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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Retrieving the age of air spectrum from tracers: principle and method" *by* Aurélien Podglajen and Felix Ploeger

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We would like to thank the reviewer for their thorough assessment and detailed comments on our manuscript. Please find below our point-by-point reply.

1. ***Reviewer*** p1 l6/7: I challenge the statement that no assumptions on the shape of the spectrum are made. The retrieval scheme presented uses a flat (all zero) a priori spectrum. I shall claim that the retrieval scheme pushes the solution towards a flat spectrum. No evidence is provided that the result is indeed independent of the chosen a priori spectrum. Setting all elements of the a priori vector zero is not equivalent with not using any a priori information. Thus, this

statement in the abstract is not supported by the paper.

   Authors We agree with the reviewer. The sentence was meant to emphasize the contrast between our method and approaches which fit parameters of a given function (such as the one used by Schoeberl et al., 2005), but it is not exact to state that no a priori information is used. We have replaced this sentence by: "An inversion methodology is introduced, which does not assume a prescribed shape for the spectrum."

   We would like to emphasize that regularization is necessary in our case to rule out unrealistic oscillations. In a certain way, regularization helps to find solutions G that are close to satisfy the constraints of a frequency distribution $G > 0$ and $\int_0^{+\infty} G(\tau;t)\mathrm{d}\tau = 1$

2. ***Reviewer*** p4 l5: Why doesn't lambda depend on r and t? Can this simplification be justified? On the previous page, this dependency is still acknowledged. And for an air parcel - or fluid element - containing a tracer like, say, CFC-12, it makes a major difference, concentration-wise, if its trajectory towards higher latitudes follows the shallow or the deep branch of the Brewer-Dobson circulation. I think this issue needs some discussion, and all related caveats need frankly to be conceded. The applicability of the method proposed needs to be critically discussed in the light of this.

   Authors We agree with the reviewer that this assumption is an important caveat which had not received proper attention in our original manuscript. The assumption is now clearly acknowledged, and the limitations are discussed in Sect. 4.3.

3. ***Reviewer*** p8/9 As stated above, the choice of Ga = 0 does not mean that there are no prior assumption on the shape of the spectrum made. Instead, the prior assumption does affect the solution. As described by Eqs 22 and 23, the retrieval will give the smallest frequencies still compatible with the measurements. The constraint term pushes the solution towards zero. The integral over the age spectrum will not even be unity. Renormalization is not discussed in the paper, but if the integral over all possible ages is not unity, the result cannot be conceived as a frequency or probability distribution. Even after renormalization, the spectrum would be flatter (less structured) than a maximum likelihood solution of the inverse problem, simply because the a priori profile is flat. Thus, it is not fair to state that no a priori assumptions on the shape of the age spectrum are made.

   Authors We thank the reviewer for this comment. We had overlooked the renormalization problem, which is now addressed in Sect.3.2.4. We agree that the choice of G_a influences the retrieval and have rephrased the abstract and the main body of the paper accordingly (see the end of Sect. 3.1.2 : "A second point is that setting $G_a = 0$ implicitly includes a priori information regarding G, albeit limited compared to the parametric approach described above. The effect of setting $G_a = \mathbf{0}$ is to favor smooth functions and implicitly penalize unphysical oscillatory solutions which would deviate significantly from the characteristics expected for a distibution (i.e. $G > 0$ and $\int_0^{+\infty} G(\tau; t)\, \mathrm{d}\tau = 1$).").

4. ***Reviewer*** By the way, I am not particularly happy with the normalization of the averaging kernels in Figure 4 to the maximum, because with this all information on the area under the averaging kernels is thrown away. This would be useful information to judge what the impact of the prior information is.

   Authors In Fig. 4, we show both the normalized (right) and non-normalized (left) averaging kernels. The information of the area under the averaging kernels is depicted on the left panel.

