalls (United States). In order to increase the signal-to-noise ratio, the diurnal average of the spectra is used. These spectra were chosen according to the following criteria: coverage of different seasons, various solar zenith angle and water vapour content.
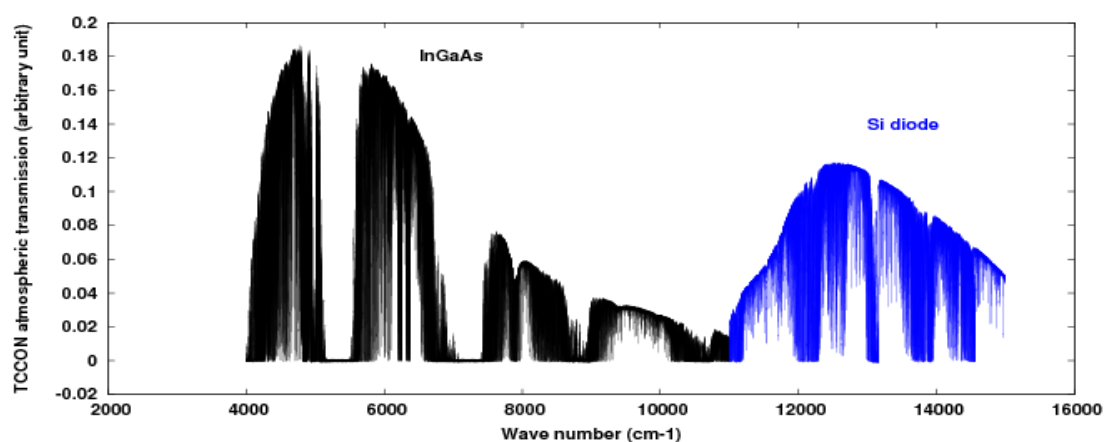

FIG. 2. Spectrum (arbitrary unit) observed by TCCON at Parkfalls for the 15th of September 2008 for the 2 detectors (InGaAs in black, Si-diode in blue).

3. The SPARTE chain

3.1. General principle

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The SPARTE chain relies on hundred of comparisons between clear sky calculated and observed radiances, so called 'calc - obs' residuals. The general principle is described in Fig. 3. The first step consists in co-locating in time and space (based on given criteria) observed radiance spectra with well-described atmospheric situations. This yields the collocation database. For each radiance-atmosphere pair, the atmospheric state, which typically includes atmospheric profiles of temperature, water vapour and several trace gases, as well as surface characteristics (surface temperature, pressure, emissivity, etc.), are used as input to the radiative transfer code 4A (see Section 2.2) to compute simulated radiances. This simulation takes into account instrumental and observation characteristics (such as the instrument spectral response function (ISRF), angle of observation, solar angle, etc) and can use any given spectroscopic database as input. The last step consists in computing the mean and standard deviation of the difference between calculated and observed radiances, the 'calculated-observed' residuals, over the hundreds of situations contained in the collocation database.

Assuming that the observed spectra come from an instrument characterized by high radiometric and spectral stabilities, averaging hundreds of situations together minimizes the simulation errors linked to the non perfect description of the atmospheric state and enables the detection of spectral signatures of errors that are related to the spectral line calculation (spectroscopic parameters, line shape, line mixing, continua, etc).

In order to highlight the strength of analyzing hundreds of situations together compared to a one-by-one analysis, two spectral regions will be considered in the following: the 645-2760 cm^{-1} interval of the IASI instrument (cf Section 3.2) and several intervals covered by TCCON FTS instruments in the short wave infrared (Section 3.3). It has to be noted that in this study, the SPARTE chain has only been applied to the validation of the line parameters, even if it could also be possible to apply it to the validation of the cross-sections and aerosols (thanks to the taking into account of diffusion in 4A/OP) sub-databases of GEISA.
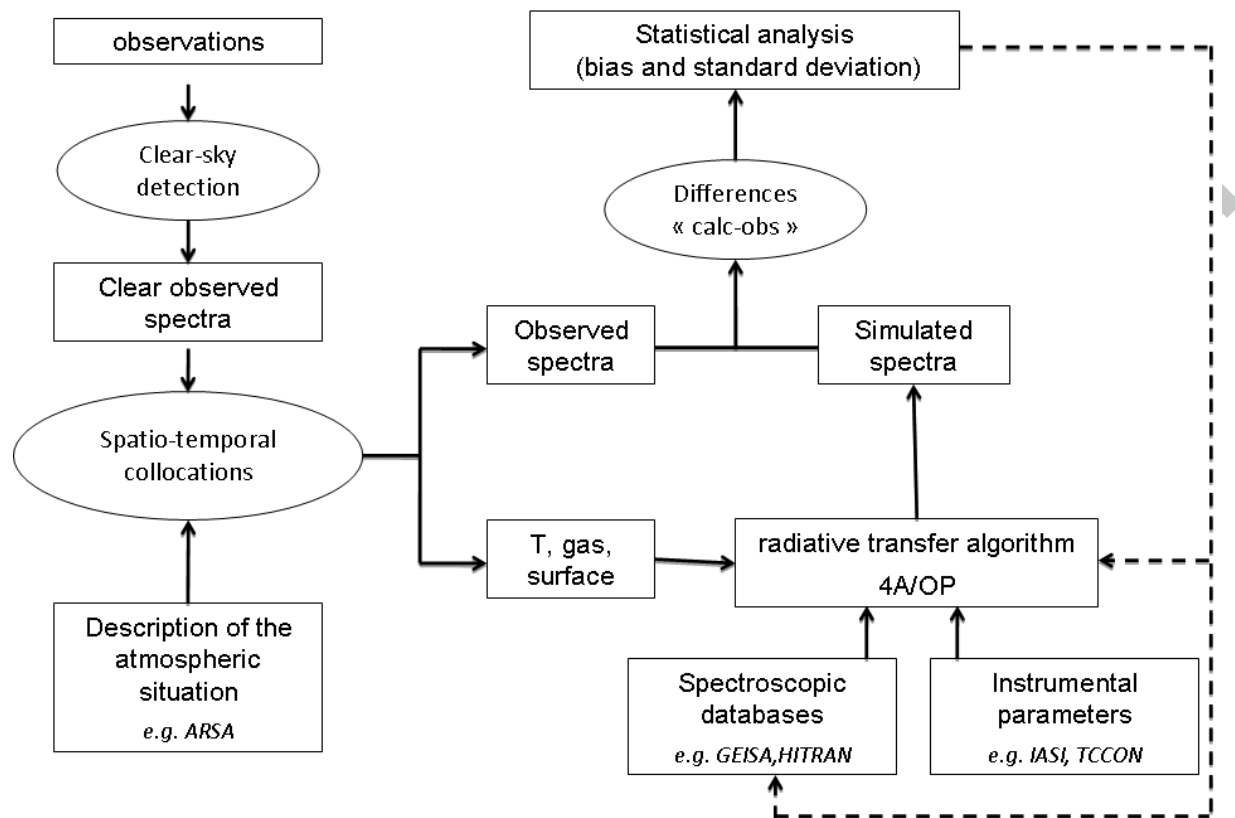
FIG 3. General principle of the Spectral Parameters And Radiative Transfer Evaluation (SPARTE) chain.

