Infrared radiation in the middle and upper atmosphere of planets: current problems in calculation and interpretation

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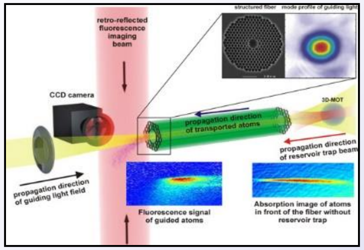
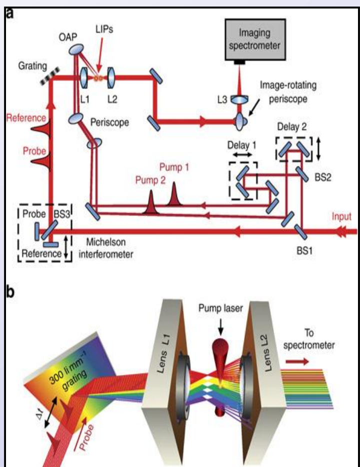
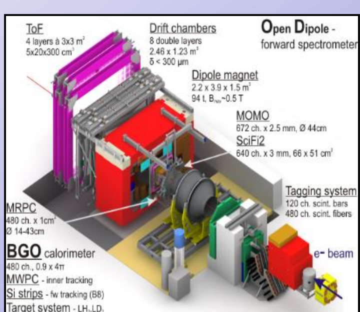
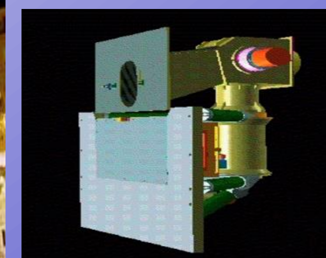
Motivation

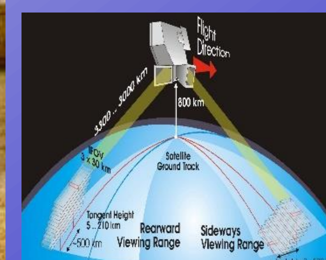
"The purpose of the workshop is to … reinforce the interactions between experts … to meet the challenges and requirements of future space observation…"











Middle and upper atmosphere (MUA): why bother?

- MUA is a “gateway” between the lower atmosphere and space.

- MUA is sensitive to solar influence and to the inputs from below.

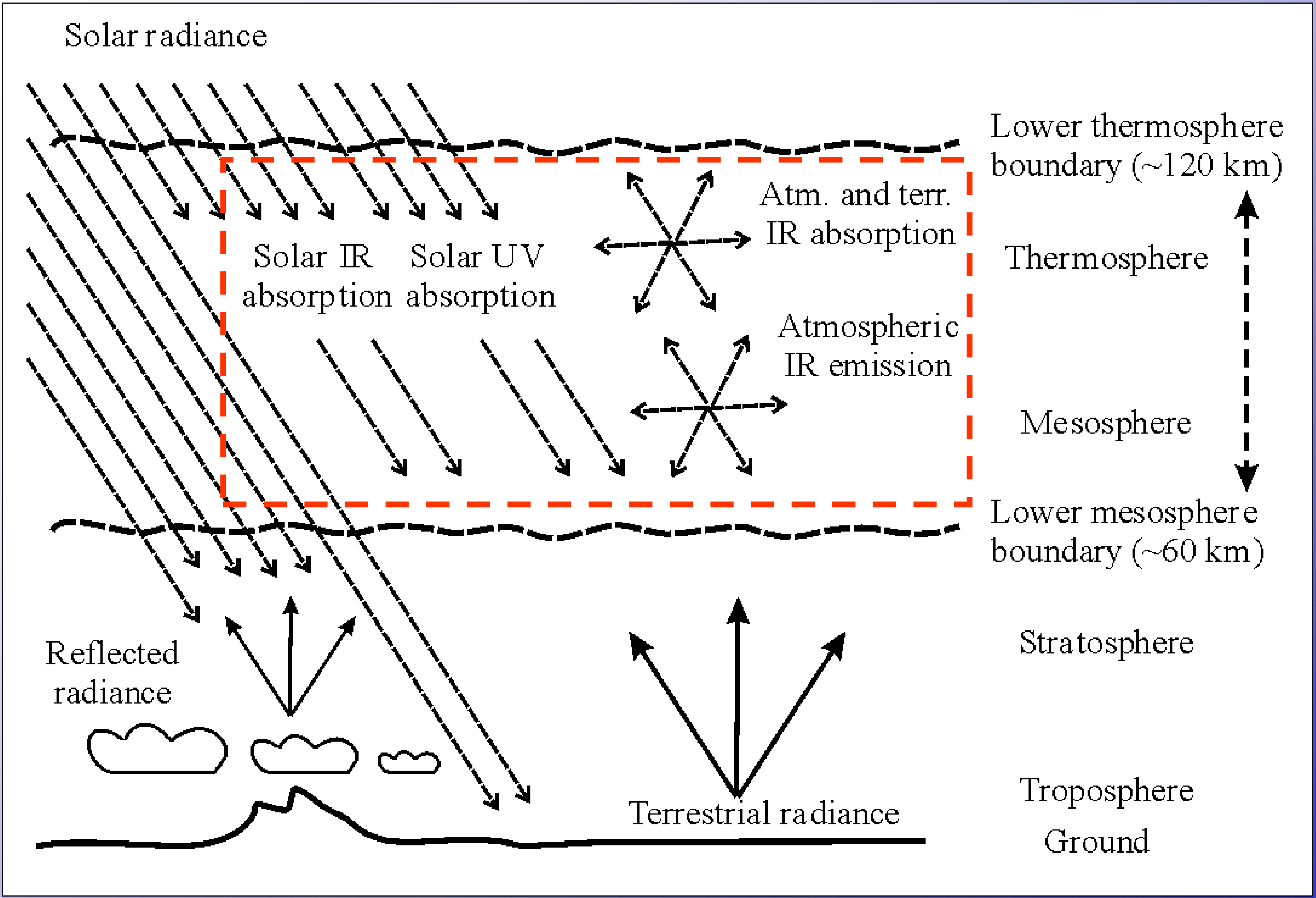
- MUA absorption and emission in molecular bands affect the atmospheric observations of other areas.

On Earth

- Anthropogenic changes in greenhouse gases may change the composition of MUA and the input from below.

- Noctilucent clouds observed in polar summer mesosphere are very sensitive to temperature changes.

