

Interactive comment on “On the relationship between tropospheric CO and CO₂ during KORUS-AQ and its role in constraining anthropogenic CO₂” by Wenfu Tang et al.

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We would like to express our gratitude to the anonymous reviewer for providing helpful comments. We certainly appreciate your time in reviewing this manuscript.

We apologize if this manuscript fails to convey succinctly our main points from the perspective of the reviewer. We will try to address each of the comments as best as we can. Certainly, we value your comments in improving this manuscript, as we also recognized the prospects of joint inversions in future studies.

————— Please see our responses to your comments below.

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(General Comment). The manuscript "On the relationship between tropospheric CO and CO₂ during KORUS-AQ and its role in constraining anthropogenic CO₂" by Tang et al. presents analysis based on the CO, CO₂ and ¹⁴CO₂ data collected during the KORUS-AQ campaign over South Korea. It compares simulations of CO, CO₂ and FFCO₂ concentrations from a global transport model to the data and presents inversions of the FFCO₂ emissions in East Asia (Eastern China, Korea and Japan) using this transport model and these data.

The prospect of the joint analysis of CO₂, CO and ¹⁴CO₂ data supported by transport model simulations and of their joint assimilation in an inversion system is very promising. Sometimes, the manuscript nearly reaches interesting insights on this topic. However, in a general way, the study and the manuscript fail to exploit the potential of such analysis. I think that more work and thoughts are needed to produce a paper that deserves publication and that this goes beyond what is usually done for "major revisions".

Response: Yes, this is a paper about the relationship between CO and CO₂. We also emphasize that while our inverse analysis is the final key point of this work, the two other important key points that we are highlighting in this work are the following:

a) evaluation of CAM-Chem with CO₂ and CO tags using prior fluxes from a posteriori fluxes from state-of-the-art global CO₂ inversions (i.e., CAMSv117r1, CT2017, CTE2018). To our knowledge, this is the first development of a joint CO₂ and CO modeling in community Earth system model, CESM/CAM-Chem. This is also one of the few studies in recent years that takes advantage of flux/source inversion products as priors in the model. We note that we have also compared our simulations to Carbon Tracker CO₂ fields to ensure that our simulations are reasonable.

b) detailed description on CO and CO₂ error statistics. Our intention is to highlight the need to evaluate the statistics of the CO and CO₂ relationship using aircraft measurements, prior to any inverse analysis. We highlight, in particular, the need to pro-

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vide spatiotemporal context on these relationships, which in previous inversion studies heavily rely on sensitivity tests. In this work, we highlight the importance of having knowledge of 'spatial error covariance localization length scales' prior to inversion.

————— 1a) It is often very difficult to follow because the writing and the reasoning are not structured and rigorous enough. A large amount of sentences are confusing because of the lack of clarity, precision and explanations. The reading of the long series of statistics lacks of hierarchy. The use of simulations or datasets that are not much exploited in the analysis does not help. For instance, efforts are needed to follow, in section 3, whether or why under or over estimations of concentrations are supposed to highlight the under or over estimation of local, regional or global sources and sinks. The reading of section 4 is even more difficult.

Response: We are not quite clear as to what the reviewer is referring to about structure and hierarchy. As for Section 3, the aim is to evaluate the modeled abundance against aircraft measurements, which we summarized in Table 2. Our intention is to lay out the basis of error statistics as any typical model evaluation would conduct and provide some physical context on these errors. We recognized that these statistics (bias, correlation, rmse, as well as error correlation, enhancement ratio), while common, have direct relevance to Section 4 where we expound more on the spatial variability of these errors. We note that a more accurate representation of both the a priori error and observation error covariance matrices is in fact the crux of the inverse analysis, as with any other inversion methods that account for these uncertainties. We do emphasize the need to investigate the covariances and error covariances (and sensitivities) as these are the terms in the analysis that 'transfer information' to unobserved parameters that we are interested in estimating. We see this as an important step before any inverse analysis is undertaken.

The spatial data groupings correspond to different sampling regimes which help us understand issues of the CTM (i.e., representativeness, boundary layer, transport, mixing, emissions, sinks). Here, we provide a spatial and temporal context on the fidelity of the

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model to capture these statistical measures, as these measures represent different components of the error characteristics.

Our emphasis is also to understand the vertical structure of these errors (Figure 4) and associate them to potential model deficiencies. As such, we used this section to compare not only with CarbonTracker (which we used for CO₂ initial condition) but also to CAMS, based on our previous work also published in this journal. As we mentioned above, we recognize the need to provide an evaluation of CAM-Chem to establish its fidelity (relative to state of the art models of CO₂) since this is the first development of such a system. We also note that this system is different from most models as we are taking advantage of 3 different a posteriori fluxes (CAMS, CT2017, CTE2018) and used these as our priors. This is the reason why there is an added dimension to our model evaluation.

