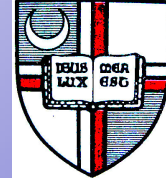
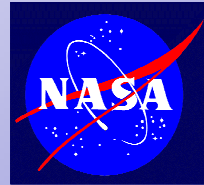


Infrared radiation of ozone in the mesosphere and lower thermosphere: energetic effects and remote sensing

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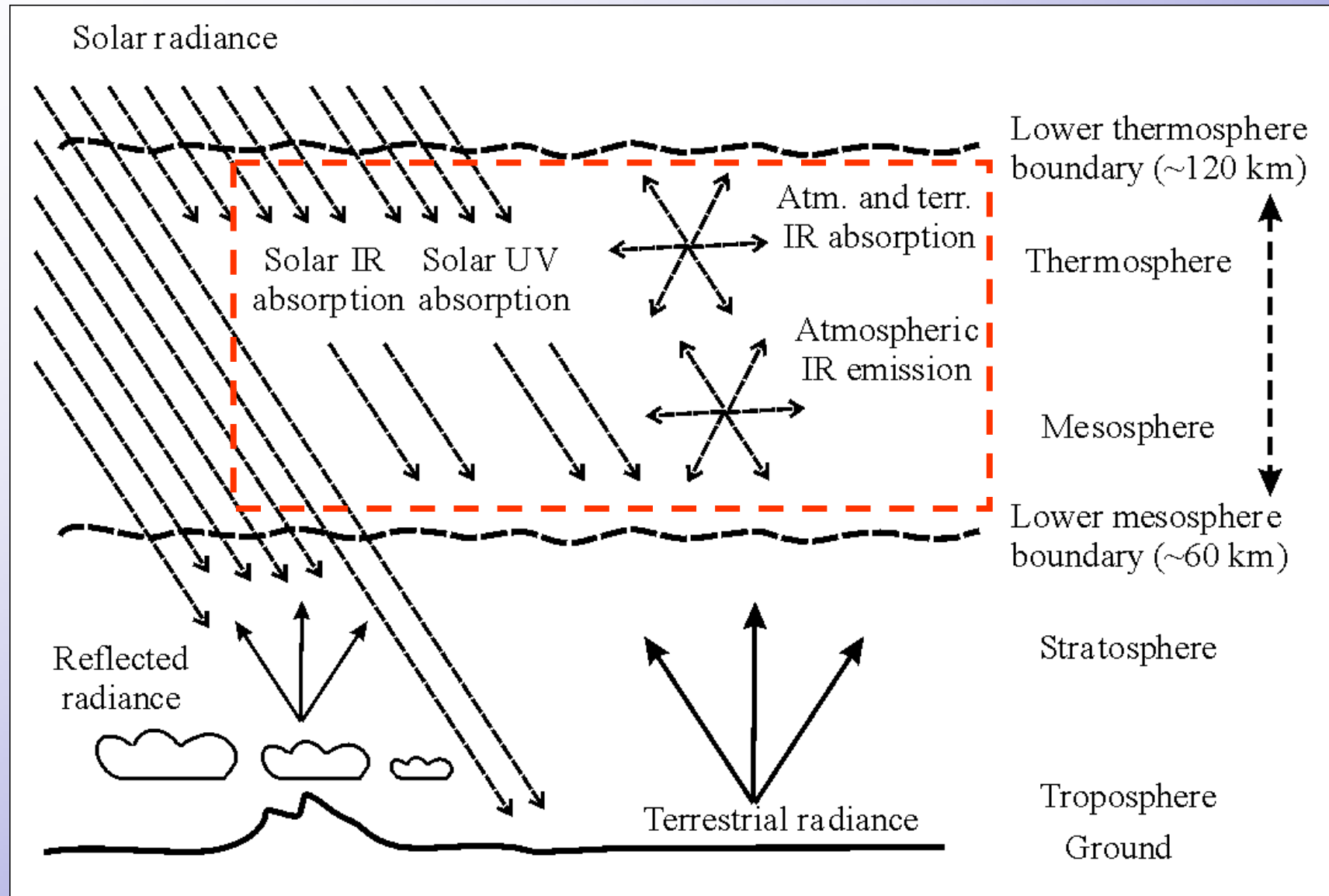
Ozone Workshop, October 2-4, 2013, Reims

MLT - why bother?

- MLT is a “gateway” between the lower atmosphere and space.
- MLT is sensitive to solar influence and to the inputs from below.
- Anthropogenic changes in greenhouse gases may change this input.
- Noctilucent clouds observed in polar MLT in the summer time are very sensitive to temperature changes and there are debates on their role as a “miner’s canary” for global changes.
- MLT area absorption and emission in molecular bands affects the atmospheric observations of other areas.



Object of Study: IR radiance in the MLT



Cooling/heating rates

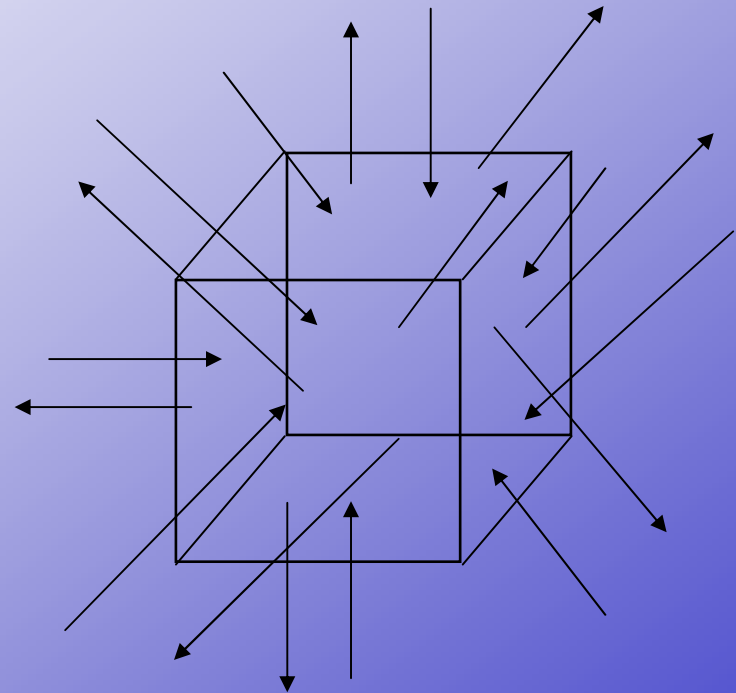
Radiative cooling/heating = radiative flux divergence

in W/m³:

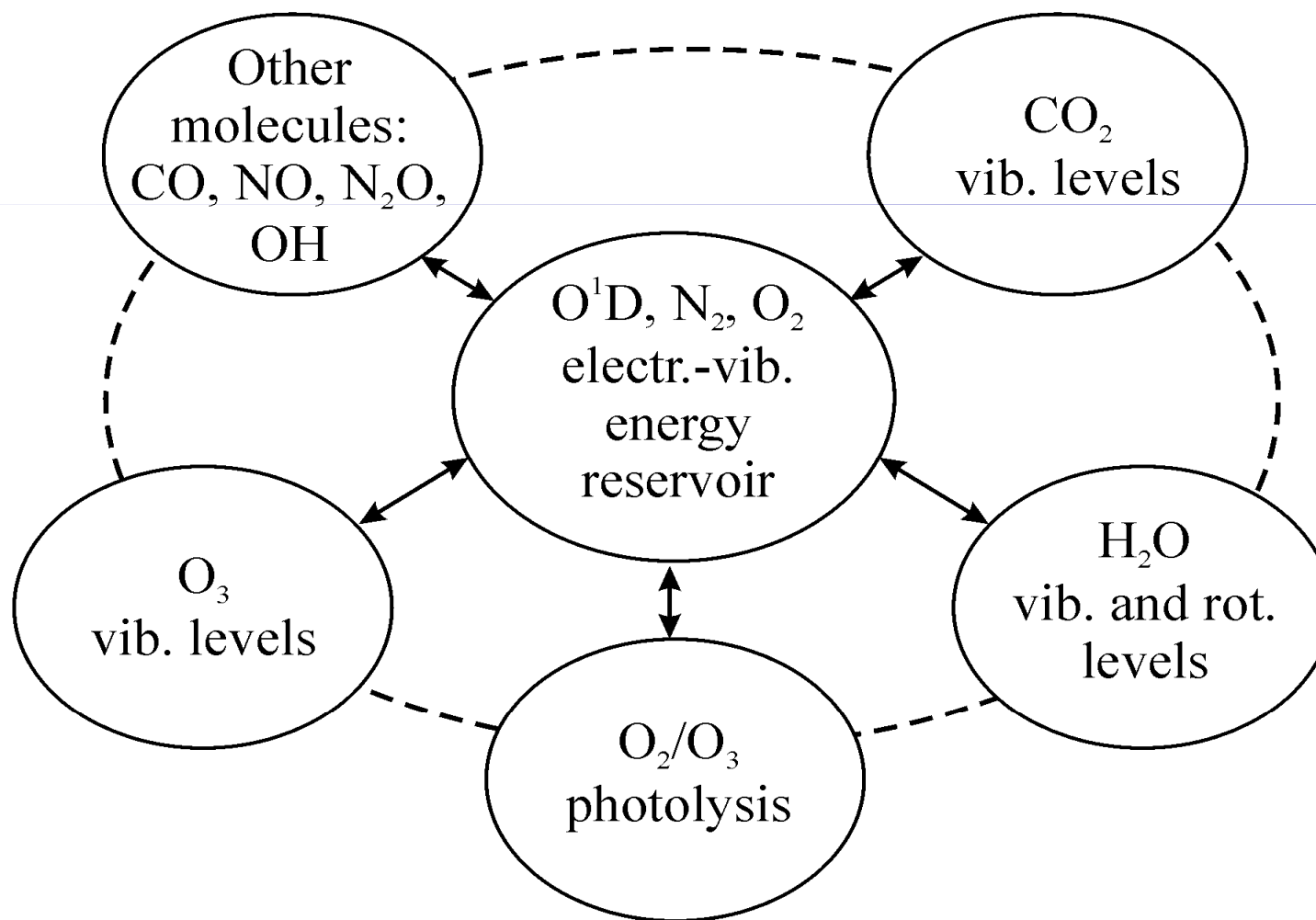
$$H(z) = \frac{1}{4\pi} \int_{\Omega} \int_{-\infty}^{+\infty} \mu \frac{dI_{\mu\nu}(z)}{dz} d\nu$$

in K/day:

$$h(z) = H(z) \cdot \frac{24 \cdot 60 \cdot 60}{C_p(z) \rho(z)}$$



Energy Exchange Between Atmospheric Molecules



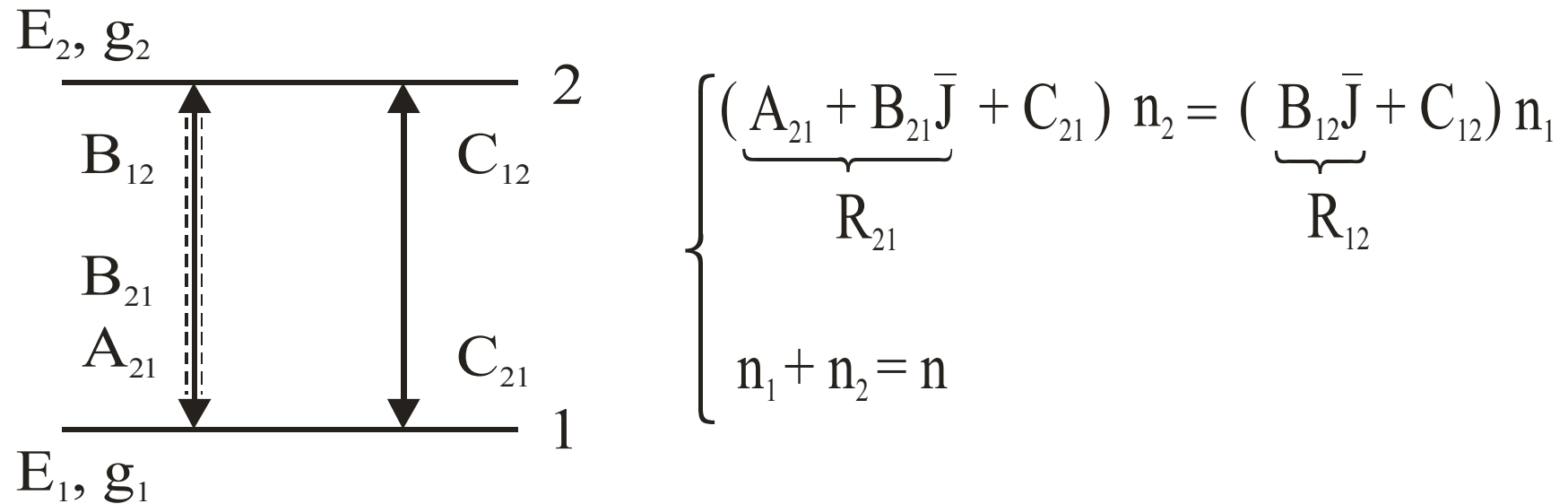
An important peculiarity of the MLT

- 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 MLT 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