IR radiance in the MUA



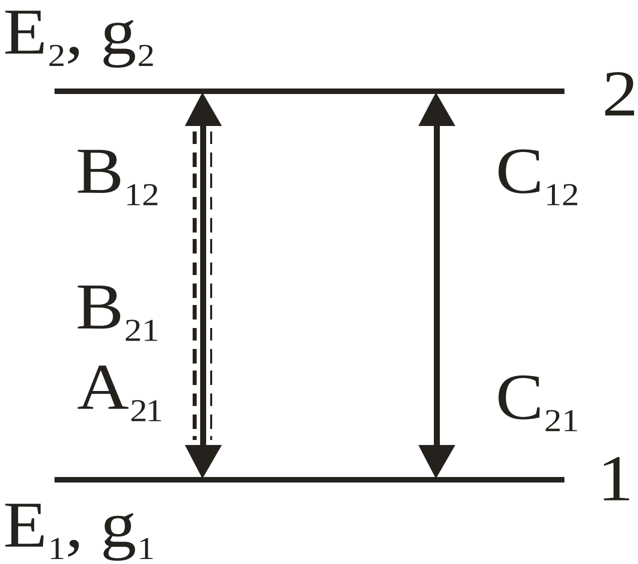
An important peculiarity of the MUA

- Infrared radiance absorption/emission corresponds to vibrational excitation/de-excitation of the molecules.
- To estimate the energetic characteristics of the given area or to interpret the infrared observations in MUA one needs to know the vibrational levels populations.

BUT !!!

- In the upper atmosphere, the collisions between the molecules are **not** frequent and the populations are **not** defined by local temperature.
- Breakdown of Local Thermodynamic Equilibrium (LTE).
- Special methodology is applied (non-LTE modeling).

LTE and non-LTE: two-level atom



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

In the lower atmosphere:

$R \ll C$

$R_{21}n_2 \sim R_{12}n_1$

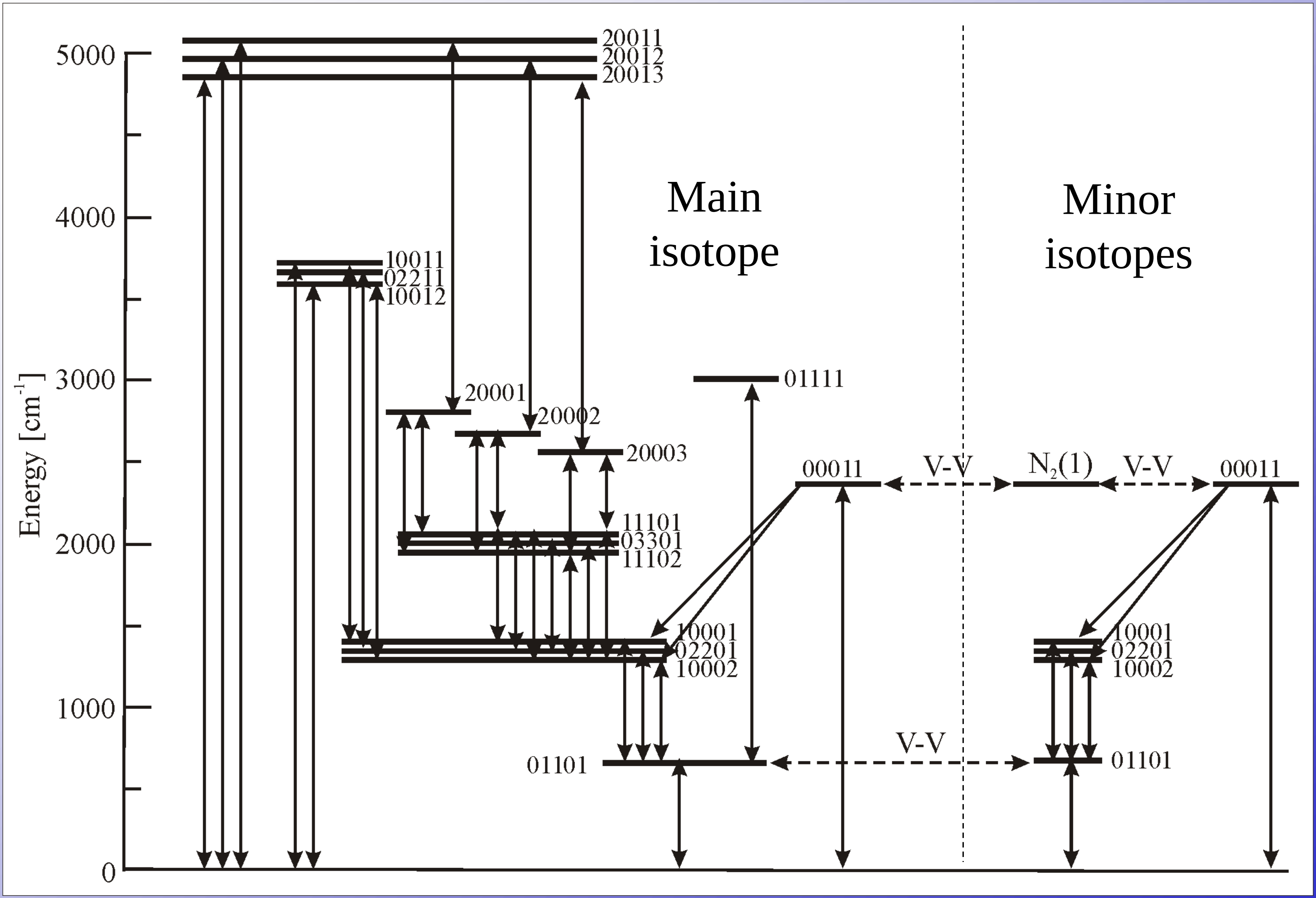
$$n_2C_{21} = n_1C_{12}$$

$$\frac{n_2}{n_1} = \frac{g_2}{g_1}\, e^{-\frac{E_2-E_1}{kT}}$$

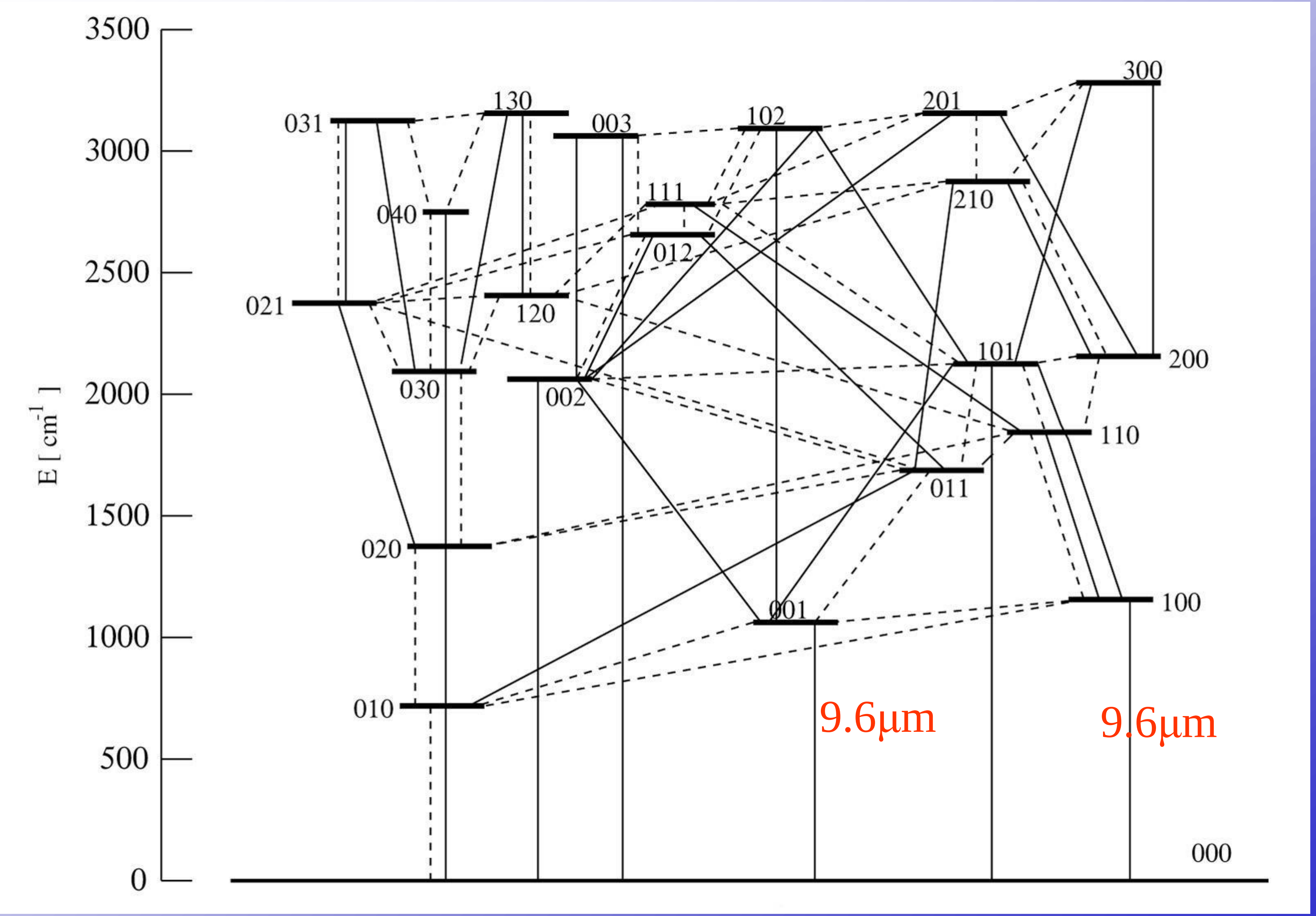
In the upper atmosphere:

Collisions are less frequent, and the populations are not defined by local temperature anymore = non-LTE