5. ***Reviewer*** p9 l24: It is not true that the accuracy of trace gas mixing ratios from measurements are proportional to their content. The error due to measurement noise (in absolute terms) is at first order independent of the amount. See Rodgers (2000), Eq 3.19, insert G from Eq 2.45, and you will see that the only term which might depend on the amount is K; within linear theory, the sensitivity K in Eq. 2.45 is assumed independent of the amount, thus the related uncertainty of the retrieved amount is independent of the amount. Otherwise the whole concept of detection limits would be un-understandable. If uncertainties were proportional to amounts, even infinitesimal amounts could be detected. Going beyond linear theory, we have to consider the non-linearity of radiative transfer. It is only the parameter errors (Rodgers, 2000, Sect 3.2.2) which tend to be proportional to the content of the target trace gas.

   Authors In general we agree with the reviewer, although this depends on the type of measurements considered. Actually, our choice there is mainly motivated by the fact that, to be useful for a given inversion, the noise in the tracer measurement should be only a fraction of the content in that tracer (i.e., the measured tracer concentration should be significantly differ from 0). We have rephrased this.

6. ***Reviewer*** p10 l7: The L-curve approach is not as objective as it may appear. This is because the tacit assumption is made that the optimal alpha is a scalar. This is an ad hoc decision which is not based on any traceable rationale. It is equivalent to the assumption that our a priori knowledge that the frequency of

a fluid element of age xy is zero is equally justified for all ages. As soon as individual constraint strengths are allowed for each age bin, the L-curve method is not particularly helpful. With this I do not want to challenge the method implemented (which I like very much, aside from my comment on p4 l5) but its description. The method uses much more a priori information on the shape of the age spectrum than it admits.

Authors We agree with the reviewer. The relative arbitrarity of our choice of $\mathbf{S}_\epsilon$ and $\mathbf{S}_a$ is now stated explicitly in the text: "Finally, the structure of $\mathbf{S}_\epsilon$ and $\mathbf{S}_a$ are merely chosen here because of their simplicity in the absence of rationale to do otherwise. One advantage is that then an unique α value needs to be tuned to perform the inversion. More complicated forms of $\mathbf{S}_\epsilon$ and $\mathbf{S}_a$ may be required in practical applications, especially if the error in tracer measurementts exhibit covariance structures."

7. ***Reviewer*** p12 l10: I am confused here. How can one expect that the resolution should be better than the sampling (The text reads as if the authors did)? I assume that the averaging kernels are evaluated on the retrieval grid, and then it is analytically impossible that the resolution can be better than the bin width used for the retrieval. Even in a maximum likelihood setting, where the averaging kernel matrix is unity, the resolution corresponds to the bin width.

   Authors The formulation was indeed confusing. We have rephrased the sentence for: "the resolution is better for short transit times, although even for those the effective resolution does not reach the 1-month-transit-time bin size chosen for the retrieval, as can be seen from the overlap of the averaging kernels"

8. ***Reviewer*** p1 l2/3: I would prefer commas over parentheses here (but this might be a question of personal preference).

   Authors We discussed this and prefer to keep the parentheses.

9. ***Reviewer*** p1 l5: "tracer" is a qualitative term and thus cannot depend linearly on anything. I suggest "the concentration of tracers", or, more specific, "the mixing ratios of tracers".

   Authors changed

10. ***Reviewer*** p1 l8: the term 'model output' is a bit too vague. Perhaps better 'output of a circulation model' or 'output of a chemistry-transport model' or whatever is adequate here.

    Authors Changed for 'output of a chemistry-transport model'

11. ***Reviewer*** p1 l22: I think 'frequency distribution' would be more adequate than 'probability distribution'. If a concept of probability is used in this context, it must be objective rather than subjective probability (because we want to describe the air parcel and not our knowledge about the air parcel). However, post factum objective probability makes nos sense, because the characteristics of the air parcel are already determined. Conversely, to describe the air parcel by the frequency of fluid elements of a certain age does make sense. The same applies to p3 l21,

Authors Changed

12. ***Reviewer*** p2 l24/25: I suggest a footnote after conceptually, saying "we write "conceptually", be- cause it is clear that physically an air parcel obviously cannot be decomposed into an "infinity of infinitesimal...". This physical restriction, however, is not a conceptual re- striction because at scales considered here this issue has no bearing" or something similar. By the way, since you later provide age spectra at finite resolution only, the concept of infinitesimal fluid elements (and a sum running to infinity in Eq 1) are actually not needed. It is sufficient to postulate that the fluid elements are small enough to be considered fairly homogeneous.

    Authors We have added the footnote.

