“Altitude Variation” of the $CO_2(\nu_2)$-O Quenching Rate Coefficient in Mesosphere and Lower Thermosphere

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Formulation of the problem

The atmospheric cooling in mesosphere and lower thermosphere is dominated by CO_2 15 μm emission:

First, CO_2 molecule is collisionally excited:

$$CO_2\ (\nu_1,\nu_2,\nu_3) + O(^3P) \leftrightarrow CO_2\ (\nu_1,\nu_2+1,\nu_3) + O(^3P) \quad (1)$$

then it emits a 15 μm quantum:

$$CO_2\ (\nu_1,\nu_2+1,\nu_3) \rightarrow CO_2\ (\nu_1,\nu_2,\nu_3) + h\nu\ (667\text{cm}^{-1})\ (2)$$

The probability of (1) is defined by $k_{VT}\{CO_2\text{-}O\}$ rate coefficient…

But !!! (see the next slide) →

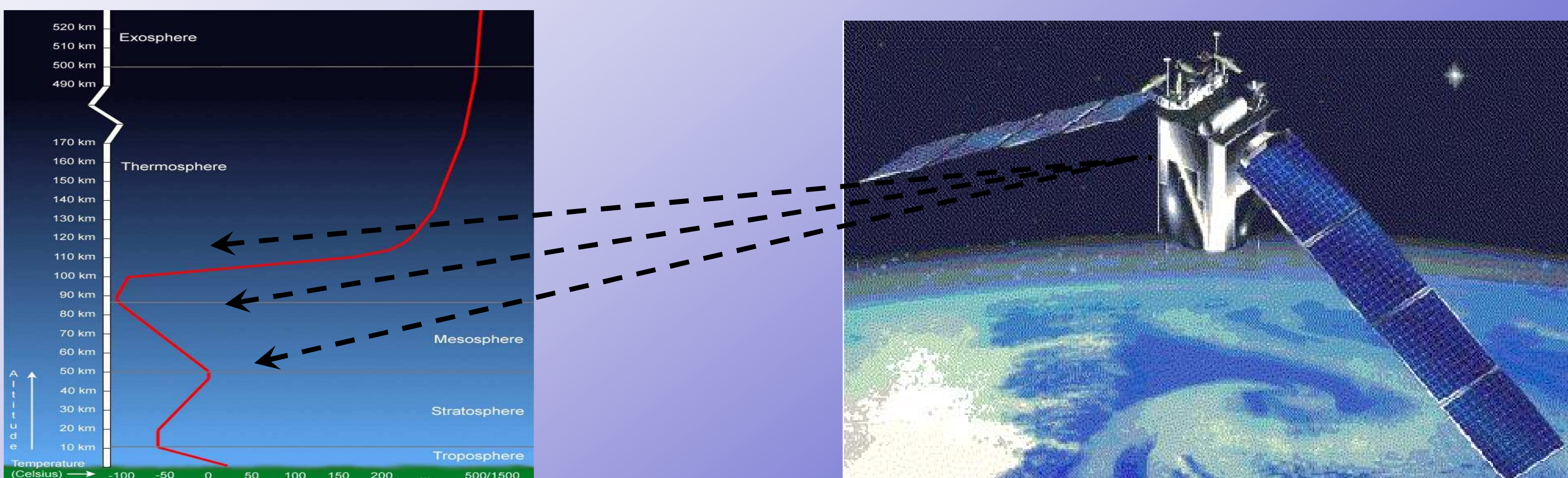
$k_{VT}\{CO_2\text{-}O\}$ measurements and estimates