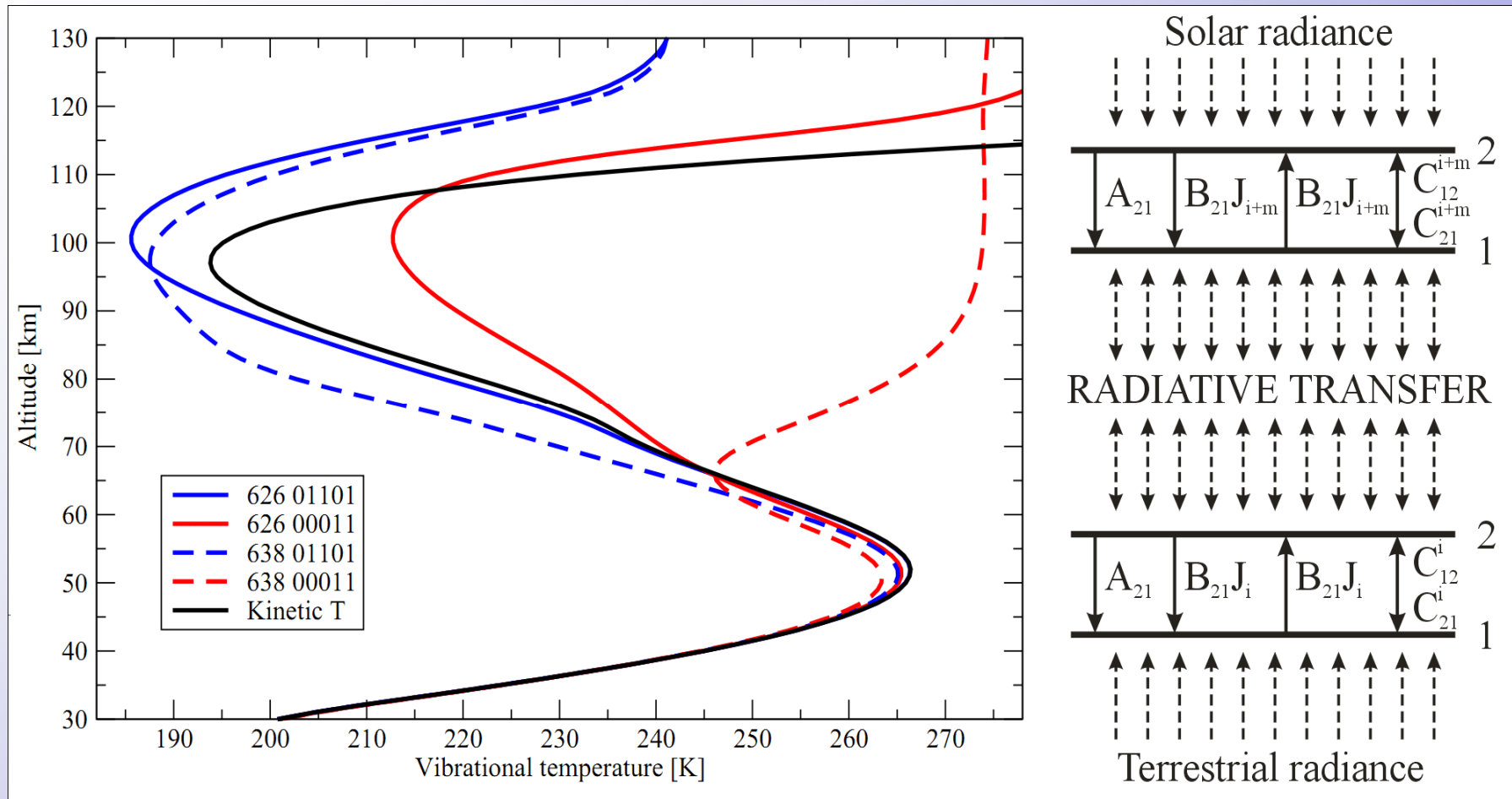
In the lower atmosphere:

$$R \ll C \quad R_{21}n_2 \sim R_{12}n_1 \quad n_2 C_{21} = n_1 C_{12} \quad \frac{n_2}{n_1} = \frac{g_2}{g_1} e^{-\frac{E_2 - E_1}{kT}}$$

In the upper atmosphere:

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

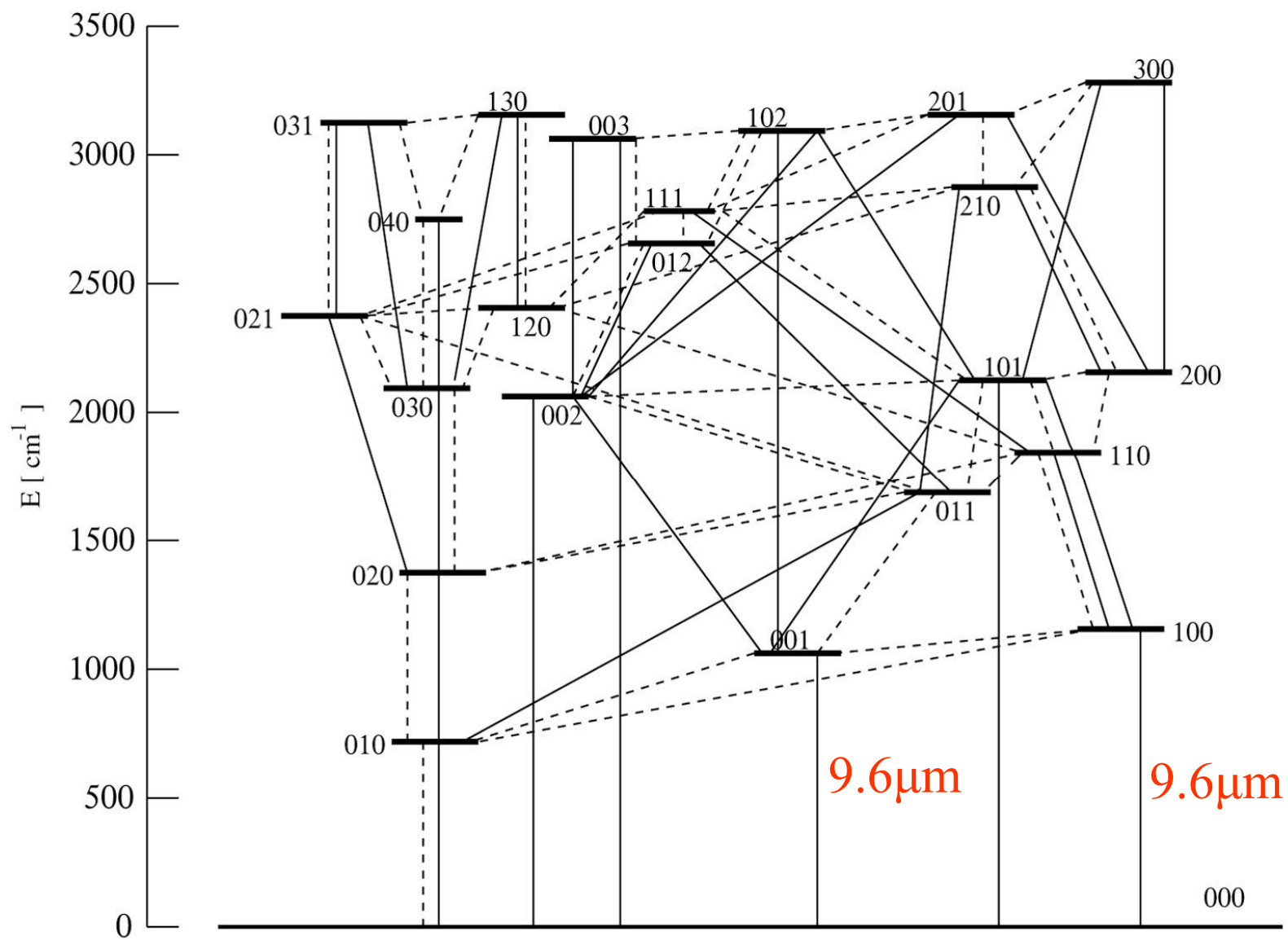
Non-LTE populations: vibrational temperatures