3.2. The thermal infrared domain: collocations with IASI

The comparison between IASI radiances and 4A/OP simulations are based on collocated clear sky atmospheric situations with the ARSA database described in Section 2.3. The space and time collocation criteria between any IASI observation and an ARSA atmospheric situation are respectively 100km and 3 hours. Following the time coverage of IASI onboard Metop-A, the available collocations span the period July 2007 to present. The related statistics (mean and standard deviation) of the residuals are separated into land/sea, day/night, satellite zenith angle and air mass type cases. The averaged number of situations per month is in the range of 200 to 1500, depending of the land/sea/night/day/airmass configuration. Due to the remarkable radiometric stability of the IASI instrument and the continuous coverage of the spectral range 645 to 2760 cm^{-1}, the resulting systematic structure of the residuals can be used to detect and potentially correct errors related to the spectral line characterization.

To illustrate the strength of considering thousands of situations, the 'calculated-observed' residuals, expressed in terms of brightness temperatures, obtained for five 'IASI observation − ARSA

atmospheric situation' pairs are shown in Fig. 4b (colored lines) together with the corresponding IASI spectra (Fig. 4a). Also shown in black is the averaged residual computed over the period July 2007 - June 2015 (8 years). The individual residuals corresponding to each collocation are strongly affected by 2 noises: the radiometric noise of the instrument and the noise coming from an incomplete characterization of the atmospheric situations. This is particularly well seen in Fig. 4b: between 2200-2400 cm^{-1}, which is a region where IASI is characterized by a particularly large radiometric noise, averaging a sufficient number of situations cancels out the radiometric noise (black line in Fig. 4b); between 1300 and 2100 cm^{-1}, which is a region dominated by H_2O absorption, averaging the residuals cancels out the improper characterization of water vapour profiles stemming from the large variability of atmospheric humidity and the large collocation criteria used in the chain. All together, averaging hundreds of residuals enables extracting significant features much smaller than the instrumental noise itself, and not affected by the incomplete characterization of the atmosphere.
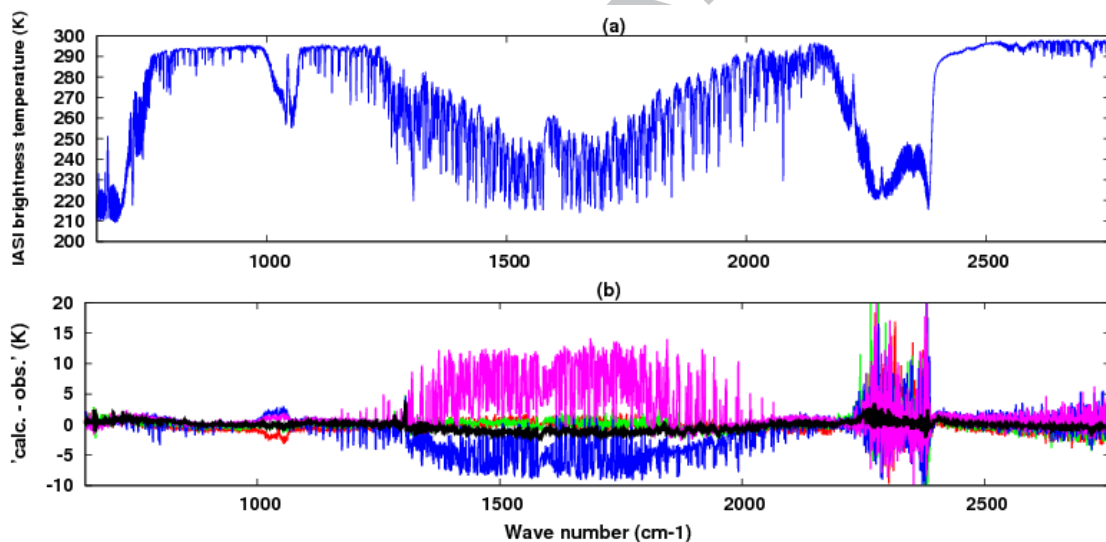

FIG. 4. (a) IASI spectrum averaged over the July 2007-June 2015 for every ARSA tropical situations over sea at night (~ 12000 collocations). (b) calculated-observed residuals for five 'IASI observation-ARSA situations' pairs randomly taken (colored lines) and averaged over the whole period (black curve).

3.3. The short-wave infrared domain: collocations with TCCON

For every selected TCCON spectrum, the atmospheric state used as inputs to 4A comes from co-located NCEP (National Centers for Environmental Prediction) reanalysis, which includes: temperature, water vapour, CO_2, N_2O, CO and CH_4 profiles. In order to take into account the diurnal

variability of gases, NCEP profiles are scaled by matching the corresponding total column abundances with the one retrieved from the spectrum itself using the GFIT retrieval code. In addition, surface pressure, altitude and solar angle are taken from TCCON dataset. The instrument spectral response function used to generate the computed spectra is taken from Wunch et al. (2011). It is a convolution of a "*sinc*" and rectangular functions representing the finite length of the interferogram and the finite field of view (FOV) of the FTS, respectively.