$k_{VT}\{CO_2\text{-}O\}$ [cm^3s^{-1}]	Reference	Comments
$3–30 \times 10^{-14}$	Crutzen, 1970	First guess
2.4×10^{-14}	Taylor, 1974; Center,1973	Laboratory measurements
5.0×10^{-13}	Sharma and Nadille, 1981	Atmospheric retrieval
1.0×10^{-12}	Gordiets et al., 1982	Numerical experiment
2.0×10^{-13}	Kumer and James, 1983	Atmospheric retrieval
2.0×10^{-13}	Dickinson, 1984; Allen, 1980	Laboratory measurements
5.2×10^{-12}	Stair et al., 1985	Atmospheric retrieval
3.5×10^{-12}	Sharma, 1987	Atmospheric retrieval
$3–9 \times 10^{-12}$	Sharma and Wintersteiner, 1990	Atmospheric retrieval
1.5×10^{-12}	Shved et al., 1991	Laboratory measurements
1.3×10^{-12}	Pollock et al., 1993	Laboratory measurements
5.0×10^{-12}	Ratkowski et al., 1994	Atmospheric retrieval
5.0×10^{-13}	Lilenfeld, 1994	Laboratory measurements
1.5×10^{-12}	Vollmann and Grossmann, 1997	Atmospheric retrieval
1.4×10^{-12}	Khvorostovskaya et al., 2002	Laboratory measurements
1.8×10^{-12}	Castle et al., 2006	Laboratory measurements
6.0×10^{-12}	Gusev et al., 2006	Atmospheric retrieval
1.5×10^{-12}	Huestis et al., 2008	Recommended value
4.0×10^{-12}	Dodd et al., 2009	Laboratory measurements

The values differ by factor of **3-4 (!)**

General idea

to minimize the difference between
simulated and measured radiances
by varying $k_{VT}\{CO_2\text{-}O\}$



simulated = calculated from the known atmospheric
profile (T(z), P(z), $[CO_2](z)$, [O](z))

General idea

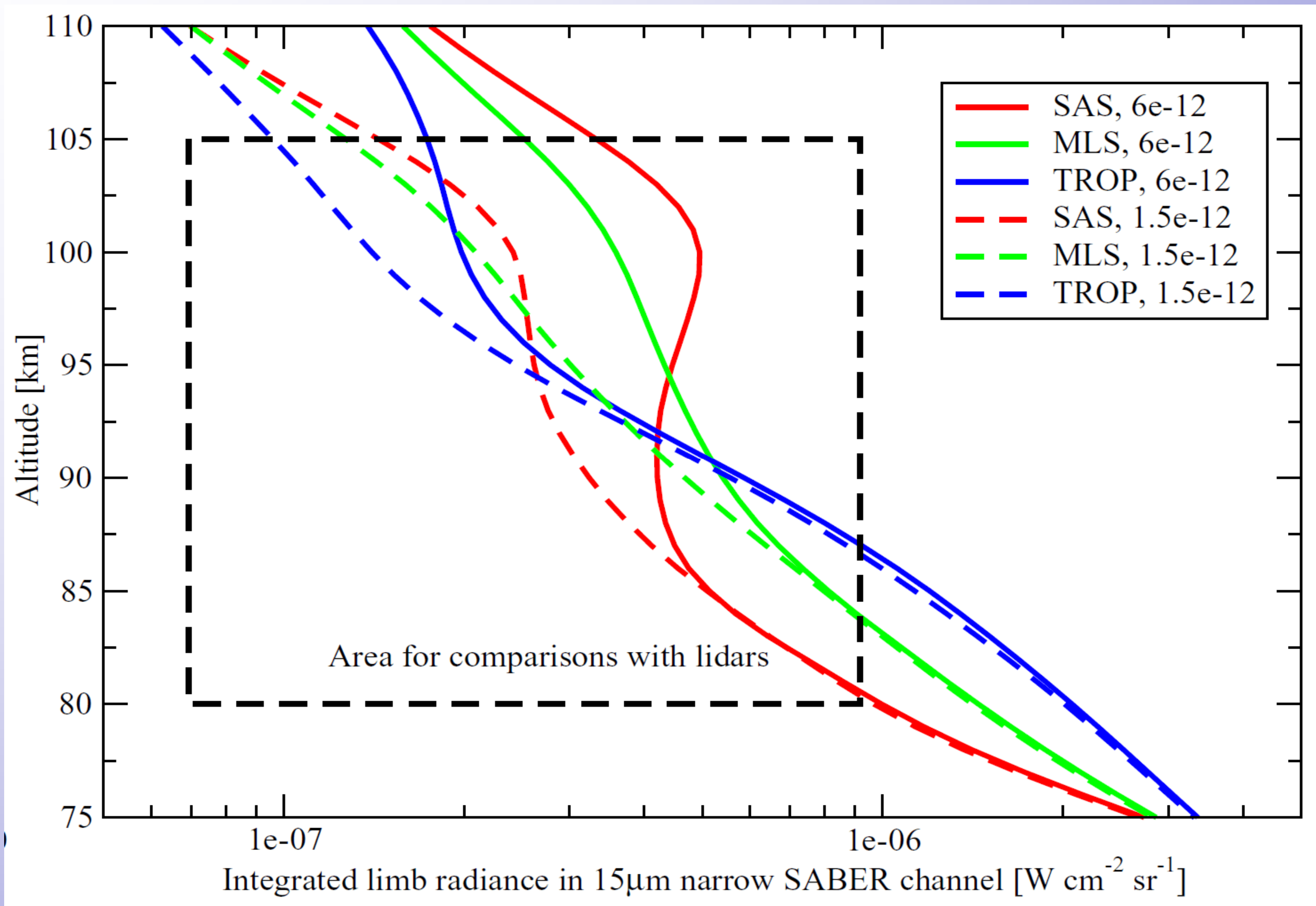
- $k_{VT}\{CO_2\text{-}O\}$ can be retrieved from 15 μm radiance.

