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## Comment on acp-2022-303

Daven Henze (Referee)

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Referee comment on "Methane emissions from China: a high-resolution inversion of TROPOMI satellite observations" by Zichong Chen et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-303-RC2>, 2022

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The manuscript by Chen et al. presents valuable and timely estimates of sector-specific CH<sub>4</sub> emissions in China inferred from TROPOMI data using an inversion with the GEOS-Chem model. Overall, the topic, scope, and presentation are well suited for ACP, and the results are of value to the community. The paper emphasizes the importance of using log-normal distributions to describe CH<sub>4</sub> emissions, and so I focused on this a bit in my review. In this regard, though, I'm concerned about one issue described in detail below. Given the importance of this part of their approach, as it related to identification of sector-specific emissions, I think it warrants thoughtful revisions. I've provided comments on some other points as well, again focusing mostly on aspects of the inversion method.

Major comments:

I do have a question about the overall framework as presented here, and in prior work from this group applying the same approach. To start with, I would appreciate seeing what equation is being minimized by Equation 5. In other words, can the cost function for the non-Gaussian case be written out, explicitly? There is a bit of literature on this topic, and results one could adopt readily, starting from this point, but without seeing what cost function they are actually minimizing, I'm inferring a bit.

It does though appear that equation (5) is derived from minimization of a cost function based on  $\ln(x)$  but not the log of  $y$ . This is potentially concerning for the following reason. The authors claim that they can substitute  $Sa'$  and  $Ka'$  for  $Sa$  and  $K$ , and then directly use the equations derived in Gaussian space for the posterior error (Eq 3) and averaging kernel (Eq 4). Now, I've seen and used this sort of Gaussian anamorphosis before, but only in the case wherein both the variable and observations are transformed from log-normal to Gaussian space. In this situation, indeed Eq 3 and 4 are applicable, as the solution is the BLUE solution (see equations 5.58 - 5.60 of Cohn 1997). Also see for example the application studies of Brioude 2011, Saide 2012, Cui 2019, or the theoretical works of Fletcher 2010 and Bocquet 2010, none of which, I note, are cited in the present

manuscript.

However, with only  $x$  transformed but not  $y$ , I'm not sure they have arrived at a BLUE solution. What is the basis for the assertion that Eq. (3) gives the posterior error of the solution to (5)? I believe it is up to the authors to show that is indeed the case, or find a paper that shows this, or if not then to demonstrate that the approximation is in some way tolerable. Citing Lu 2022 is not sufficient, as that paper makes the same assumption without discussion. Alternatively, the authors might choose instead an ensemble approach to calculating their posterior error, as we did in Cui 2019 (using  $\sim 1000$  samples).

That being said, I appreciate the authors' ensemble / sensitivity approach to error estimation, and it could be the distinction I'm making has little impact on total error given that their error ranges from the ensemble of sensitivity runs was often larger than their analytic posterior error. But the interpretation of the error correlations and averaging kernel is rather critical to the results in the manuscript regrading sector-specific emissions, and it seems these are based entirely on the analytical calculations. Though that brings me to my last point on this topic..

Lastly, and separately, that the ensemble standard deviation is typically larger than the analytic standard deviation (line 331 / Fig S1) also makes me wonder about the adequacy of the analytic posterior and  $A_k$ , even if calculated correctly. Did the authors consider updating  $\hat{S}$  using information from the ensemble, rather than just the diagonal? If they in fact did do this (i.e., if Fig 6 incorporated the spread from the ensemble), it might be worth stating more clearly, as I didn't catch that.

Minor comments:

97: I know what you mean, but technically it isn't "no additional" cost, though indeed it is quite minimal relative to additional CTM runs. Still, the language could be more precise.

201: The previous global inversion of Qu 2021 used for the boundary conditions was

based off an earlier version of TROPOMI data. Are the biases in that version (1.02) different from the one currently used (2.01)? I know the paper discusses how biases with respect to GOSAT have changed substantially between these two, by about 20 ppb, and that is in part impacted by the regional scope and higher spatial resolution of this analysis. Still, I wonder if the use of boundary values optimized by v1.02 data that is generally biased higher than the v2.02 data contributes to in part to the emissions increases found within this region inversion. Or is this compensated for by the boundary values mostly being scaled down (Table S1)?

215: What is the reason for choosing 600 basis functions? This comes across as a bit arbitrary. Later we learn the inversion DOFs is 167, so 600 does seem like a safe dimension for the inversion. Still, there's no guarantee with this framework that all modes of variability in the emissions at 0.25 degree resolution captured by the observations are spanned by the 600 element GMM basis.

230: The use of the word "optimal" here is ambiguous. More precisely, the equation presented below provides the maximum likelihood estimate of the posterior (which for normally distributed errors is also the mean), and is better described as such.

238: More precisely, additional regularization is used because both the observation and prior error covariance may not be well known. It's not as if gamma only corrects for unknowns in the former. In fact, your results somewhat support this. If gamma were really only compensating for misspecification of  $S_o$ , then we would wonder why your estimate of  $S_o$  is off by nearly two orders of magnitude. In contrast, if we assume some of this is owing to underestimation of  $S_a$ , that helps explain why the increments found for individual posterior  $x$  values relative to  $x_a$  can exceed the prior uncertainty of 50%.

241: Actually, the relationship between  $x$  and concentrations is affine, not linear, since you don't include the initial concentrations in  $x$ . But I'm guessing that the timescale of the inversion is long enough that the role of the initial conditions is minimal.

269: Since your emissions don't actually follow a normal distribution and thus  $(x-x_a)^2$  isn't chi-squared, does this still hold? Shouldn't it be based on  $\ln(x)$  and  $S_a'$  to be chi-squared? I doubt it impacts your results though, and as you've already checked your sensitivity to gamma the overall conclusions likely remain unchanged. Still, in this case one might consider other methods for estimating gamma. By construction of  $S_o$ , I think your observation term is order  $m$  to begin with, so that method wouldn't work. It looks to me like the value identified in Lu 2021 Fig 4b might be at the corner of an L-curve, as the red line started to flatten again where the blue line rises dramatically.

291: It's not clear to me how the authors arrived at  $k = 10$ . The cited work of Lu 2022 does not explain either, and only cites Rodgers 2000. Levenberg-Marquardt schemes usually adjust  $k$  dynamically, letting it be smaller when the minimization is progressing well (i.e., approaching taking Gauss-Newton steps) and making it be larger (i.e.,

approaching steepest-descent steps) if the minimization starts to become unstable. How many iterations were typically required?

281 - 305: I note that discussion of Equation 5 (lines 281 - 289, then again 295-305) conceptually follows Lu 2022 sentence by sentence, which while paraphrased, is not original content. Would it be better to just cite Lu 2022 here rather than repeat? Otherwise we have about a dozen instances in sequence like the following:

Lu 2022: The boundary conditions are still optimized with normal error distributions, assuming an error standard deviation of 10 ppb.

Chen 2022: The normal error assumption is retained for the boundary conditions elements of the state vector, with a prior error standard deviation of 10 ppb.

Perhaps this is a moot point though, if the authors do revise this section given some of the questions raised earlier.

Throughout: could use some more proofreading, for example

14: as a prior

23: the uncertainty range

34: unaccounted for (?)

84: extra space

85: missing spaces

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