Vibrational levels and transitions for CO_2

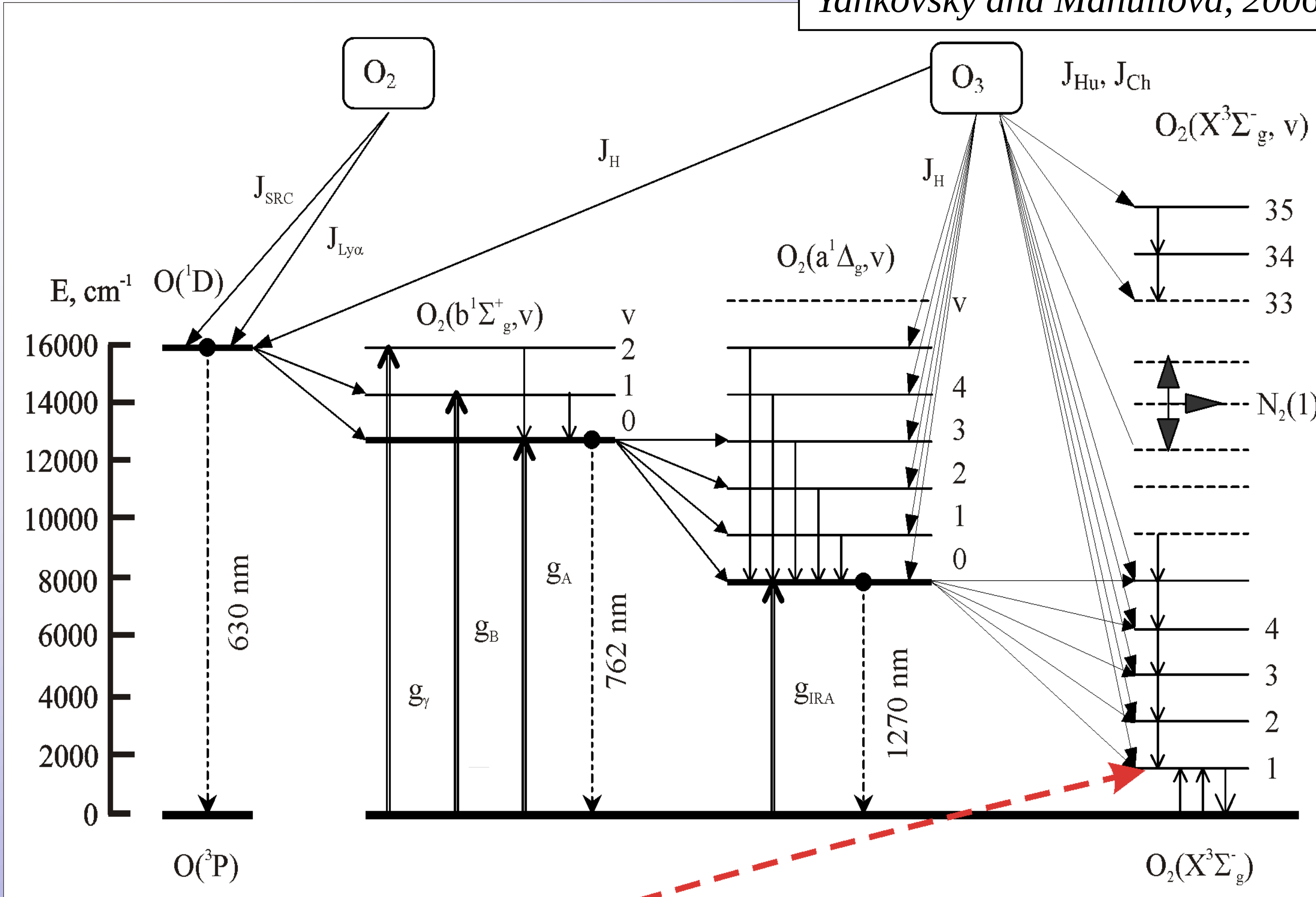


O_3 levels and transitions



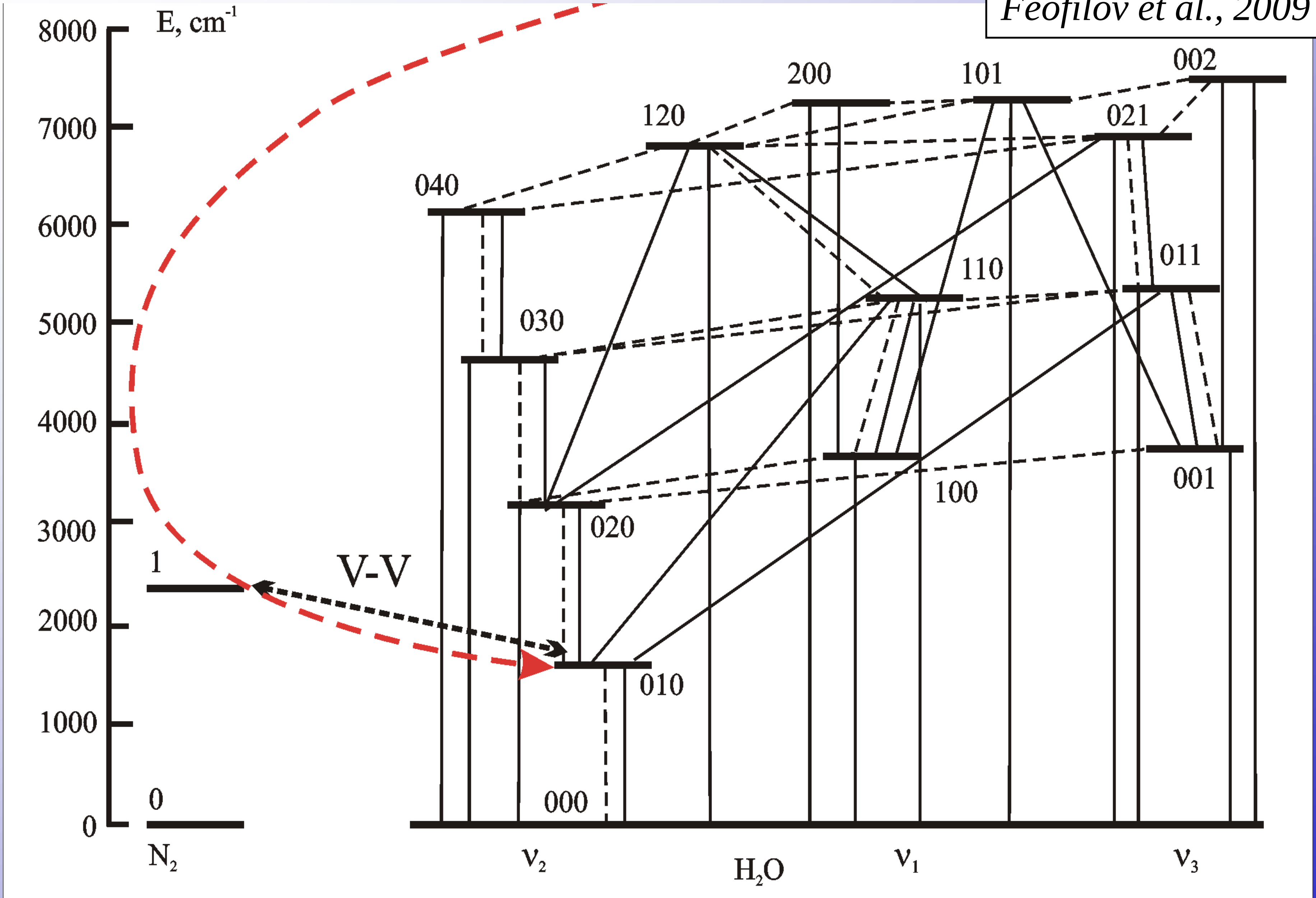
O_2/O_3 photolysis products

Yankovsky and Manuilova, 2006

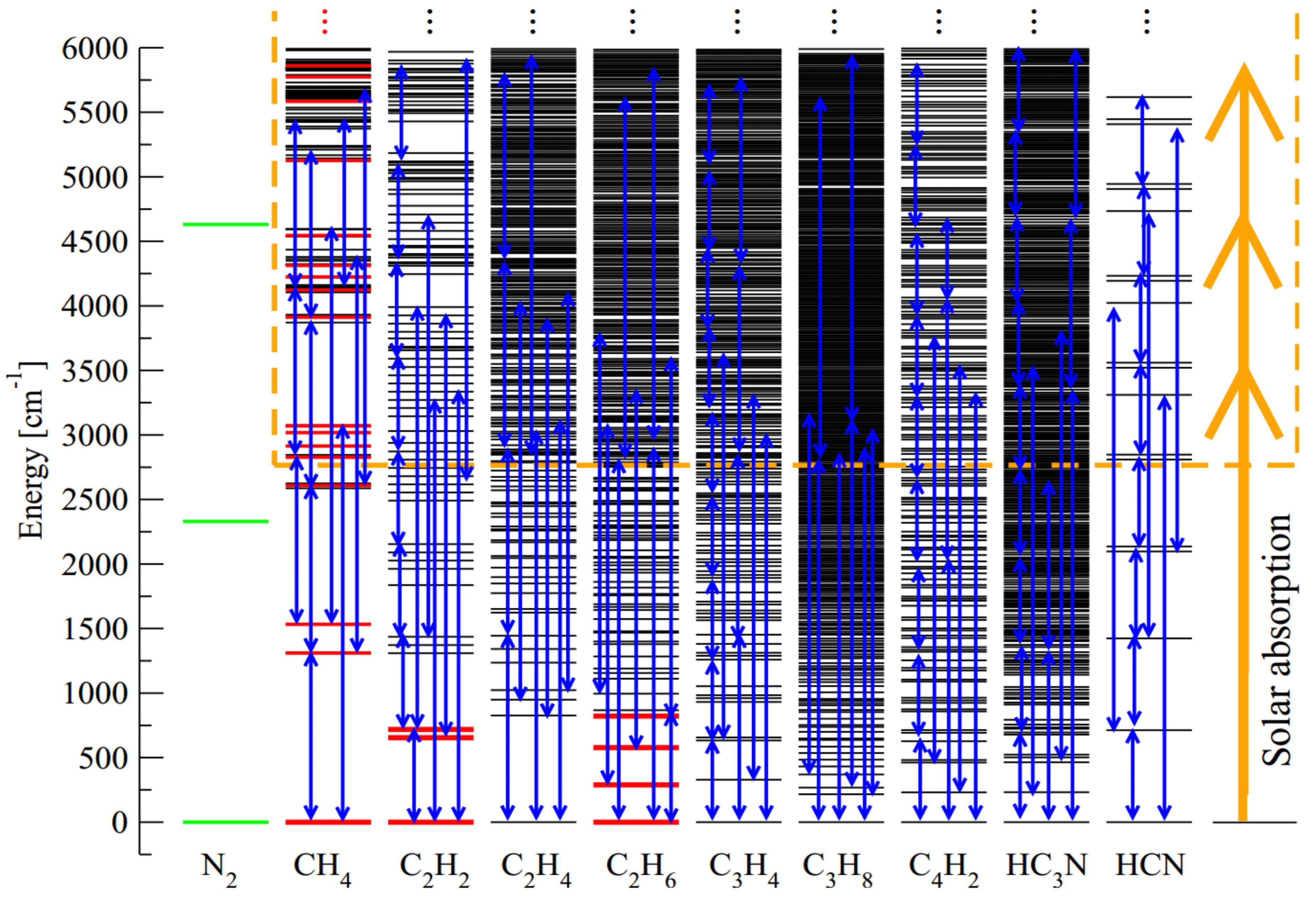


H_2O levels and transitions

Feofilov et al., 2009



Hydrocarbons in Titan's MUA



LTE breakdown for different species and atmospheres

