Executive Summary:

STUDY ON THE SIMULATION OF ATMOSPHERIC INFRARED SPECTRA

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1 INTRODUCTION

The breakdown of local thermodynamic equilibrium (NLTE) in the atmosphere is an issue of major importance in infrared limb emission spectroscopy of the atmosphere. A strategy is needed to handle NLTE during retrieval of temperature and constituents' abundancies from limb emission spectra as recorded by a high resolution limb sounder such as the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS). The simplest and most efficient strategy certainly is to neglect NLTE, while inclusion of NLTE is computationally and algorithmically more demanding. The appropriate way to handle NLTE is closely connected to proper selection of microwindows (i.e. subsets of the measurement vector actually used for analysis). In this work we propose and justify strategies for handling of NLTE for retrieval of pressure, temperature, O_3, H_2O, CH_4, N_2O, HNO_3, CO, NO, NO_2, N_2O_5 and $ClONO_2$, along with lists of optimized microwindows which are compatible with the proposed strategy.

2 MIPAS INSTRUMENT SPECIFICATION

The spectral resolution of the MIPAS instrument is 0.035 cm−1 (unapodized). The spectral bands of the instrument are 685 - 970 cm−1 1020 - 1170 cm−1, 1215 - 1500 cm−1; 1570 - 1750 cm−1, and 1820 - 2410 cm−1. the noise equivalent spectral radiances (NESR) for the unapodized spectra are 50, 40, 20, 6, and 4.2 nW/(cm^2 sr cm^{-1}), respectively. Smaller values are assumed for apodized spectra: 30.35, 24.28, 12.14, 3.64, and 2.549 nW/(cm^2 sr cm^{-1}). The spacing of tangent altitudes is assumed at 3 km, from 8 km to 83 km.

3 NLTE MODELLING

Spectroscopic Data: A precondition to appropriate assessment of NLTE is precise NLTE modelling, which relies on spectroscopic line parameters. Since under NLTE hotbands may become significant, which are of no importance under local thermodynamic equilibrium, the available spectroscopic linelist has been extended by spectroscopic data of 199 bands of the ozone molecule involved in 17 polyads.

Vibrational Temperatures: For four different atmospheric conditions (midlatitude daytime, midlatitude nighttime, polar winter, polar summer) vibrational temperatures for bands of CO_2, H_2O, CH_4, N_2O, O_3, CO, and NO have been calculated in the framework of this study, while vibrational temperatures for HNO_3 and NO_2 have been included from other sources. The altitude range vibrational temperatures have been calculated for extends up to 120 km for all species under consideration, except for NO, where the altitude range has been extended up to 200 km. Furthermore uncertainties of vibrational temperatures were estimated. Vibrational Temperatures have been calculated for the following vibrational states:

- CO_2: 47 vibrational levels from 6 isotopes from energies from 667 to 4200 cm^{-1} emitting in the 15, 10 and 4.3 μm regions.
- CO: The fundamental transitions around 4.6 μm
- H_2O: 5 vibrational levels from 3 isotopes emitting in the 6.3 μm region.
- CH_4: 4 vibrational states from 2 isotopes emitting at 7.7 and 6.5 μm
- N_2O: 16 vibrational states from 4 isotopes
- HNO_3: 9 vibrational levels

A visualization tool for vibrational temperatures has been developed.

Radiance Spectra: Using the "Reference Forward Model" (RFM) for all four atmospheric conditions and all 26 tangent altitudes between 8 and 83 km, radiance spectra have been generated for LTE and NLTE conditions as well as minimum and maximum NLTE atmospheres (i.e. nominal vibrational temperatures minus, respectively plus, 2-sigma uncertainty of vibrational temperatures). Furthermore Jacobian (partial derivatives of spectral radiances with respect to numerous atmospheric state parameters have been provided for use in microwindow optimization and error analysis. In order to handle this large amount of spectra, a dedicated visualization tool has been provided.