- At each altitude $I_{15\mu m}$ is a function of T, P, $[CO_2]$, and [O].

- Due to non-LTE effects and atmospheric absorption, $I_{15\mu m}$ is a function of T(z), P(z), $[CO_2](z)$, [O](z).

- Knowing these ingredients → $CO_2(\nu_2)$ levels populations → $k_{VT}\{CO_2\text{-}O\}$ rate coefficient variation → → optimal $k_{VT}\{CO_2\text{-}O\}$.

Sensitivity study: k_{VT}=1.5x10^{-12} vs 6.0x10^{-12} cm^3s^{-1}



Datasets used in the study: Fort Collins lidar and SABER/TIMED

Colorado State University Lidar

- Sodium lidar, 80-110 km temperature measurements.
- Fort Collins, Colorado at (40.59N, 105.14W)
- In operation since 1990, since 2002 - both night- and daytime measurements.
- Two telescopes pointed at 30° off zenith east and north.
- 50 shots/sec; accumulation over 2 min intervals.

SABER instrument:

- Designed for studying Mesosphere/Lower Thermosphere
- Limb scanning infrared radiometer
- 10 broadband channels (1.27-17 μm)
- Products: P, T, CO_2, O_3, H_2O, NO, O_2, OH, NO, O, H
- Pressure/temperature retrieval depends on $k_{VT}\{CO_2\text{-}O\}$