$$\frac{n_l}{n_0} = \frac{g_l}{g_0} \cdot e^{-\frac{(E_l - E_0)}{kT_{vib}}}$$

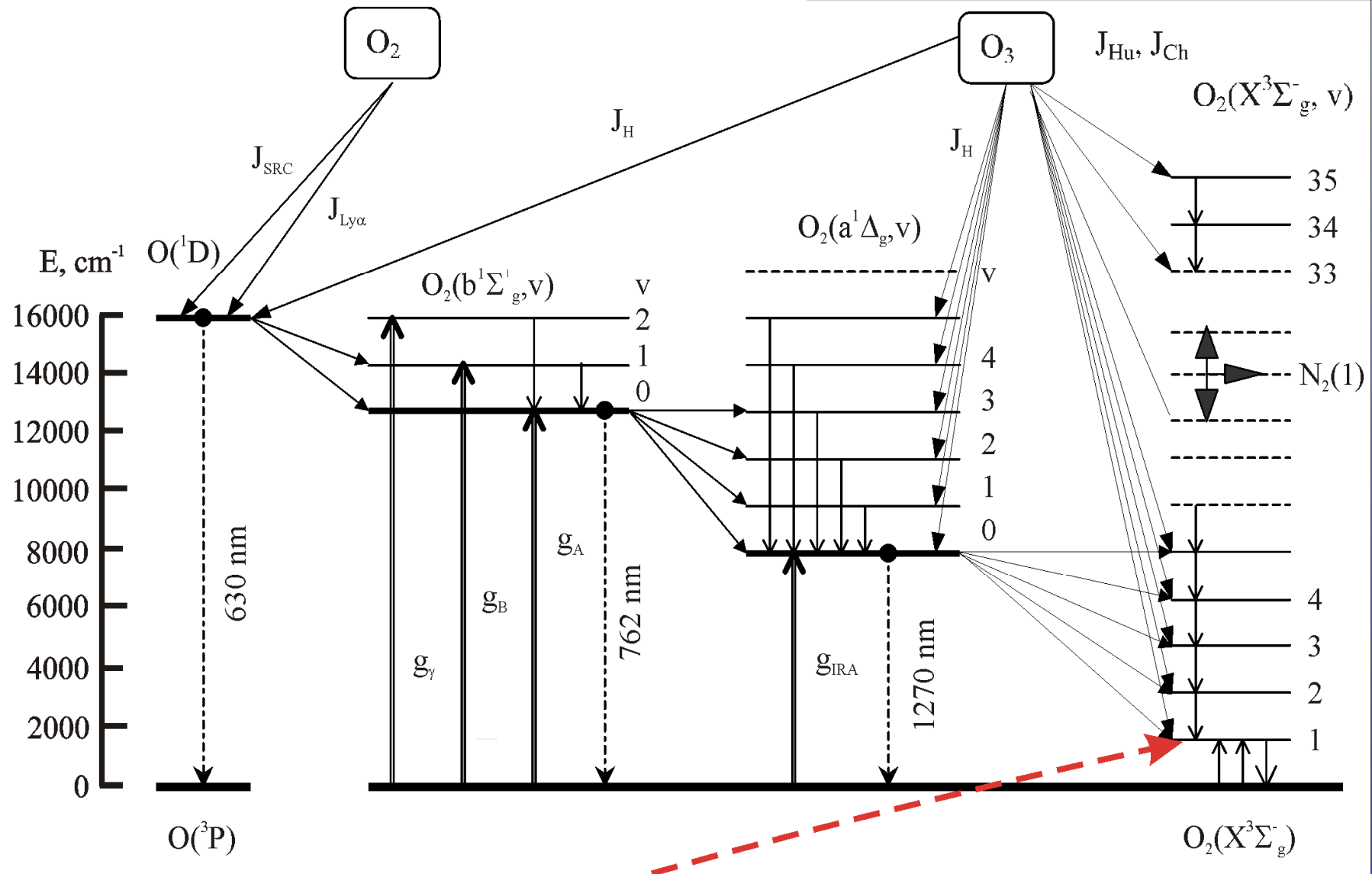
$$\begin{aligned} T_{vib} &= T_{kin}: \text{LTE} \\ T_{vib} &\neq T_{kin}: \text{non-LTE} \end{aligned}$$

