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Spectra calculations in central and wing regions of CO_2 IR bands between 10 and 20 μm. III: atmospheric emission spectra

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Abstract

A theoretical model for the prediction of CO_2 absorption in both central and wing regions of infrared absorption bands was presented in the companion paper I. It correctly accounts for line-mixing effects and was validated by comparisons with laboratory spectra in the 600–1000 cm^{-1} region. This quality was confirmed using atmospheric transmissions measured by solar occultation experiments in the second paper. The present work completes these studies by now considering atmospheric emission in the 10–20 μm range. Comparisons are made between computed atmospheric radiances and measurements obtained using four different Fourier transform experiments collecting spectra for nadir, up-looking, as well as limb (from balloon and satellite) geometries. Our results confirm that using a Voigt model can lead to very large errors that affect the spectrum more than 300 cm^{-1} away from the center of the CO_2 ν_2 band. They also demonstrate the capability of our model to represent accurately the radiances in the entire region for a variety of atmospheric paths. This success opens interesting perspectives for the sounding of pressure and temperature profiles, particularly at low altitudes. Another benefit of the quality of the model should be an increased accuracy in the retrieval of atmospheric state parameters from broad features in the measured spectra (clouds, aerosols, heavy trace gases).

Keywords: CO_2; Infrared; Absorption; Shape; Model; Atmospheric; Emission

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1. Introduction

The need for correct forward modeling of CO_2 absorption for the treatment of atmospheric spectra is now well known and large efforts have been devoted to this subject (see Refs. [1–3] and those cited there in). Whereas precise models have been proposed for isolated transitions and Q branches [3,4], the line and band wings have received a much poorer treatment based on empirical line-shape correction factors χ. These are approximate, have very little physical basis, and, due to lack of data, the wings of the $\Pi \leftarrow \Sigma\, \nu_2$ band have to be computed using values of χ determined in the $\Sigma \leftarrow \Sigma\, \nu_3$ band. This situation, which has led to discrepancies between measured and calculated atmospheric spectra in the 15 μm region (e.g. [5]), also called for a precise and unified approach permitting the treatment of all CO_2 absorption features, including local transitions, dense Q branches, and the line wings.

Such a model was proposed in a preceding companion paper [1]. It uses the energy corrected sudden approximation in order to construct the line-mixing relaxation matrix. It correctly and automatically accounts for the symmetry of the vibrational levels and for the coupling of angular momenta thus permitting calculation of line-coupling between any (P, Q, and R) lines within any band. Comparisons between predictions of this model and temperature-dependent laboratory measurements for CO_2–N_2 mixtures [1] in the 15 μm region have demonstrated the quality of this approach. The shape of the central part of the intense bands at elevated pressure is well described and absorption in the wings is satisfactorily modeled in a wide range of wavenumbers around the band centers. Further tests of this approach have been made in Ref. [2] by comparisons between forward calculations and atmospheric transmissions measured using balloon-borne and ground-based solar occultation experiments. They have confirmed the quality of the model and the importance of errors induced by the use of purely Voigt line shapes. Furthermore recall that, as explained in [1], the present model is an extension of that initially developed for Q branches [3,6] whose quality was widely demonstrated using laboratory and atmospheric measurements.

The present paper extends the test of the model by making comparisons between forward calculations and atmospheric radiances measured by different Fourier transform instruments. A number of emission spectra are used which correspond to various geometries including nadir, up-looking, and (balloon and satellite borne) limb viewings. The results confirm the quality of the approach proposed with which all measurements are correctly reproduced. In particular, whereas use of the Voigt model can lead to very large errors, deviations between measurements and our calculations show no specific broad features over the entire spectral ranges studied. Among various remote sensing possibilities offered by the quality of the model, that of retrieving species that make small contributions highly contaminated by CO_2 emission is demonstrated.

2. Atmospheric spectra

The atmospheric emission spectra used have been recorded by four different Fourier transform instruments that have all been carefully calibrated and provide absolute radiances.

The high-resolution interferometer sounder (HIS) [7] was developed by the University of Wisconsin to measure thermal radiation emitted by the atmosphere. It is a Fourier transform instrument with a spectral resolution of about 0.3 cm^{-1} (unapodized). The radiance spectra used in the present