Assessing Spectroscopic Parameter Archives for the Second Generation Vertical Sounders Radiance Simulation: Illustration through the GEISA/IASI Database

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Introduction

Line-by-line compilations of spectroscopic parameters are used for a vast array of applications and especially for terrestrial atmospheric remote sensing. Related with the radiance simulation for actual or near future atmospheric sounders, reviews of the current state and recent developments of public spectroscopic databases, such as GEISA-03 (Jacquinet-Husson et al. 2003, 2005b), HITRAN-04 (Rothman et al. 2005), MIPAS (Flaud et al. 2003), etc…., are continually made.
Actually, the performance of instruments like AIRS (*http://www.airs.jpl.nasa.gov*), in the USA, and IASI (*http://earth-sciences.cnes.fr/IASI/*) in Europe**,** which have a better vertical resolution and accuracy, compared to the presently existing satellite infrared vertical sounders, is directly related to the quality of the spectroscopic parameters of the optically active gases, since these are essential input in the forward models used to simulate recorded radiance spectra. Consequently, a strong demand exists for a highly comprehensive, well validated, efficiently operational, and desirably interactive computer-based spectroscopic database. In order to meet demands such as these, the ARA group at LMD (Chédin et al. 1982, Husson et al. 1992; 1994, Jacquinet-Husson et al. 1999) has been engaged during the past three decades in the development of the GEISA database.

The purpose of this paper is to present selected results of a critical assessment, in terms of spectroscopic line parameters archive, of GEISA and HITRAN, in the context of comparisons between recorded and calculated experimental spectra.

GEISA-03 and GEISA/IASI-03 Overview

GEISA, a computer-accessible spectroscopic database, was designed to facilitate accurate and fast forward, calculations of atmospheric radiative transfer using a line-by-line and (atmospheric) layer-by-layer approach. This effort has proven to be beneficial to the atmospheric scientific community participating in direct and inverse radiative transfer studies. In its 2003 edition, GEISA-03 is comprised of the following three independent sub-databases devoted, respectively, to:

- ***infrared spectral line parameters,*** related with 42 molecules (98 isotopic species), corresponding to 1,668,371 entries in the spectral range from 10^{-6} to 35,877 cm^{-1}. A summary of GEISA-03 line parameters sub-database content is given in Table 1 (portioned in two parts), where the archived molecular species are listed in the first columns with the number of associated line transitions in the second columns.

Table 1: GEISA-03 line transition parameters sub-database content summary

MOLECULE	# TRANSITIONS	MOLECULE	# TRANSITIONS
H2O	58726	HF	107
CO2	76826	HCl	533
O3	319248	HBr	1294
N2O	26681	HI	806
CO	13515	ClO	7230
CH4	216196	OCS	24922
O2	6290	H2CO	2701
NO	99123	C2H6	14981
SO2	38853	CH3D	35518
NO2	104224	C2H2	3115
NH3	29082	C2H4	12978
PH3	11740	GeH4	824
HNO3	171504	HCN	2550
OH	42866	C3H8	8983
42 MOLECULES		96 ISOTOPIC SPECIES	
SPECTRAL RANGE: 0 – 35,877 CM^{-1}		TOTAL # TRANSITIONS : 1,668,371	

- ***absorption cross-sections,*** in the spectral range of the infrared (32 molecular species, mainly CFC's) and of the UV/Visible (11 molecular species). A summary of the GEISA-03 database on UV/Visible absorption cross-sections is given in Table 2. The molecular species names are listed in columns 1. In the three following columns are the experimental conditions associated with the data files, i.e.: the spectral coverage (nm), in column 2; the overall temperature range (K), in column 3; the total pressure range (hPa), in column 4. For each file, the number of associated temperature-pressure sets is in column 5 and the related references and number of entries, in columns 6 and 7, respectively.
- ***microphysical and optical properties of basic atmospheric aerosols***.

The GEISA/IASI database was derived from GEISA, as described in Jacquinet-Husson et al. (1998). It has been created for the purpose of assessing the capability of IASI, within the ISSWG, in the framework of the EPS preparation of CNES/EUMETSAT. The assessment will be done by simulating either high-resolution radiance spectra or experimental data, or both, as the situation demands. In order to bring about an improvement in our knowledge of spectroscopic parameters and to ensure the continuous upgrade and maintenance of GEISA/IASI during the fifteen years of operation of the IASI instrument, EUMETSAT and CNES have created the GIDSC. EUMETSAT is planning to implement GEISA/IASI into the ground segment of EPS. There have been three versions of GEISA/IASI since its creation. GEISA/IASI, in its current edition GEISA/IASI-03 (Jacquinet-Husson et al. 2005a), is both an extract, which has been devised to suit the needs of IASI or AIRS within the 599-3001 cm^{-1} spectral range from a more extensive GEISA database and a continuing update of GEISA-03, with a similar structure, including three sub-databases related with:

Table2: GEISA-03 UV/VIS cross-sections sub-database Overview

Molecule	Spectral range (nm)	Temperature range (K)	Pressure range (hPa)	# T,P sets	References	# entries
O3	231 - 794	202 - 293	undef	5	BU	16,500
	230 - 830	203 - 293	100 - 1000	10		200,000
	230 - 1070	203 - 293	50 – 900	5		21,000
NO2	231 - 794	221 - 293	undef	4		13,600
	230 - 830	223 - 293	100 - 1000	10		1,358,000
	230 - 900	203 - 293	100 - 150	5		16,800
	238 - 1000	294	0	1	BIRA/IASB	13,500
	238 - 1000	220 - 294	0 – 1.33	13		225,000
	385 - 725	220 - 294	0 - 1013.23	27		9,855,000
BrO	300- 385	203 - 298	undef	5	BU	25,000
OClO	325 - 425	213 - 293	undef	10		132,500
	290 - 460	293	900	1		1,200
OBrO	385 - 616	298	undef	1		40,500
SO2	240 - 395	203 - 293	100	5		7,000
	250 - 330	294	undef	2	BIRA/IASB	9,600
H2CO	250 - 400	293	4.10	1	BU	1,400
O2	235 - 800	203 - 293	900 - 945	6		5,990
	627 - 1290	223 - 290	100 - 1000	24	RAL/MSF	390,360
	246 - 266	296	200	5		517,495
	240 - 330	287 - 289	undef	2	BIRA/IASB	15,560
O2 + N2	1000 - 1333	132 - 295	668 - 1009	12	RAL/MSF	50,800
O4	333 - 666	294	undef	1	BIRA/IASB	12,200
	454 - 667	223 - 294	1000	8	RAL/MSF	29,040
CS2	290 - 350	294	undef	1	BIRA/IASB	908