O_3 levels and transitions



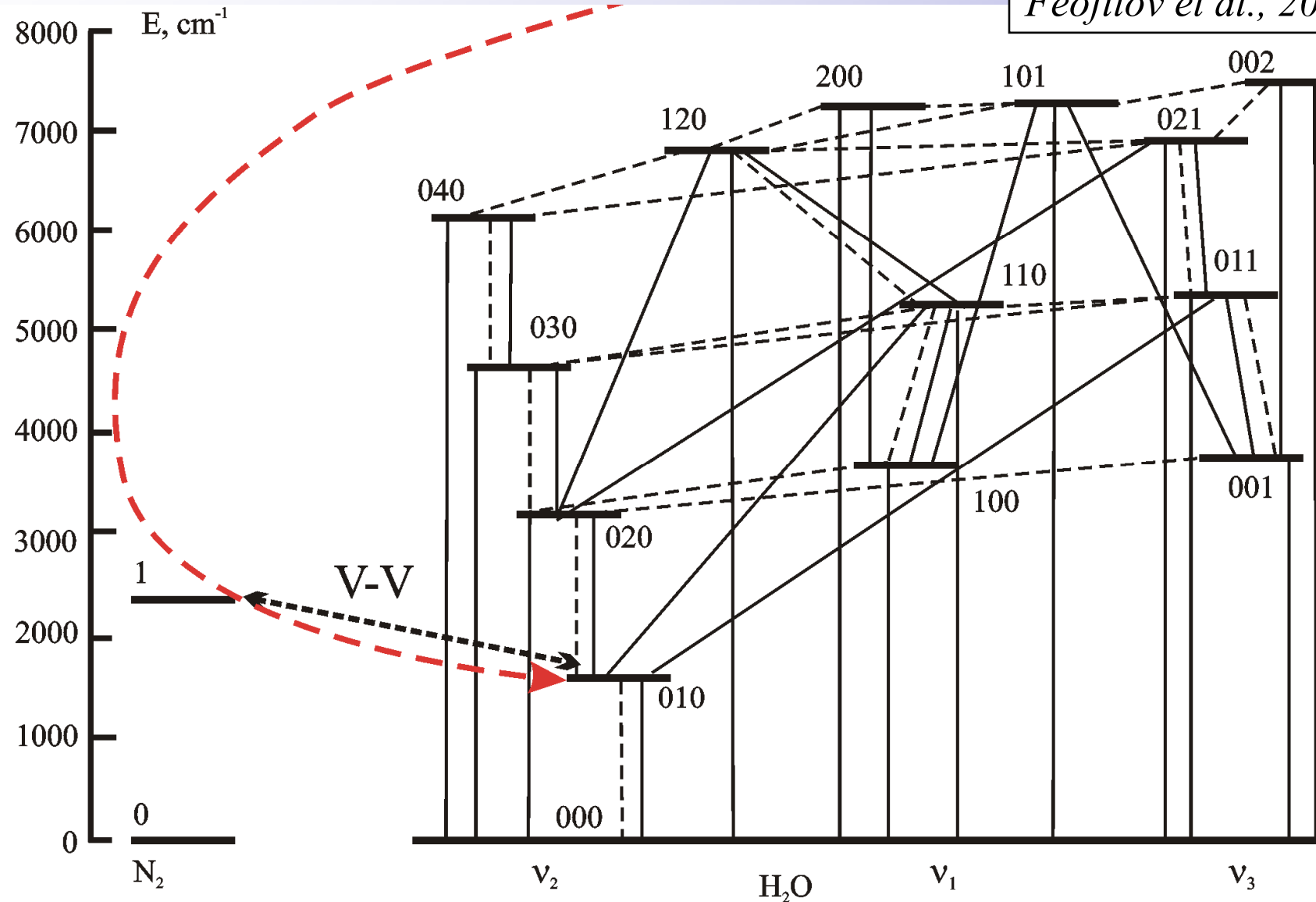
O_2/O_3 photolysis products

Yankovsky and Manuilova, 2006



H_2O levels and transitions

Feofilov et al., 2009



Energy exchange processes for O_3



Intra-molecular
energy transfer



Vibrational-
translational
exchange



Vibrational-
vibrational
exchange

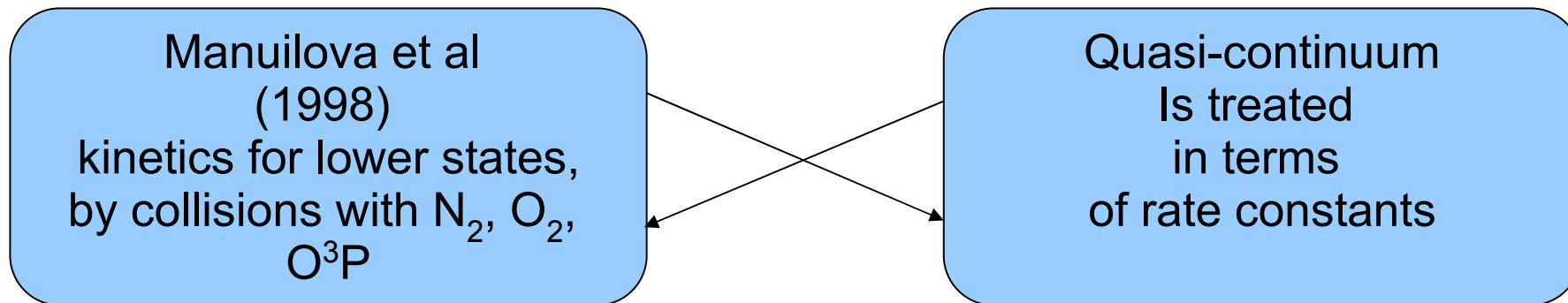


Three-body
recombination

Plus emission / absorption of radiation

Three-body recombination and nascent population model

A hybrid of models



A) Gil-Lopez et al., (2005): Zero surprisal model of nascent population, E_D is the dissociation energy

$$P(v) = (1 - E(v)/E_D)^{1.5} / \sum_v (1 - E(v)/E_D)^{1.5}$$

B) Kaufmann et al. (2006): Zero surprisal model or all excitation goes on 00v3, with v3=3,5 or 8

C) Fernandez et al. (2009, 2010): single level nascent population at O₃ (006), assuming that about 70% of recombination energy goes to vibrations

An inconsistency between O_3 retrieved from 9.6 μm and simulated/measured 4.8 μm radiance

4.8 microns emission
from 200, 101, 002
for known O_3
was 2-3 times
lower than measured
for ~ 75 km

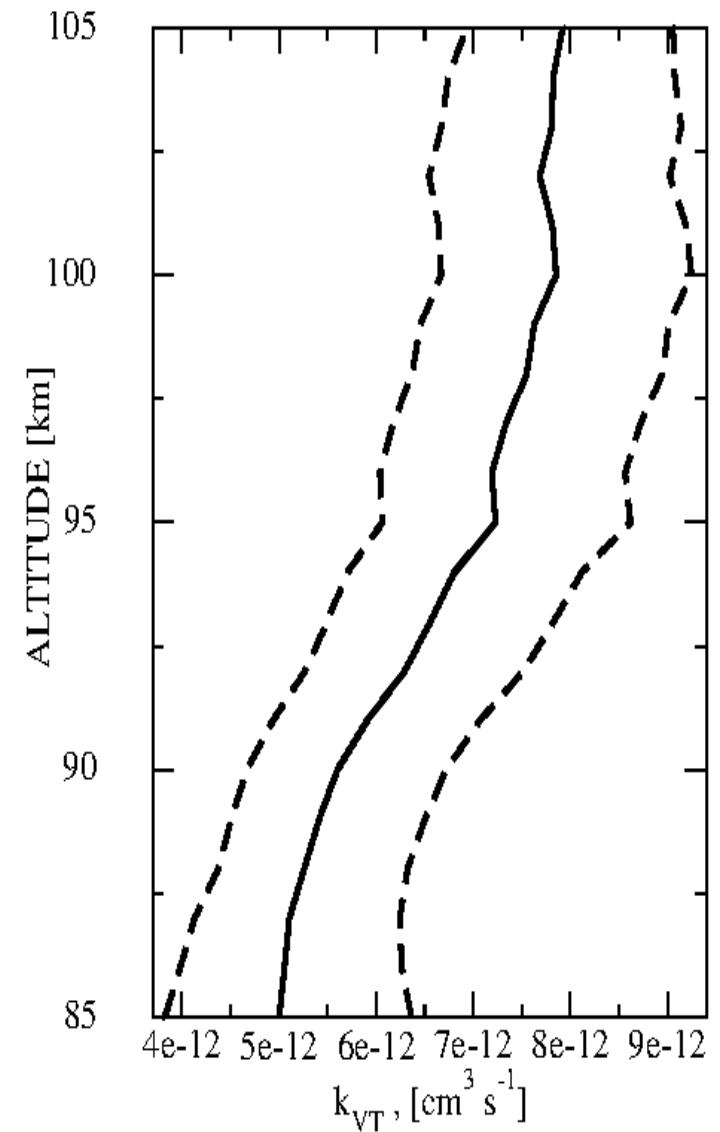
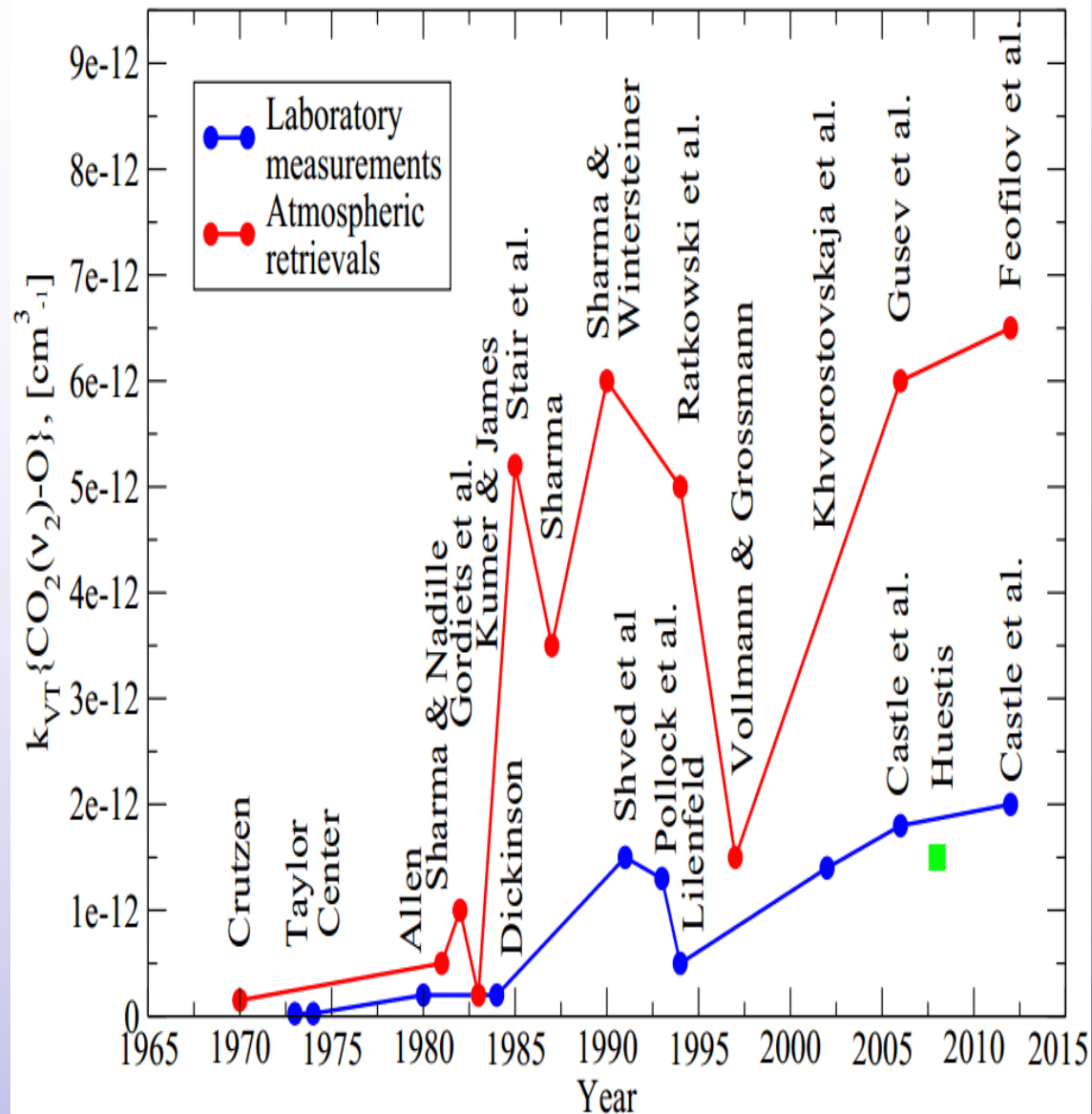
Kaufmann et al, 2006:

**The rate of stretching to bending mode transition
001,100- \rightarrow 010 K_{D_2} must be 3-4 lower
than well known measured
value**

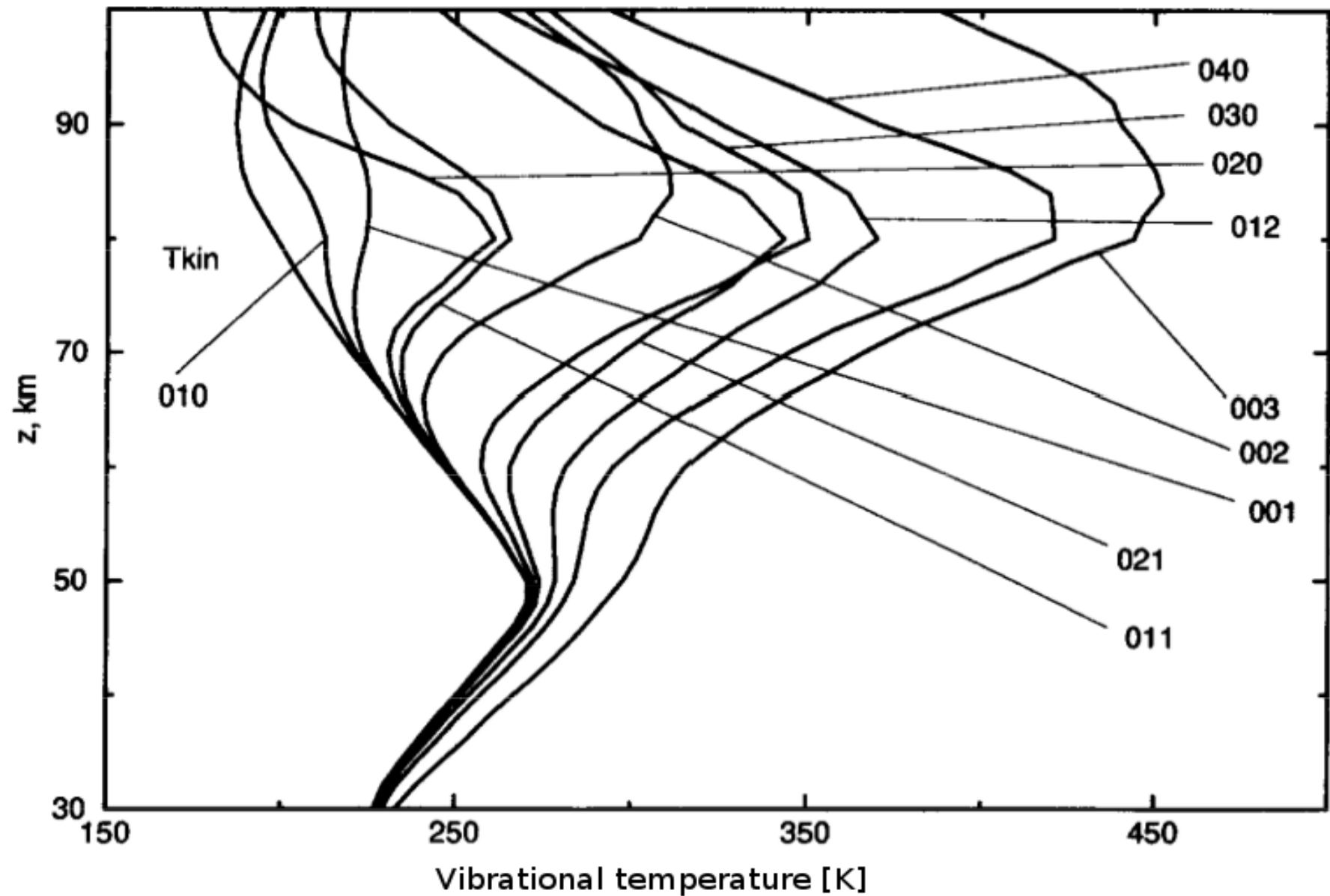
Fernandez et al., 2010:

**new bending to stretching mode transitions,
for lower levels 020- \rightarrow 100,001 SSH estimated K_{2D}
For higher levels – harmonic oscillator rule**

