

Interactive comment on “A new time-independent formulation of fractional release” by Jennifer Ostermüller et al.

Anonymous Referee #2

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This is an interesting paper that promises an improved method to compute fraction release factors (FRF). It includes results that suggest great promise but as presently written it does not make a convincing case. The paper is unclear and confusing in its derivations, which are at the heart of the paper. In addition, it makes exaggerated claims as to the expected properties of FRFs (e. g., their time-independence). Thus, I cannot recommend publication in ACP without major revisions.

Specific Comments

P3 L1-2: It is expected (even if undesired) that ODPs can change with time so not all time variation in the FRFs is incorrect. So a bit more discussion of this point is needed.

P4 L3: Do you mean Newman et al. (2007) here?

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P4 eq (5): Should be $G(r, t')$ here.

P4 L11-23: I find this "derivation" sloppy, misleading, and possibly incorrect. The $f(t)$ defined in eq (1) is a function of time (i.e., model time). The $f(t')$ in eq (6) is a function of transit time. These are completely different and cannot be replaced by each other. You should write the f in eq (6) as $f(r, t, t')$ and it would be better to use a new function name as well. f^* may fit in well with your later discussion.

However, I strongly feel that L11-19 should just be removed from this section and fleshed out in more detail in section 5. Eq (8) is trivial to derive from eq (1) and (5) and without more discussion your alternative derivation is just confuses the matter. For example, why would $f(r, t, t')$ be independent of transit time t' ? Clearly it should be strongly dependent on transit time. But you confuse the matter by saying just "time" on L18 and thus make it seem you are talking about model time, which are completely unrelated.

P4 eq (8): Should be $f(r, t)$ on the left side of equation.

P5 L2-3: You changed the meaning of the sentence in the Laube et al. paper. The full sentence is "Experimentally derived correlations of FRFs with mean age of air should thus only be considered as time-independent as long as there are no major changes in stratospheric transport or relative tropospheric growth rates." Seems reasonable enough. Not being time-independent is expected (e.g., from circulation changes) and does not render the concept invalid. This is not to say that all sources of time-dependence are ok, but just that a more nuanced discussion is needed.

P5 L6-7: There is no such inherent assumption. One can derive eq (8) from eq (1) and (5).

P5 L9-10: Again, this sentence is too absolute. Eq (8) is not expected to produce time-independent values (as stated many times already). The approach of eq (8) can only hope to remove time-dependence from source gas trends (which of course it may or

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may not).

P6 L9: You do not state how you compute the age of air. Please discuss.

P6 L32 Mention you are starting to discuss figure 2. Took me a bit to figure out the change and figure 2 is never mentioned in the text.

P7 L6-17: This discussion needs to be moved up to the beginning of the section or perhaps to Section 3 since it affects all of the calculations shown in this section.

Section 4: I'd be interested in seeing the difference between eq (1) and (8), where in eq (1) a reasonable lag is used to account for the transit time. I get the feeling looking at your results that they may not be much worse using (1). I think this comparison should be added to the paper (it could be in a supplement).

Section 5: This derivation seems awfully convoluted. Certainly it could be cleaned up. And it will help if you move and expand the discussion around eq (6) and (7) to here, as discussed above. And please explicitly state that f is a function of both t and t' , $f(r, t, t')$, so it is more clear what is going on.

P10 L1: \bar{f} does not depend on t' but it is not a constant.

P10 L21-23: Now I'm confused. If you use the same function for G_N^* as G , then eq (8) and eq (19) are identical. Thus, you should reproduce the results in figure 3 in figure 4. What have you gained in the derivation?

P11 L4-5: How are you using Γ^* ? This is not explained at all. Perhaps this is at the heart of my confusion on the last point. I'm suspecting that your definition of age differs in figure 3 and 4. More description is greatly needed.

P11 L27-29: Seems like these details should have been mentioned in Section 3.

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