Non-LTE radiative transfer in the context of infrared satellite observations of the lower atmosphere

A. Feofilov[1], L. Rezac[2], A. Kutepov[3]

artem.feofilov@lmd.polytechnique.fr

1: Laboratoire de Météorologie Dynamique, Fx-Conseil, Ecole Polytechnique, France;
2: Max Planck Institute for Solar System Research, Goettingen, Germany
3: The Catholic University of America / NASA Goddard Space Flight Center, Greenbelt, MD, USA

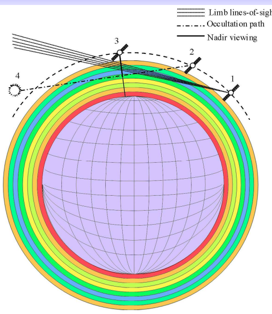
Abstract. In recent decades, more and more advanced satellite sensors have been measuring the Earth's atmosphere, clouds, aerosols, and surface characteristics to enable enhancements in weather prediction, climate monitoring, and environmental change detection. In particular, hyperspectral instruments measuring the thermal infrared part of the spectrum provide a plethora of information about atmospheric temperature, trace gas concentration, clouds, aerosols, and surface in narrow spectral channels. These instruments are usually flying onboard a series of meteorological platforms and are therefore predestined for climate monitoring.

The results obtained in this field are already impressive: mid-tropospheric temperature and humidity profile retrieval with an accuracy of up to 1 K and 10%, respectively; near real-time mapping of chemical species and aerosols; tracking the greenhouse gases, cloud properties retrieval, and the list is not exhaustive.

However, there is a limitation, which does not allow a full usage of all advantages these instruments offer. The problem is linked with the radiation of the atmospheric layers, for which the conditions of local thermodynamic equilibrium (LTE) do not hold, and the populations of vibrational levels deviate from the Boltzmann distribution for the local kinetic temperature. The non-LTE effects strongly limit the exploitation of the channels sensitive to the contribution of the middle and upper atmosphere. For instance, about a quarter of IASI channels are not currently used due to this issue.

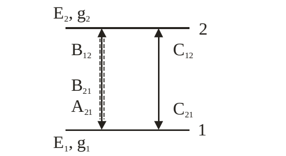
Even though current retrievals take into account the atmosphere up to the mesopause level, the complexity of the processes governing the vibrational level populations in the upper atmosphere precludes using exact non-LTE calculations. As a result, some channels can not be used at all or empirical corrections are introduced. This issue has been clearly identified by the radiative transfer community, as well as by atmospheric composition and climate communities. In this work, we show an approach for applying complex non-LTE models developed in recent years to the operational retrievals in the lower atmosphere.

Atmospheric observations from space



- Nadir observations: shorter optical paths, information about atmospheric layers comes from different spectral channels.
- Limb viewing geometry: longer optical paths, vertical resolution is defined by the instrument's field of view
- Occultation measurements: high vertical resolution, absorption from the ground state, observations are limited in time and space.
- All three types of measurement involve line of sights passing through the middle and upper atmospheric layers.
- The contribution of the latter is the highest for limb observations, but is not negligible for nadir viewing instruments and in some cases even for occultation observations.

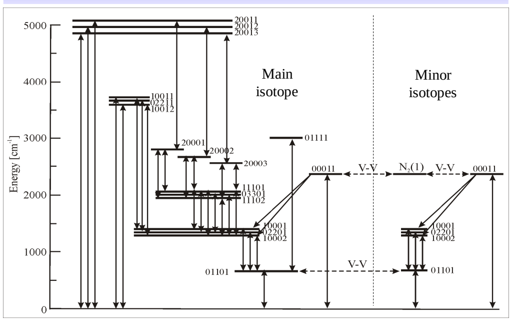
Explaining LTE and non-LTE using 2-level atomic gas



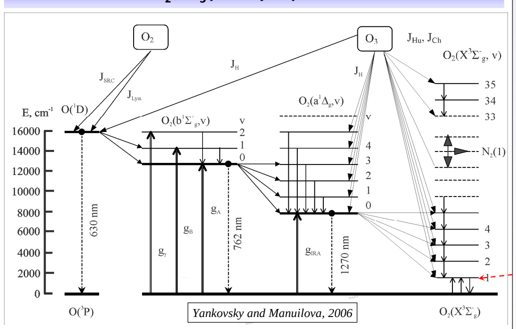
$$\begin{cases} (\underbrace{A_{21} + B_{21}\bar{J}}_{R_{21}} + C_{21})\, n_2 = (\underbrace{B_{12}\bar{J}}_{R_{12}} + C_{12})\, n_1 \\ n_1 + n_2 = n \end{cases}$$

- Let's consider an atmosphere of 2-level atomic gas with known $P(z)$, $T(z)$, A_{21}, B_{12}, B_{21}, C_{12}, C_{21}
- We are interested in calculating the full state of the atmosphere, that means n_1, n_2 at all z.
- At each atmospheric level, the collisions are local
- Radiation comes from above and from below
- $C >> R \rightarrow$ local thermodynamic equilibrium, LTE (lower atm.)
- $R \approx C \rightarrow$ **non-LTE**
- The problem becomes **non-local and non-linear.**

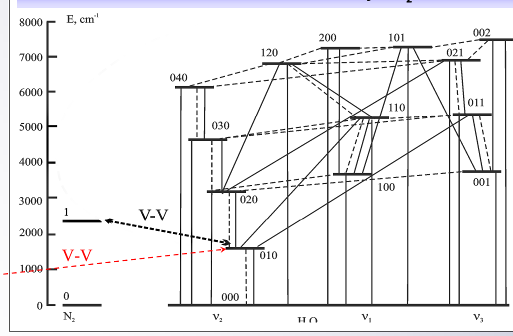
Vibrational levels and transitions for CO_2