- Earth:

  $CO_2(\nu_2)$ – 70 km; 15μm channel used for T retrieval
  $CO_2(\nu_3)$ – 40 km for the daytime; 4.3μm CO_2 retrieval
  $O_3(\nu_3)$ – 30 km daytime, 70 km nighttime; 9.6μm O_3 retr
  $H_2O(\nu_2)$ – 50 km, 6.3μm; solar pumped levels: down to 0km

- Mars:

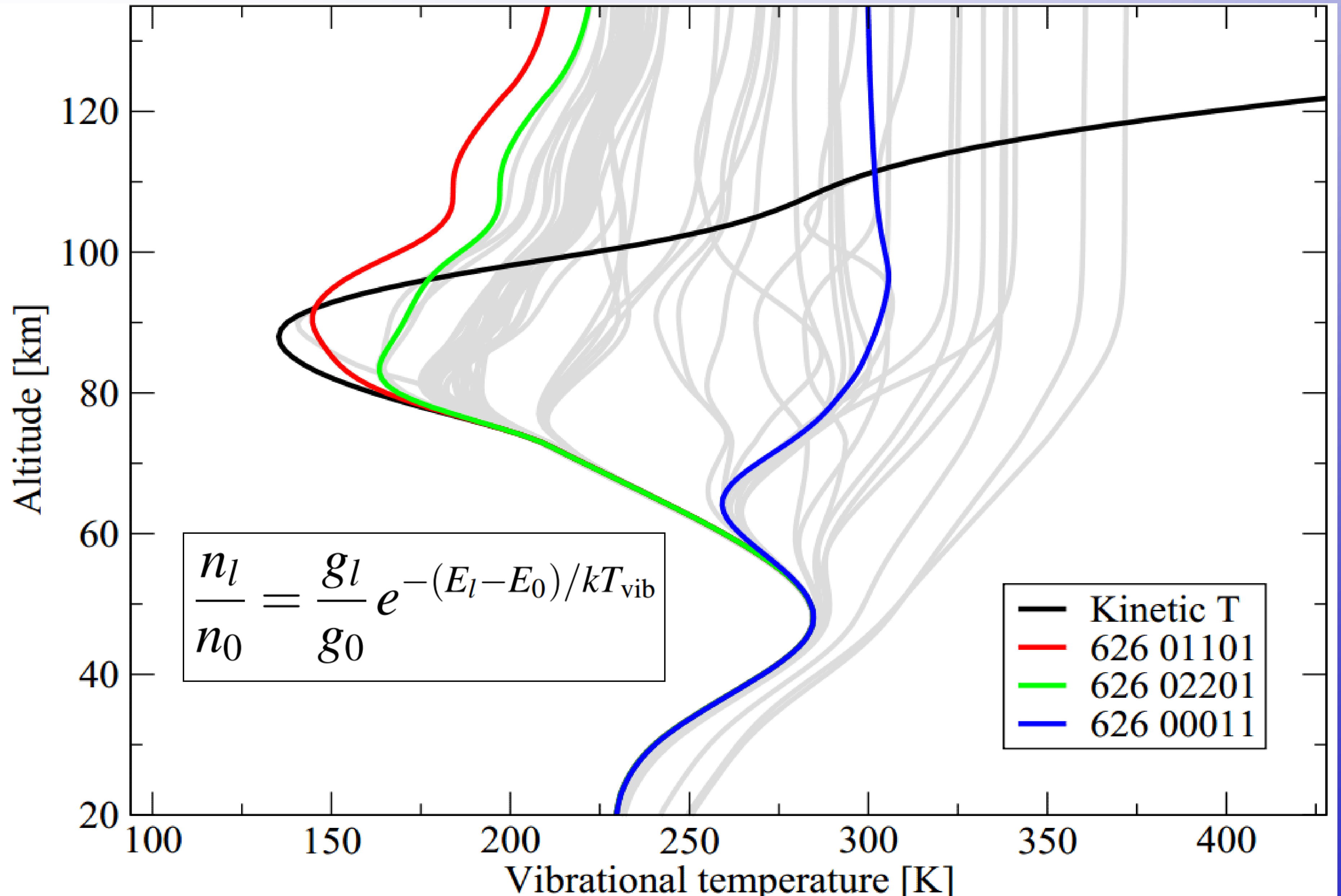
  $CO_2(\nu_2$, 15μm) – 80 km; 15μm channel used for T retrieval