TCCON spectra are usually uncalibrated and given in photons s^{-1} m^{-2} sr^{-1} cm unit, while 4A spectra are radiances given in W m^{-2} sr^{-1} cm^{-1}. Since the FTS instrument ISRF is not known (especially the filter response function), TCCON spectra thus need to be normalized in order to get two products that can be compared with each other. First, in a limited spectral region of ~100 cm^{-1}, quasi-transparent channels (i.e. with an atmospheric transmission greater than 0.97) are selected in 4A spectra. Then, from these channels, a baseline is computed for the whole region for both 4A and TCCON spectra. Finally, by dividing both spectra by their own baseline, 4A and TCCON transmissions can be compared.

This normalization procedure suffers from two limitations: (i) a transmission of 1 is artificially attributed to channels characterized by transmissions greater than 0.97; (ii) in spectral regions where no quasi-transparent channels can be found, the procedure can induce a global bias in the residuals. However, these limitations do not impact the detection of line parameters issues, which display low transmission and are characterized by local spectral signatures.

The 'calculated-observed' residuals obtained for five "TCCON observation – atmospheric situation" pairs are shown in Fig. 5 together with the corresponding TCCON spectrum. Also shown in black (Fig. 5b) are the averaged spectra computed over the whole 465 pairs considered here. As discussed in Section 3.2, considering the averaged spectrum cancels out the radiometric noise and levels out the influence of atmospheric situations and observation characteristics.
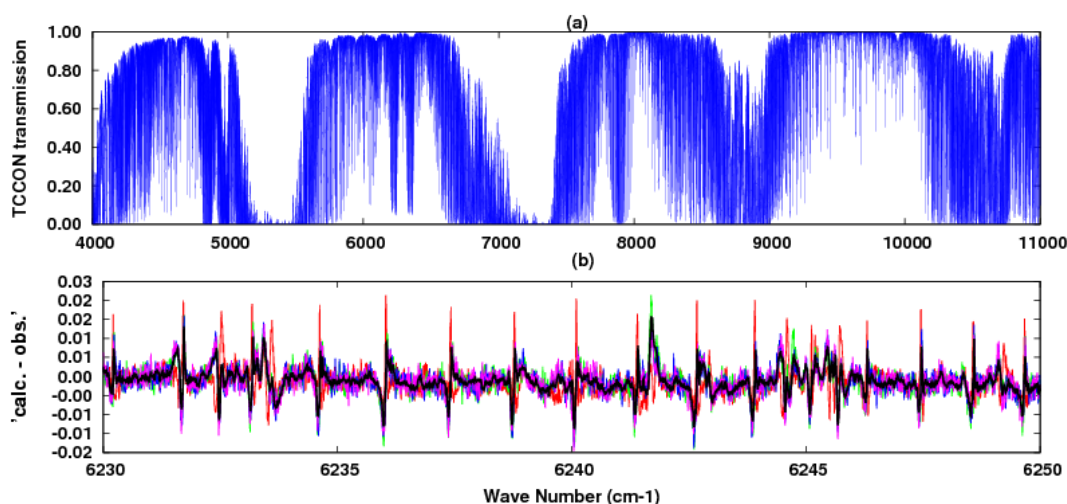
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FIG. 5. (a) TCCON transmission spectrum averaged over 465 spectra acquired between May 2008 and April 2011 at Parkfalls (United States). (b) calculated-observed residuals for 5 individual 'TCCON observation-atmospheric situations' pairs (colored lines) and averaged over 465 spectra (black line).

4. Application to the validation of spectroscopic parameters and radiative transfer modelling

4.1 Distinguishing HDO from its H_2O main isotope in the radiative transfer modelling and in GEISA-2015

This section gives an example of how the SPARTE chain has contributed to successively improve the forward radiative transfer modelling and consolidate the content of the GEISA spectroscopic databank. Since the launch of IASI onboard Metop-A, the careful monitoring of 'calc-obs' residuals, which is mandatory for successfully solving inverse radiative transfer problems, has revealed persistent large values of the residuals near the end of band 3 of IASI. In particular, in the 2170-2730 cm^{-1}, values as high as -1.5 K of the residuals were found (Fig. 6b). Based on the GEISA-2011 line list, this pronounced spectral feature was traced back to HDO, since most of the "H_2O" lines in this interval are actually HDO lines (Fig. 6c). Now, the 'calc-obs' residuals displayed in Fig. 6b were computed assuming the same concentration profile for each isotopologue fo H_2O. The origin of the large residuals could thus come from either an incoherent description of the water vapour atmospheric profile possibly, or a wrong description of the spectroscopic parameters in GEISA-2011, or a combination of both.

The HDO/H_2O ratio profile is commonly given in form of a δD value, which is the relative difference of the actual HDO/H_2O ratio to a standard HDO/H_2O ratio:

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$$\delta D = 1000 \times ([HD^{(16)}O] / [H_2^{(16)}O] / SMOW - 1),$$