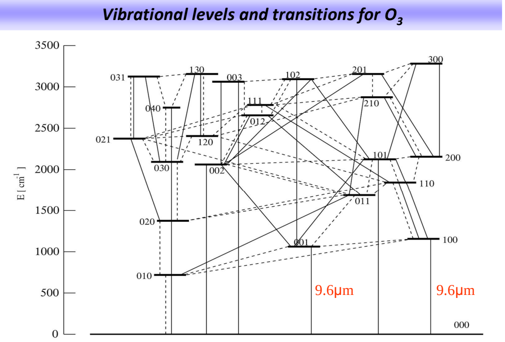
O_2 / O_3 photolysis products



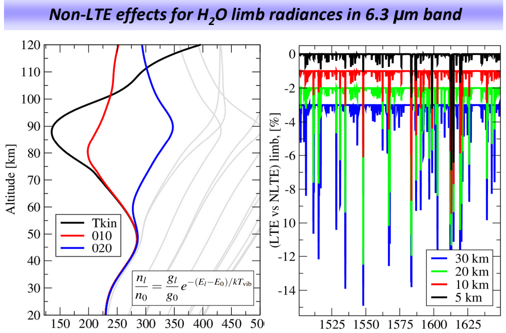
Vibrational levels and transitions for H_2O



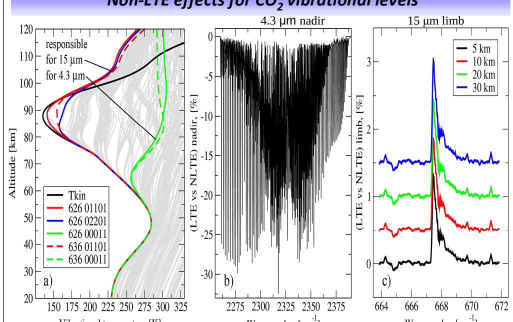
Vibrational levels and transitions for O_3



Non-LTE effects for H_2O limb radiances in 6.3 μm band

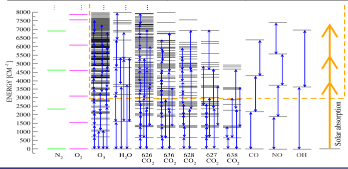


Non-LTE effects for CO_2 vibrational levels



ALI-ARMS non-LTE research code

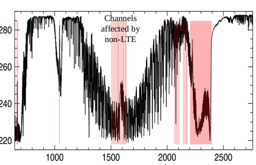
Retrieved/analyzed parameter(s)	Spectral band	Experiment/Model	References
CO_2 VMR, Earth	4.3 μm	CRISTA spectrometer	Kaufmann et al., 2002
CO_2 emissions, Mars	10 μm	MGS TES spectrometer	Maguire et al., 2002
O_3 VMR, Earth	9.6 μm	CRISTA spectrometer	Kaufmann et al., 2003
IR cooling/heating, Mars	15 μm	Mars GCM	Hartogh et al., 2005
Temperature, Earth	15 μm	CRISTA spectrometer	Gusev et al., 2006
Temperature, Earth	15 μm	SABER radiometer	Kutepov et al., 2006
H_2O VMR, Earth	6.3 μm	SABER radiometer	Feofilov et al., 2009
CO_2 VMR, temperature, Earth	4.7, 15 μm	SABER radiometer	Rezac, 2011, 2014
Temperature, Mars	15 μm	MGS TES spectrometer	Feofilov et al., 2012
HCN rotational cooling, Titan	100-1000 μm	Theoretical study	Rezac et al., 2013

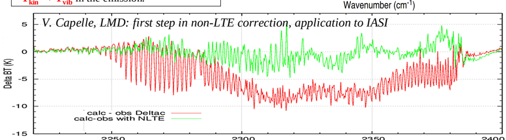


Vibrational level diagram for molecules included in the current ALI-ARMS reference model (CH_4 and N_2O not shown). Blue: major fundamental and hot transitions. Dashed orange: vibrational level manifold pumped by near-IR solar radiation (thick orange). The levels are also coupled throuhg V–T and V–V energy exchanges (not shown).

Including the non-LTE effects to radiative transfer codes

Approach
- Non-LTE code ALI-ARMS, used for atmospheres of Earth, Mars, Titan, Saturn.
- Non-LTE models (=sets of processes) for CO_2, O_3, H_2O, CH_4, CO, C_xH_y.
- Can be used as a source of corrections for each channel (like in [DeSouza-Machado et al, 2007]) – technical, large LUTs.
- Another way is to incorporate vibrational level populations to existing RT codes – smaller LUTs, real radiative transfer: $T_{kin} \rightarrow T_{vib}$ in the emission.





Conclusions and outlook

- The vibrational level populations of atmospheric molecules are not in LTE at pressures lower than ~1 Pa. For some solar-pumped levels, LTE breakdown continues down to the ground.
- Satellite observations in limb, nadir, and occultation modes are sensitive to contribution of the non-LTE layers.
- Ignoring the non-LTE effects can lead up to 30% difference in spectrally resolved radiance in nadir.
- The non-LTE codes can treat this physics given that the set of spectroscopic and collisional parameters is given, which is the case for most of atmospheric molecules.
- A new method of including the non-LTE effects to existing radiative transfer codes has been suggested.
- For operational purposes, the method requires calculation of LUTs for vibrational temperatures with the help of non-LTE code ALI-ARMS.
- The method will be applied and validated using the IASI channels (645−2760 cm⁻¹)