We approach this model evaluation similar to Tang et al. (2018) in order to be consistent with our previous work. We want to show across all the sections, the main essence of this paper: to show the relationship between CO and CO₂ and ffCO₂. For our comparison with 14CO₂ data, we note that to our knowledge this is the first publication of this data for this campaign with comparison to model equivalents. This is the reason why we have a separate sub-section. This is also very timely as there have been recent publications (also in this journal) about 14CO₂ in nearby region.

It is unfortunate that the reviewer finds this section to be confusing. We do recognize that this manuscript is long but we chose to elaborate more on these statistics. This is the main reason why prior to our inverse analysis (Section 5), we expanded our discussion on error statistics to make sense of error covariances used in inverse analysis and to come up with reasonable 'localization' through the use of a subset of measurements closer to the sources (section 4.1 and 4.2 leading to section 4.3). We emphasize that this is not a manuscript of an inverse analysis of ffCO₂ but more of elucidating the need to investigate the relationships prior to any inversions.

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————— 1b) Furthermore, despite being relatively long, the text goes too fast on some of the critical parts of the reasoning like the rationale for the study in the introduction and the justification for the use of specific inversion configurations and parameters.

I think that many assumptions and analysis are debatable and that the manuscript shows a lack of hindsight on the topic and results of this study.

Response: We regret that the reviewer finds that many of our assumptions and analysis are debatable. We do recognize the length of this manuscript. As we mentioned above, we emphasize that this is not an inverse study per se. We intend to show the role of CO, CO₂ and ffCO₂ relationship on the inverse analysis (section 5) but only to briefly make an example. In this manuscript, we focus on discussing the spatial variability of the terms used in the inverse analysis and highlight the need to evaluate these relationships in CTMs. Again, the approach we have taken on this paper (i.e., revisiting Palmer et al. 2006) could open up discussion on the methodology for joint inverse analysis. Our intention is to bring to light some of the issues that need to be considered for future joint inversions, especially on the use of satellite retrievals for inversions, which lacks the vertical information that is critical in identifying transport-related errors that may confound any inverse analysis. This is not only true in single-species inversions but in fact is an added complication with multi-species inversions that should be carefully considered. There is relevance to adding CO observations in reducing uncertainties on top-down estimates of ffCO₂; especially with the availability of atmospheric composition retrievals (CO, NO₂) nowadays (i.e., TROPOMI, GOSAT-2). We emphasize the need to better understand the joint inversion, as this has yet to be investigated rigorously and that the species relationships (and associated errors) should not be taken for granted. As mentioned in the manuscript, our approach is based on widely used Bayesian synthesis inversion introduced by Enting (2002) and used by Baker et al. (2006), and more recently applied for multi-species inversion by Boschetti et al. (2018). While Wang et al. (2018) used a similar method (for single-species CO₂ inversion), they focused on investigating the impact of reduced carbon component of CO₂

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based on CO on their estimates. We note that this approach, while with disadvantages, is still used in global inversions.

————— 2a) The specific scope and the objectives of sections 3 and 4 are not clear. This manuscript is the 5th one analyzing the CO₂ and/or CO data from the KORUS-AQ campaign using transport models (after Tang et al. 2018, Halliday et al. 2019, Tang et al. 2019a and Gaubert et al. 2020, ACPD). How do section 3 and 4 draw on these previous publications and bring new learning?

Response: We thank the reviewer for pointing this out. As we mentioned above, this is the first evaluation of a community Earth system model (CESM/CAM-Chem) for joint CO₂ and CO simulations. Tang et al. 2018 is about CAMS not CAM-Chem. We draw insights to this paper by comparing CAMS to CAM-Chem given the relatively different model configuration between these models. Halliday et al. 2019 is about enhancement ratios mostly focused on data. We draw insights to this paper by comparing their analysis to model equivalents especially with regards to the vertical distribution of enhancement ratios and correlations. As we mentioned in the Introduction, we view this as complementary (i.e., data and model comparison with inverse analysis). Tang et al. 2019a is about CO not CO₂ using CAM-Chem. We draw insights on this work by using the optimized CO and error statistics (model vs data) which we used in our inverse analysis. Gaubert et al. 2020 is about data assimilation (with EnKF). We draw insights to this work by comparing tagging simulations that we did versus EnKF emission estimation that they conducted.

