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ESTIMATING ATMOSPHERIC RADIATIVE FORCINGS USING SENSITIVITY MONTE CARLO METHODS

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ABSTRACT. One aspect of climate change analysis is the quantification of radiative forcings, i.e., the change of top-of-atmosphere (TOA) net radiative flux induced by an isolated, instantaneous change in surface or atmospheric constitution. In this paper, we discuss recent advances in path-integral formulations for producing reference estimates of radiative forcings, in the form of partial derivatives we call "sensitivities". We present the theoretical framework and highlight the role of computer science acceleration techniques in making the computational cost insensitive to the system's multidimensional and multiphysics complexity. The approach is demonstrated by estimating the flux sensitivity to the concentration of two greenhouse gases.

1. INTRODUCTION AND CONTEXT

Monte Carlo (MC) methods produce unbiased estimates of integral quantities and their uncertainties, making them suitable for providing reference results in atmospheric radiative transfer studies. Although being extensively used for benchmarking faster 1D radiative transfer codes to guide improvements in remote sensing, weather forecast and climate modeling, the high computational cost of MC 3D codes, such as the I3RC community model [1] or MYSTIC [2], usually prevents their direct employment in operational contexts. However, recent advances in transferring computer science acceleration techniques to atmospheric radiative transfer science—to handle complex surfaces and detailed cloud fields for instance [3–5]—open new perspectives for using MC methods beyond reference simulations. These methods have recently been shown to be key in addressing multiphysics and multidimensional integrals [6] resulting in computationally efficient simulations that are insensitive to the size and complexity of the integration domains [5,7,8].

A recent example in this respect is a novel work that couples a line-by-line spectroscopic model to

a radiative transfer model, resulting in a multiphysics—spectro-radiative—model first described in [9], that has recently benefited from a close collaboration between multiple communities (computer science, atmospheric science, spectroscopy and energetics physics). This collaboration has resulted in a MC radiative transfer code designed for the climate community. Developed during the PhD work of Nyffenegger-P  r  , it provides reference results for estimating atmospheric radiative fluxes integrated over any spatial, temporal and wavenumber domain, using the atmosphere and the surface properties’ description as input data, typically derived from a global and multi-decadal GCM simulation. A concrete application of this tool in climate science is estimating the global broadband flux at TOA, that is, radiances averaged temporally over a climate period and spatially over the whole globe, as well as integrated over all frequencies and outgoing directions, without any compromise on the radiative or the spectroscopic physics description. The combination of MC methods with computer graphics acceleration techniques [10,11] results in computation times of just a few seconds that are insensitive to the input data complexity. This means that the computational cost of estimating the radiative flux at a particular time, location, and wavenumber is comparable to that of averaging over an entire century, over the entire globe, and integrating over the entire infrared spectrum.

In the present work, we argue that in addition to estimating radiative flux at TOA, we can also estimate unbiased radiative forcings in the form of partial derivatives (sensitivities). Radiative forcing with respect to a parameter is defined as the variation in the outgoing radiative flux at TOA when this parameter value is changed while keeping the other model parameters fixed. Indeed, analyzing the physical mechanisms of global warming requires characterizing its evolution as a function of the climate system state, in which a first step is often to evaluate the radiative forcing resulting from a change in surface or atmospheric constitution (e.g., gas, cloud, aerosol or surface properties) which are parameters in the spectro-radiative model. Instead of computing two estimates based on different sets of parameter values, sensitivities can be estimated using straightforward MC methods that differentiate the spectro-radiative model. First developments for sensitivities have been disposable for about 20 years [12] and have the benefit of providing the estimates’ uncertainties. As statistical convergence issues often arise in a manner that is specific to each application, this method is at the heart of very active fields of research, including in computer graphics science [13].

In section (2.1), we present a succinct examination of the difficulties associated with calculating radiance. Next, we outline the foundations of its efficient MC estimation through the null-collisions technique [14]. In section (2.2), we establish a transport model for sensitivities that shares strong physical similarities with the transport of radiance. As a result, it is shown that the prior theoretical advancements presented for estimating radiance can now also be applied to estimate sensitivities and thus radiative forcings.