BU: Bremen University *Molecular Spectroscopy and Chemical Kinetics Group* **J. Orphal**
BIRA/IASB: Belgium Institute for Space Aeronomy **A.C. Vandaele**
RAL/MSF: Rutherford Appleton Laboratory Molecular Spectroscopy Facility Spectroscopy Group **K.M. Smith**

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- ***infrared spectral line parameters***, of 14 molecules (53 isotopic species): H2O, CO2, O3, N2O, CO, CH4, O2, NO, SO2, NO2, HNO3, OCS, C2H2, N2. A summary of GEISA/IASI-03 line parameters sub-database content is given in Table 3. The items listed for each molecular species given in column 1, are: the molecule and its various isotopes identification codes (defined for the GEISA management software), in columns 2 and 3 respectively, and the number of associated line transitions, in column 4 (702,550 lines in total).
- ***IR absorption cross-sections*** (mainly CFC's) for 6 molecular species: CFC-11, CFC-12, CFC-14, CCl4, N2O5, HCFC-22
- ***microphysical and optical properties of basic atmospheric aerosol components*** (similar with the GEISA-03 archive)

Table 3: GEISA/IASI-03 line transition parameters sub-database content summary

MOLECULE	CODE	ISOTOPES	# TRANSITIONS
H2O	1	161-162-171-181-182	13278
CO2	2	626-627-628-636-637-638-728-828-838	50840
O3	3	666-668-686-667-676	195102
N2O	4	446-447-448-456-546	18966
CO	5	26- 36- 28- 27- 38- 37	3674
CH4	6	211-311-212 *(ch3d)*	121281
O2	7	66- 67- 68	435
NO	8	46- 48- 56	29608
SO2	9	626-646	22301
NO2	10	646	71687
HNO3	13	146	152586
OCS	20	622-624-632-623-822-634-722	19768
C2H2	24	221-231	2904
N2	33	44	120

14 MOLECULES **51 ISOTOPIC SPECIES**

SPECTRAL RANGE: 599 – 3001 CM^{-1} **TOTAL # TRANSITIONS: 702,550**

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The parameters of each spectral line or molecular vibrational-rotational transition are stored in the new "format field standard" of GEISA and GEISA/IASI, with an extended number (from 16 to 30) of the selected line parameters, with the associated error estimation, as described in Jacquinet-Husson et al. 2005a. The spectroscopic parameters of importance for radiative transfer modeling, and used in the GEISA general management associated software are listed in Table 4, representing a part of the total file structure (9 format fields). These parameters, are: the wavenumber (A field; cm^{-1}) of the line associated with the rovibrational transition; the intensity of the line (B field; cm molecule^{-1} at 296K); the Lorentzian collision halfwidth (C field; cm^{-1}atm^{-1} at 296K); the energy of the lower level of the transition (D field; cm^{-1}); the transition quantum identifications for the lower and upper levels of the transition(E field); the temperature dependent coefficient n of the halfwidth (F field). Identification codes for isotope, molecule and GEISA data identification are in fields G-J, respectively.
Default values have been chosen for missing data values, i.e.: -1.0 cm^{-1} for the energy of the lower transition level, -0.9999 cm^{-1}atm^{-1} at 296K for the Lorentzian collision half-width and 0.75 for n coefficient.
All the archived data of GEISA and GEISA/IASI can be handled through a user friendly associated management software, which is posted on the ARA/LMD group web site at *http://ara.lmd.polytechnique.fr*

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Table 4: Nine first fields of the format for the line transition parameters in GEISA-03 (30 format fields in all)

Fortran format descriptor	**F12.6**	**D11.4**	**F6.4**	**F10.4**	**A36**	**F4.2**	**I3**	**I3**	**A3**
Field name	**A**	**B**	**C**	**D**	**E**	**F**	**G**	**I**	**J**

GEISA-03 and GEISA/IASI-03 line transition parameters update assessment

Updated molecules and related spectral intervals for the 2003 edition of GEISA and GEISA/IASI are presented in Tables 5a) (portioned in two parts) and 5b) respectively, where the archived molecular species are listed in the first columns with the corresponding spectral intervals in the second columns.

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Table 5a): Updated molecules and spectral intervals in GEISA-03

Molecule	Updated spectral intervals (cm^{-1})	Molecule	Updated spectral intervals (cm^{-1})
H2O	**500 – 2819** **9603 – 11399** **13184 – 25232**	**OH**	**29808 - 35877**
CO2	**436 - 2826**	**HBr**	**17 – 396** **2124 - 2790**
O3	**600 – 3391**	**HI**	**13 – 320** **1951 - 2403**
N2O	**872 – 1243**	**C2H6**	**2975 - 2978**
CH4	**0 – 6184**	**CH3D**	**0 – 6184**
O2	**7665 – 8064** **11484 - 15928**	**C2H2**	**605 - 3374**
NO	**1487 - 3799**	**HOCl**	**1179 - 1320**
NO2	**2719 – 3074**	**CH3Cl**	**1261 - 1646**
NH3	**0 - 5294**	**COF2**	**1857 - 2001**
PH3	**18 - 2479**	**HO2**	**0 - 908**

Table 5b): Updated molecules and spectral intervals in GEISA/IASI-03

Molecule	Updated spectral intervals (cm^{-1})
H2O(Toth)	**599.681 - 2819.848**
H2O (RAL)	**700.032 - 1299.980**
CO2	**599.007 - 2826.650**
O3	**600.179 - 3000.971**
N2O	**872.399 - 1243.755**
CH4	**855.753 - 3000.997**
NO	**1487.366 - 2188.447**
NO2	**2719.056 - 2992.323**
C2H2	**604.774 - 2254.963**

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Three updated molecules (H2O, O3 and CH4) have been selected, in the following, giving examples on how critical evaluations of the impact of newly archived spectroscopic parameters on IASI direct sounding modeling, may be made. The simulation, in the spectral range 645-2760 cm^{-1}, of an IASI spectrum (brightness temperature (K), in green) is shown on figures1a), 1b), 1c), where H2O

(1a)), O3 (1b)) and CH4 (1c)) are highlighted in red. The computation has been processed with the current 4A-2000 version of the ARA/LMD 4A fast line-by-line model (Scott and Chédin 1981; Tournier et al. 1995), an advanced version of the nominal line-by-line code STRANSAC (Scott 1974), relying on GEISA/IASI-03. A non apodized IASI apparatus function (Phulpin/CNES 2003) has been used, with a spectral resolution of 0.25 cm^{-1}.