Note that this is the first time we provide model-data synthesis for CAM-Chem on both CO₂ and CO. Here, we highlight the correlation (and error correlation) as this is really the crux of any inversions and clearly there is lack of detailed investigation reported in literature with regards to these covariances for joint inversions. And if we are to proceed with joint global inversions, we ought to be aware of the deficiencies of the CTMs in capturing these relationships. And what better way to do this but by comparison with aircraft data. Also, note that 14CO₂ during KORUS-AQ is first reported in this

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manuscript. If there's new learning on this manuscript, it would be the synthesis of all these in the context of inverse analysis.

————— 2b) Furthermore, opposed to what is claimed repeatedly (e.g. in the title, the abstract, the beginning of section 1.1, the beginning of the conclusion...), I hardly see how these sections feed the configuration (in particular the error covariance matrices) or the analysis of the inversions in section 5.

The introduction does not help much. Until the beginning of section 1.1, this is a collection of very general (and sometimes misleading) statements about the monitoring CO₂ anthropogenic emissions using atmospheric data. Section 1.1 fails to bring clear specific context, rationale and objectives for this study. Lines 150-154 look like a summary of the activities rather than a list of objectives. Lines 154-172 attempt to distinguish this new study from the previous ones by pointing to practical differences that are hardly convincing. Lines 769-770 and 775-776 claim that the set-up of matrix S_e follows values of errors on CO and CO₂ and err_{RCO,CO_2} from section 4 but I do not see how. Anyhow, the errors and the correlation of errors in CO and CO₂ modeled concentrations are driven by the atmospheric transport, the surface fluxes (and other source and sinks), and to errors in both the transport model and in the modeled sources and sinks. I do not see why it should be used to characterize transport model errors only. The authors seem to miss the links between their statistics of model-data differences and S_a (see below my general comment about the key role that this matrix should have played).

Response: We thank the reviewer for what appears to be a thorough read of our manuscript. We value your comments. We again regret that the reviewer fails to find a clear understanding of the flow. This may be due to the length of the manuscript. It is quite unfortunate that the reviewer views the Introduction as a "collection of very general (and sometimes misleading) statements". Our intent was to provide context on the role of CO to GHG and $ffCO_2$ in particular. Why CO? We apologize if our interpretation of literature is inaccurate from the reviewer's perspective. We would ap-

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preciate it though if the reviewer points out some of these inaccuracies and misleading statements.

To reiterate, Section 3 is about comparison with data and models. Section 4 is about error correlation and enhancement ratios. Section 5 is about a simple demonstration of a joint inverse analysis. Section 4 feeds to Section 5 through the analysis of the spatial extent of these errors (especially in the vertical). We use the error correlations from Section 4 in particular and localized the dataset we used in the inversion to account for potential confounding factors (transport errors) especially aloft. Again, this paper is not all about joint inverse analysis.

We would like to repeat our response to Reviewer 2 with regards to S_a and S_e as this is also relevant in later comments of the reviewer:

Palmer et al., (2006), as well as Boschetti et al. (2018), relied on tagging CO₂ and CO components using a CTM (linearizing both CO₂ and CO) to conduct a suite of Bayesian synthesis inversions. Boschetti et al. (2018), however, utilized STILT to obtain these Jacobians.

Within the CTM (where both CO₂ and CO are simulated), there is, however, a physically-based coupling between these runs through the use of emission ratios in prior emissions for $ffCO_2$ and CO. That is, they are both emitted to the atmosphere in the model by sector-specific assumptions of emission factors per species, and associated emission ratio between species. As we showed in Tang et al. (2019b), for a particular combustion process, we can estimate the combustion products by stoichiometry:

— please see figure 1 for equations and figures —

And so, if we are to look at the equivalent atmosphere abundance of these emission tags (as they are emitted to the atmosphere), they are mostly correlated near the sources, with correlations diminishing downwind of the source. Obviously, observ-

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ing the relationship downwind does not imply that the correlations (or the lack thereof) are created by this co-emission alone, but typically also through mixing, transport, and associated sinks. These are further quantified in the inverse analysis through the a priori error covariance matrix (i.e., errors in emission ratios) and observation error covariance matrix (i.e., errors in transport, mixing, among others).

We note here that a more accurate representation of both the a priori error and observation error covariance matrices is in fact the crux of the analysis, as with any other inversion methods that account for these uncertainties. We do emphasize the need to investigate the covariances and error covariances (and sensitivities) as these are the terms in the analysis that ‘transfer information’ to unobserved parameters that we are interested in estimating.

We agree that it is worth comparing with what happens in nature. In fact, this is one of our key points – i.e., can we evaluate this ‘modeled’ relationship? Are these relationships that can be derived from observations of the species abundance in nature reasonably captured by CTMs? We agree with Peter regarding the expression on sources. Please see our response to Comment 3). In essence, we are looking at α (effective emission factor in our notation), which we argue should be consistently represented in the a priori inventory for both CO and CO₂. At present, with few exceptions, ffCO₂ is derived from energy statistics separate from air pollution emission inventories.