2. THEORY AND METHODS

2.1 Estimating the global outgoing radiative flux at TOA

The global outgoing radiative flux at TOA, $\bar{\Phi}$, is the monochromatic radiance L_ν averaged over a time period Δt (typically 1 to 30 years), over the entire globe of surface S , integrated over all frequencies ν and outgoing directions \vec{u} . The expression for this multiple path-integral is:

$$\bar{\Phi} = \int_{\Delta t} p_T(t) dt \int_{TOA} p_S(\vec{x}) dS(\vec{x}) \int_0^{+\infty} p_N(\nu) d\nu \int_{2\pi} p_U(\vec{u}) d\vec{u} \sigma T_{\max}^4 \frac{L_\nu(\vec{x}, \vec{u}, t)}{L_\nu^{eq}(T_{\max})}, \quad (1)$$

with $p_T(t) = \frac{1}{\Delta t}$, $p_S(\vec{x}) = \frac{1}{S}$, $p_N(\nu) = \frac{\pi L_\nu^{eq}(T_{\max})}{\sigma T_{\max}^4}$ and $p_U(\vec{u}) = \frac{\vec{u} \cdot \vec{n}}{\pi}$ the respective probability density functions associated with sampling a time, a position at TOA, a wavenumber and an outgoing

direction over the upper hemisphere. We can read Equation (1) as follows: assuming the monochromatic radiance $L_\nu(\vec{x}, \vec{u}, t)$ is known at all times, positions, frequencies and directions, the average flux $\bar{\phi}$ is simply obtained by successively sampling a time according to $p_T(t)$, a position according to $p_S(\vec{x})$, a frequency according to $p_N(\nu)$, and an outgoing direction according to $p_U(\vec{u})$, and averaging the associated realization weights $\omega = \sigma T_{\max}^4 \frac{L_\nu(\vec{x}, \vec{u}, t)}{L_\nu^{eq}(T_{\max})}$. Each successive sampling is a MC realization. According to the Central Limit Theorem, drawing a large number N of realizations, their sample mean $\bar{\omega}$ approaches a normal distribution of variance $var(\bar{\omega}) = \frac{var(\omega)}{N}$ that tends towards zero when N tends towards infinity. The sample mean $\bar{\omega}$ is used as a statistical estimator for $\bar{\phi}$.

In reality, the main difficulty lies in computing $L_\nu(\vec{x}, \vec{u}, t)$ in a heterogeneous and anisothermal participating medium. Neglecting the sphericity of the atmosphere, the transport of radiance can be described by the stationary form of the monochromatic Radiative Transfer Equation (RTE):

$$\begin{cases} \forall \vec{x} \in \Omega, \forall \vec{u} \in \mathbb{S}^2 : \\ \vec{u} \cdot \text{grad}_X L_\nu(\vec{x}, \vec{u}) = -k_{ext,\nu}(\vec{x}) L_\nu(\vec{x}, \vec{u}) + k_{a,\nu}(\vec{x}) L_\nu^{eq}(T(\vec{x})) \\ \quad + k_{s,\nu}(\vec{x}) \int_{4\pi} p(\vec{u}', \vec{u}) d\vec{u}' L_\nu(\vec{x}, \vec{u}') \\ \forall \vec{y} \in \partial\Omega, \forall \vec{u}_+ \in \mathbb{S}_+^2 : \\ L_\nu(\vec{y}, \vec{u}_+) = L_{\nu\partial\Omega}(\vec{y}, \vec{u}_+) \end{cases} \quad (2)$$

Ω is the geometrical domain and $\partial\Omega$ its boundary (the space and the land or oceanic surface) at which the monochromatic radiance $L_{\nu\partial\Omega}$ is known for all locations \vec{y} and all directions \vec{u}_+ of the incoming hemisphere \mathbb{S}_+^2 at the boundary. The coefficients $k_{a,\nu} \equiv k_{a,\nu}(\vec{x})$, $k_{s,\nu} \equiv k_{s,\nu}(\vec{x})$ and $k_{ext,\nu} = k_{a,\nu} + k_{s,\nu}$ are absorption, scattering and extinction coefficient respectively. $L_\nu^{eq}(T(\vec{x}))$ is equilibrium Black Body radiance (following Planck Black Body radiance function of temperature $T(\vec{x})$ at location \vec{x}) and $p(\vec{u}', \vec{u})$ is the single scattering phase function, i.e. probability density that the scattering direction is \vec{u}' for this incoming direction \vec{u} .