where SMOW= $3.1152 \times 10-4$ (with SMOW: Standard Mean Ocean Water). Several measurements of δD (e.g. Schneider et al, 2010 and references herein) have been performed over the years, pointing out the importance of isotopes to assess the relative importance of moistening or dehydrating processes as well as the role that quantification of δD can play as markers for the source and transport of the water vapour as well as for the description of high atmospheric stable or unstable conditions. Based on these measurements of δD profiles, it has been decided to disentangle, in our simulations, the vertical profiles of HDO mixing ratios from the H_2O ones. A first test was performed using 3,000 clear sky collocations of observed and simulated spectra for 4 months (January, April, July, October) of each year of IASI observations from July 2007 to October 2011. This led to considerably reduce the 'calc-obs' residuals, and enabled a more confident way to validate the update of both molecules in the 2015 version of GEISA.
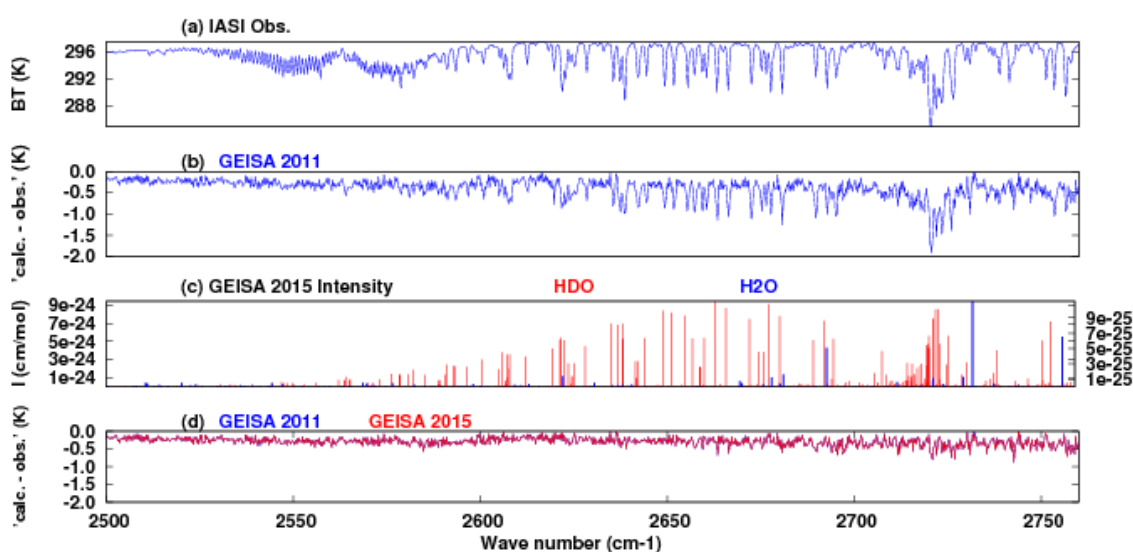

FIG. 6. Application of the SPARTE chain to IASI observation between 2500 and 2760 cm^{-1} for January 2008 (200 situations): (a) Averaged IASI observed spectrum (blue). (b) 'calculated-observed' residuals between IASI and 4A/OP for GEISA-2011 (blue), H_2O and HDO having the same concentration profile. (c) HDO (left Y-axis) and H_2O (right Y-axis) line intensities from GEISA-2015. (d) 'calculated-observed' residuals between IASI and 4A/OP for GEISA-2011 (blue) and GEISA-2015 (red) (HDO vertical profile modulated from H_2O profile by δD (Schneider et al., 2010)).

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Figure 6d shows the residuals obtained considering GEISA-2011 (blue) and GEISA-2015 (red). As compared to Figure 6b, the residuals are now close to 0 throughout the considered spectral region. Associated to the taking into account in the radiative transfer model of a variable $HD^{(16)}O/[H_2^{(16)}O$ δD value, the validation of the update of the spectroscopic parameters as well as the consideration of HDO as an independent molecule in both GEISA-2015 and 4A, has been confidently validated.

The residuals are quite similar between both versions of GEISA confirming an expected minor update of the database in this spectral range. In the section 4.2, we will see that in the near infrared, the update of the H_2O/HDO spectroscopic parameters can have a more important impact.

The remaining small spectral variations could be explained by the natural seasonal and spatial variability of δD, which was not taken into account in the simulations (information not available in the traditional radiosoundings nor in the reanalyses products), as well as by remaining problems in the spectroscopic parameters. Ultimately, the final verification could be achieved by introducing in the SPARTE chain real HDO profiles, coming for instance from measurement campaigns collocated in space and time with IASI.

4.2. Evaluation of line parameters using different spectroscopic databases as inputs

This section focuses on the use of the SPARTE chain for evaluating individual line parameters archived in spectroscopic databases. An error in a given line parameter usually induces a specific shape of the 'calc-obs' residuals. This is illustrated by Fig. 7 which shows the residual obtained when a few parameters of a given CH_4 line at 6026.2271 cm^{-1} (Fig. 7f) are shifted one after the other by: -1% in intensity (Fig. 7a), +0.002 cm^{-1} in line position (Fig. 7b), -10% in the air-broadened Half-Width at Half-Maximum (HWHM) (Fig. 7c), +0.002 cm^{-1} in pressure shift (Fig. 7d). These typical 'calculated-observed' values will of course vary with the line considered, but the shapes themselves will remain. Now, most of the time, uncertainties can simultaneously affect several parameters of the same line. Figure 7e shows the residual obtained when an error is made in both the line position (0.002 cm^{-1}) and the HWHM (-10%). These typical residual shapes are used in the following to identify the parameters which are the best candidates for explaining spectral artefacts in radiative transfer simulations.
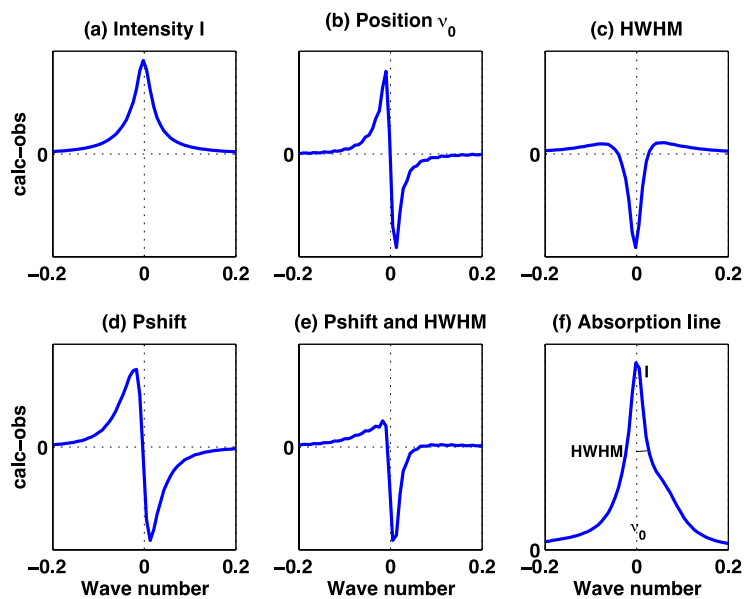
FIG 7. Typical transmission sensitivities of the 'calculated-observed' residuals to variation of several parameters of the CH_4 line at 6026.2271 cm^{-1}: (a) -1% in line intensity ; (b); +0.002 cm^{-1} in line position (c) -10% in HWHM; (d) +0.002 cm^{-1} in pressure shift; (e) +0.002 cm^{-1} of the position and -10% of the HWHM. (f) Illustration of the name convention of the main parameters used.

