Energy transfer rates between CO2 vibrational states. Lessons from atmospheric measurements at 4.3-um by Virtis/VEx

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The VIRTIS spectrometer on board Venus Express has been performing an extensive mapping of the Venus atmospheric emissions between 1 and 5 um, including a systematic survey of the strongest CO2 transitions, in both nadir and limb geometries. This work is based on the analysis of the important ro-vibrational band of CO2 at 4.3 um in the upper atmosphere of Venus during daytime, which is known to be emitted under non-local thermodynamic equilibrium (non-LTE) conditions. The spectral resolution of one of the signals, Virtis-H, is about R=1200, enough to identify different spectral signatures with clarity, and therefore to separate contributions from different CO2 excited states. These emission bands supply a valuable dataset for validating our theoretical non-LTE models. In particular, the correct simulation of the detailed spectral shape of the whole 4.3 um band represents a strong test for these models.

It is well known that the non-LTE situations are complex conditions where diverse microscopic processes compete to establish the given populations of the excited states of the emitting gas. In the case of CO2, their strong emissions at 4.3 um are initially produced by direct solar absorption in a number of ro-vibrational bands extending in the near-IR. Then, radiative and collisional quenching, and thermal (vibrational-translational) and non-thermal (vibrational-vibrational) energy transfer processes determine the state of the gas at each altitude and local time. The precise modelling of all these processes is subject to a number of uncertainties. Of particular relevance are those related to the important non-thermal V-V collisions between excited states of CO2, poorly constrained by laboratory measurements, but essential to explain the CO2 spectra..

A non-LTE model developed at the IAA/CSIC in Granada was used to simulate the Virtis-H spectra, revealing a systematic discrepancy between the emission at two different wavelengths, 4.40 and 4.28 um. A revision of the possible collisional quenching routes, within their uncertainty brackets, was carried out and it was found that an increase of the V-V relaxation rates of high energy CO2 states by a factor between 2 and 4 could reproduce the observed band's spectral shape. This determination reduces the previous bracket of values from laboratory determinations by a factor of 2, although it is still subject to uncertainties such as the unknown thermal state of the atmosphere. It is interesting to note that a similar anomaly occurred in the simulation of Mars atmospheric CO2 spectra taken by PFS/Mars Express which could be resolved by the revision of the V-V rate coefficients mentioned above. .

We will discuss briefly other rate coefficients whose current uncertain determinations have an impact on the simulation and analysis of infrared measurements by Venus Express and would be interesting targets for new laboratory determinations.