The path-integral corresponding to system (2) is:

$$L_\nu(\vec{x}, \vec{u}) = \int_0^{+\infty} p_{\mathcal{L}}(l) dl \left[\begin{array}{l} \mathcal{H}(\vec{x}' \notin \Omega) L_{\nu\partial\Omega}(\vec{x}', \vec{u}') \\ + \mathcal{H}(\vec{x}' \in \Omega) \left[\begin{array}{l} P_a(\vec{x}') L_\nu^{eq}(T(\vec{x}')) \\ + P_s(\vec{x}') \int_{4\pi} p(\vec{u}', \vec{u}) d\vec{u}' L_\nu(\vec{x}', \vec{u}') \end{array} \right] \end{array} \right], \quad (3)$$

with $p_{\mathcal{L}}(l) = k_{ext,\nu}(\vec{x}') e^{(-\int_0^l k_{ext,\nu}(\vec{x}') dl')}$ the probability density function for sampling a free path of length l , following which a collision occurs at position $\vec{x}' = \vec{x} - l\vec{u}$. Equation (3) can be translated into a MC algorithm to estimate $L_\nu(\vec{x}, \vec{u})$: at each collision, the new position \vec{x}' is either outside the medium (as it reaches the boundary conditions: $\mathcal{H}(\vec{x}' \notin \Omega)$), in which case the retained weight is $L_{\nu\partial\Omega}$, or \vec{x}' is still in the atmosphere ($\mathcal{H}(\vec{x}' \in \Omega)$). In the latter case, two types of collision events can take place according to probabilities $P_a(\vec{x}) = \frac{k_{a,\nu}(\vec{x})}{k_{ext,\nu}(\vec{x})}$ and $P_s(\vec{x}) = \frac{k_{s,\nu}(\vec{x})}{k_{ext,\nu}(\vec{x})}$ for an absorption event or a scattering event, respectively. In the case of an absorption event, the MC weight is the Black Body equilibrium radiance $L_\nu^{eq}(T(\vec{x}'))$ at the collision position. In the case of a scattering event, the photon trajectory continues in a new propagation direction \vec{u}' sampled according to the phase function $p(\vec{u}', \vec{u})$, and the radiance $L_\nu(\vec{x}', \vec{u}')$ at the new position and the new direction is calculated; it is a recursive algorithm.

The model above presents two shortcomings. The first one is encountered when sampling the extinction length l , as $L_\nu(\vec{x}, \vec{u})$ depends non-linearly (through an exponential function) on the heterogeneous extinction coefficient $k_{ext,\nu}(\vec{x})$. The second one is encountered when sampling the collision type; the absorption coefficient field is expressed as a sum over millions of molecular transitions

$h_{a,\nu,j}(\vec{x})$; $k_{a,\nu}(\vec{x}, t) = \sum_{j=1}^{N_t} h_{a,\nu,j}(\vec{x})$ which is computationally expensive to estimate. Approximation methods for the calculation of the absorption coefficient field exist, but their model errors can be difficult to quantify. Interestingly, using the null-collisions method makes both of these limitations vanish [12]. Not only does introducing fictive colliders homogenize the extinction coefficient field [15,16], but it also enables a coupling between the line-by-line spectroscopic model and the radiative transfer model [6,14], so that the resulting multiphysics path-integral formulation is treated statistically through double randomization, which will become more obvious through the algorithmic illustration at the end of the next section. The path-integral is reformulated using the null-collision method as:

$$L_\nu(\vec{x}, \vec{u}) = \int_0^{+\infty} \hat{p}_\mathcal{L}(l) dl$$

$$\left[\begin{array}{l} \mathcal{H}(\vec{x}' \notin \Omega) L_{\nu\partial\Omega}(\vec{x}', \vec{u}') \\ + \mathcal{H}(\vec{x}' \in \Omega) \left[\begin{array}{l} \hat{P}_s(\vec{x}') \left\{ \begin{array}{l} P_s(\vec{x}') \int_{4\pi} p(\vec{u}', \vec{u}) d\vec{u}' L_\nu(\vec{x}', \vec{u}') \\ + (1 - P_s(\vec{x}')) L_\nu(\vec{x}', \vec{u}) \end{array} \right\} \\ + (1 - \hat{P}_s(\vec{x}')) \left(\sum_{j=1}^{N_t} P_J(j, \nu) \left\{ \begin{array}{l} P_{a,j}(\vec{x}') L_\nu^{eq}(T(\vec{x}')) \\ + (1 - P_{a,j}(\vec{x}')) L_\nu(\vec{x}', \vec{u}) \end{array} \right\} \right) \end{array} \right] \end{array} \right] \quad (4)$$

