

# High-Resolution Spectroscopic Database for the NASA Earth Observing System Program

## Annual Progress Report for NASA Grant NAG5-8420

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### 1. Introduction

The purpose of this project is to develop and enhance the HITRAN molecular spectroscopic database and associated software to support the observational programs of the Earth Observing System (EOS). In particular, the focus is on the EOS projects: the Atmospheric Infrared Sounder (AIRS), the High-Resolution Dynamics Limb Sounder (HIRDLS), Measurements of Pollution in the Troposphere (MOPITT), the Tropospheric Emission Spectrometer (TES), and the Stratospheric Aerosol and Gas Experiment (SAGE III). The data requirements of these programs in terms of spectroscopy are varied, but usually call for additional spectral parameters or improvements to existing molecular bands. In addition, cross-section data for heavier molecular species must be expanded and made amenable to modeling in remote sensing. The effort in the project also includes developing software and distribution to make access, manipulation, and use of HITRAN functional to the EOS program.

### 2. Data Acquisition and Data Improvement

A new edition of the HITRAN database and compilation was made available in December 2000. Major additions and improvements have been made for the line-by-line portion (HITRAN), the infrared cross-sections for heavy molecular species, and for the indices of refraction of aerosols.

#### 2.1 Line-by-line Parameters

For the line-by-line spectroscopic parameters, the most prominent improvements have been to water vapor, oxygen, nitric oxide, ammonia, nitric acid, hydroxyl radical, carbonyl sulfide, acetylene, hydrogen sulfide, and ethylene. Table 1 summarizes molecules that have been updated since the previous public edition of HITRAN.

The H<sub>2</sub>O parameters have been enhanced in two spectral regions, the region of the bending mode at 6  $\mu\text{m}$ , and the near-IR and visible region. The 6  $\mu\text{m}$  data come from the work of R.A. Toth of JPL.<sup>1</sup> Independent comparisons with observations by the TES team show that this update is a major improvement (C.P. Rinsland, NASA Langley Research Center and S.A. Clough, AER Inc, private communication). In the region above 8000  $\text{cm}^{-1}$ , an amalgamation of new data and analysis has been accomplished. L.R. Brown of JPL has measured and analyzed positions, intensities, broadening, and line-shifts in the 9650 to 11400  $\text{cm}^{-1}$  region.<sup>2</sup> We have processed these new data

with corrections and improvements recently made by the group at NASA Ames Research Center<sup>3</sup> that covers the overall region from 8000 to 25000 cm<sup>-1</sup>. The principal investigator of this project analyzed these latter data and removed duplicate lines, erroneous assignments, and made other corrections. This region is particularly important for resolving some of the discrepancies in atmospheric radiative budget modeling, for example, as well as serving specifically for **SAGE III**.

<b>Table 1. HITRAN Line-by-line Updates</b>	
<b>Molecule</b>	<b>Number of Transitions</b>
H <sub>2</sub> O	51930
N <sub>2</sub> O	26175
O <sub>2</sub>	6289
NO	17666
NH <sub>3</sub>	29084
HNO <sub>3</sub>	171504
OH	40055
HBr	1294
HI	806
OCS	19921
C <sub>2</sub> H <sub>2</sub>	1620
H <sub>2</sub> S	20788
HO <sub>2</sub>	38808
C <sub>2</sub> H <sub>4</sub>	12978

<b>Table 2. IR Cross-section Updates</b>	
<b>Molecule</b>	<b>Common Name</b>
C <sub>2</sub> F <sub>6</sub>	Hexafluoroethane
CCl <sub>4</sub>	Carbon tetrachloride
CCl <sub>3</sub> F	CFC-11
CCl <sub>2</sub> F <sub>2</sub>	CFC-12
CF <sub>4</sub>	CFC-14
CHCl <sub>2</sub> CF <sub>3</sub>	HCFC-123
CHClF <sub>2</sub>	HCFC-124
CH <sub>3</sub> CCl <sub>2</sub> F	HCFC-141b
CH <sub>3</sub> CClF <sub>2</sub>	HCFC-142b
CHClF <sub>2</sub>	HCFC-22
CHCl <sub>2</sub> CF <sub>2</sub> CF <sub>3</sub>	HCFC-225ca
CClF <sub>2</sub> CF <sub>2</sub> CHClF	HCFC-225cb
CHF <sub>2</sub> CF <sub>3</sub>	HFC-125
CHF <sub>2</sub> CHF <sub>2</sub>	HFC-134
CFH <sub>2</sub> CF <sub>3</sub>	HFC-134a
CF <sub>3</sub> CH <sub>3</sub>	HFC-143a
CH <sub>3</sub> CHF <sub>2</sub>	HFC-152a
CH <sub>2</sub> F <sub>2</sub>	HFC-32
SF <sub>6</sub>	Sulfur hexafluoride