- Titan:

  CH_4, C_2H_2, C_2H_6 – 450 km

- Rule of thumb: $P < 1$Pa – non-LTE

Example of LTE breakdown for CO_2 levels and T_{vib}



Radiative and collisional terms: uncertainties

$$n_l \sum (R_{ll'} + C_{ll'}) = \sum n_{l'} (R_{l'l} + C_{l'l}) + P_l - L_l$$

$$\sum n_l = n$$

Atmospheric applications:

- Calculating of radiative cooling/heating rates for GCMs
- Retrieval of pressure/temperature and trace gas concentrations from satellite observations

Main contributors:

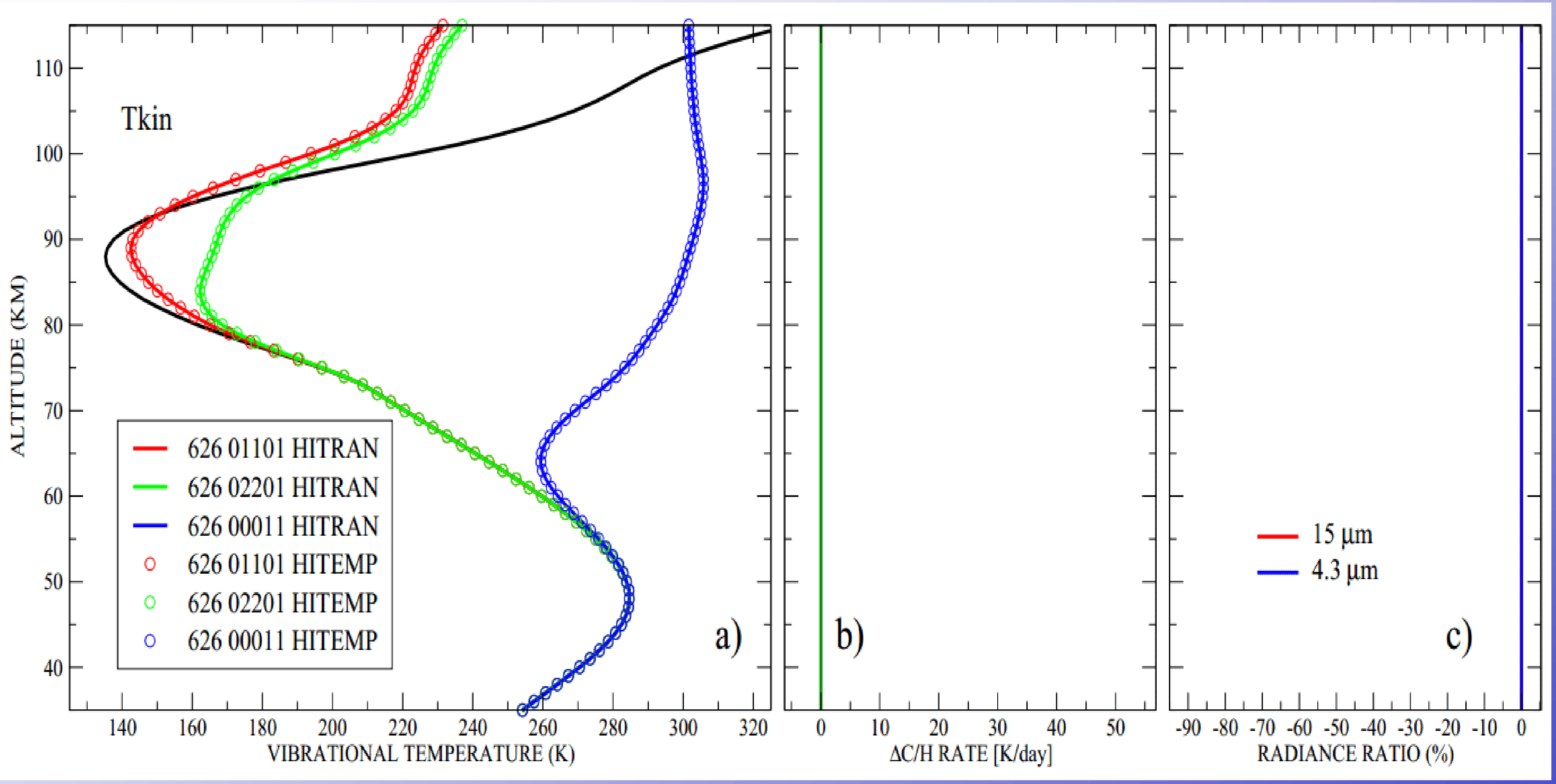
- Fundamental/first hot bands of main isotopes

Uncertainties:

- Radiative terms (solar absorption, IR emission)
- Collisional terms (vibration↔vibration, vibration↔translation)