Equation (4) results in an algorithm in which the photon can propagate in the medium without knowing upfront the real extinction coefficient field, needed only locally at each collision location; instead, only upper bound free parameters $\hat{k}_{a,\nu} \equiv \hat{k}_{a,\nu}(\vec{x})$, $\hat{k}_{s,\nu} \equiv \hat{k}_{s,\nu}(\vec{x})$ and $\hat{k}_{\text{ext},\nu} = \hat{k}_{a,\nu} + \hat{k}_{s,\nu}$ are needed to define probability functions $\hat{p}_\mathcal{L}(l) = \hat{k}_{\text{ext},\nu}(\vec{x}) e^{-\int_0^l \hat{k}_{\text{ext},\nu}(\vec{x}) dl'}$, $\hat{P}_s(\vec{x}) = \frac{\hat{k}_{s,\nu}}{\hat{k}_{\text{ext},\nu}}(\vec{x})$ and $\hat{P}_a(\vec{x}) = (1 - \hat{P}_s(\vec{x})) = \frac{\hat{k}_{a,\nu}}{\hat{k}_{\text{ext},\nu}}(\vec{x})$. At each absorption event, instead of using a pre-computed value of $k_{a,\nu}$ to determine the nature -null or real- of the event, one of the state transitions that contribute to the absorption coefficient is sampled according to the probability density function $P_J(j, \nu)$, after which, whether it is a true transition or a null-transition is sampled according to $P_{a,j}$ and $(1 - P_{a,j})$ respectively. In the case of a scattering event, whether it is a true scattering or a null-scattering is sampled according to P_s and $(1 - P_s)$ respectively. The upper bound free parameters do not necessarily ensure an efficient sampling of transitions for a highly varying absorption spectra in frequencies, which can lead to a significant computational cost if chosen too high compared to the true absorption spectra, such as the number of null-collision events become too large. This is when computer scientists' expertise in structuring and processing data come into play. The frequency dimension is partitioned by a hierarchical grid to obtain a field of upper bounds adapted to the absorption spectra for an efficient sampling of the large spectral data. This makes the computational cost insensitive to the size (number of transitions) and the complexity (shape of absorption spectra) of the spectroscopic database. The next section details a similar algorithm that will be used to estimate radiative forcings.

2.2 Estimating radiative forcings as global radiative flux sensitivities

The previous section establishes the MC approach for evaluating the global outgoing radiative flux at TOA using statistical methods. In this section, we show how to wield this framework to estimate radiative forcings as flux sensitivity estimates, hence retaining the benefits associated with the use of the null-collision technique.

Radiative forcing for a given parameter $\bar{\pi}$ is defined as the partial derivative of the flux at TOA with respect to $\bar{\pi}$. Given that the sensitivity with respect to $\bar{\pi}$ is $s_\nu(\vec{x}, \vec{u}, t, \bar{\pi}) = \partial_{\bar{\pi}} L_\nu(\vec{x}, \vec{u}, t, \bar{\pi})$, the radiative forcing is:

$$\partial_{\bar{\pi}} \bar{\Phi} = \int_{\Delta t} p_T(t) dt \int_{\text{TOA}} p_S(\vec{x}) dS(\vec{x}) \int_0^{+\infty} p_N(\nu) d\nu \int_{2\pi} p_U(\vec{u}) d\vec{u} \sigma_{\text{max}} T_{\text{max}}^4 \frac{s_\nu(\vec{x}, \vec{u}, t, \bar{\pi})}{L_\nu^{eq}(T_{\text{max}})}, \quad (5)$$

which requires a path-integral formulation of s_ν , similar to the path-integral formulation that was required for L_ν in the previous section. In addressing the question of sensitivity estimation using MC methods, there exists two different approaches. The first one would consist in differentiating the path-integral formulation of $L_\nu(\vec{x}, \vec{u})$ in Equation (4), which preserves the same random samplings between the quantity and its derivatives [10], but this can be difficult to implement in practice in the case of optimized null-collision algorithms as pointed out in [17]. The second approach would consist in differentiating the radiative transport model of $L_\nu(\vec{x}, \vec{u})$ in Equation (2) to establish a transport model for the sensitivity. This method was originally developed for geometric sensitivities (derivatives with respect to parameters that modify the geometry [18,19]) but will be used hereafter on parametric (non-geometric) sensitivities. Using the second approach, we trade the benefit of preserving the sampling path between the quantity and its derivative for 1/ improved convergence in our optimized null-collision algorithm, and 2/ an interesting framework for modeling sensitivity as a transported quantity, and thus analyzing the physics of its propagation in the participating medium. Differentiating Equation (2) with respect to $\vec{\pi}$ according to the second approach, a transport model for the sensitivity can be expressed by a new RTE:

$$\left\{ \begin{array}{l} \forall \vec{x} \in \Omega, \forall \vec{u} \in \mathbb{S}^2 : \\ \vec{u} \cdot \vec{\text{grad}}_{\mathcal{X}} s_\nu(\vec{x}, \vec{u}, \vec{\pi}) = -k_{ext,\nu}(\vec{x}, \vec{\pi}) s_\nu(\vec{x}, \vec{u}, \vec{\pi}) + k_{a,\nu}(\vec{x}, \vec{\pi}) \left[\frac{\partial \vec{\pi} k_{a,\nu}(\vec{x}, \vec{\pi})}{k_{a,\nu}(\vec{x}, \vec{\pi})} (L_\nu^{eq}(T(\vec{x})) - L_\nu(\vec{x}, \vec{u}, \vec{\pi})) \right] \\ \quad + k_{s,\nu}(\vec{x}, \vec{\pi}) \int_{4\pi} p(\vec{u}', \vec{u}) d\vec{u}' (s_\nu(\vec{x}, \vec{u}', \vec{\pi}) + \left[\frac{\partial \vec{\pi} k_{s,\nu}(\vec{x}, \vec{\pi})}{k_{s,\nu}(\vec{x}, \vec{\pi})} (L_\nu(\vec{x}, \vec{u}', \vec{\pi}) - L_\nu(\vec{x}, \vec{u}, \vec{\pi})) \right]) \\ \forall \vec{y} \in \partial\Omega, \forall \vec{u}_+ \in \mathbb{S}_+^2 : \\ s_\nu(\vec{y}, \vec{u}_+) = 0 \end{array} \right. \quad (6)$$

It is now obvious that the transport of sensitivity is very similar to that of radiance, only the sources via absorption and scattering are different. Moreover, via these sources, the transport model for sensitivity is coupled to the transport model of radiance, which results in an additional coupling (in addition to the spectro-radiative one) to the system to be solved. Up to this point, the transport model we obtain for $s_\nu(\vec{x}, \vec{u}, \vec{\pi})$ can be solved using any model for the absorption and scattering coefficients $k_{a,\nu}(\vec{x}, \vec{\pi})$ and $k_{s,\nu}(\vec{x}, \vec{\pi})$. It is however possible to establish a sensitivity model that couples line-by-line spectroscopy and radiative transfer, following the same formal developments established as in the construction of the multiphysics model for radiance. We obtain the following path-integral formulation for sensitivity :

$$s_\nu(\vec{x}, \vec{u}, \vec{\pi}) = \int_0^{+\infty} \hat{p}_{\mathcal{L}}(l) dl$$

$$\left[\begin{array}{l} \mathcal{H}(\vec{x}' \notin \Omega) \cdot 0 \\ + \mathcal{H}(\vec{x}' \in \Omega) \\ \left\{ \begin{array}{l} \hat{P}_s(\vec{x}') \left\{ \begin{array}{l} P_s(\vec{x}') \int_{4\pi} p(\vec{u}', \vec{u}) d\vec{u}' \left[s_\nu(\vec{x}', \vec{u}', \vec{\pi}) + \left[\frac{\partial \vec{\pi} k_{a,\nu}(\vec{x}, \vec{\pi})}{k_{a,\nu}(\vec{x}, \vec{\pi})} (L_\nu(\vec{x}', \vec{u}', \vec{\pi}) - L_\nu(\vec{x}', \vec{u}, \vec{\pi})) \right] \right] \right. \\ \left. + (1 - P_s(\vec{x}')) [s_\nu(\vec{x}', \vec{u}, \vec{\pi})] \end{array} \right\} \\ + (1 - \hat{P}_s(\vec{x}')) \left(\sum_{j=1}^{N_t} P_J(j, \nu) \left\{ \begin{array}{l} P_{a,j}(\vec{x}') \left[\frac{\partial \vec{\pi} h_{a,\nu,j}(\vec{x}, \vec{\pi})}{h_{a,\nu,j}(\vec{x}, \vec{\pi})} (L_\nu^{eq}(T(\vec{x}')) - L_\nu(\vec{x}', \vec{u}, \vec{\pi})) \right] \right. \\ \left. + (1 - P_{a,j}(\vec{x}')) [s_\nu(\vec{x}', \vec{u}, \vec{\pi})] \end{array} \right\} \right) \end{array} \right\} \end{array} \right] \quad (7)$$

Algorithm 1 illustrates the statistical procedure for sampling a Monte Carlo weight ω for sensitivity. When L_ν appears in ω , using double randomization, we sample one realization of the algorithm associated to Equation (4). In this algorithm, the coupling of the radiative transfer model and the line-by-line spectroscopic model also occurs through double randomization. Therefore, the successive sampling of collisions' positions, the scattering and molecular transitions are all contained in a single sampled path to evaluate the MC weight ω .

