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Atmospheric Chemistry and Physics (ACP)

Dear Editor,

Here we address the latest review of our manuscript titled "A CO₂ - Δ¹⁴CO₂ inversion setup for estimating European fossil CO₂ emissions"

We sincerely appreciate feedback on our manuscript and are committed to improve it further.

Below, we provide a detailed response (in regular font) to each of the referee's comments (in italics), indicating how we have addressed them in the revised manuscript. We hope this clarifies any misunderstandings and demonstrates our commitment to meeting the high standards of the journal.

Referee's comments

L151: In line 142, the authors state "the background mixing ratios are calculated by computing a smoothed and detrended average of real observations" but in line 151, they still state that the background is modelled. The authors state that they changed this sentence according to the same comment I made in the first review, but they changed "modelled" to "assumed" for the mixing ratio "y" but not the background "y^b". If I understand correctly the sentence should be e.g.: "y is the modelled mixing ratio, and y^b is the background mixing ratio derived from smoothing real observations..."

We agree with the reviewer, we made a mistake when modifying the sentence in the last rebuttal. We have modified the sentence in L151 as follows:

"where y_{CO_2} and $y_{\text{C}\Delta^{14}\text{C}}$ represent the modeled mixing ratios of CO₂ and CΔ¹⁴C, respectively, and $y_{\text{CO}_2}^b$ and $y_{\text{C}\Delta^{14}\text{C}}^b$ denote their background mixing ratios (i.e., the boundary condition), derived from smoothed real observations (see Section 3.3)."

L151 and Eq.1b: It would be clearer to simply state that y is the modelled mixing ratio, and y^b is the modelled background mixing ratio. And then I suggest changing the name of the variable y_C_delta^14C to simply e.g. y_14CO2 since this is not a delta value any more since it represents the 14CO2 mixing ratio. Then below, the authors should state that the y_14CO2 mixing ratio is calculated from the delta14C value and that mixing ratio is modelled (and not delta14C) because this is additive.

L170: *There appears to still be some confusion about the meaning of the mixing ratio of $^{14}\text{CO}_2$. Mixing ratios are calculated with respect to the volume (volume mixing ratio) or mass (mass mixing ratio) of air. Since fossil fuel emissions contain no $^{14}\text{CO}_2$ then these emissions will not affect the mixing ratio of $^{14}\text{CO}_2$ (as far as these emissions do not affect the total mass of air). They will only affect the mixing ratio of CO_2 . So although fossil emissions affect the ratio of $^{14}\text{CO}_2$ to $^{12}\text{CO}_2$, and thus $\delta^{14}\text{CO}_2$, they do not affect the mixing ratio of $^{14}\text{CO}_2$ as this is relative to air (not CO_2).*

Here we address the points raised related to L151, Eq. 1b and L170. The referee is correct that emissions of fossil CO_2 do not affect $^{14}\text{CO}_2$ mole fractions, which is precisely why it is not useful to convert measurements of $\Delta^{14}\text{CO}_2$ into $^{14}\text{CO}_2$ mole fractions for an inversion. Emissions of fossil CO_2 show up as strong depletions in $\Delta^{14}\text{CO}_2$ space, but $\Delta^{14}\text{CO}_2$ cannot be transported because it is not additive. For instance, $\Delta^{14}\text{CO}_2$ cannot be summed across grid cells to construct a “global total $\Delta^{14}\text{CO}_2$ ”. Although equation (1b) of Miller et al. (2012) is convenient for expressing the different forcings on atmospheric $\Delta^{14}\text{CO}_2$, it is not useful for transport modeling.

Instead, equations (1a) and (1b) of Miller et al. (2012) can be combined to derive equation (1b) of Basu et al., (2016), which serves as the basis for Equations 1-4 of our manuscript. In this formulation, the quantity $C \times \Delta_{\text{atm}}$ or $\text{CO}_2 \times \Delta^{14}\text{CO}_2$ is additive and therefore *can* be transported. Note that $\text{CO}_2 \times \Delta^{14}\text{CO}_2$ is *not* the mole fraction of $^{14}\text{CO}_2$, as per the definition of $\Delta^{14}\text{CO}_2$ (Stuiver, 1980; Stuiver and Polach, 1977). It is simply a made-up tracer whose emissions can be calculated given emissions of CO_2 , $^{14}\text{CO}_2$ and $\Delta^{14}\text{CO}_2$ source signatures, and whose atmospheric observations can be derived from measurements of CO_2 and $\Delta^{14}\text{CO}_2$. The formulation of the tracer $\text{CO}_2 \times \Delta^{14}\text{CO}_2$ lends itself to mass balance equations that can be coded up in an atmospheric inverse model.

Eq. 7: The authors state that the matrices and vectors have the following dimensions:

$x_c : (n_{\text{popt}}, n_{\text{topt}})$ but since x_c is a vector presumably the authors mean $(n_{\text{popt}} \times n_{\text{topt}})$
 $H : (n_{\text{obs}}, n_{\text{popt}} \times n_{\text{topt}})$
 $T_H \otimes T_T (n_{\text{pmod}} \times n_{\text{tmod}}, n_{\text{popt}} \times n_{\text{topt}})$

but then the dimensions of $T_H \otimes T_T$ do not conform with those of H , which has $n_{\text{popt}} \times n_{\text{topt}}$ number of columns. And since H is the Jacobian matrix corresponding to the optimized state vector, x_c , why is there any need for $T_H \otimes T_T$ because the mapping from the original to the optimized resolution appears to be already taken into account with H .

We agree with the referee. Indeed, the Kronecker product $\mathbf{T}_H \otimes \mathbf{T}_T$ is the part of \mathbf{H} used to map the fluxes from the modeling space to the optimization space. We replaced \mathbf{H} by \mathbf{K} (the transport operator in Equations 1, 2, and 4, with dimensions $(n_{\text{obs}}, n_{\text{pmod}} * n_{\text{popt}})$), such that $\mathbf{H} = \mathbf{K}(\mathbf{T}_H \otimes \mathbf{T}_T)$.

We have removed the Kronecker product from the equation and revised the explanatory paragraph as follows:

“Equation 7 can be rewritten as:

$$\delta_y = \sum_c \mathbf{H} \mathbf{x}_c$$

where \mathbf{H} is the Jacobian matrix of the observation operator with dimensions $(n_{obs}, n_{p_{opt}} * n_{t_{opt}})$, and \mathbf{x}_c , with dimensions $(n_{p_{opt}} * n_{t_{opt}})$, represents the portion of the control vector \mathbf{x} that contains offsets for the optimized categories c . Thus, \mathbf{x}_c is built from the relative contribution of each model time step t_{mod} (1 hour) and of each grid cell p_{mod} ($0.5^\circ \times 0.5^\circ$) to each optimized time step t_{opt} and cluster p_{mod} . Here, $n_{t_{opt}}$ and $n_{p_{opt}}$ represent the number of optimized intervals (weekly) and grid cell clusters (e.g. 2500 for biosphere), respectively.”

References

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