FINAL REPORT PROGRAM LEFE

Program LEFE/ CHAT	Project Title Towards a better interpretation of atmospheric phenomena based on an improved modeling of the radiative transfer in the middle and upper atmospheres	Year 2016

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Context :

In the middle and upper atmospheres of planets, the infrared (IR) radiation is unbound from the local kinetic temperature (Local Thermodynamic Equilibrium (LTE) breakdown). This affects both the energetic properties of the atmosphere and the observations of the lower atmospheric layers by space borne infrared instruments and requires special treatment.

Objectives / scientific questions :

Improving the quality of the cooling/heating rates calculation and the interpretation of the IR space borne observations requires (a) updating the non-LTE model; (b) including a large number of spectral transitions including the weak ones; (c) implementating sophisticated non-LTE modeling into operational LTE-based codes.

Main results:

(1) The model of 4.3 μm nighttime emissions has been updated using recently found mechanism of energy transfer from vibrationally excited OH(v) through N_2(v) to CO_2(v3) vibrational levels. Adding this mechanism helps to resolve long-standing discrepancy between measured and simulated 4.3 μm emissions.
(2) A methodology for implementing the non-LTE populations to LTE-based radiative transfer codes has been suggested and the first calculations for molecular spectra measured by IASI have been performed with its help, showing an improvement between the simulated and measured radiances (Fig. 1).
(3) The look-up tables with vibrational level populations of the CO_2 molecule have been calculated for typical atmospheric scenarios.
(4) The breakdown of local thermodynamic equilibrium for rotational levels in 4.3 μm rovibrational band was modeled for the first time and the results showed the necessity of this modeling for the interpretation of observed Martian spectra.
(5) A powerful system (TheoReTS) for theoretical calculations of molecular spectra predictions has been developed for atmospheric molecules, including methane and ethylene.

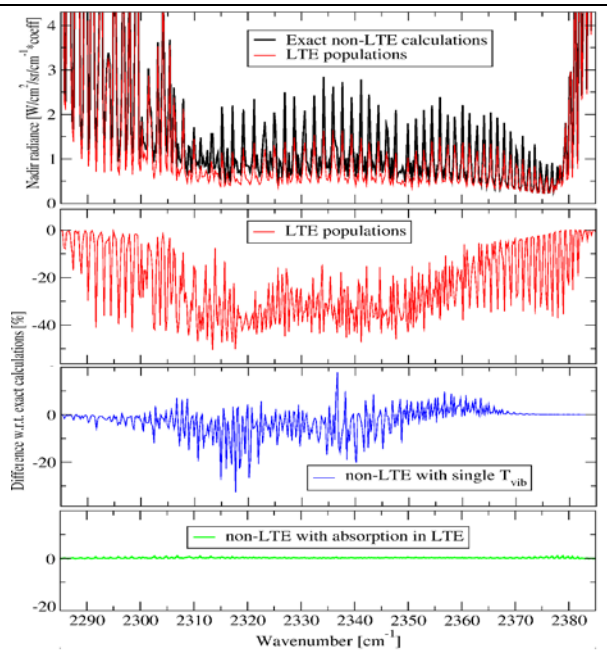

Figure 1

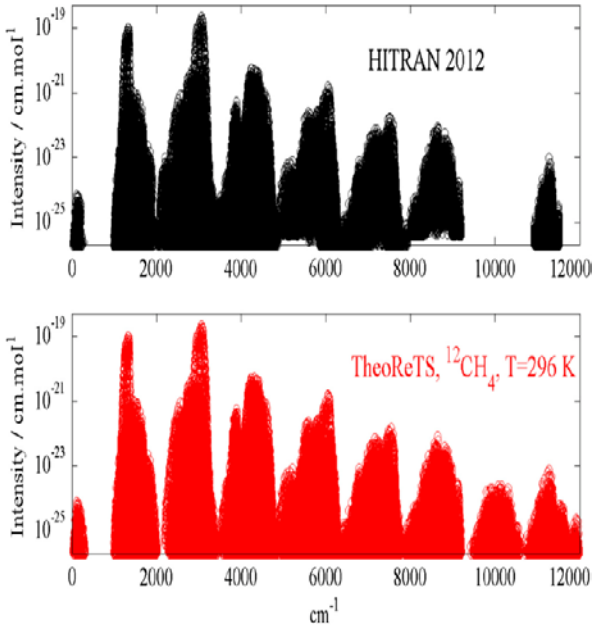

Figure 2

The Fig. 1. illustrates the necessity of proper modeling the vibrational level populations for the interpretation of spectrally resolved nadir-looking observations performed by IASI instrument in 4.3 µm CO_2 band: not taking the non-LTE into account results in 40% error in the simulated radiance (the upper two panels) whereas the optimized non-LTE model (lower panel) ensures less than 1% error. The current implementation of the LUT-based non-LTE populations to the 4A/OP LMD LTE-based radiative transfer code results in ~15% error (not shown) and needs to be improved. In Fig. 2 we show an example of comparison of $^{12}CH_4$ absorption coefficients between TheoReTS (Reims-Tomsk information system) and HITRAN2012 for simulations performed at a resolution of 0.5 cm^{-1} and a pressure of 10 Torr. This figure shows that the GSMA produced spectra are more complete in some intervals than HITRAN databases particularly for weak lines which were not yet assigned in published spectra analysis. These and other improvements ensure the quality of the atmospheric molecular infrared spectra interpretation.

Future of the project:

The model of nighttime 4.3 µm radiance will be implemented to the multi-channel retrieval algorithm, which will be able to retrieve simultaneously CO_2, OH, atomic oxygen and kinetic temperature from 4.3, 1.6, 2.0, and 15 µm radiances. The methodology of embedding the non-LTE populations in the LTE-based radiative transfer codes needs to be improved to eliminate the remaining residuals. When done, the technology will be transferred to a number of radiative transfer codes for planetary atmospheres (e.g. for Mars, Titan, and Saturn), which suffer from a lack of this kind of modeling. The molecular spectra database produced by the GSMA group will be used in the interpretation of atmospheric radiation of Earth and other planets, including the extraterrestrial ones.

Publications *:*

1. Panka, P. A., A. A. Kutepov, K. S. Kalogerakis, D. Janches, J. M. Russell, L. Rezac, A. G. Feofilov, M. G. Mlynczak, and E. Yigit, "Resolving the mesospheric nighttime 4.3um emission puzzle: comparison of the CO2(v3) and OH(v) emission models", Atmos. Chem. Phys. 17, 9751-9760, https://doi.org/10.5194/acp-17-9751-2017, (2017).

2. Kutepov,. A. A., L. Rezac, and A. G. Feofilov, "Evidence of a significant rotational non-LTE effect in the CO2 4.3 um PFS-MEX limb spectra", Atmos. Meas. Techn. 10, 265-271, doi: 10.5194/amt-2016-188, (2017).

3. Rey, M., A.V. Nikitin, Y.L. Babikov, V.G. Tyuterev, "TheoReTS – An information system for theoretical spectra based on variational predictions from molecular potential energy and dipole moment surfaces", J. Mol. Spectr., 327, 138-158, (2016).