For oxygen parameter updates, we had discussed the source<sup>4</sup> of the new atmospheric A-band data in the previous annual report. These data, in the 760-nm region, were particularly examined and updated with **SAGE III** in mind. Line positions, intensities, air- and self-broadening halfwidths, and pressure-shift parameters were obtained. The work confirmed that the older HITRAN intensities were good, but that the positions required updating (normally the reverse situation of many improvements). We have also updated the intensities of the infrared band of oxygen at 1.27  $\mu\text{m}$ . The new data come from laboratory work performed at NIST.<sup>5</sup> This band is very important to a number of NASA atmospheric remote-sensing programs, especially as it has been used as a benchmark for ozone retrievals. Previously, the intensities in this band relied on a very few measurements with a wide spread in values. We have compared the NIST values with independent laboratory measurements at the Rutherford Appleton Laboratory in the UK, and find excellent agreement.

Various improvements have been made for nitric oxide, NO. Most of the recent work comes from our collaboration with the group at the University of Paris.<sup>6</sup> There still remains work to be finished on obtaining a consistency of the halfwidths for the different bands.

A major update and revision of ammonia has been accomplished for this edition. The line list is a culmination of the effort of L.R. Brown.<sup>7</sup> Besides more consistency for the whole line list, the positions and intensities are significantly better for the 1200 to 2100 and 3000 to 3600  $\text{cm}^{-1}$  regions. New air-broadened widths have been applied throughout, from 0 to 5300  $\text{cm}^{-1}$ .

For nitric acid,  $\text{HNO}_3$ , there has been a significant improvement in the 11- $\mu\text{m}$  region, the atmospheric window. There has been an ongoing analysis<sup>8</sup> here that is complicated by the hot bands and resonances. The NASA Upper Atmospheric Research Satellite experiment CLAES (Cryogenic Limb Array Etalon Spectrometer) data, for example, show the impact of the improvement for this band region.

A major improvement for the intensities of the hydroxyl radical, OH, has been achieved by the work of Goldman et al.<sup>9</sup> We have revised all the bands on the HITRAN database as a result of their analysis. These data also serve for non-local thermodynamic equilibrium and for high-temperature applications.

Carbonyl sulfide, OCS, has had a major update throughout the infrared, due to the efforts of the group at the University of Louvain, Belgium (A. Fayt, private communication). The dynamic range and number of bands and isotopomers has increased substantially on this new edition of HITRAN.

For acetylene,  $\text{C}_2\text{H}_2$ , groups in both France and Belgium<sup>10</sup> have provided us with new results for fundamental, hot, and combination bands.

Hydrogen sulfide,  $\text{H}_2\text{S}$ , has had major revisions.<sup>11</sup> A whole new band system has been added to the HITRAN database, which now extends the database for  $\text{H}_2\text{S}$  to well beyond 3000  $\text{cm}^{-1}$ .

A new species has also been added to HITRAN: ethylene,  $\text{C}_2\text{H}_4$ . The extensive work for this species was the result of combined works in Belgium, France, and the University of Tennessee.<sup>12</sup>

Improvements and corrections have also been made for some of the hydrogen halides and nitrous oxide. We discuss later in this report some of the line-by-line data that are close to being ready for inclusion into HITRAN.