- **H2O GEISA/IASI-03 update assessment**

Two different series of updated spectroscopic parameters of H2O are catalogued. They stem from the measurements by Toth (2000) and from the EUMETSAT/RAL archive (Stewart 2003). Toth's data enter into the entire database, while the EUMETSAT/RAL data appear in a supplemental list. A choice cannot been made as to a final selection until the results of validation campaigns are used to study the respective accuracies in a detailed manner

Fig. 2 details the H2O line coverage of the updated GEISA/IASI-2003 content. There are two panels, one a) corresponding to Toth's data and the other one b) to EUMETSAT/RAL data. It has to be noticed that the spectral coverage of Toth's updated data is larger than the EUMETSAT/RAL one. On each panel, the Logarithms of the intensities of the updated or added lines, averaged over 10 cm^{-1} intervals, are plotted versus the total GEISA/IASI spectral range (from 599 to 3001 cm^{-1}).
Illustrations of the differences, in spectroscopic parameter values, resulting from the Toth's (TOT) or EUMETSAT/RAL (RAL) choice for H2O update in GEISA/IASI-2003 (GS) are presented in Fig.3a) to Fig.3c):

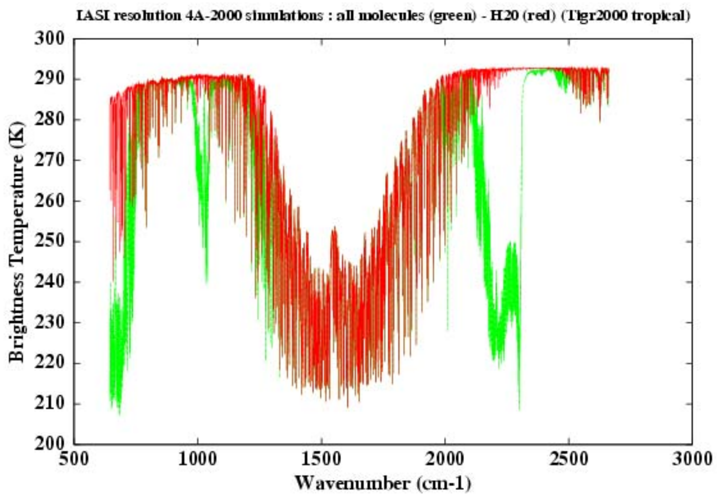
- Fig 3a) is split in two panels, i. e.: the left hand panel is related to the line air collision half-widths (HW) differences (HWdif) and the right one to the line intensities (I) differences (Idif). These differences, in percent, using GEISA/IASI-03 archived data value (GS+TOT) as reference, have been evaluated with the formulas:
  HWdif (%) = HW(RAL)-HW(GS+TOT)/HW(GS+TOT)
  Idif (%) = I(RAL)-I(GS+TOT)/I(GS+TOT)
  For each parameter, the % differences (Y axis) are expanded from 700 to 1300 cm^{-1} (X-axis)
  It is clear from this figure that important differences exist between the two available sources of data: up to 200% for the HW and over 100% for the absorption line intensities.

The objective of figures 3b) and 3c) series is to illustrate the impact of differences in H2O spectroscopic data on forward modeling computations. Simulations of IASI spectrum are obtained with 4A-2000, using, either Toth's (in green) or RAL (in red) H2O spectroscopic parameters, in GEISA/IASI-03.

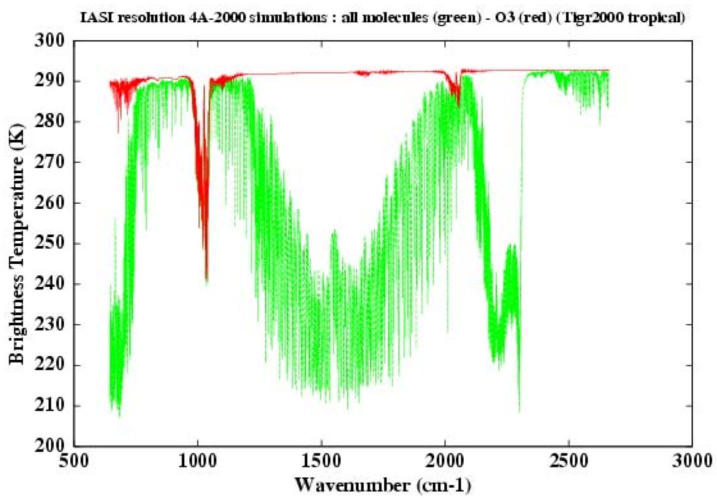
- Fig. 3b) presents comparisons of IASI spectrum simulations with upwelling spectral measurements made by the HIS (Smith et al. 1983) instrument (similarity in viewing geometry with IASI) during the CAMEX-1 (Griffin et al. 1994) field campaign, on 29 September 1993, from an altitude of approximately 20 km, on board of the ER-2 aircraft. The data were prepared and distributed by CIMSS (Knuteson et al. 2000).
  On the same left hand graphic, are plotted, the two HIS brightness temperatures as simulated by 4A-2000, in the spectral interval 850-900 cm^{-1} (part of IASI band-1). The right hand graphic shows the brightness temperature differences between the HIS measurements and 4A-2000 modeling, using the same color codes, as previously, to identify the origin of H2O data.

These differences are expanding from -1.2K to 0.6 K. The largest discrepancies appear around, 850, 855, 871 and 887 cm^{-1} and present some more important spikes in the case of RAL data (especially at 887 cm^{-1})

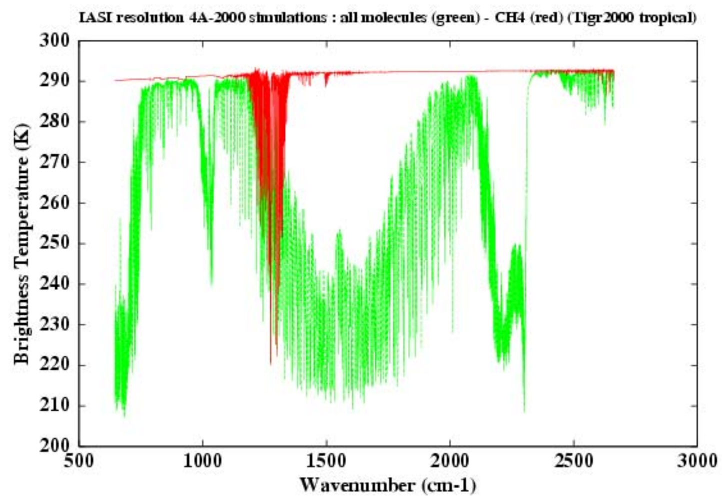
- Fig. 3c) shows the differences obtained in IASI brightness temperatures, for a US Standard Atmosphere tropical situation (1976) 4A-2000 modeling. Similar different H2O alternative data, as in Fig. 3b), have been used with similar colors in the simulation plots of the left hand figure. On the right hand figure is the graphic (in red color) of the brightness temperature differences related with the use of Toth's or RAL data in the spectroscopic database. In this figure, important difference spikes appear in the 1300 cm^{-1} and in the 1700-2000 cm^{-1} spectral regions. These differences may be as large as nearly 2K.