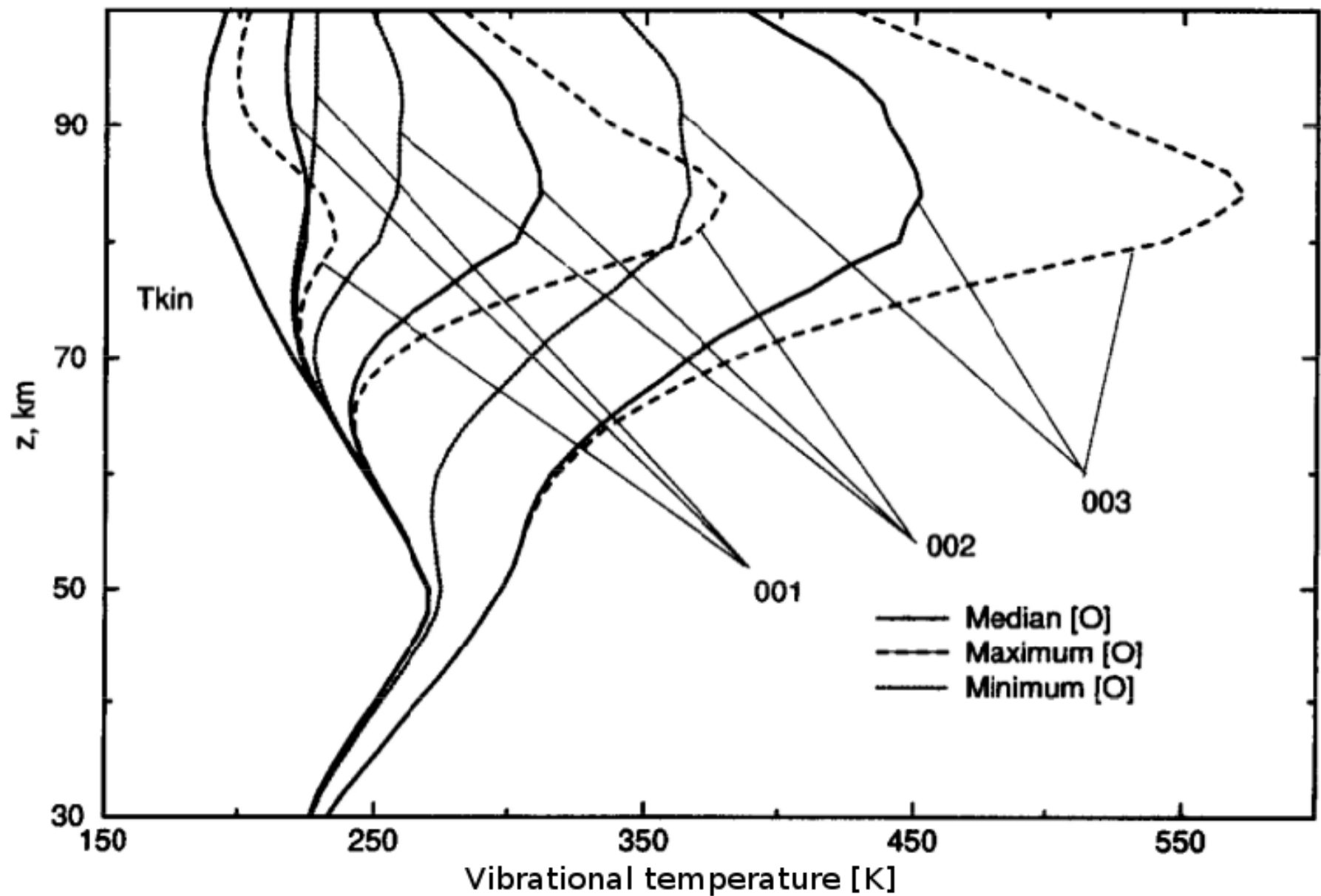
$k_{VT}\{CO_2-O\}$ measurements: lab vs atmosphere



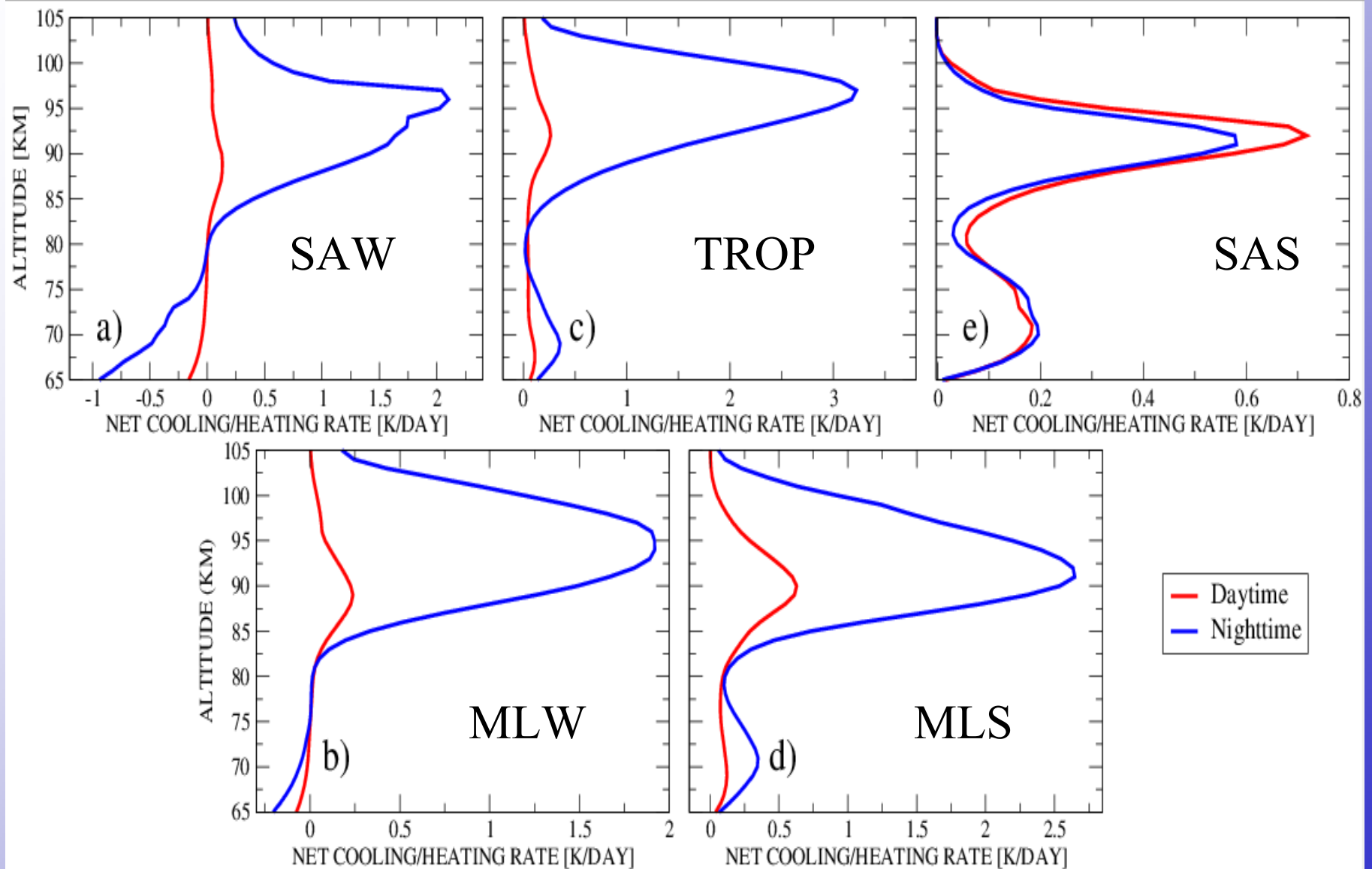
O_3 vibrational level populations – midlatitude day