3. RESULTS

The theoretical developments presented are applicable to any parameter of interest in the model. Using Algorithm 1, Figure 1 (a) and (b) illustrates the variations in radiative forcings associated to a change in carbon dioxide and water vapor concentrations. The sensitivity parameter $\tilde{\pi}$ acts on the concentration fields and then on the radiative fluxes through the molecular transitions $h_{a,v,j}(\vec{x})$ as a multiplier factor of the reference molar fraction of gas, i.e. $x_{gas} = \tilde{\pi} * x_{gas,ref}$. The radiative forcings are estimated in a purely absorbing atmosphere, averaged spatially over the entire globe and temporally over a month, and integrated over the infrared spectrum, as the average of 640,000 realization weights. For a given gas and for all values of $\tilde{\pi}$, the perturbation of $\tilde{\pi}$ corresponds to the injection of the same infinitesimal amount of this gas in the atmosphere, and the corresponding sensitivity is the resulting variation in global outgoing flux at TOA. As expected, the higher the concentration of gas (increasing values of $\tilde{\pi}$), the smaller the impact of its perturbation, as it tends towards zero. Moreover, radiative forcings are negative, such as the increase of greenhouse gas concentration decreases the outgoing flux, which is consistent with the greenhouse effect. As a result, the terrestrial radiative budget increases. Comparing the two gases, the higher the absorption of a gas, the greater the expected impact of increasing its concentration, which is consistent with the obtained sensitivities; water vapor is more absorbing than carbon dioxide, resulting in larger radiative forcings. The MC sensitivity method yields unbiased statistical uncertainty, while the uncertainty in finite differences (FD) stems from flux estimation uncertainties. However, the uncertainty due to discretization choice in FD remains unaccessible. Poor discretization choices lead to oscillations in finite differences, as we observe. Nonetheless, optimizing discretization for the same number of realizations would increase finite differences' uncertainties, thereby underscoring the importance of the MC method.

Figure 1 (c) illustrates the insensitivity of computational cost to the integration domain complexity; that is enlarging the integration domains does not impact the computational time. Its first graph represents radiative forcing of CO_2 for a given profile and date, spectrally integrated over a band of increasing width. In the middle graph and the last graph, the radiative forcing is additionally integrated over spatial and time domains of increasing size. We observe that it is as costly to estimate the radiative forcing for a given date and position on a small frequency interval (first red point) as it is to estimate the result integrated over the entire thermal infrared, over the entire Earth, and over a month (last green point), for the same level of data resolution.

4. CONCLUSIONS

We propose a practical MC approach for estimating radiative forcings integrated over any spatial, temporal and wavenumber domains. The use of the null-collision method along with computer graphics acceleration techniques are demonstrated to be key in making the computations not only fast, but also insensitive to the path-integral complexity. Efficient radiative forcings also require a careful choice of methods: differentiating the transport equation instead of the path-integral formulation, due to convergence issues that specifically arise in the second approach for optimized null-collision algorithms. The approach is illustrated through greenhouse gas concentration radiative forcing in a purely absorbing medium.