a)



b)



c)

Fig. 1: IASI resolution 4A-2000 simulation

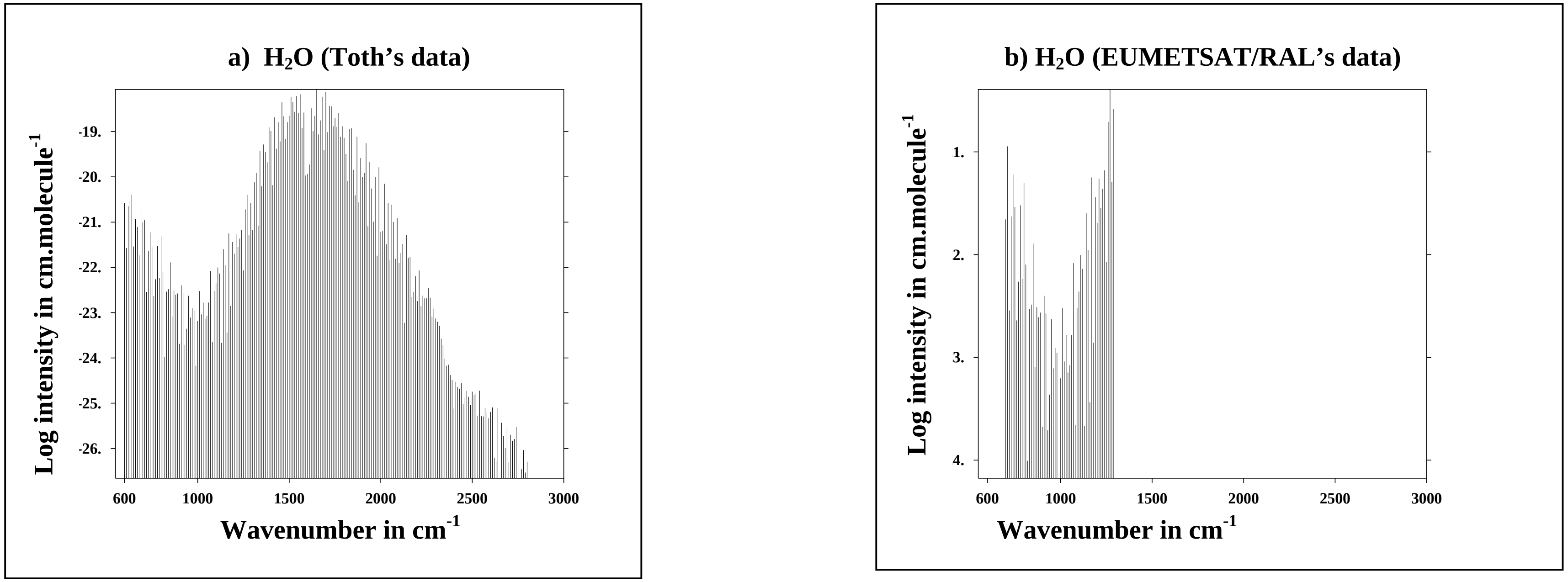
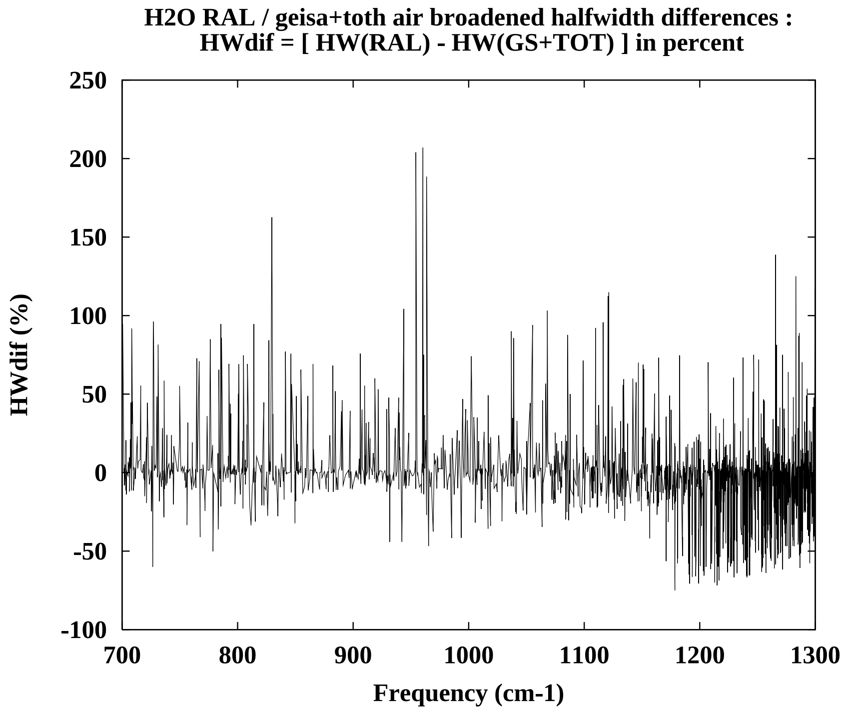