In the following, we focus on the contribution of the SPARTE chain to the GEISA-2015 update of CH_4 and H_2O in the short-wave part of the spectrum. Figure 8 summarizes the comparisons of the 'calculated-observed' residuals between TCCON and 4A/OP when three different spectroscopic databases are used: GEISA-2011 (blue), HITRAN-2012 (black) and GEISA 2015 (red). Figure 8a shows the spectrum of atmospheric transmission measured by TCCON in the region 6026-6032 cm^{-1}. The corresponding residuals (differences of transmissions) are shown in Fig. 8b.

Over this spectral range, three mains features can be observed in Fig. 8b around 6026.2, 6029.7 and 6031.9 cm^{-1}, with pronounced differences between the residuals obtained with the 3 databases. These differences in the residuals stem directly from the differences in the values of several line parameters of CH_4 and H_2O archived in each database. To ease the interpretation of these differences, the location and intensities of H_2O and CH_4 absorption lines absorbing in this region are shown in Fig. 8c. Values of the major spectral line parameters of both gases found in GEISA-2011, GEISA-2015 and HITRAN-2012 are given in Table 1. Each parameter is listed with the number of digits given by the providers.

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Around 6031.9 cm^{-1}, the differences between the residuals come from the line parameters of an H$_2$O line. In this case, the worst residual is obtained with HITRAN-2012 for which the shape of the residual is similar to the one given in Fig. 7c, which seems to indicate that the spectroscopic parameter at the origin of this feature is HWHM. This is confirmed by Table 1 (lines 7 to 9): the HWMH value stored in HITRAN for this line differs significantly from the value archived in GEISA-2011 or even from the updated value in GEISA-2015. It is also quite different from the usual value of HWHM for similar water vapour lines.

Another parameter to consider here is the pressure shift. As seen in Table 1, the value of this parameter may largely differ between the three databases. While for GEISA-2011, the value was not given (set to zero), a factor of two was observed between HITRAN-2012 and GEISA-2015.

This case highlights the difficulty that has to be faced when dealing with complex line parameters. Indeed, contributing laboratories usually provide the database management teams with 4 parameters: position, intensity and ground level energy. For most of the other parameters, such as the HWHM value of this H$_2$O line, values have to be filled in, in order to allow radiative transfer modelling (see Jacquinet et al, 2016, this issue). This example also illustrates the interest of having complementary updates not done at the same time for both databases in order to evaluate the choices made by each management team. In the case of this H$_2$O absorption line, the SPARTE chain clearly validates the choice made in GEISA.

Close to the same line, it can be seen in Fig. 8b that an issue still remains above 6032 cm^{-1} for all databases, with residuals of ~0.015 K that still need to be studied.

Between 6029.4 and 6030.0 cm^{-1}, absorption by CH$_4$ or H$_2$O is weak (Fig. 8a, transmission close to 1). The corresponding 'calculated-observed residual' (Fig. 8b) is very close to zero, but we can clearly see a different feature between GEISA 2011 on the one hand, and both HITRAN-2012 and GEISA-2015, on the other. For this spectral range, there was no line in the GEISA-2011 database. In both HITRAN-2012 and GEISA-2015 updates, 15 lines with small intensities (between 10e-24 to 10e-27) have been added compared to previous versions (five of the strongest lines shown in Table 1). Even if the residuals are close (even smaller) to the instrument noise (estimated to 0.0015 in transmission for this spectral range), significant improvement of the residuals is seen, which validates the quality of the new lines added. It can be noticed that for these absorption lines, both HITRAN-2012 and GEISA-2015 have globally used the same update.

Around 6026.2 cm^{-1}, the strongest 'calculated-observed' residuals are found for both GEISA-2011 and HITRAN-2012 (Fig. 8b). In that case, several parameters could be involved, as the temperature dependence "n" which is quite different. After studying the impact of most of them, we have concluded that the residuals are, in a first order, related to an improper characterization of both HWHM and pressure shift of a strong CH_4 line (more than 100 times more intense than the absorption line considered previously at 6029.7 cm^{-1}, see Table 1). With GEISA-2015, the updated values of these two parameters significantly reduce spectral residuals.

Database	Species	Position (cm^{-1})	Intensity (cm/atm)	Width (cm/cm^{-1})	E_0 (cm-1)	n	P shift (cm^{-1})
GEISA-2011		6026.227400	5.7210D-22	*0.0634*	10.4816	0.85	*-0.011113*
GEISA-2015	CH_4	6026.227100	5.7680D-22	*0.0650*	10.4817	0.68	-0.012052
HITRAN-2012		6026.226861	5.7680D-22	*0.0682*	10.4817	0.82	*-0.008500*
GEISA-2011		-	-	-	-	-	-
GEISA-2015		6029.557600	4.7340D-24	0.066	62.8758	0.73	-0.012059
		6029.655800	2.0490D-24	0.066	62.8768	0.73	-0.012059
		6029.832500	1.3560D-24	0.060	999.9999	0.85	-0.012060
		6029.845200	4.9590D-24	0.060	662.1400	0.85	-0.012060
	CH_4	6029.873100	2.1220D-24	0.060	516.2800	0.85	-0.012060
HITRAN-2012		6029.557600	4.7340D-24	0.0637	62.8758	0.73	-0.01090
		6029.655800	2.0490D-24	0.0637	62.8768	0.73	-0.01090
		6029.832500	1.3560D-24	0.0493	999.9999	0.62	-0.01200
		6029.845200	4.9590D-24	0.0523	662.1400	0.64	-0.01200
		6029.873100	2.1220D-24	0.0550	516.2800	0.65	-0.01200
GEISA-2011		6031.899390	7.3020D-26	0.0300	2042.31	0.36	-0.000000
GEISA-2015	H_2O	6031.899400	7.1300D-26	0.0380	2042.31	0.41	-0.011060
HITRAN-2012		6031.899390	7.3020D-26	*0.0078*	2042.31	0.36	-0.022000