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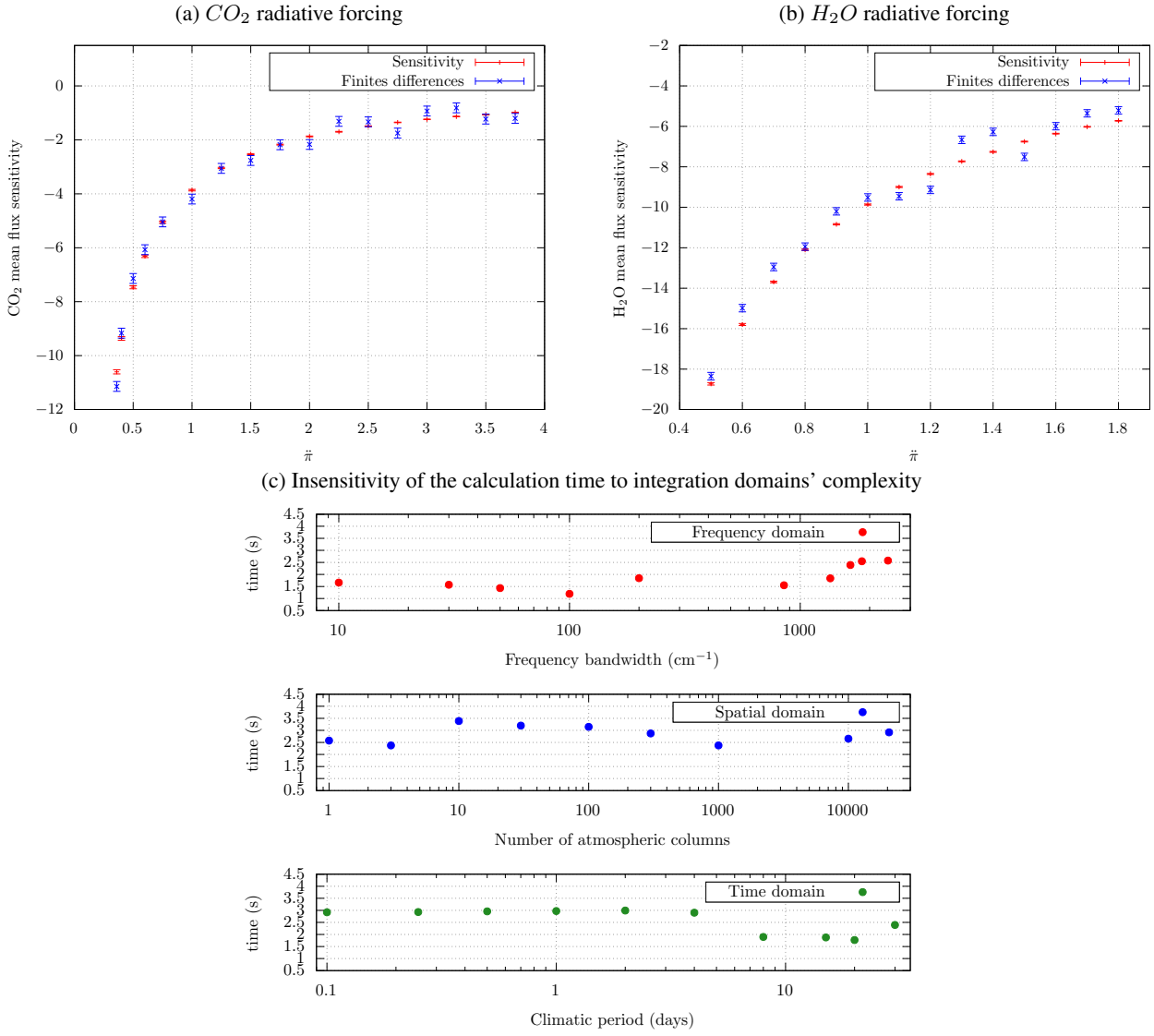


Figure 1. Radiative forcing associated with a linear increase of gas concentration, as a function of the multiplier coefficient $\bar{\pi}$, is displayed for two gases: (a) CO_2 and (b) H_2O . Sensitivity estimates are obtained using Algorithm 1, whereas finite difference is based on the difference between two standard MC flux estimates obtained for different values of $\bar{\pi}$. Insensitivity to the dimensions of the main integration domains (spectral, spatial, and temporal) of the calculation time required to estimate the sensitivity of an outgoing radiative flux to CO_2 concentration is displayed in (c).