Fig. 2: H2O GEISA/IASI-03 update and alternative archive



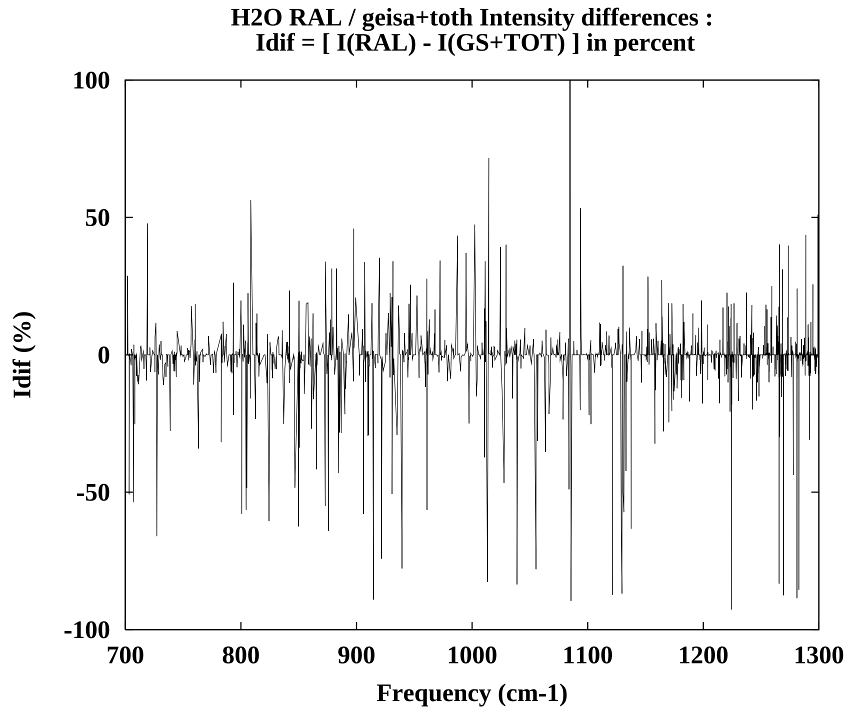


Fig. 3a): Impacts of differences in choices for H2O update in GEISA/IASI-03: left hand figure half-width differences (in %); right hand figure, intensity differences (in %)

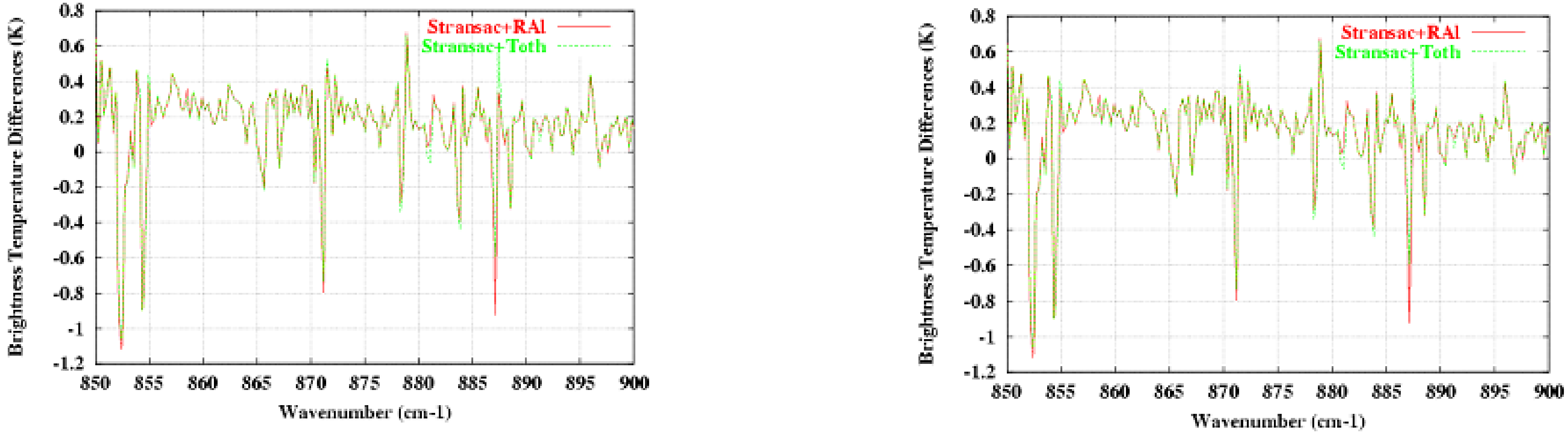


Fig. 3b): CAMEX(HIS) 29/09/93 campaign. Differences (ΔTB K) in 4A-2000 simulations using RAL or Toth's H_2O spectroscopy in GEISA/IASI-03

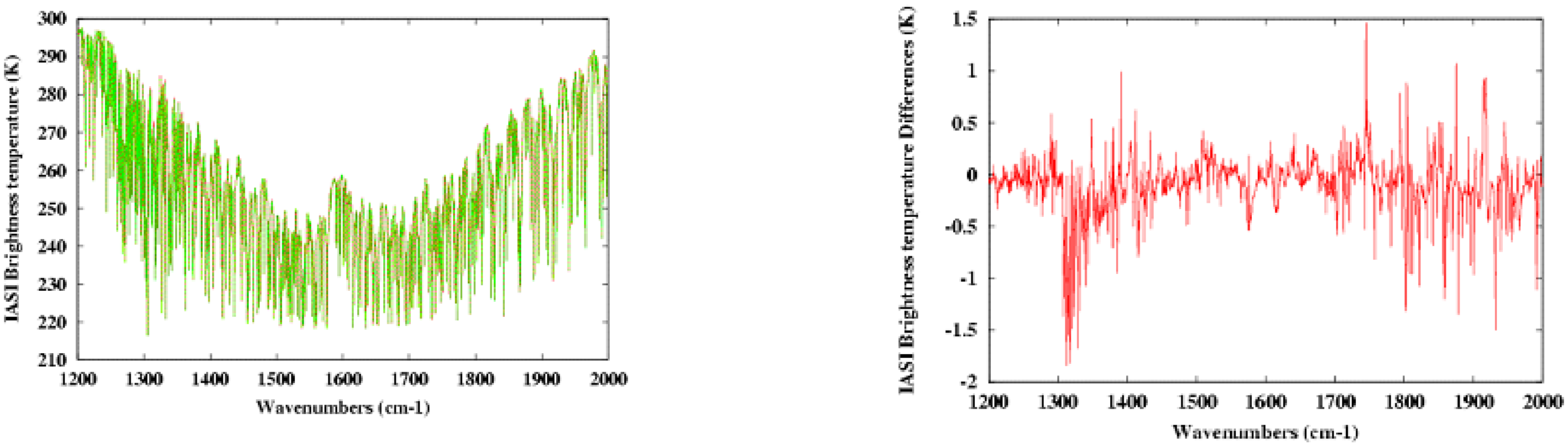


Fig. 3c): Tropical U.S. Standard Atmosphere 1976. Differences (ΔTB K) in 4A-2000 simulations using RAL or Toth's H_2O spectroscopy in GEISA/IASI-03

- **O_3 GEISA/IASI-03 update assessment**

An example of the impact of differences in O3 spectroscopic parameters, on IMG total column retrieval, is presented in Fig. 4 (Coheur et al. 2005). Four different databases, i.e.: GEISA-97 (Jacquinet-Husson et al. 1999), GEISA-03, MIPAS, HITRAN-2000 (Rothman et al. 2003), have been used in the evaluation of the errors between the observations and the theoretical calculations (obs.-calc., on the Y-axis) in the spectral range (980-1100 cm^{-1}, on the X-axis). Depending on the database, the RMS value varies from 1.34 10^{-7} to 5.45 10^{-8}, and the ozone total column value from 306.3 Dobson to 312.0 Dobson.