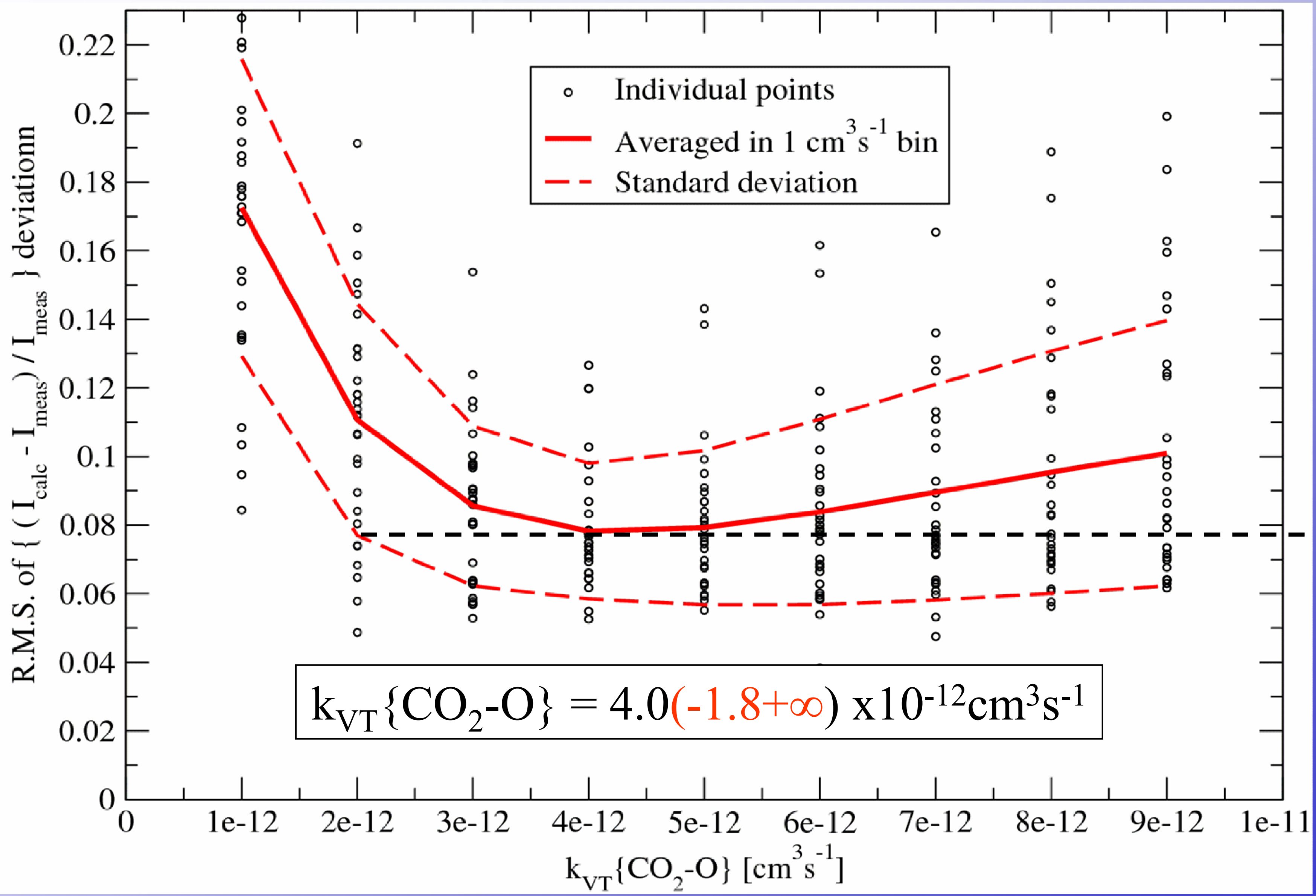
Retrieval approach

- Searching for simultaneous common volume measurements performed by SABER and lidar.
- Using lidar temperatures in 80-110km as reference data.
- Using SABER retrieved P(z), T(z) (<80km), CO_2(z), O(z).