## 2.2 Cross-sections

The cross-section data compliment the HITRAN line-by-line data. They have been established to cover molecules whose spectra are so dense as to make a line-by-line transition representation either extremely difficult or impractical from the user's standpoint. Examples of these situations are heavy molecular species (generally of molecular weight more than ozone or nitric acid). The spectra of these gases are also complicated by low frequency fundamentals whose hot bands cannot be ignored. There are also significant pressure effects, which are implicitly represented by the pressure-temperature sets incorporated into the HITRAN compilation. The cross-sections are the result of direct experiment, not calculation as is often the case for the line-by-line parameters. Table 2 gives the species that have been added to the HITRAN compilation since the last release. It should be remarked that in terms of storage, the cross-sections now take up more space than the conventional HITRAN database. The update for this edition has enabled us to remove some older cross-sections that were found to be deficient; we have retained some older redundant, but nevertheless accurate sets and placed them into a supplemental directory.

It should be mentioned here that in the line-by-line portion, we have also relegated two molecules, SF<sub>6</sub> and ClONO<sub>2</sub>, also to a supplemental directory. These species are far better represented by their corresponding cross-section files, but may be of some use in special simulations, so we have retained them in this manner which avoids inappropriate use in atmospheric modeling.

There is also the addition to HITRAN of nitrogen dioxide, NO<sub>2</sub>, cross-sections in the ultraviolet. These data come from the group of M. Carleer in Belgium.<sup>13</sup>

## 2.3 Aerosol Indices of Refraction

A very extensive tabulation for various aerosol indices of refraction has been compiled and submitted to the HITRAN project by S. Massie of the National Center for Atmospheric Research. The sets were summarized in the last report, and a more complete table is given in both the HITRAN web-site and ftp-site.

## 3. HAWKS Software Development, Upgrades, and Distribution

The majority of the software planned for the new HITRAN edition was completed during this period. It has been written in the Java language for cross-platform applicability. The functionality of the previous HAWKS (HITRAN Atmospheric Workstation) software have been retained. The plotting capabilities have been improved and extended; it is now possible to plot the cross-section data in addition to HITRAN line-by-line parameters. The Ontar Corporation, under the direction of J. Schroeder, have performed this task, as well as been highly involved with all aspects of database management.

Figure 1 shows the file structure of the new HITRAN edition. The new edition, labeled v11.0, has been made available on an anonymous ftp-site located at the Harvard-Smithsonian Center for Astrophysics. We have asked users to first fill out the request form in the web-site, and then instructions for accessing the compilation are sent to them. We had finally exhausted our supply of CD-ROMs with the 1996 edition in November 2000. Well over 3000 copies had been distributed. Our surveys had shown that more and more users preferred a web distribution

