Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-990-RC1, 2018

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

Anonymous Referee #1

Received and published: 25 October 2018

The manuscript by Aurelien Podglajen and Felix Ploeger presents a novel method to obtain information about the age of air spectrum of air parcels from the concentrations of multiple tracers. Without any doubt this topic lies within the scope of ACP. The highly original method proposed seems adequate and basically sound to me (perhaps excempt from p4 l5). The presentation of the information is adequate for the most part of the paper. I have, however, spotted some issues that need clarification.

Major issues:

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

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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.

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.

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. Re-normalization 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 re-normalization, 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. 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.

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.

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.

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.

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

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'.

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.

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,

p2 l24/25: I suggest a footnote after conceptually, saying "we write 'conceptually', because 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 restriction 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.

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.

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.

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).

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).

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.

p7 l18: Since there is nothing unclear in the notation, I suggest 'In order to simplify the notation'.

p8 l28: What is presented here is not 'THE' Tikhonov approach. The Tikhonov approach 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.

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 forward 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.

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 distributions, and which provide dense global tracer distributions, have not been mentioned here (see references mentioned above, or Kellmann et al. 2012)

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