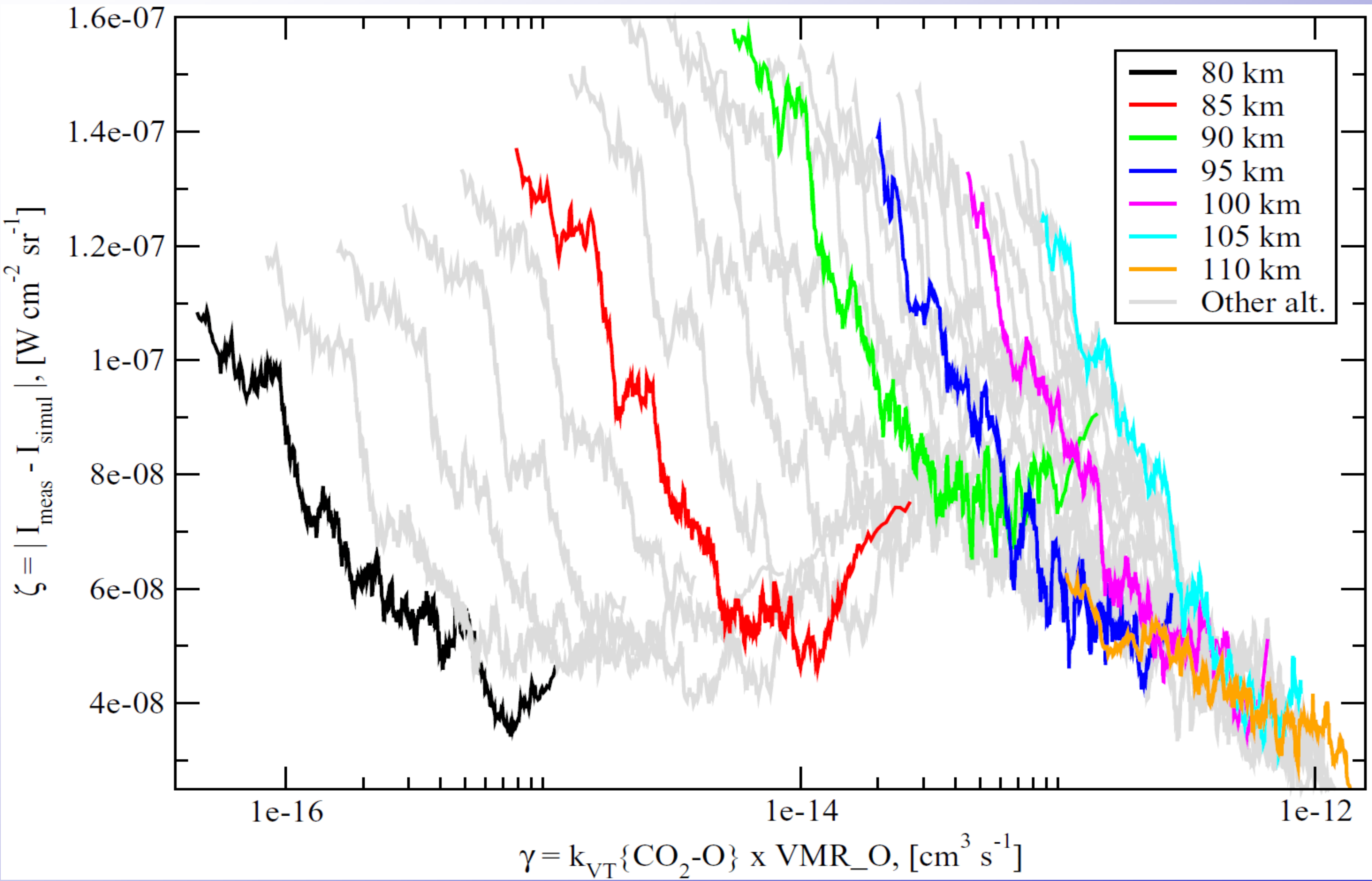
Treating all altitudes together, minimizing the difference
$I_{simul} – I_{meas}$
by varying $k_{VT}\{CO_2\text{-}O\}$
(AGU 2009)

Treating altitudes separately, minimizing the differences
$I_{simul}(z) – I_{meas}(z)$
by varying $k_{VT}\{CO_2\text{-}O\}(z)$
(AGU 2010)

Treating all altitudes together: large uncertainty



Treating altitudes separately



γ is a combined variable. Individual [O](z) profiles are not involved here (!)