Table 1. Line parameters contained in the 3 spectroscopic databases GEISA-2011, HITRAN-2012 and GEISA-2015 for 3 absorption lines located in the spectral range considered in Fig. 8. Parameters

written in italic-bold are those identified as responsible for strong departures in the 'calculated-observed' residuals (see text for details). The parameter E_0, n and Pshift (columns 6 to 8) corresponds respectively to the energy of the lower level energy, the temperature dependence coefficient of the air broadening half-width and the pressure shift.
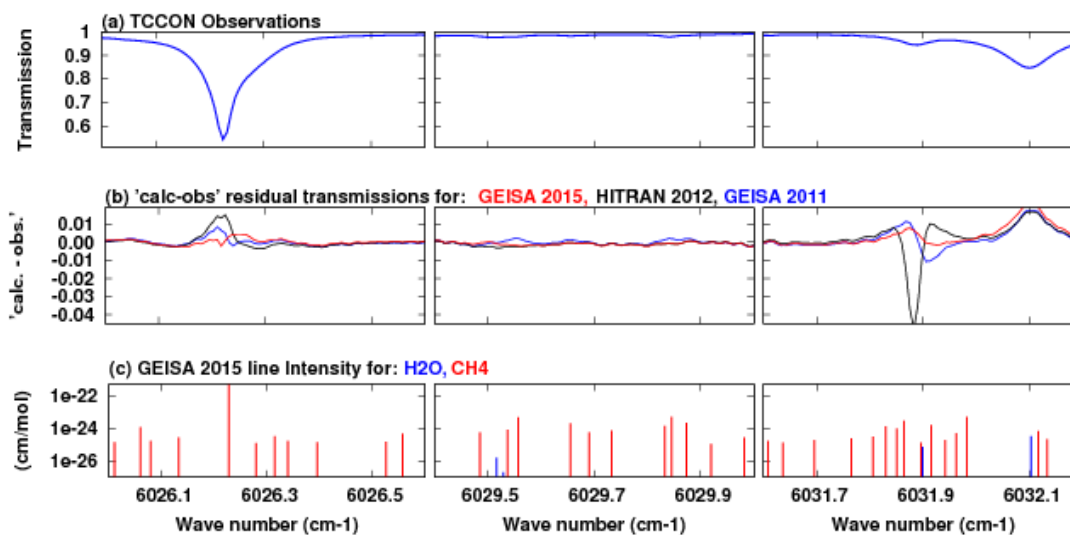

FIG 8. Comparison between GEISA-2011, GEISA-2015 and HITRAN-2012 using the SPARTE chain. (a) Transmission spectrum observed by TCCON, as input. (b) 'calculated-observed' residuals for GEISA-2015 (red), GEISA-2011 (blue) and HITRAN-2012 (black). (c) Intensities of CH_4 (red) and H_2O (blue) lines from GEISA-2015.

Overall, these 3 examples demonstrate that the comparison of the shape of the "calculated-observed" residuals provided by the SPARTE chain applied to various spectroscopic databases enables: (i) the identification of the parameters responsible for a significant deviation from 0 of the residuals; (ii) the validation of the update of each database. This is the case for the major line parameters, such as line position, intensity, or HWHM, but also for more complex parameters such as pressure shift. They also demonstrate that SPARTE is able to validate line intensities with radiance features that can be smaller than the instrumental noise (factor 2.5 often observed).

4.3 Evaluation of spectroscopic parameters in a Non Voigt configuration: example of CO_2 at 2 μm

In this section, we focus on the evaluation of updated line parameters for CO_2 at 2 μm. One of the difficulties in validating the spectroscopy of CO_2 is that, instead of assuming a classic Voigt line shape, the possible redistribution of the energy between some of the transitions, called 'line mixing', has to be taken into account. Most of the current RTMs, including 4A, now use the 'global' approach developed by the Laboratoire Interuniversitaire des Systèmes Atmosphériques (LISA) laboratory (Lamouroux et al.,2015; Tran et al., 2006; Tran et al., 2008, Tran et al., 2010). Over time, successive versions have been developed, from the simplest one which takes into account only the Q branches at 15μm and 4.3 μm, to the most complicated one which takes into account all the P, Q and R branches from the infrared to the near infrared. Until recently, each of these versions was based on a frozen version of the HITRAN database, rendering the validation of CO_2 updated lines in GEISA challenging. The last official release of the line mixing in 4A/OP was done with the coefficients calculated with the HITRAN-2008 databases (Lamouroux et al., 2010).

During the 2015 update of GEISA, for the first time, the modelling of the line-mixing has been fully adapted by LMD to work with any spectroscopic database available. The resulting code has been implemented in 4A and used in the SPARTE chain to validate the update of GEISA-2015 CO_2 line parameters.

This has enabled identifying a remaining issue with line parameters of some specific isotopologues of CO_2. Figure 9 shows the results of applying the SPARTE chain to the validation of GEISA-2015 in the 2 μm absorption band of CO_2. Figure 9a shows the averaged spectrum measured by TCCON. Figure 9b shows the residuals between TCCON spectra and calculated spectra by 4A/OP with GEISA-2015 as input in the range 4790-4930 cm^{-1}. Two kinds of variation of the residuals can be observed; they are plotted in different colors in Fig. 9b. Figure 10a and 10b show a zoom of these residuals over a few cm^{-1} for each mode. Between 4820 and 4880 cm^{-1}, the residuals are characterized by a kind of sinusoidal shape, with a slow increase and a fast decrease (Fig. 10a). Between 4790 and 4820 cm^{-1}, and then 4880 and 4930 cm^{-1}, the residuals are more characterized by a succession of well-pronounced peaks (Fig. 10b). These 2 behaviours are strongly linked to the isotopologues 626 ($^{16}O^{12}C^{16}O$) and 628 ($^{16}O^{12}C^{18}O$) of CO_2 as seen by comparing the shape of the residuals (Fig. 9b) with the positions of their corresponding lines (Fig. 9c).The shapes of the residuals thus clearly indicate some issues with the line parameters specific to each isotopologue.