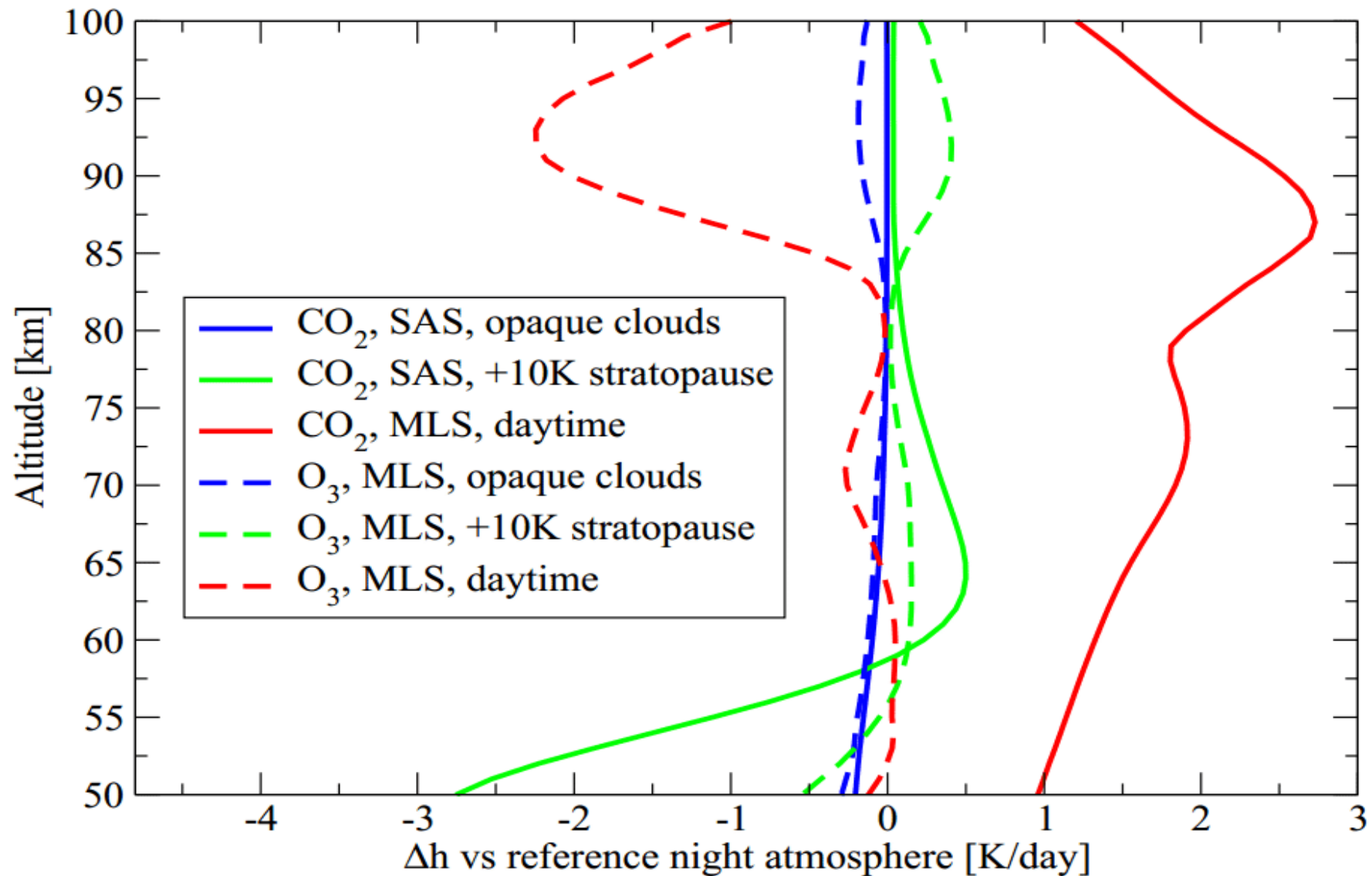
Sensitivity of ν_3 -levels to atomic O



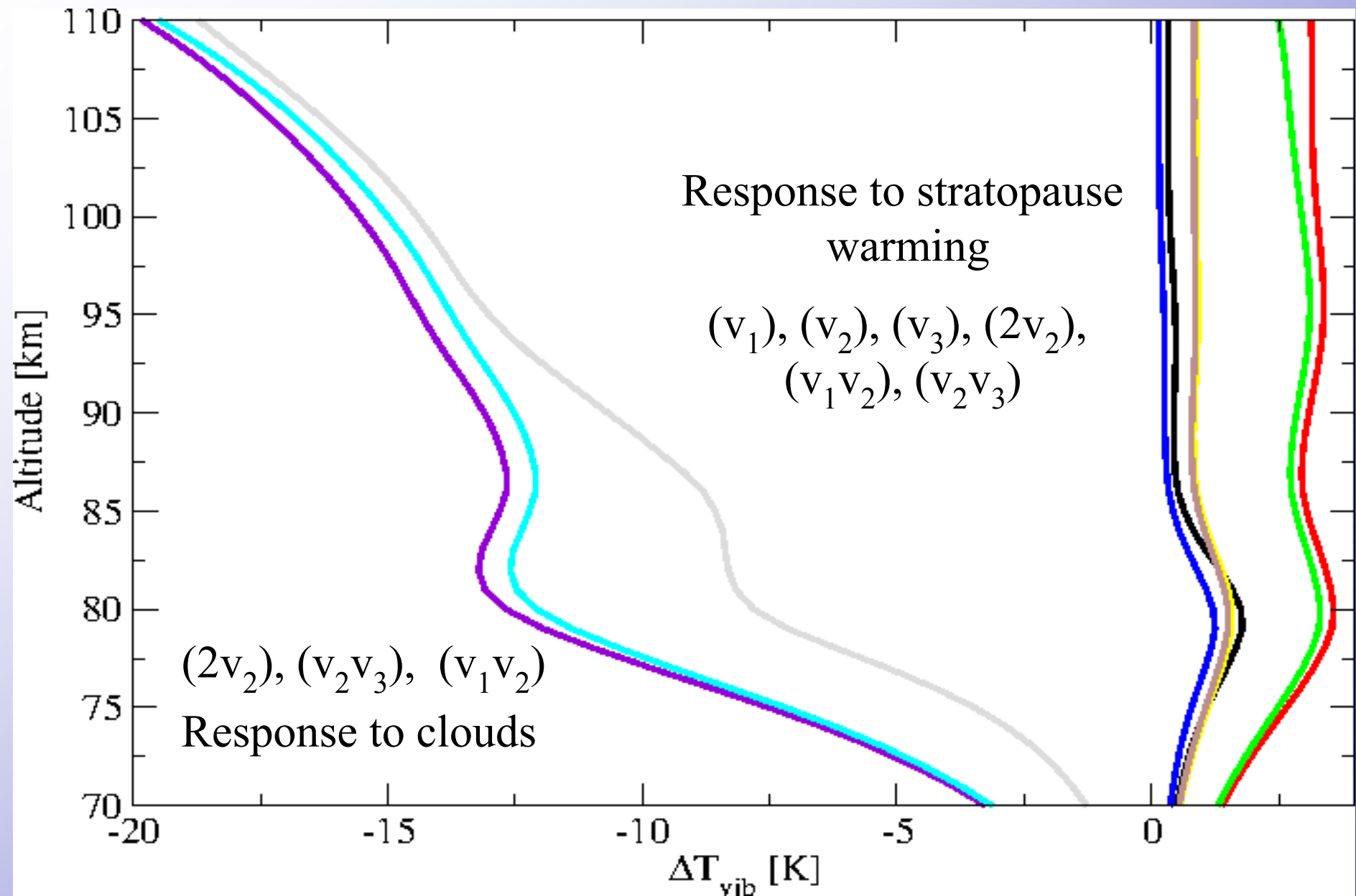
O_3 cooling/heating rates



MLT sensitivity to clouds, stratopause T, and solar pumping changes



O_3 vibr. levels, sensitive to changes in other layers



Take home messages

- ✓ O_3 is an important component of the MLT
- ✓ $O_3(9.6\mu m)$ IR cooling/heating in 65-105 km altitude range is -1...+3 K/day
- ✓ Energetic effects of direct radiative coupling with tropo- and stratosphere are less than 1 K/day
- ✓ Changes in lower atmosphere affect vibrational levels, which are pumped by radiation coming in optically thin lines.
- ✓ Adequate model of nascent population of O_3 molecule formed in three-body recombination is still required
- ✓ Collisional rate calculations are needed.