We note, however, that as we previously mentioned the yield is quantified as effective emission factor, and the emission ratio is the quantity we can evaluate with observations by looking at the derived enhancement ratio ($\Delta\text{CO}/\Delta\text{CO}_2$) from observations of CO and CO₂ abundance. Note that this can only be ‘appropriately’ evaluated with observations near the emission sources, just like studies using field campaign measurements to estimate emission ratio. Otherwise, the enhancement ratio is confounded by mixing and transport and hence cannot be taken and interpreted as the abundance equivalent of emission ratio. See for example Mauzerall et al. (1998) Figure 4 (shown below):

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— please see figure 2 for equations and figures —

where, the regression line of [X] and [Y] (i.e., $(\Delta[Y])/(\Delta[X])$) downwind is steeper than over a fresh plume. This complication arises from various sources and sinks, other than combustion which in reality are ubiquitous. And so, here we take the enhancement ratio only as a key diagnostic on the bulk relationship between CO and CO₂ (resulting from a combination of emission ratio, mixing and sinks).

Palmer et al. (2006) investigated the sensitivities of different assumptions of activity, and emission factors and concluded that errors in emission factors largely contribute to the uncertainty estimate. Rather than ‘reinventing the wheel’, we took a more conservative value of 0.5 (Boschetti et al. 2018 used 0.7, while Palmer et al. 2006 suggested >0.7). As we noted in the manuscript, there is little information from flux measurements to verify this error correlation. Also, note that these correlations, when used in the inversion, is not correlation between CO and CO₂ abundance near the source but the correlation of the errors between modeled CO and CO₂ abundance.

We agree on the proper interpretation of observation error covariances, R. We also agree on the influence of R on the Bayesian synthesis inversion mean estimate and a posteriori error covariance estimate. In fact, both P (or B) and R influence the first and second moment estimates of the conditional pdf. This is the main reason why prior to our inverse analysis (section 5), we expanded our discussion on error statistics to make sense of R and to come up with reasonable localization of R through the use of a subset of measurements closer to the sources (section 4.1 and 4.2 leading to section 4.3).

This influence is clearly more complex but it shows as well that the a posteriori error covariance is a function of both the a priori error and observation error covariances. While the Jacobians (tags) itself should exhibit some correlation as the species are co-emitted, we need to quantify the uncertainties, not only on error variances but more importantly on the error correlations.

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We note as well that our inverse analysis is a product of this investigation on R. By way of expressing the sensitivity (enhancement ratio) with correlation (see related response in Comment 3), we get:

— please see figure 3 for equations and figures —

The same relationship holds for the error sensitivity. In this manuscript, we focus on discussing the spatial variability of these terms, as well as the corresponding magnitudes, so as to guide us in defining the cross-species error correlation components of R (see Eq. 4 of our manuscript). In fact, we used a more conservative error correlation of 0.33 (Boschetti et al. 2018 and Palmer et al. 2006 used 0.7). We opted not to show our tests where we vary our assumptions of these values as this was already done by Palmer et al. (2006). Rather, we choose to emphasize the need to have an estimate of these values based on data, especially aircraft measurements given known issues with vertical transport/mixing in CTMs. We intend to show the role of this relationship on the inverse analysis (section 5) but only to briefly make an example. We emphasize that this is not a manuscript of an inverse analysis of ffCO_2 but more of elucidating the need to investigate the relationships prior to any inversions.

As an aside, if we are to recast the first moment of $p(x|y)$:

— please see figure 4 for equations and figures —

————— 3a) There is a critical lack of proper discussion on the spatial extent and scales that are suitable for the analysis of the data. Parts of sections 3 to 5 attempt to distinguish the influence of Eastern Asia or the rest of the world vs that of Korea, I340-350 discuss the sources overflowed during the campaign and the flights over the West Sea are said to be designed to capture “China pollution outflow”. Some sentences even raise (too late) some concerns associated to the coarse spatial resolution of CAM-chem. However, in general, and in particular in the title, introduction and section 2, there is no real reasoning regarding the observation footprints and regarding the modeling and inversion domain and resolution that are suited for these data

C11

Response: This is quite unfortunate as this is one of the main theme of this manuscript. We revisited Palmer et al. (2006) which uses a global CTM and aircraft data over similar region. We are well cognizant of the representativeness of these measurements. This is why we ended up aggregating our tags (emission regions). In particular, we only focused on East Asia and Korea+Japan. This is also the reason why we use more local data close to the source rather than using all the data. We note however that we have compared to CarbonTracker and CAMS of similar resolution. Again, the inverse analysis is intended to show an example as knowledge of enhancement ratio and correlation extends beyond inverse analysis.

