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Sampling molecular state transitions in a line-by-line Monte Carlo approach to estimate radiative forcing for climate change studies.

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A major challenge of the beginning of the century is the nation's actions over the terrestrial climate to counter the global warming. This warming is caused by the human activity and in particular by the emission of important amounts of greenhouse gases. Its close understanding and precise estimate are necessary to take appropriate actions and one of the driving concepts along this line is radiative forcing. This forcing is defined as the difference between two Top Of Atmosphere (TOA) radiative fluxes, both integrated spatially over all the Earth and temporally over a climatic period, the difference being due to a change of one single parameter. Here we focus on the forcing to carbon dioxide. So, our question is the estimation of the difference between two integrated TOA fluxes, before and after we change the CO₂ concentration field.

Our goal is therefore to model physically and estimate numerically radiative fluxes integrated over all positions at TOA and over all times in a given period. The modelling involves the main ingredients of radiative transfer physics, including multiple scattering by clouds, but the principal difficulty is the emission and attenuation of radiation by molecular gases for highly heterogeneous fields of concentrations and pressure. Indeed, the absorption coefficient is modelled as the sum of millions of spectrally sharp lines associated to molecular state transitions. This spectral model is called a line-by-line model and the corresponding spectroscopic data are available in databases such as HITRAN or GEISA. The numerical implementation of line-by-line models using deterministic schemes is commonly presented as time consuming because the spectral integration requires tens of millions of discrete frequencies. When multiplying this by the number of discrete positions and discrete times required to cover the earth in a climatic period respectively, the computation becomes unfeasible. Approximate spectral models exist and permit to bypass such difficulties, typically the CK-distributions model. But a straightforward use of these models would introduce errors that are not compatible with the accuracy level required for radiative forcing studies. The accuracy level is appreciated in terms of flux differences, these differences are small, and compared to these small values the main question becomes "is the difference accurate enough even if the fluxes are not?". Strong efforts were devoted to this question and today's radiative forcing studies are accurate enough, even when using approximate spectral models. However, this introduction of approximate spectral models is a loss of flexibility. The full quantification of the remaining uncertainty level is still an open question and each change in the spectroscopic database, or each change on the line shape assumption (line profile truncations, continuums) need to reprocess the data for tuning the parameters of the approximate models and re-assess the accuracy level. We propose here to re-open the question of numerically estimate radiative forcing, sticking to a pure line-by-line model, but using a statistical approach in which the molecular state transitions are directly sampled from the databases. We use the Monte-Carlo method, a reference method in radiative transfer, which is famous for easily handling multidimensional integrals and systematically associating uncertainty estimates to each estimated quantity. The main reason why this could not be performed until now is the fact that the transition data appear inside the exponential of Beer extinction and none of the available nonlinear Monte Carlo methods could address this specific difficulty. This point was revisited in (Dauchet et al., 2018) and we can now make the following proposition:

1) A null-collision approach is used so that the sampling of absorption free paths is possible prior to the knowledge of the true absorption optical thickness (adding fictive colliders so that the atmosphere looks homogeneous along the line of sight, Galtier et al., 2013; El Hafi et al., 2021).

2) At each absorption location (true or virtual, this is not decided yet), instead of computing the local value of the absorption coefficient, a line is sampled among all those available in the database (Galtier et al., 2016). In theoretical terms, this means that a path-integral approach is retained for the coupled problem, radiative transfer combined to spectroscopy: the absorption coefficient will never be estimated.

3) A set of probabilities are chosen, for sampling the lines, using a method based on machine learning techniques, building a hierarchical and multi-resolution representation of probabilities in the frequency domain (McCool et al., 1997).

Our results indicate that it is possible to estimate the radiative forcing at a global scale (for the entire Earth), integrated over a one-year period and integrated over all the thermal infrared range within a few minutes on a personal computer. Furthermore, we study the associated computation times and show that they are insensitive to the domain complexity (as was observed for a similar Monte Carlo approach in Villefranque et al., 2019). It is not more time consuming to estimate a radiative value integrated over a day or a month, integrated over one atmospheric column or over all the Earth, or finally over a narrow spectral band or all the infrared range.

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