Algorithm 1 Recursive algorithm for one Monte Carlo weight ω realization of $s_\nu(\vec{x}, \vec{u}, \bar{\pi})$

```

Initialize position:  $\vec{x}' \leftarrow \vec{x}$ 
Initialize direction:  $\vec{u}' \leftarrow \vec{u}$ 
Sample a length to find the next collision position according to  $\hat{p}_L(l)$ 
Update position:  $\vec{x}' \leftarrow \vec{x}' - l\vec{u}'$ 
if  $\vec{x}' \notin \Omega$  then
     $\omega = 0$ 
else
    Sample a uniform random variable  $r \in [0, 1]$ 
    if  $r < \hat{P}_s(\vec{x}')$  then
        Sample a uniform random variable  $r \in [0, 1]$ 
        if  $r < P_s(\vec{x}')$  then
            Sample scattering direction  $\vec{u}'$  according to phase function  $p(\vec{u}', \vec{u})$ 
             $\omega = s_\nu(\vec{x}', \vec{u}', \bar{\pi}) + \frac{\partial_{\bar{\pi}} k_{d,\nu}(\vec{x}, \bar{\pi})}{k_{d,\nu}(\vec{x}, \bar{\pi})} (L_\nu(\vec{x}', \vec{u}', \bar{\pi}) - L_\nu(\vec{x}', \vec{u}, \bar{\pi}))$ 
        else
            It is a null-scattering event, recurse algorithm to estimate  $s_\nu(\vec{x}', \vec{u}, \bar{\pi})$ .
    else
        Sample a transition  $j$  according to  $P_J(j, \nu)$ 
        Sample a uniform random variable  $r \in [0, 1]$ 
        if  $r < P_{a,j}(\vec{x}')$  then
             $\omega = \frac{\partial_{\bar{\pi}} h_{a,\nu,j}(\vec{x}, \bar{\pi})}{h_{a,\nu,j}(\vec{x}, \bar{\pi})} (L_\nu^{eq}(T(\vec{x}')) - L_\nu(\vec{x}', \vec{u}, \bar{\pi}))$ 
        else
            It is a null-transition event, recurse algorithm to estimate  $s_\nu(\vec{x}', \vec{u}, \bar{\pi})$ .

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REFERENCES

- [1] Pincus, R. et al., "Computational cost and accuracy in calculating three-dimensional radiative transfer : Results for new implementations of monte carlo and SHDOM," *Journal of the Atmospheric Sciences*, 66(10) :31313146, 2009.
- [2] Mayer, B., "Radiative transfer in the cloudy atmosphere," *The European Physical Journal Conferences*, 1 :7599, 2009.
- [3] Niro, F. et al., "European Space Agency (ESA) Calibration/Validation Strategy for Optical Land-Imaging Satellites and Pathway towards Interoperability," *Remote Sensing*, 13(15) :3003, 2021.
- [4] Schwaerzel, M. et al., "Impact of 3d radiative transfer on airborne N O 2 imaging remote sensing over cities with buildings," *Atmospheric Measurement Techniques*, 14(10) :64696482, 2021.
- [5] Villefranque, N. et al., "A path-tracing monte carlo library for 3-d radiative transfer in highly resolved cloudy atmospheres," *Journal of Advances in Modeling Earth Systems*, 11(8) :24492473, 2019.
- [6] Tregan, J.-M. et al., "Coupling radiative, conductive and convective heat-transfers in a single Monte Carlo algorithm: A general theoretical framework for linear situations," *PLoS ONE*, 18(4): e0283681, 2023.
- [7] Ibarrart, L. et al., "Advection, diffusion and linear transport in a single path-sampling Monte-Carlo algorithm : getting insensitive to geometrical refinement," working paper or preprint : <https://hal.science/hal-03818899v2/file/main%20%281%29.pdf>.
- [8] Villefranque, N. et al., "The teapot in a city : A paradigm shift in urban climate modeling," *Science Advances*, 8(27), 2022.
- [9] Galtier, M. et al., "Radiative transfer and spectroscopic databases : A line-sampling monte carlo approach," *Journal of Quantitative Spectroscopy and Radiative Transfer*, 172 :8397, 2016.
- [10] Novák, J. et al., "Monte Carlo methods for volumetric light transport simulation," *Computer Graphics Forum (Proceedings of Eurographics - State of the Art Reports)*, 2018.
- [11] Miller, B. et al., *ACM Transactions on Graphics (Proceedings of SIGGRAPH)*, 2019.
- [12] de Lataillade, A. et al., "Monte carlo method and sensitivity estimations," *Journal of Quantitative Spectroscopy and Radiative Transfer*, 75(5) :529538., 2002.
- [13] Zeltner, T. et al., "Monte carlo estimators for differential light transport," *ACM Transactions on Graphics*, 40(4) :116, 2021.
- [14] El Hafí, M. et al., "Three viewpoints on null-collision monte carlo algorithms," *Journal of Quantitative Spectroscopy and Radiative Transfer*, 260 :107402, 2021.
- [15] Galtier, M. et al., "Integral formulation of null-collision monte carlo algorithms," *Journal of Quantitative Spectroscopy and Radiative Transfer*, 125 :5768, 2013.
- [16] Nyffenegger-Péré, Y., "Coupler le rayonnement et la spectroscopie raie par raie dans un même algorithme de Monte Carlo : permettre le calcul de référence des forçages radiatifs", PhD thesis, Paul Sabatier University.
- [17] Tregan, J.-M. et al., "Convergence issues in derivatives of monte carlo null- collision integral formulations : A solution," *Journal of Computational Physics*, 413 :109463, 2020.
- [18] Lapeyre, P. et al., "Monte-carlo and sensitivity transport models for domain deformation," *Journal of Quantitative Spectroscopy and Radiative Transfer*, 251 :107022, 2020.
- [19] Lapeyre, P. et al., "A physical model and a monte carlo estimate for the specific intensity spatial derivative, angular derivative and geometric sensitivity," working paper or preprint : <https://arxiv.org/abs/2206.05167>.