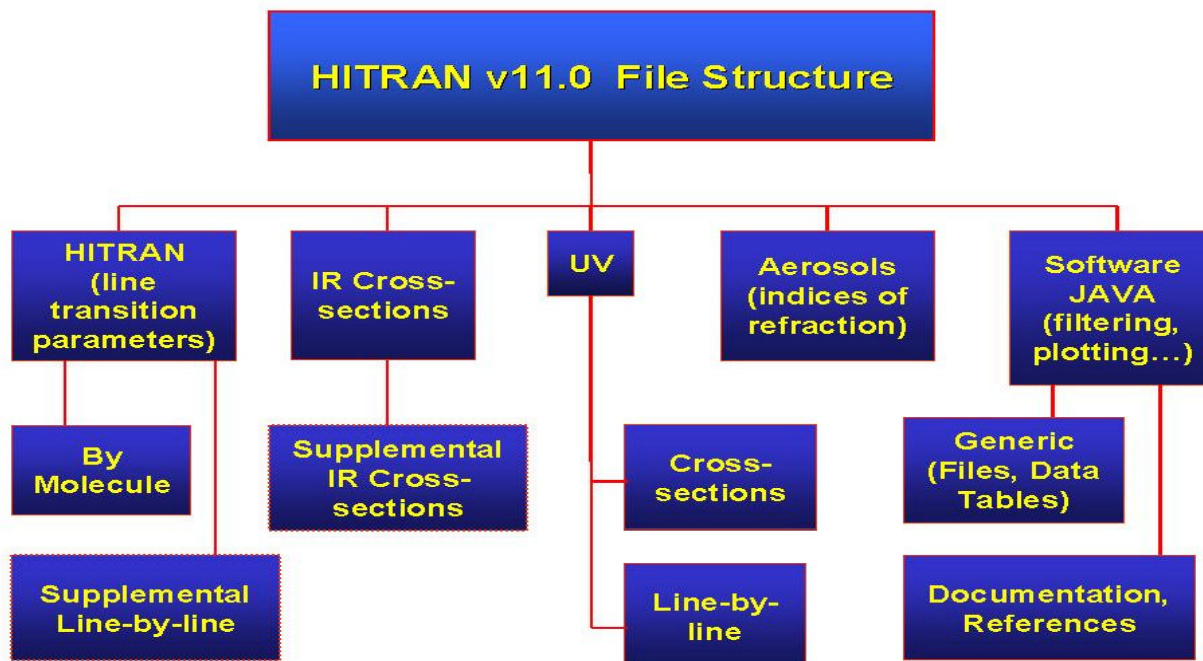


Figure 1. File Structure for HITRAN2000.

of HITRAN, and this media is certainly a more economical and time-saving method. It also allows for more frequent updates, while still allowing for an archival sense.

We have compressed the HITRAN data files (which are all ASCII) using the gzip compression scheme. The files are broken up into individual files for those users who either have slow connections or who are only interested in a sub-set of the data. Documentation is provided in the form of Adobe pdf files.

#### 4. HITRAN Web-site Development

The HITRAN web-site ([cfa-www.harvard.edu/HITRAN](http://cfa-www.harvard.edu/HITRAN)) has been substantially renovated during this period of performance. There are several useful sub-pages. The "HITRAN FACTS" page gives summary tables of all the contents of the new HITRAN compilation. The "HITRAN UPDATES" sub-page includes corrections as discovered by users and new data that has been validated since the current edition. We now have initiated a "FAQ" page to highlight frequently asked questions and our solutions. If users do not look at this, at least it provides us with rapid answers to phone calls and e-mails. We continue to provide a page with links to conferences of specific relevance to HITRAN, and also a page with links to other related database lists.

The ftp-site ([cfa-ftp.harvard.edu/pub/HITRAN/](http://cfa-ftp.harvard.edu/pub/HITRAN/)) for the new HITRAN compilation was also set up. There are security issues that have prevented a few users from accessing the site, but we have provided a set of suggested solutions with the announcement for the site that goes out to requesters. Generally the problem has been that the machines the users are using are not properly registered with a nameserver. Our computer center here has been assisting these legitimate users to solve this problem at their end.

## 5. Collaborations

The development of HITRAN relies on cooperation and collaboration with the whole spectroscopic community. As can be seen from the description of data in this report, we have continued to develop these connections. In this period in particular, the generous contributions of groups in Belgium are to be noted. We are also working closely here with scientists in the Atomic and Molecular Physics Division: K. Chance and K. Yoshino especially for updating and improving the UV portion of the compilation. Linda Brown of JPL provided much data in very complete HITRAN format (even anticipating the next expanded format). Aaron Goldman of the University of Denver also provided much data, as did Steven Massie of NCAR for the aerosols. We have a strong collaboration with Victor Dana and Jean-Yves Mandin of the University of Paris, providing us with accurate high-resolution parameters for trace gas species. Curtis Rinsland and Mary Ann Smith of NASA Langley Research Center and their colleagues at the College of William and Mary, D. Chris Benner and V. Malathy Devi, are also close collaborators. Robert Gamache at the University of Massachusetts, Lowell has been working on the partition sums and other molecular aspects of the database.

## 6. Meetings, Presentations, and Publications

The major event during this reporting period was the holding of the Sixth Biennial HITRAN Conference here at the Harvard-Smithsonian June 19-21. The Proceedings of the Conference are available in an Adobe pdf file in the HITRAN web-site. The meeting was well attended, and was comprised of five oral presentation sessions and two poster sessions. The first poster session emphasized remote-sensing issues and their relation to databases. The sessions focused on the spectroscopic database issues of line parameters and cross-sections. A whole session was devoted to water vapor.

I also was invited in February to give a seminar on HITRAN and visit the National Institute of Environmental Studies in Tsukuba, Japan. This provided me with the opportunity to learn more about their work on the ILAS satellite.

In March the PI was invited to give a presentation on atmospheric molecules at the conference on Atomic and Molecular data (ICAMDATA), held this time at Oxford University, UK. This conference had previously been held at the National Institute of Standards and Technology (NIST), and is promoted by NIST. The areas covered naturally go towards the commercial applications. During the conference K. Chance and I visited the Rutherford Appleton Laboratory and had discussions with J. Ballard, K. Smith, and D. Newnham concerning their new laboratory data for collision-induced cross-sections and infrared molecular cross-sections.