4 Microwindow Database

Microwindows for analysis of pressure, temperature, O_3, H_2O, CH_4, N_2O, HNO_3, CO, NO, NO_2, N_2O_5 and $ClONO_2$, have been defined such that the total retrieval error to be assigned to each microwindow is minimized. Sources of error under consideration were: measurement noise, temperature uncertainties, pointing uncertainties, uncertain abundancies of interfering species, gain calibration error, spectroscopic data uncertainties, variability of isotopic fractioning of species, and uncertainties of vibrational temperatures (for CO and NO only), or neglection of NLTE (all other target species). As a consequence the microwindows represent an optimum tradeoff between errors of different sources. Since during the step of micowindow optimization for one altitude it is not clear which microwindows will be selected for higher altitudes, inter-layer error correlations have to be ignored at this step. While optimized microwindows are altitude-dependent, i.e. microwindow boundaries vary from altitude to altitude, they are not compatible with the current version of the 'Optimized Forward Code

/ Optimized Retrieval Code'. Therfore suboptimal databases have been generated which include microwindows of limited altitude dependence; for the so-called 'One-Mask Microwindow Database' microwindows do not change their boundaries with altitude' while for the 'Three-Mask Microwindow Database' microwindow boundaries may change their boundaries up to two times with altitude. Most important table entries in the database are estimates of retrieval errors due error sources mentioned above, and are meant to make possible an optimized decision on which subset of microwindows to use in an actual retrieval.

5 Error estimation

Sample error estimation for CH_4 based on the full Jacobian and thus exceeding the single layer approximation leads to retrieval errors larger by a factor of two compared to the microwindow database entries. Two possible reasons have been identifies: (a) there is an inconsistency in the use of Jacobians. While partial derivatives were povided for perturbation of entire layer between adjacent tangent altitudes, they were used as if they were related to a triangular perturbation resulting from perturbation at a level and interpolation towards adjacent levels; (b) inter-layer error correlations are expected to increase the retrieval error but are not included in the single layer approximation.

6 NO retrieval study

- A major uncertainty in NO vibrational temperature is that due to knowledge of thermospheric kinetic temperature (which will not be measured by MIPAS). It can be anticipated that variability in vibrational temperatures arising from solar and geomagnetic activity, for example, will be >100K at mid-latitudes, and 100's K at polar latitudes

- The height range within which NO profiles of useful precision (<100%) could be retrieved from a single MIPAS limb-scan was found to be 35 to 48km for mid-latitude day. For mid-latitude night, information could be retrieved only in the lower thermosphere, but for polar winter, information could be retrieved (at degraded vertical resolution) in the mesosphere, provided that concentrations are enhanced sufficiently.

- Provided that the thermosphere is completely stable during a limb-scan, gross errors of thermospheric origin appear to be confined mainly to the thermosphere itself and to the lower mesosphere.

- Gross errors (> factor 10) in stratospheric NO retrievals were, however, found to result from a a horizontal gradient (factor 2) in thermospheric [NO] encountered during a limb-scan.

- For accurate stratospheric NO retrievals, it will be *essential* to:

  1. model the atmosphere up to 200km
  2. account adequately for horizontal gradients in thermospheric NO emission

  → A special processing scheme will therefore be required for MIPAS, along similar lines to that which has been developed for ISAMS.

- Due to the importance of characterising horizontal gradients, it is recommended that MIPAS view to the rear of ENVISAT rather than to the side and that a limb-scan to 200km be incorporated into the standard sequence.

- Extending the nominal limb-scan to 62km would allow improved precision on retrieval of NO in the stratopause region.

- Further work is necessary to: (i) investigate the impact on stratospheric retrievals of photochemically-induced NO NLTE emission from the upper stratosphere and the lower thermosphere and possible use of the 15 NO microwindows to retrieve an effective thermospheric temperature, to reduce downward propogation of temperature errors from the thermosphere.

7 Conclusion and Recommendation

For all target quantities where the baseline retrieval scenario was neglection of NLTE (O_3, H_2O, CH_4, N_2O, HNO_3, NO_2, N_2O_5 and $ClONO_2$), a considerable number of microwindows was found where NLTE transitions are excluded such that neglection of NLTE is not the predominating source of error. Thus the approach to neglect NLTE has been proven reasonable. In particular for O_3 this is only true for the nominal altitude range in the operational retrieval, whereas for higher altitudes a more sophisticated approach must be considered. Retrieval of CO depends largely on precise modelling of vibrational temperatures and thus is not suitable for routine analysis. NO, due to the large fraction of thermospheric signal in stratospheric limb measurements, is a very special case and therefore needs a dedicated treatment in the retrieval. We tested and recommend Optimal Estimation for this purpose.

Furthermore we recommend

- to modify the Optimum Forward Model / Optimum Retrieval Model such that it supports altitude-dependent microwindows in an efficient manner;

- to generate a fully altitude-dependent microwindow database which makes best use of optimized microwindows;

- to apply a cost function to each microwindow during construction of the microwindow database;

- to substitute the altitude-independent retrieval accuracy requirements by more realistic altitude-dependent ones.