————— 3b) The introduction mixes all inversion scales and all types of observation networks. Nothing is said about the wind fields during the campaign. The comparisons of a single global coarse resolution model to TCCON individual sites, to OCO-2, to MOPITT data or to the aircraft data are brought together without consideration for the differences between these observation datasets in terms of flux and process representativeness. The statistics obtained here are sometimes compared with results from other campaigns in different regions or with different coverage, or from “state-of-the-art” models (without mentioning whether they are global or regional). The contrast between the spatial extent of the regions for which the total emissions are rescaled by the inversion and the local (nearly vertical) reading of the data footprints at I340-350 is questioning.

Given the spatial extent of the KORUS-AQ campaign (less than $8^\circ \times 8^\circ$) and its density of data over South Korea, the use of a global 1° resolution transport model with coarse vertical resolution to analyze it is not obvious and sounds like a step backward compared to the previous publications on this campaign (which used higher resolution models). From what I understand, the model is interpolated at each observation location and all the statistics are derived over the ensemble of observation locations. What can be the meaning of such statistics at a resolution much finer than that of the model?

C12

Response: We compared our results to CarbonTracker as well. CAMS is also compared with CAM-Chem. Palmer et al. (2006) used a global CTM. There are several global inversions as well in literature that in fact used regional datasets. The intent of using TCCON, OCO-2 and MOPITT is to provide a comprehensive (albeit limited in time) evaluation on the extent of CAM-Chem (relative to other models) to capture not just the regional scale but global features of both CO and CO₂ (e.g., surface and column, synoptic and boundary layer). There's obvious (and understandable) argument about using global CTMs to estimate regional sources. But we have carefully presented all these statistics and show that at some of these scales, it is appropriate to estimate regional sources. Our evaluation of CO₂ is comparable to CarbonTracker and CAMS. Having said this, we will modify our manuscript to ensure we mention the appropriate scales of these comparisons.

It is unclear as to what the reviewer is referring to with regards to the statement "brought together without consideration for the differences between these observation datasets in terms of flux and process representativeness". Across more than 20 years of inversion literature, we are well-cognizant of the issue of representativeness especially with global models. We will try to revise this manuscript to provide more emphasis. However, providing proofs and arguments related to representativeness and aggregation errors is beyond the scope for this manuscript.

We would like to reiterate here that joint inversions are relatively new. While dealing with confounding factors is necessary, we intend to show that CTMs that do not capture even the large-scale features of the CO and CO₂ relationship would have difficulty taking advantage of the information that CO brings. In fact, we emphasize that joint inversion of CO and CO₂ has to be conducted at scales commensurate with the lifetime of CO. Our presentation of the statistics is an honest way to show the scales at which CAM-Chem has captured.

————— 4a) The usual concept for the co-assimilation of CO and CO₂ is that the signal from misfits between modeled and measured CO can be used to add constraint

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on the inversion of FFCO₂ emissions because uncertainties in the FFCO₂ emissions are connected to uncertainties in FFCO emissions. This usually translates into two options for the inversion: (a) rescaling activity levels underlying both FFCO and FFCO₂ emissions rather than FFCO and FFCO₂ emissions separately or (b) rescaling separately FFCO and FFCO₂ emissions accounting for positive correlations between their respective prior uncertainties. The key challenge for joint CO-CO₂ inversions is usually thought to be the uncertainties and high spatial and temporal variations in the CO/CO₂ emission ratios i.e. in (a) the uncertainties and variations in the CO and CO₂ emission factors to be multiplied by the common activity indices to get emissions and in (b) the level and variations of the positive correlation between the prior uncertainties in FFCO and FFCO₂ emissions. The authors of this study control separately FFCO and FFCO₂ emissions but neglect the correlations between their respective prior uncertainties or acknowledge that they have no idea about how to parameterize them, cutting the critical connection between FFCO₂ and FFCO (see lines 819-830). The configuration of Sa is actually made all the more difficult by using two different inventories for prior FFCO₂ and FFCO emissions, which undermines the ability to rely on tight connections between these emissions (especially since the CO inventory has been multiplied by 2 to better fit the data before the inversion).

Response: Please see our response to comment 2b) and to Reviewer 2 on Sa. While these inventories are taken from different sources, we argue that evaluating the bulk enhancement ratios would provide insights on their inconsistencies. We highlight in fact the need to have consistent emission inventories for GHG and air quality. This manuscript is trying to revisit Palmer et al. (2006) and open up discussion on meaningful ways on how these correlative measurements can be used effectively in ffCO₂ estimation.