CO_2

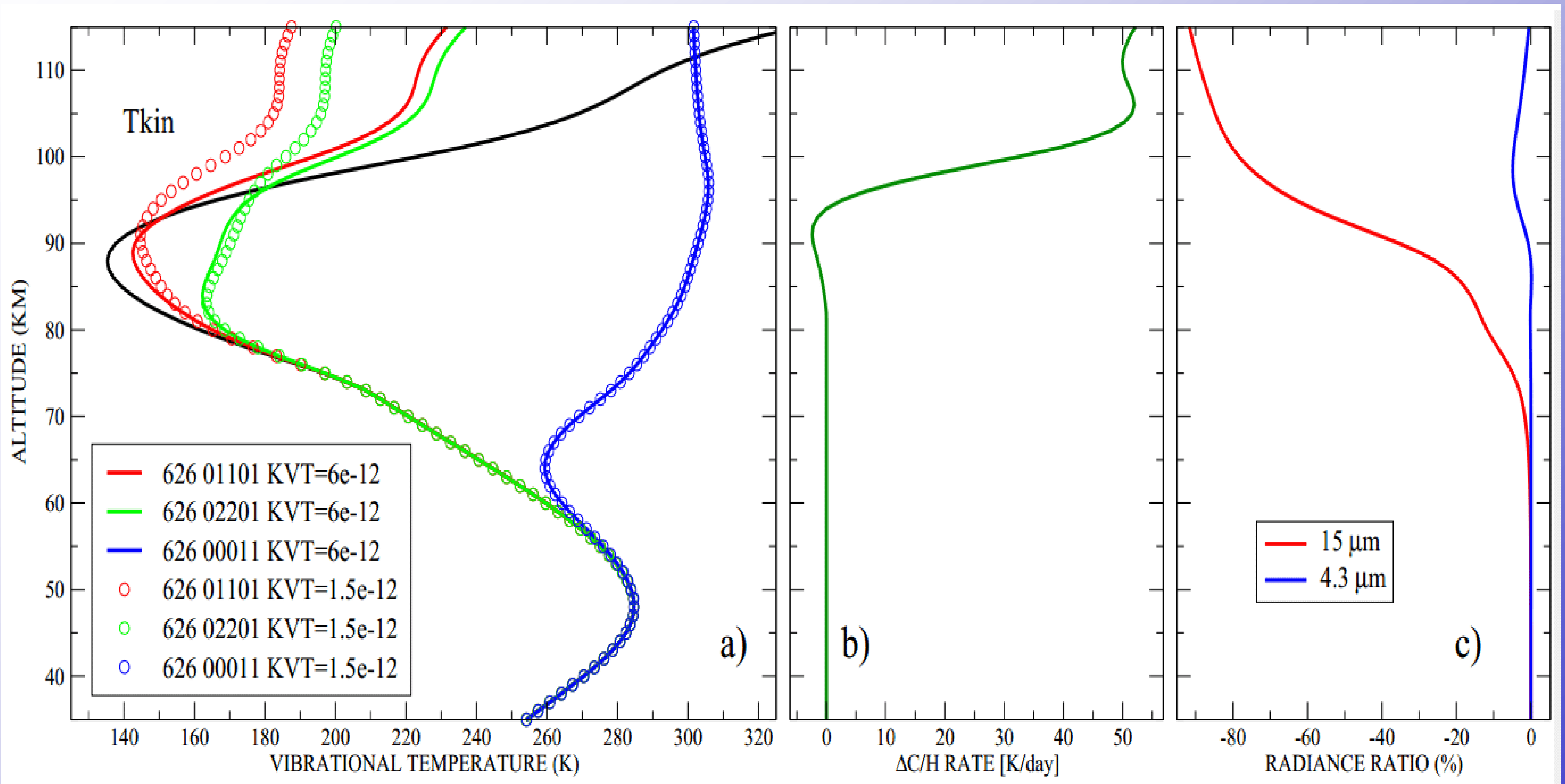
Uncertainties in radiative terms, $R_{ll'}$



- Changing the number of ro-vibrational lines from 20000 (HITRAN) to 40000 (HITEMP) does not affect low ν_2- and ν_3- level populations
- Cooling/heating rates and 15 µm and 4.3 µm radiances remain the same. No changes in GCM or retrieval is expected.

CO_2

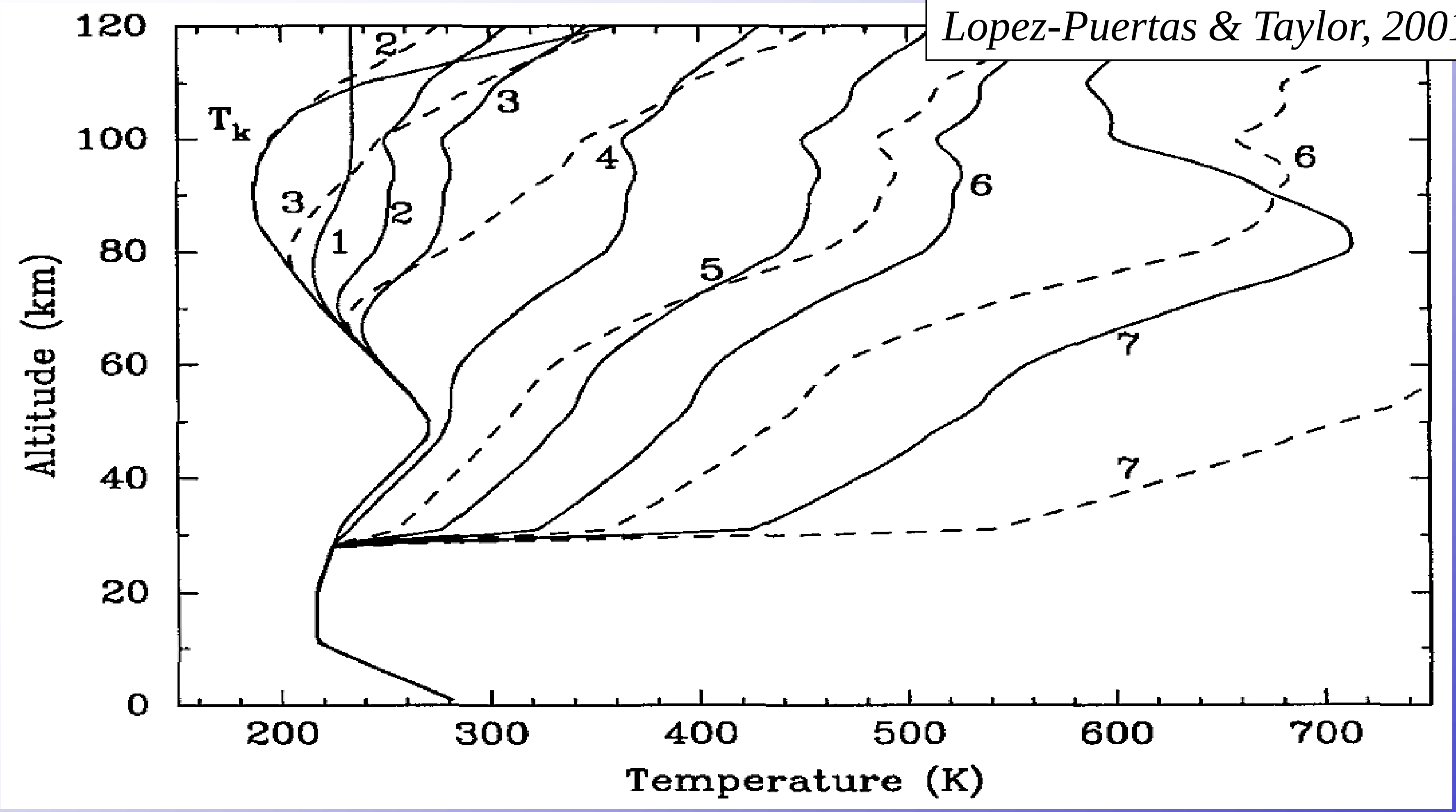
Uncertainties in quenching rate coefficient, $C_{ll'}$



- Current uncertainty in $k_{VT}(CO_2(v_2)\text{-}O)$ is a factor of 3-4 [Feofilov et al, 2012]
- This uncertainty affects T retrievals above ~60km and C/H rates above ~80km
- Laboratory measurements do not match atmospheric observations.