13. ***Reviewer*** p3 l13 loss of radioactive tracers or photochemical loss are exponential, not linear. I concede that the loss RATE is linear in concentration (and thus the statement in the paper is formally correct) but it is very easy to misunderstand this sentence. Rewording would be appreciated.

    Authors Rephrased for "Another example is that of tracers whose loss/growth rate is a linear function of their concentration"

14. ***Reviewer*** p4 l17/18: I think that your construal of the age spectrum still contains the weight of the boundary condition history. If I understand your construal correctly, in your case this boundary condition history is modulated by the loss term. If you inserted the word "only" before the closing parenthesis, I think the

statement would be clearer.

    Authors That is not what we meant. Usually the age spectrum is seen as the weighting function of the boundary condition history to get the tracer content, while here we consider the boundary condition history as the weighting function of the age spectrum. We have rephrased that sentence: "note that this perspective is reversed with respect to the general view that the age spectrum is a weighting function of the tracer boundary condition history modulated by the loss terms"

15. ***Reviewer*** p5 l5-7: Observational evidence of non-stationarity of stratospheric transport is also available, see, e.g., Stiller et al. (2012, Fig 9), Haenel et al. (2015, Figs 8/9).

    Authors Thanks for those references, which we have added.

16. ***Reviewer*** p6 l26: I suggest to replace "expectation" with "assumption", because "expectation" is ambiguous. It is occasionally used as a short cut wording for "expectation value". Since the manuscript deals quite extensively with distribution functions, I suggest to avoid the use of terms which can be misunderstood as statistical technical terms (although the correct connotation should be clear from the context).

    Authors Changed

17. ***Reviewer*** p7 l17 and elsewhere: It is a bit uncommon to use bold capital letters for vectors (I understand G is a vector, not a matrix). If I remember the

ACP author guidelines correctly, bold face capital letters are understood to be matrices. Please check the ACP author guidelines, and change to lower case bold face g if adequate.

Authors We have changed it following to bold italic ACP author guidelines: "Matrices are printed in boldface, and vectors in boldface italics.". We chose to keep the capital for consistency with the literature.

18. ***Reviewer*** p7 l18: Since there is nothing unclear in the notation, I suggest "In order to simplify the notation".

    Authors Changed

19. ***Reviewer*** p8 l28: What is presented here is not "THE" Tikhonov approach. The Tikhonov ap- proach includes a large class of families of constraints, often involving squared nth order finite difference operators as regularization matrix. In remote sensing, squared 1st order difference operators are particularly common. The use of a diagonal matrix does belong to the class of Tikhonov schemes but it is formally equivalent to what you present in Eq 20 and ignore covariance information. Thus I consider the wording as a bit misleading.

    Authors We have reworded those sentences: "We will follow an empirical approach here for the regularization, which belongs to the class of Tikhonov regularization schemes."

20. ***Reviewer*** p19 l3: Not sure if it is so clear that the uncertainties in radiative transfer are larger than those of the forward model used here. Doesn't the for-

ward model used here include (at least implicitly) all the uncertainties of the sink terms, i.e. all the uncertainties related to photo-chemistry (incl. self absorption in layers above; uncertainties in T-dependencies of absorption cross-sections etc) and, depending on the trace gases considered, the OH concentrations along the trajectories etc? Also it is not clear why the Tikhonov approach is more adequate for simple problems than other approaches.

    Authors We agree that the uncertainties related to the chemistry are large and poorly constrained. However, they are absent for inert tracers. We have added a footnote: "This is at least the case for inert tracers; for chemically active tracers the sources of uncertainties are many and more difficult to quantify."

21. ***Reviewer*** p19 l24: It comes a bit as a surprise that just those measurements which have actually provided information on non-stationarity of age-of-air dis- tributions, and which provide dense global tracer distributions, have not been mentioned here (see references men- tioned above, or Kellmann et al. 2012)

    Authors Thank you for those references, which are now included.

References

Schoeberl, M. R., Douglass, A. R., Polansky, B., Boone, C., Walker, K. A., and Bernath, P.: Estimation of stratospheric age spectrum from chemical tracers, Journal of Geophysical Research: Atmospheres, 110, doi:10.1029/2005JD006125, https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2005JD006125, 2005.

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