In order to identify the parameters that could influence the shapes of the residuals, the SPARTE chain is now applied to the 4840-4845 cm^{-1} range, within the vibrational transition from the level ' 0 0 0 01' to the level ' 2 0 0 13' of CO_2 isotopologue 626. Comparing the residuals obtained with either the

previous version of the line mixing coefficients (black) or the new one obtained with GEISA-2015 (blue), Fig. 11b shows that the updated line parameters for CO_2 626 have led to a better agreement between the calculated and observed spectra in terms of averaged over the spectral range. The same has been done with HITRAN-2012 and no significant differences (less than 0.001 % in transmission) with GEISA-2015 have been observed in this spectral range. However, the shape of the residuals is the same for both versions of GEISA. This indicates a remaining issue with the line parameters of this isotopologue. Several tests have led us to the conclusion that a major part of the residual shape could be explained by an error in the position of the line (or pressure shift) of approximatively 0.002 cm^{-1}. For this line position, the estimated accuracy is around 0.001 cm^{-1} (Perevalov V., personal communication). Therefore, the source of the residual may be attributed to the pressure shift. Figure 11c shows the residual (less than 0.3-0.4% in transmission) when the pressure shift of CO_2 626 is shifted by -0.002 cm^{-1} from its original value. The resulting slope of the residual is lower and closer to zero. To make these results as exhaustive as possible, an extensive study, covering the entire spectral range 4790-4930 cm^{-1} presented in Fig. 9, will have to be performed in order to determine which values of the pressure shift minimize the residuals for each isotopologue. Final decision will be made in close collaboration with data providers.
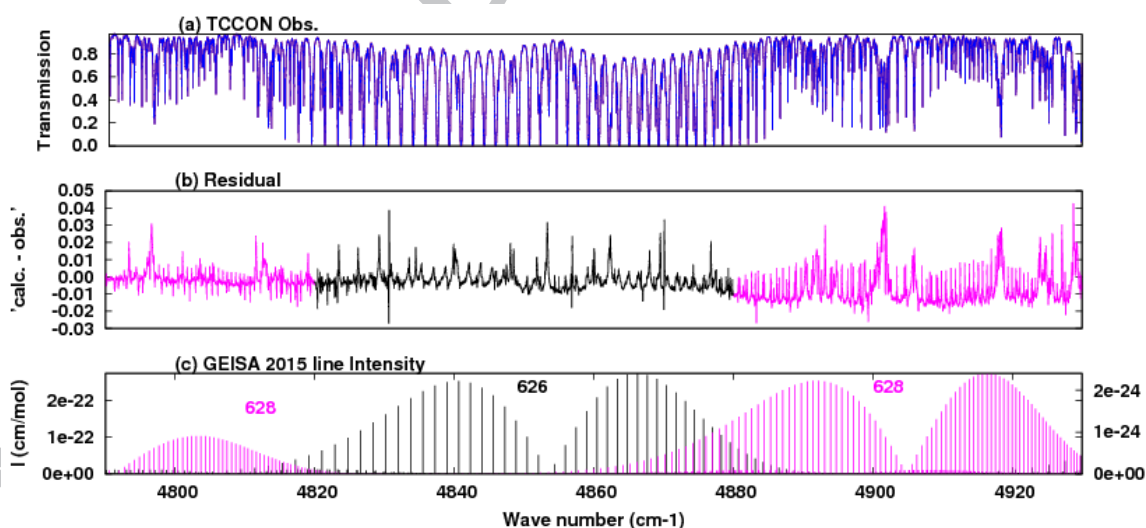

FIG 9. Application of the SPARTE chain to the 2μm band of CO_2. (a) Transmission spectrum observed by TCCON (red) and simulated with 4A/OP using GEISA-2015 as input (blue). (b) Corresponding 'calculated-observed' residuals. The colors highlight the isotopologue mainly responsible for the absorption: 626 (black) and 628 (pink). (c) Intensities of 626 (black, left Y-axis) and 628 (pink, right Y-axis) CO_2 lines from GEISA-2015.
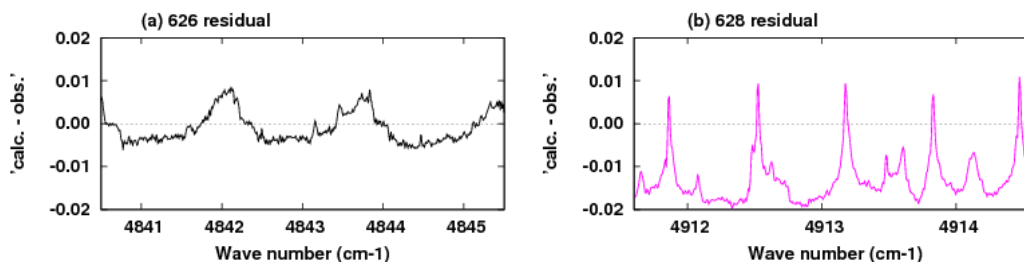
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FIG 10. Zoom over 4840.4 and 4845.5 cm-1 (a) and over 4911.5 and 4914.5 cm^{-1} (b) of Fig. 9b.
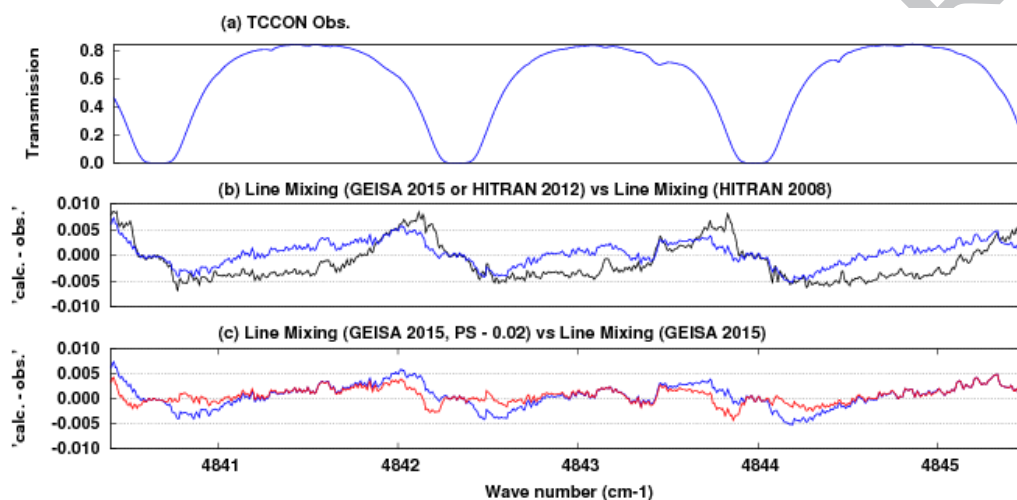

FIG 11. (a) Transmission spectrum observed by TCCON between 4840.4 and 4845.5 cm^{-1}. (b) 'calculated-observed' residuals with line mixing coefficients calculated for GEISA-2015 or HITRAN-2012 (blue) and for HITRAN-2008 (black); (c) 'calculated-observed' residuals with line mixing coefficients calculated for GEISA-2015 with few lines of the "2 0 0 13 --   0 0 0 01" transition of the 626 isotopologue of CO_2 of GEISA-2015 shifted (red) or not (blue, same as b)) by -0.002 cm^{-1} in pressure shift.