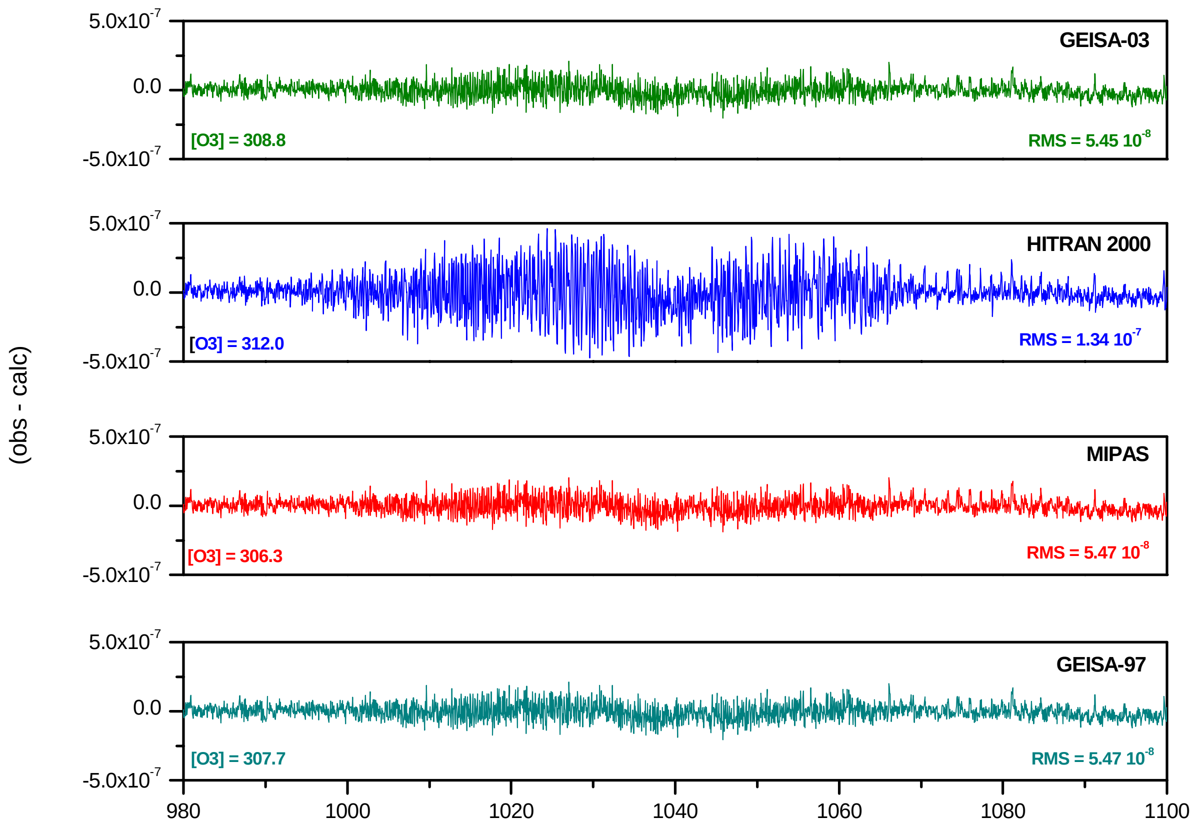


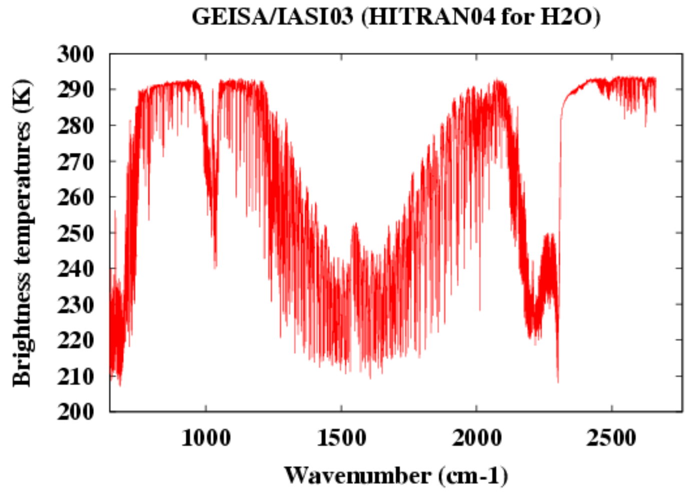
Fig.4: Spectroscopy impact on IMG O3 total column retrieval (Courtesy of P.F. Coheur –ULB)

Differences between GEISA-03 and HITRAN-04: Examples of impacts evaluation

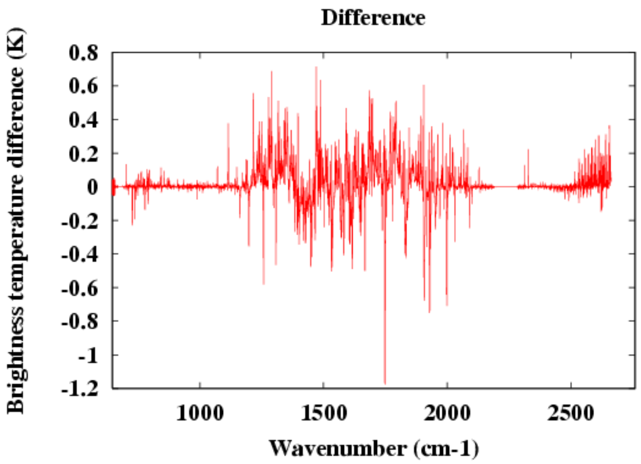
The two last editions of GEISA and HITRAN, described extensively in Jacquinet-Husson et al. (2003, 2005b) and in Rothman et al. (2005) exhibit differences related with the choices made for new or updated spectroscopic parameters (see related database descriptions for details). IASI sounding simulations have been made to evaluate impacts of differences between both database archives. Two molecular species, i.e.: H2O (mainly intensities and air broadened halfwidths differences) and CH4, have been retained. Computations were made using 4A-2000 in the case of a mean tropical atmospheric profile from the Thermodynamic Initial Guess Retrieval (TIGR) dataset (Chédin et al. (1985), Achard (1991), Chevallier et al. 1998), in its latest version (TIGR-2000).