————— 4b) The authors even assume that these correlations are negative, i.e. that the combustion efficiency could be the main source of uncertainty in both FFCO₂ and FFCO emissions. However, the prior FFCO and FFCO₂ emissions are based on differ-

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ent inventories (and FFCO emissions have been multiplied by 2), so that, in principle, the ratios between these emissions should not correspond to an assumption of this efficiency. In a more general way, such an assumption is quite surprising. Uncertainties in the level of activity at national, and even more at regional to local scales should be a dominant source of uncertainty in FFCO₂ emission inventories (especially at sub-annual temporal scales). Such a driver of uncertainty in FFCO₂ emissions raises correlations with uncertainties in FFCO emissions. Even if uncertainties in CO emission factors are one of the main sources of uncertainties in FFCO emissions, their counterpart in FFCO₂ emissions (the generation of plus or minus CO₂ depending on the combustion efficiency) can hardly balance this driver. The assumption of negative correlations between uncertainties in FFCO₂ and FFCO prior emissions is further weakened by various discussions in sections 3 and 4 that point to common underestimation or overestimation of sources in both the CO₂ and CO inventories.

Response: Please see our response to comment 2b) and to Reviewer 2 on Sa as well. Based on Palmer et al. (2006), they found that the emission factor contributes largely to the uncertainty. Looking at emission ratios, which to an extent (Eq A3) can be proportional to enhancement ratio especially near the source, activity cancels out. The utility of CO lies not on the magnitude alone but its sensitivity to CO₂ (i.e., its relationship). Please see our recasting of the a posteriori estimates in our response to comment 2b) as well. We agree with the reviewer that it is non-trivial to attribute the mismatch on specific sectors or to activity alone. We cannot differentiate this with the data so far nor can we differentiate this with CO alone. Our intention here is to highlight the information content of CO on ffCO₂ estimation. But in order to do this, we need to ensure that we understand the error statistics and that the CTMs capture this relationship. Again, the inverse analysis is an example of showing this information content.

————— 4c) The idea that Se could ensure the expected connection between FFCO₂ and FFCO is not relevant. Se prevents the inversion from overfitting the data and limits

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the corrections to the prior emissions. Correlations between CO₂ and CO transport modeling errors help the inversion better filter such errors when deriving FFCO₂ and FFCO emissions but this has a very indirect and weak impact on the connection between these emissions. One could think that the results from the inversions in section 5 contradict these statements. However, I assume that the consistency between FFCO₂ and CO₂-CO inversions and their divergence from CO₂ inversions is intrinsically linked to the very low dimension of these inversions (with 4 or 8 unknowns). Various assumptions and settings of these inversion raise issues (see below) and I thus have the feeling that this result is not robust.

The general topic of the CO-CO₂ correlations that should be at the heart of this paper is complex and I feel that the authors missed it.

Response: We beg to disagree. As with Palmer et al. (2006), in the Bayesian synthesis inversion framework, Se does not only represent observation error covariances, but more dominantly represent (supposedly) model errors including transport, mixing and sinks. Hence, the mismatch (innovation) reflects errors in capturing the relationship – perhaps not on the emission level but on the representation of the variability of the enhancement ratio due to differences in lifetime of the species (e.g., sources and sinks). The main issue with these inversions is the fact that we treat the model as perfect and it is extremely difficult to quantify model errors. Errors on the Jacobian (which we are trying to assess) should not be taken for granted.

Yes, the consistency is due to lower dimension; mainly as a result of our effort to understand the scales at which the information content of CO can be utilized effectively.

It is again unfortunate that the reviewer sees the general essence of this work as something that we missed. We will continue on this work and refine our statements to improve on clarity.

We note that this concept, while old, is already difficult to grasp. How much more by introducing this to inverse analysis. We emphasize that there are only very few studies

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about joint inversion, mainly due to the misconception that the inherent variability of these relationships is far greater than the information content. We argue that we need to start thinking about extracting information more effectively.

————— 5a) I don't understand how global inversions (even if they focus on East Asia) assimilating CO₂ (with or without CO) can behave correctly when inverting scaling factors for FFCO₂ emissions only and keeping other CO₂ fluxes fixed. The "posterior" biogenic fluxes from CTE and CAMS bear large uncertainties. The problem is enhanced by the lack of spatial resolution of the inversion, which does not help distinguish the patterns from anthropogenic vs biogenic fluxes in the data.

Response: This is a valid point. We do not have a concrete answer to this. We view this as a signal-to-noise issue: i.e., we should see a large source of ffCO₂ over East Asia and that expect to be able to 'track' this with CO. We also think that posterior errors are smaller than prior errors especially for CESM/CAM-Chem, which is an Earth system model with dynamic vegetation. We view this as well as a two-step approach (or iterative), where the expectation is that posterior fluxes fit atmospheric abundance better than the prior fluxes (but obviously perhaps for the wrong reason).