5. Conclusion and perspectives

The need to improve and consolidate the spectroscopic databases and the RTMs that use them remains and will remain a priority in order to: (i) fully exploit the high spectral resolution and radiometric accuracy of the new atmospheric instruments for an improved restitution of various geophysical variables; (ii) reanalyze the existing time series of previous satellites and establish fundamental climate data records. Concerning the validation of radiative transfer modelling, many intercomparison exercises are done on a regular basis at an international level to evaluate the performances of available models. In general, the conclusions are not easy to analyze for 2 main

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reasons: (i) the spectroscopic databases used as input are not the same. This has led to a repeated recommendation from several international working groups such as the RTSP-ITSC that RTMs should be able to use any spectroscopic database as inputs, at least GEISA and HITRAN; (ii) different sources and their uncertainties remain in the content of each database.

In this paper, we have described a quasi automatic chain, named SPARTE (Spectroscopic Parameters And Radiative Transfer Evaluation), which enables the evaluation of each step of the radiative transfer modelling, as well as the identification of remaining issues with line parameters archived in spectroscopic databases. This chain relies on the comparison between spectra simulated by the regularly validated operational radiative transfer algorithm 4A using a given spectroscopic database (such as GEISA or HITRAN) as input, and spectra measured by any ground-based, airborne or space borne instruments. For every single month of observations, hundreds of comparisons between simulated and observed clear-sky radiances (so called 'calculated-observed' residuals) in various conditions (land, sea, night, day, airmass, observation geometry) are performed, from which global and robust statistics, independent of each situation, can be derived. All the elements belonging to the SPARTE chain - the radiative transfer model (4A) and the various databases (ARSA, GEISA, TIGR) - are freely accessible or distributed upon simple request (see http://ara.abct.lmd.polytechnique.fr).

We have demonstrated that, thanks to the availability of spectrally well resolved instruments, characterized by both high spectral resolution and spectral and radiometric stabilities, it is possible to assess the validity of several spectroscopic line-parameters, including 'major' parameters (e.g. intensity, position, …), often measured, but not only (as air-broadened HWHM), and some "minor" ones, such as pressure shift. In particular, spectral signature of errors - related to the spectral line calculation (spectroscopic parameters, line mixing, etc), in some cases much lower than the noise of the instrument itself, can be detected. In a number of cases, the analysis of the residuals enables the identification of the required corrections to the parameters.

Thanks to the adaptation of the line-mixing code of (Lamouroux personal communication) for use with any spectroscopic database, we have shown that the SPARTE chain is also well suited to evaluate the spectroscopic parameters of molecules such as CO_2 for which interferences between the lines have to be taken into account before evaluating each individual parameter.

In the context of the 2015 update of GEISA (Jacquinet et al., 2016, this issue), the SPARTE chain has been used to evaluate the major revision of line-parameters of H_2O, CO_2 and CH_4, over a large spectral domain (between 645 and 11000 cm^{-1}). In particular, for H_2O, the use of the SPARTE chain has enabled: (i) to validate the positions and intensities of the lines, with overall significantly lower residuals for GEISA-2015 than for GEISA-2011; (ii) to validate the choice of complementary parameters such as pressure shift and air-broadened HWHM for the spectral range covered by IASI and TCCON; (iii) in comparison with HITRAN-2012, to generally validate GEISA-2015 for which the corresponding residuals are smaller for a large part of the spectral range. In particular, comparisons between residuals obtained with GEISA-2015 and HITRAN-2012 have highlighted a specific issue with some air-broadened HWHM values in the latter, that can be clearly identified on the 'calculated-observed' residuals; (iv) to validate the important update of H_2O and HDO, as well as the orientation chosen in GEISA-2015 to consider HDO as an independent molecule from the other isotopologues of H_2O (as it has been done in the past with CH_3D and C_2HD) and to bring the attention of radiative transfer modelers to the importance of using a pertinent representation of a pressure dependent behaviour of HDO/H2O concentrations. For CH_4 and CO_2, the SPARTE chain has allowed to select, among the various updates proposed by various contributors, the list of parameters that delivered most of the time the lowest residuals. The SPARTE chain has also revealed that some specific hot bands of CH_4 could have a lower precision than the other bands in position and/or in intensity (Jacquinet et al., 2016, this issue). In that case, the SPARTE chain has led us to keep the previous version of the transitions.

Using infrared sounders such as IASI gives the possibility to validate the spectroscopy (line parameters, continua, line-mixing) of the main absorbers (H_2O, CO_2, O_3, N_2O, CO and CH_4) as well as some trace gases characterized by smaller absorptions in the thermal infrared domain (HNO_3, OCS, CFCs, etc). In the future, the chain will be extended to the processing of other instruments, covering various spectral ranges. In particular, owing to the geometry of the observations, the use of limb-viewing instruments should provide signatures of molecules characterized by lower signatures (OCS, HCl) and allow the study of the pressure-temperature dependence of the parameters. The availability of modelling line-mixing effects for any spectroscopic database will also enable the study of a large number of molecules such as O_2 for which the actual line mixing modelling shows large residuals in the 0.76 µm band, which is commonly used by mission such as OCO-2 or GOSAT.

Acknowledgements

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Highlights

- A chain to validate line parameter lists for spectroscopic databases is proposed.

- It compares thousands of simulated and observed spectra.

- Averaging the residuals reveals well-defined signatures of spectroscopic issues.

Graphical abstract