In April, I attended the EOS Working Group (IWG) Meeting in Tucson. In May, I visited with Linda Brown and Robert Toth at JPL. The discussions were fruitful and enabled HITRAN to acquire the vastly improved water-vapor data in the 6- $\mu$ m region, for example.

Besides giving the opening presentation at the HITRAN Conference, I attended the XVI<sup>th</sup> International Conference on High Resolution Infrared Spectroscopy, Prague, Czech Republic in September. This conference is the pre-eminent meeting of high-resolution spectroscopy in Europe in the even years between the well established West-European conference usually held in France on the odd-numbered years. Like the annual Ohio State meeting, these conferences afford an unusual opportunity to gain information and data prior to publication.

*Publications during this period:*

“Atmospheric Molecules,” L.S. Rothman and M. ĀivkoviĀ-Rothman, *Atomic and Molecular Data and their Applications*, eds. K.A. Berrington and K.L. Bell, AIP Conference Proceedings **543**, 92-103 (2000).

Chapter “Atmospheric Ultraviolet Spectroscopy,” K. Chance and L.S. Rothman in *Ultraviolet Spectroscopy and UV Lasers*, eds. P. Misra and M.A. Dubinskii, Marcel Decker, New York (2000).

*Presentations during this period:*

“HITRAN Database,” Seminar, National Institute of Environmental Studies, Tsukuba, Japan (February 2000). (Invited)

“Atmospheric Molecules,” International Conference on Atomic and Molecular Data (ICAMDATA), Oxford, UK (March 2000). (invited)

“HITRAN2000,” The Sixth Biennial HITRAN Database Conference, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts (June 2000); XVI<sup>th</sup> International Conference on High Resolution Infrared Spectroscopy, Prague, Czech Republic (September 2000).

“The Venus 1- $\mu\text{m}$  CO<sub>2</sub> Bands and the O<sub>2</sub> (0-1) X <sup>3</sup>G<sub>g</sub><sup>-</sup> - a<sup>1</sup>)<sub>g</sub> and (1-0) X <sup>3</sup>G<sub>g</sub><sup>-</sup> - b<sup>1</sup>G<sub>g</sub><sup>+</sup> Bands in the Earth’s Atmosphere,” The Sixth Biennial HITRAN Database Conference, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts (June 2000); XVI<sup>th</sup> International Conference on High Resolution Infrared Spectroscopy, Prague, Czech Republic (September 2000).

## 7. Future Effort

We are on the verge of making available a vastly improved line list for methane which will have an impact on the **MOPITT** and many other programs of EOS. This effort is in collaboration with L.R. Brown. Other line-parameter data that will soon be made available include bands of NO<sub>2</sub>, CH<sub>3</sub>Cl, HOCl, H<sub>2</sub>O<sub>2</sub>, and the numerous UV data sets mentioned in the last report.

We are evaluating new data on water vapor in the near IR through visible region that has come from a collaboration of laboratories directed out of the University College London supported by the European Space Agency.

We are collaborating with the group at NASA Langley and R. Hawkins at AFRL to update the CO<sub>2</sub> parameters based on energy level observations in the 3- $\mu\text{m}$  region and new intensity observations by NASA Ames. This effort requires a whole new global fit of the energy levels, and then a calculation of all carbon dioxide parameters for a self-consistent set.

We are expecting to receive new parameters for carbon monoxide from NASA Ames. These data as well as the CO<sub>2</sub> improvements are expected to impact **MOPITT**, **TES**, etc.

We are planning to introduce collision-induced absorption (CIA) parameters to the HITRAN compilation. These can be formatted similarly to the current cross-section data.

The plans continue for an enhanced format for HITRAN that will include new parameters for each line: the Einstein-A coefficient, the error and references for self-broadened, temperature dependence of halfwidth, and pressure shift parameters. There will also be a flag for treatment of lines with line-coupling parameters and the statistical weights of the upper and lower states of a transition. It should be remarked that the improved header format for the cross-sections has already been implemented.

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