————— 5b) I do not understand the rationale behind removing observations above 3 km (the "localization purposes": l791-792 refer to "previous section" but I do not see where sections 3 and 4 help understand this choice). The inversions are global, and data above 3 km should be helpful for constraining ROW.

Response: We selected the data where ffCO₂ signal is relatively strong (Figure 8 of Section 4). In essence, we view this as reducing the noise. For example, data above 3km do not really have ffCO₂ signal from both East Asia and Korea. You can also think about this as tapering the radius of influence of a given datapoint by localizing its impact – similar to error covariance localization in EnKF. Here, we are effectively localizing the Jacobians and use of data to lower than 3km.

————— 5c) I have some doubts regarding the way the FFCO₂ simulations are com-

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pared to gradients of FFCO₂ from gradients in 14CO₂ observations: I believe that the authors should have computed, in the FFCO₂ simulations, the gradients to the location of the 14CO₂ background sites rather than introduce a "ffCO₂ offset".

Response: This is a valid point. We think that our global ffCO₂ is under-estimated and that spatial gradient (variability) is ffCO₂ offset is small. We view this to be similar to a constant in a regression analysis. However, we acknowledge this issue. Thank you for the suggestion.

————— 5d) There is a lack of consideration for the inventories. As mentioned above, the fact that the FFCO₂ and FFCO inventories are quite independent strongly hampers the interpretation of the model-data comparisons and the reading of CO/CO₂ ratios in the model. The EDGAR inventory, which plays a critical role in this study (this is the FFCO₂ emission inventory behind the simulations in sections 3 and 4 and the prior emission inventory in section 5) is not even named in the main text, the authors speaking about CTE and CAMS fluxes (which combine natural fluxes optimized through global inversions with the EDGAR inventory) only. Differences between the CAMS analysis and CAM-Chem simulations are not supported by insights on the emission maps behind these two simulations. The FFCO₂ emissions from the inversion are hardly compared to official inventories (and the CO emissions from the inversions are not discussed).

Response: We mentioned it in Table 1 and Table S1. We showed a regional budget for ffCO₂ in Table S2. In addition, we showed the spatial distribution of the fluxes in Figure S1.

We do acknowledge that we did not choose to compare rigorously with emission inventories at this stage of our work, given that we are only starting to get a handle of this relationship and full, comprehensive inversion is beyond the scope of this work at this point.

————— 5e) I could draw a long list of secondary issues in specific paragraphs or

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sentences (and even in the mathematical notations and equations) but I restrain myself to the main ones raised above

Response: Your input is always welcome. We follow the notations by Rodgers 2001 and Palmer et al. 2006. Similar notations are used by Boschetti et al. (2018) as well.

————— 5d) In conclusion, I think that the scope and configuration of the analysis should be rethought, and that the presentation of such a study should be strongly improved. I thus recommend this manuscript to be rejected and re-submitted after a deep revision.

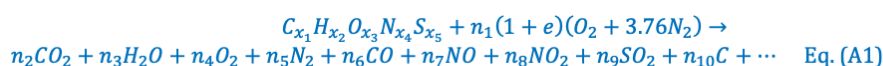
Response: Thank you for your comments. We will revisit the manner in which we presented our study for this manuscript.

References (not cited in the manuscript):

Mauzerall, D.L., Logan, J.A., Jacob, D.J., Anderson, B.E., Blake, D.R., Bradshaw, J.D., Heikes, B., Sachse, G.W., Singh, H. and Talbot, B., 1998. Photochemistry in biomass burning plumes and implications for tropospheric ozone over the tropical South Atlantic. *Journal of Geophysical Research: Atmospheres*, 103(D7), pp.8401-8423.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2020-864>, 2020.

C19



where emissions of these intermediate products are typically expressed as:

$$\begin{aligned} E_x &= \sum_s [A_s \cdot EF_{x,s} \cdot (1 - CE_{x,s})] \\ &= \sum_s [A_s \cdot EEF_{x,s}] \end{aligned} \quad \text{Eq. (A2)}$$

Here, E_x is the total mass of emissions for species x , $EF_{x,s}$ is its associated emission factor for a specific source/sector s , and A_s is the activity level of the source. $CE_{x,s}$ corresponds to effectiveness of control measure and $EEF_{x,s} = EF_{x,s} \cdot (1 - CE_{x,s})$ is the effective emission factor. When we take the ratio of emissions (Eq. A2) of co-emitted species x and y , we obtain

$$\begin{aligned} \frac{E_y}{E_x} &= \frac{\sum_s [A_s \cdot EEF_{y,s}]}{\sum_s [A_s \cdot EEF_{x,s}]} \\ &= \sum_s \left(\frac{EEF_{y,s}}{EEF_{x,s}} \right) \left(\frac{E_{x,s}}{E_{x,total}} \right) \end{aligned} \quad \text{Eq. (A3)}$$

This ratio can be expressed as the sum of the products of the ratio of effective emission factors ($R_{x,y,s}^{EEF}$) and the fractional contribution of emission sector f for species ($f_{x,s}$).