Illustrations of the impact of these differences in IASI sounding modeling are presented in the following figures:

- Figs. 5 a) shows an IASI simulation (brightness temperatures (K)) as a function of the wavenumber (IASI spectral range) using the GEISA/IASI-03 database, where the H2O archive has been replaced by the HITRAN-04 one. Previous Fig.1a) shows a similar simulation with the original GEISA/IASI-03 H2O content. Related brightness temperature differences are on Fig. 5 b), showing spikes between nearly -1.2 and 0.8 K.



a)



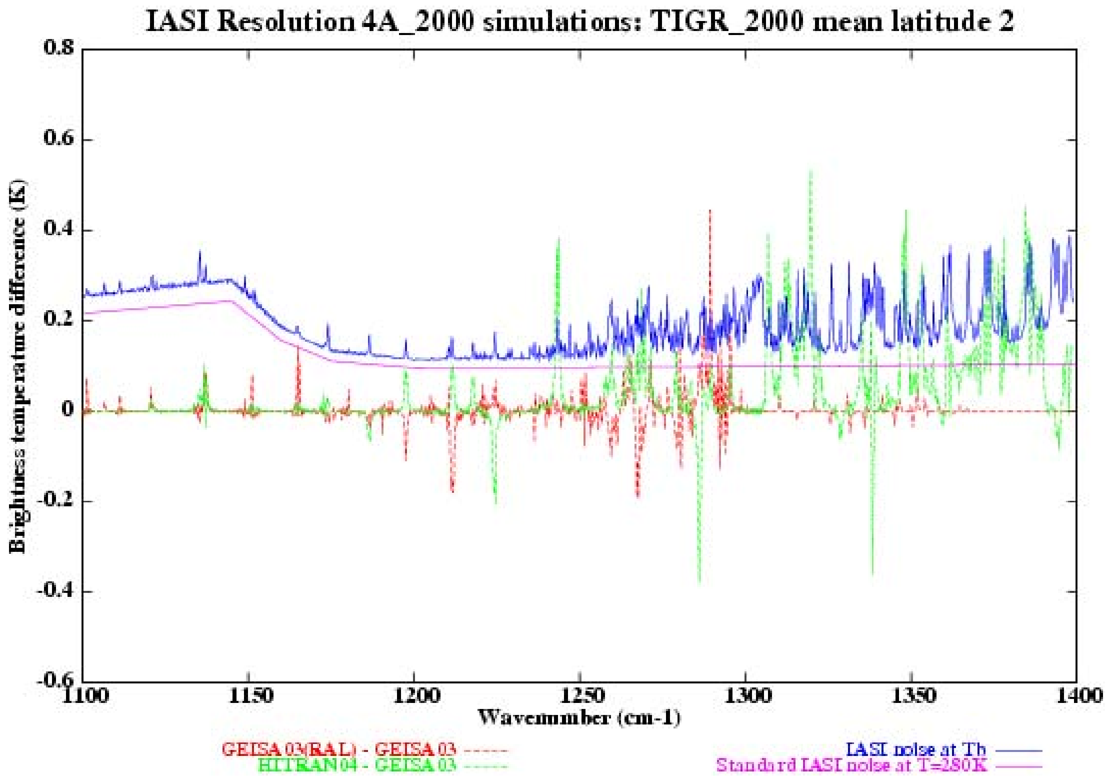
b)

Fig. 5: GEISA/IASI-03 and HITRAN-04 H2O archive differences and impact on H2O 4A-2000 IASI simulations

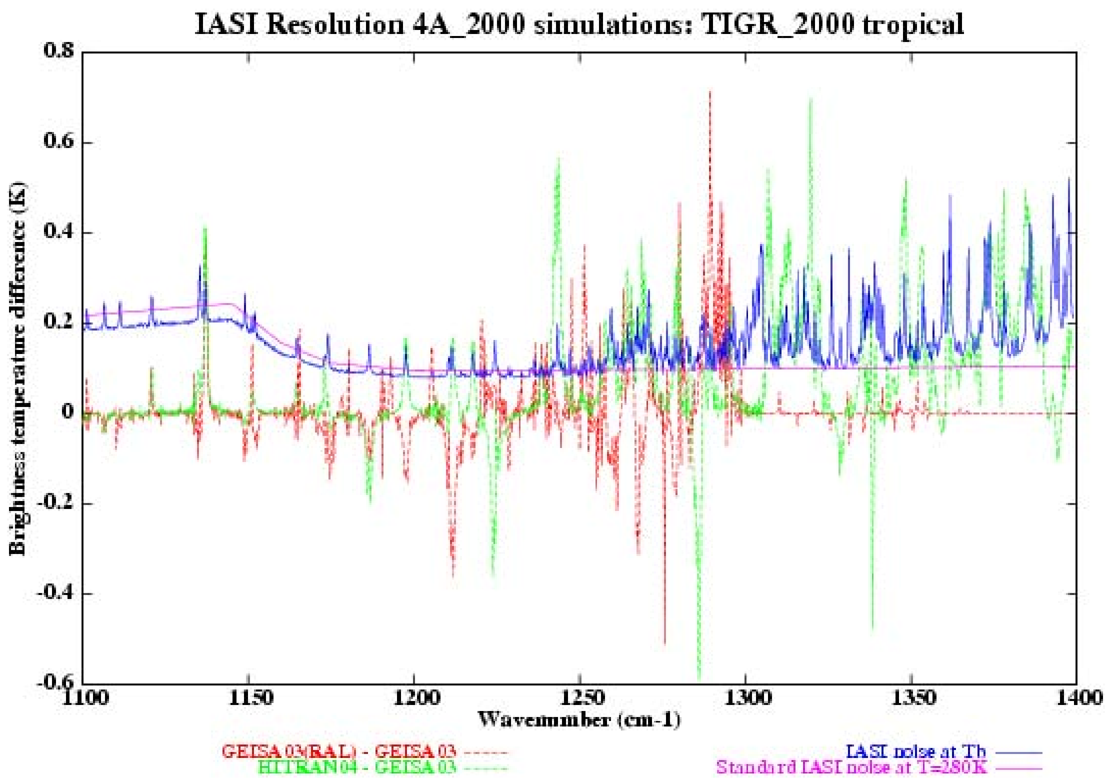
Figs. 6 a) and 6 b) present IASI 4A-2000 simulations brightness temperature differences (K), for two case studies related with the evaluation of the impacts of differences in spectroscopic databases archive, for H2O and CH4. The red curve corresponds to the differences corresponding, either to the choice of RAL (GEISA-03(RAL) id) or to the choice of Toth's H2O data (GEISA-03 id), in GEISA/IASI -03. The green curve corresponds to the differences in using either HITRAN-04 or GEISA-03 in 4A-2000 computation. Associated with these two curves are two IASI noise curves for two temperatures: the first one (in violet) corresponds to a temperature of 280K (CNES definition, Phulpin 2003) and the second one (in blue) to the noise converted to the real temperature of the scene. Along the X-axis, the spectral interval is 1100-1400 cm^{-1}, in both figures. Fig. 6 a) corresponds to a TIGR-2000 mean latitude-2 atmospheric situation and Fig. 6 b) to a tropical one.