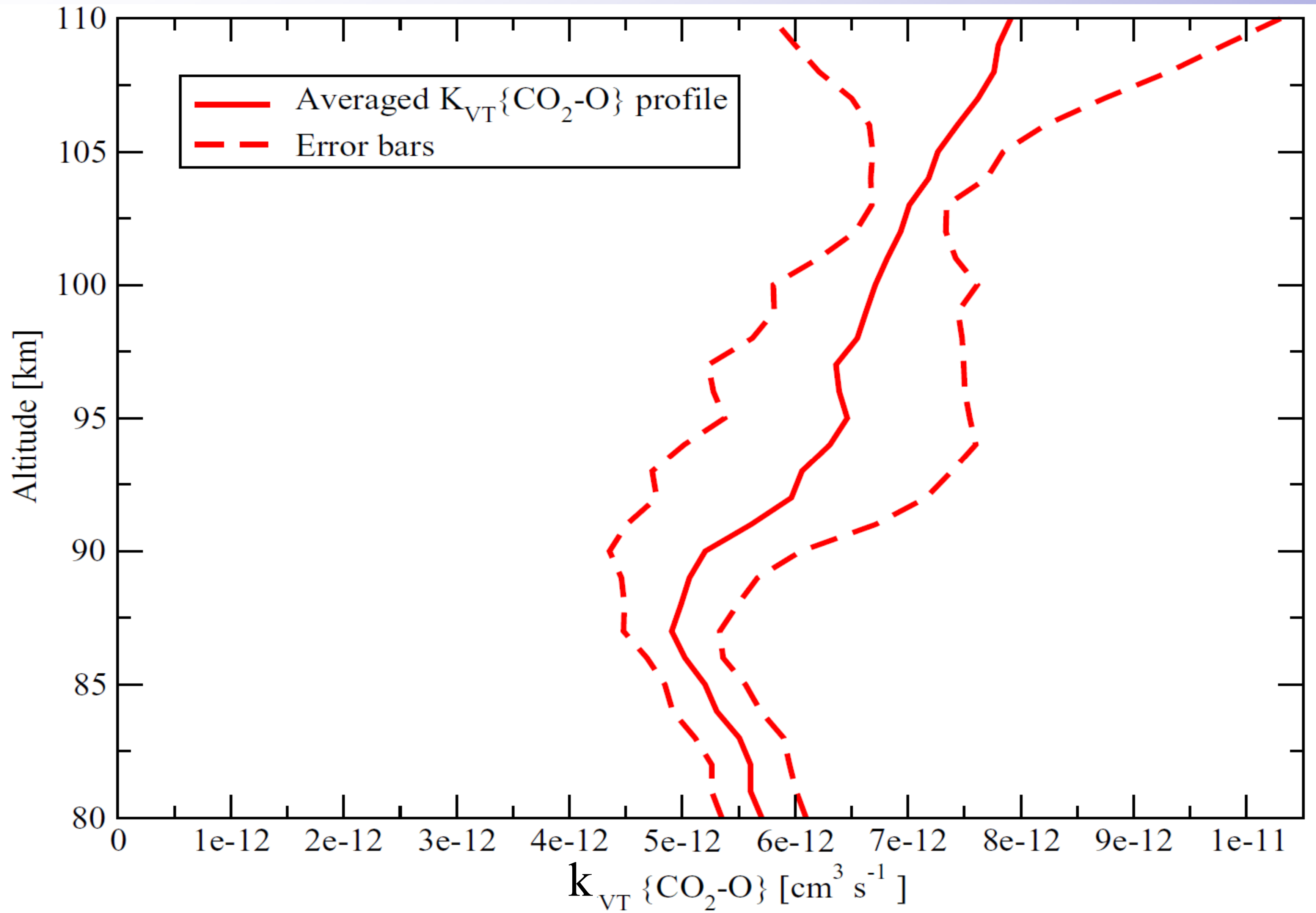
Treating altitudes separately

- Each $|I_{meas} - I_{simul}|$ ($k_{VT}\{CO_2\text{-}O\}x[O]$) = $|I_{meas} - I_{simul}|$ (γ) curve built for a given altitude z has a minimum at $\gamma_{min}(z)$.

- $k_{VT}\{CO_2\text{-}O\}(z) = \gamma_{min}(z) / [O](z)_{aver}$

See next slide for $k_{VT}\{CO_2\text{-}O\}(z)$….

Treating altitudes separately: resulting $k_{VT}\{CO_2\text{-}O\}(z)$



Possible reasons for $k_{VT}\{CO_2\text{-}O\}(z) \neq const$

- Lidar temperatures offset, SABER radiances offset? - Unlikely.

- SABER $[CO_2](z)$ offset?   not found.

- SABER $[O](z)_{aver}$ offset? It's possible but the altitudinal behavior of $[O](z)_{aver}$ will remain the same.

- Temperature dependence of $k_{VT}\{CO_2\text{-}O\}$ other than $(T/300)^{½}$? It's possible since low-temperature laboratory measurements are sparse.

- Some physics we miss that does not happen in the laboratory but is present in the atmosphere:
  - non-thermal O atoms?
  - other pumping source?

Conclusions

- The methodology of $k_{VT}\{CO_2\text{-}O\}$ retrieval from overlapping SABER infrared radiance measurements and lidar temperatures has been developed.
- We base our analysis on $\gamma = k_{VT}\{CO_2\text{-}O\}$ x [O] variation and obtained $\gamma_{optimal}(z)$is a separate scientific product.
- Retrieved $k_{VT}\{CO_2\text{-}O\}$ values are close to most of atmospheric based estimates of this rate coefficient, however:
- Treating altitudes separately leads to retrieving $k_{VT}\{CO_2\text{-}O\}(z) \neq$ const altitude profile that may reveal an unknown component of MLT energy budget and help solving the mystery of discrepancy between laboratory and atmospheric measurements of quenching rate coefficient.
- More comparisons are needed including polar summer and polar winter conditions.

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