O_3

Uncertainties in nascent population model, P_l

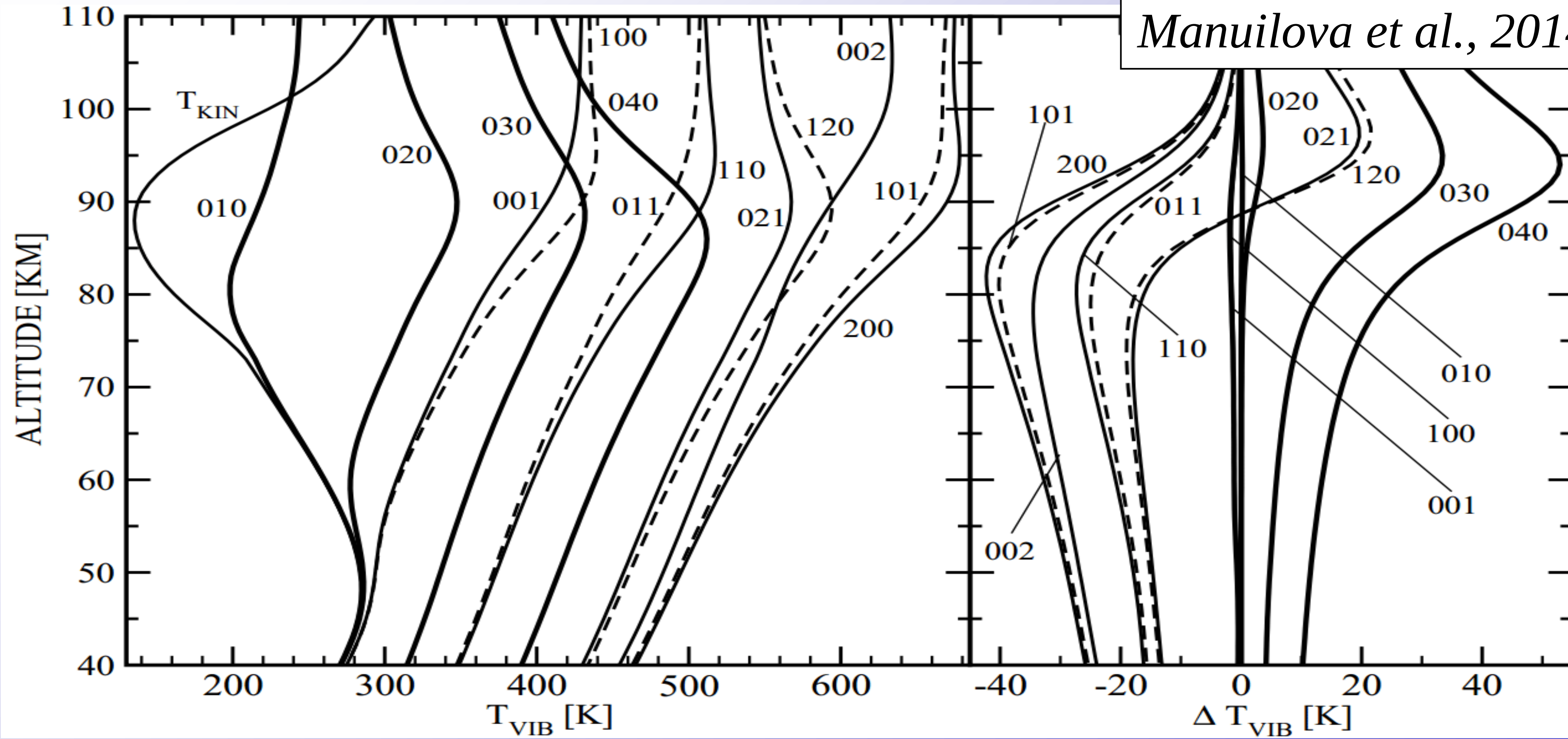


- Three-body recombination reaction $O_2+O+M \rightarrow O_3(\nu_1, \nu_2, \nu_3)+M$
- Where does the energy excess go? ($E_{vib}+E_{transl} = 8900 cm^{-1}$)
- "Zero-surprisal" (solid) vs "Top-8" (dashed) model plot shows the sensitivity of daytime ν_3 level population to nascent distribution.

H_2O

Uncertainties in $C_{ll'}$

Manuilova et al., 2014



- 1) $H_2O(\nu_1; \nu_2; \nu_3) + O_2(0) \leftrightarrow H_2O(\nu_1; \nu_2{-}1; \nu_3) + O_2(1)$

  2) $H_2O(\nu_1; \nu_2; \nu_3) + M \leftrightarrow H_2O(\nu_1; \nu_2{-}1; \nu_3) + M$

  3) $H_2O(\nu_1; \nu_2; \nu_3) + M \leftrightarrow H_2O(\nu_1{-}1; \nu_2{+}2; \nu_3) + M$

  4) $H_2O(\nu_1; \nu_2; \nu_3) + M \leftrightarrow H_2O(\nu_1; \nu_2{+}2; \nu_3{-}1) + M$
- The rate constants for 3) and 4) are known only for (001,100 →020).
- Manuilova et al., [2014] have estimated ([Barnes et al., 2004]) the rate constants for #3 and #4 for the upper vibrational levels to be **4 times larger** than currently used (!)

Conclusions and suggestions

- ✓ The vibrational level populations of atmospheric molecules are not in LTE at pressures lower than ~1 Pa
- ✓ This affects both radiative cooling/heating rates calculation and interpretation of satellite observations in the infrared
- ✓ The non-LTE codes can treat this physics given that the set of spectroscopic and collisional parameters is given.
- ✓ Accuracy and completeness of the spectroscopic parameters is sufficient for most of planetary atmosphere applications.
- ✓ Some crucial rate coefficients are known with large uncertainty
- ✓ The effects in observations can propagate downwards (!)
- ✓ GEISA community expertise in quantum mechanic calculations is highly welcome.