Fig. 1.

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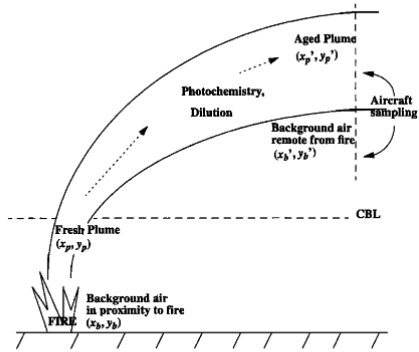


Figure 4a. Schematic of plume formation and transport. The (x_p, y_p) and (x_b, y_b) indicate mixing ratios within the fresh plume at the time of sampling and in background air in proximity to the fire, respectively. The (x_p', y_p') and (x_b', y_b') indicate mixing ratios within the plume sampled by the aircraft and in background air in the vicinity of the sampled plume, respectively.

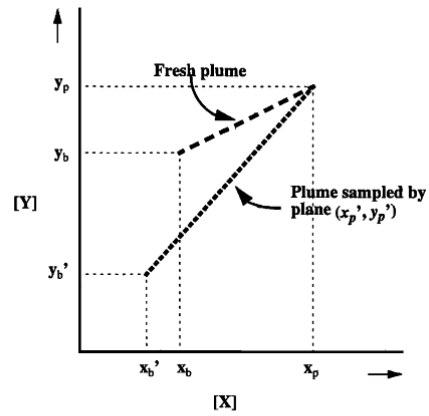


Figure 4b. Schematic of plume sampling. Long-dashed line indicates a fresh plume with enhancements $(x_p - x_b, y_p - y_b)$ relative to background mixing ratios near the fire (x_b, y_b) . Dotted line indicates a plume remote from the fire which the aircraft measures relative to local background mixing ratios (x_b', y_b') . Enhancement ratios are obtained from the slopes of these lines.

Fig. 2.

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$$\frac{\Delta CO}{\Delta CO_2} = \frac{cov(CO, CO_2)}{var(CO_2)} = \rho_{CO, CO_2} \frac{\sigma_{CO}}{\sigma_{CO_2}}$$

Fig. 3.

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$$\mathbf{x}^a = (\mathbf{H}^T \mathbf{R}^{-1} \mathbf{H} + \mathbf{B}^{-1})^{-1} (\mathbf{H}^T \mathbf{R}^{-1} \mathbf{H} \mathbf{y} + \mathbf{B}^{-1} \mathbf{x}^b)$$

or

$$\mathbf{x}^a = \mathbf{x}^b + \mathbf{B} \mathbf{H}^T (\mathbf{H} \mathbf{B} \mathbf{H}^T + \mathbf{R})^{-1} (\mathbf{y} - \mathbf{H} \mathbf{x}^b)$$

to:

$$\mathbf{x}^a - \mathbf{x}^b = \mathbf{B} \mathbf{H}^T (\mathbf{H} \mathbf{B} \mathbf{H}^T)^{-1} (\mathbf{H} \mathbf{B} \mathbf{H}^T) (\mathbf{H} \mathbf{B} \mathbf{H}^T + \mathbf{R})^{-1} (\mathbf{y} - \mathbf{H} \mathbf{x}^b)$$

we can easily recognize that the increment $\mathbf{x}^a - \mathbf{x}^b$ is a function of three terms:

- 1) Innovation: $(\mathbf{y} - \mathbf{H} \mathbf{x}^b)$, which is the mismatch of observed and modeled abundance
- 2) Least squares fit $(\mathbf{H} \mathbf{B} \mathbf{H}^T) (\mathbf{H} \mathbf{B} \mathbf{H}^T + \mathbf{R})^{-1}$ which is the ratio of the error variances
- 3) Linear regression: $\mathbf{B} \mathbf{H}^T (\mathbf{H} \mathbf{B} \mathbf{H}^T)^{-1} = \text{cov}(\mathbf{H} \mathbf{e}_x, \mathbf{e}_y) / \text{var}(\mathbf{e}_y)$ where $\mathbf{B} = \text{cov}(\mathbf{e}_y, \mathbf{e}_y^T)$ which is the slope of a line.

In this manuscript, we highlight the need to investigate 3).

Fig. 4.