- Fig. 7 shows similar comparisons when, either HITRAN-04 or GEISA-03 are used in 4A-2000 IASI resolution simulations, in the 2000-2180 cm^{-1} spectral region.

From these illustrations of IASI simulation comparisons for different archived spectroscopic parameters, it appears that the HITRAN-04 archive is significantly different from the RAL and from the Toth's/GEISA-03 one, for H2O, especially in the 1100 – 1400 cm^{-1} spectral region. Considering the actual values of IASI theoretical noise, we are confident that, when IASI real sounding data will be available, they will allow a validation in the choice of the spectroscopic parameters. For example, in the 1290 cm^{-1} spectral region, the differences between simulations, using either GEISA-03 with RAL H2O data or GEISA-03 with Toth's H2O data, show discrepancies, in brightness temperature values, of almost four times the IASI noise value.



a)



b)

Figs. 6a) b): IASI 4A-2000 Simulations using TIGR-2000 atmospheric mean latitude 2 a) and tropical b) profiles (1100-1400 cm^{-1} spectral region)

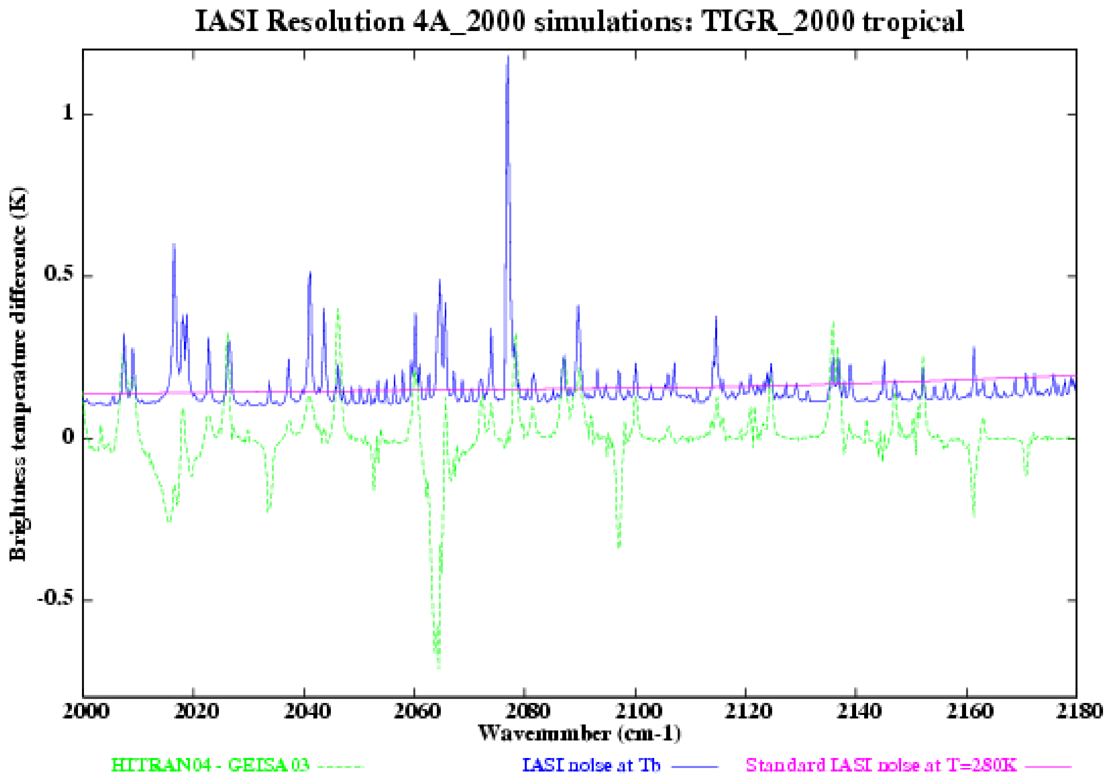


Fig. 7: IASI 4A-2000 Simulation using TIGR-2000 atmospheric tropical profiles (2000-2180 cm^{-1}spectral region)

GEISA and GEISA/IASI management software and database availability

The GEISA management software facilities are interfaced on the ARA/LMD group web site at: *http://ara.lmd.polytechnique.fr*. They are also accessible at the GEISA restricted free access ftp site:*http://ara.lmd.polytechnique.fr/ftpgeisa*. Previously, the potential user required a login and a password, at the ARA/LMD web site.

Acknowledgements

This research funded by CNES is supported by EUMETSAT in the frame of the EPS/METOP project. Both institutions are acknowledged for their encouragements.

List of Acronyms:

AIRS: Advanced InfraRed Sounder; **ARA:** Atmospheric Radiation Analysis; **4 A**: Automatized Atmospheric Absorption Atlas; **CAMEX-1**: first Convection And Moisture Experiment campaign; **CIMSS**: Cooperative Institute for Meteorological Satellite Studies; **CNES**: Centre National d'Etudes Spatiales, France; **EPS**: European Polar System; **EUMETSAT**: EUropean organization for the exploitation of METeorological SATellites; **GEISA**: Gestion et Etude des Informations Spectroscopiques Atmosphériques: Management and Study of Atmospheric Spectroscopic Information; **GIDSC**: GEISA/IASI Database Scientific Committee; **HIS**: High-resolution Interferometer Sounder; **HITRAN**: HIgh resolution TRANsmission spectroscopic database; **IASI**: Infrared Atmospheric Sounder Interferometer; **IMG**: Interferometric Monitor for Greenhouse gases; **ISSWG**: IASI Sounding Science Working Group; **LMD**: Laboratoire de Météorologie Dynamique ;**METOP**: METeorological OPerational Satellite ; **MIPAS**: Michelson Interferometer for Passive Atmospheric Sounding;**RAL**: Rutherford Appleton Laboratory;**RMS**: Root-Mean Square; **TIGR**: Thermodynamic Initial Guess Retrieval ; **ULB**: Université Libre de Bruxelles

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