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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A Bayesian inversion estimate of N₂O emissions for western and central Europe and the assessment of aggregation errors" *by* R. L. Thompson et al.

Anonymous Referee #1

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1 General comments

This paper addresses a challenging problem of improving spatial and refining temporal resolution of the fluxes of N_2O emissions over Europe with the help of merely one observational site. In a series of synthetic experiments the authors give evidence for the necessity of a correct specification of the correlation parameters and illustrate how the aggregation errors could be handled in the inversion procedure. Both these aspects are crucial for ensuring a correct constraint of the inverse problem of this paper. Finally, the findings on correlations between N_2O emissions and the meteorological patterns are noteworthy results.

This reviewer has examined this well structured and written paper with interest. I recommend the article for publication in ACP after the authors have addressed the points raised in the review, with an emphasis on eliminating the sources of possible confusion in the presentation of the inversion scheme.

A broader context and importance of the problem are well presented in the Introduction to the paper. I find the methodology description somewhat not as clear as it could be. We have here an inverse problem defined by a matrix F which in itself can be produced using a forward or backward (as it is reasonably the case in the paper) mode of an atmospheric transport model. Therefore, I consider it to be important to keep the explanations precise and concise. What to me seems particularly confusing is to mention STILT in the context of the forward problem formulation in a way leaving room for doubt if STILT was executed in a forward or a backward mode (for example, p.26085, l.23-24). I know there is 'Time-Inverted' in the model acronym but I still find it disturbing. Also swapping between 'footprint' and 'surface influence term' is disturbing. One needs to wait until p.26092 to make sure that they both stand for (almost) the same notion, footprint being the influence term averaged over a period of time.

I appreciate a series of synthetic experiments preceding the inversion of the real observations. Could the authors comment on the (im)possibility of the transfer of the studies on finding optimal correlation lengths from synthetic to a real data experiment? The synthetic experiments clearly illustrate the importance of having a good specification of those correlation lengths.

For a series of inversion experiments I think that a a reader would be helped if some details of each inversion set-up could be added to Table 2. I wished a reader could quickly realise what is the number of the estimated state variables (in space and time) versus the number of the observations used in the inversion.

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Interactive Comment

A list of comments presented below is meant to point to the authors the statements which this reviewer finds confusing and to help the authors in the revision process. Some corrections of the English language are proposed in the Technical corrections. Please do check them with a native speaker of English.

2 Specific comments

1. Introduction, p.26076, l.11, 'In an ideal inversion ...'
   I would not call the circumstances described in this sentence an 'ideal inversion '. The fact of having multiple observations of each state variable at each resolved time-step replaces an under-determined problem with an over-determined one, which is not necessarily ideal. Moreover, if each state variable is observed at each resolved time-step, then one would need to ask oneself a question about what inversion has to bring to such a complete picture. To make a long story short I would suggest starting this paragraph with 'Ideally, there would be multiple observations ...'.

2. Atmospheric transport model, p.26078, l.20-21
   At the end of the sentence 'The density of particles ...' the authors could add a short definition of a footprint, please.

3. Atmospheric transport model, p.26079, l.3-4
   The sentence 'The STILT model itself has a dynamic resolution ...' is somewhat perturbing as a Lagrangian model does not really have a resolution. I suppose the authors may mean the resolution of its output and it this is the case I would suggest then something of the sort: 'The output of the STILT model can be obtained with a dynamic resolution ...'.

4. Simulation of N_2O, p.26079, l.11-17

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I am a bit confused by the first three sentences of this section and by the words 'advect a tracer' in particular. Could it clearly be stated that advection is backwards in time. Or the first sentence removed?

5. Simulation of N_2O, p.26080, l.3
   I somehow have a problem with a good understanding of what $c(x_i, y_j, z_k, t_0)$ is. I do suspect it is a mixing ratio at the domain boundary which impacts the observation(s) of N_2O. But what does t_0 really mean in this context? Could the authors add/reformulate their explanation. The caption of Fig.6 is much clearer than the formulation in the body of the article. Is the word 'influence' fortunate in this context? It would also help if 'trajectory' could be replaced with 'back-' or 'backward trajectory', should this indeed be the case.

6. Bayesian inversion set-up, p.26081, l.13-14
   Maybe I am missing something but, to me, 'the surface influence term' $F(x_r, y_r, z_r, t_r|x_i, y_j, t_m)$ of section 2.4 is the footprint of 2.3. I would not rename it 'surface influence term' to avoid confusion. Or, introduce the notion of 'surface influence term' in parallel to footprint on p.26078, l.20-21. Please, clarify.

7. Bayesian inversion set-up, p.26081, l.18
   I believe that 'n' is the number of grid-boxes in the spatial domain times the number of time intervals for the retrieved sources. Could this be specified, please?

8. Bayesian inversion set-up, p. 26081, l. 21
   I may be missing something here but it is not clear to me why the mixing ratio at the lateral boundary at the time of an observation would influence the observation itself. This remark goes together with one of the previous ones.

9. Bayesian inversion set-up , p.26083, l.2
   Could the authors specify in which sense this is a 'best' estimate for a reader unfamiliar with the linear estimation theory.

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10. Synthetic data generation, p.26085, l.23
I may be biased but when I read 'We advected the synthetic fluxes with STILT' I tend to see forward trajectories in my head. I am suddenly unsure if F was computed in a receptor oriented approach. I mean, I am sure because it is stated in 2.2 but I find that the formulation used may be confusing.

11. Reference inversion, p.26087, l.13
Could the authors specify the number of the degrees of freedom estimated in the inversion and the number of the observations used to constrain them?

12. Temporal resolution ..., p.26089, l.22
What is the value of T for the correlation between the time intervals, please?

13. Temporal resolution ..., p.26090, l.18
Do I understand correctly that the chi-squared value here is closer to 1 than in the experiment with fine resolution of both synthetic and retrieved fluxes? Do the authors consider this difference as significant?

14. N_2O flux estimates, p.26091, l.24-26
I understand that for real flux estimates the authors do not have true fluxes. Could, nevertheless, any of the criteria used for the synthetic measurements be translated to the studies of real observations (e.g. error reduction) in an attempt of verifying the values of the spatial and temporal correlations.

15. N_2O flux estimates, p.26092, l.1
I am a bit lost here. What is the temporal resolution of inversion for real observations?

16. N_2O flux estimates, p.26092, l.3-4
Could the authors comment shortly on the error estimation in order to satisfy reader's curiosity? For atmospheric transport, in particular.

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17. N_2O flux estimates, p.26092, l.8-9
Could the authors clarify what is the role of the flask sampling site? Does it provide some measurements against which inversion has been tested. If so, could it be stated clearly.

18. p. 26109, caption Fig.2
It is suddenly not clear to me from this caption if STILT was used in a forward or in a backward mode. It would already be much better if the authors removed the word 'forward' from this caption.

19. p.26110, caption Fig.3
Please add to the caption the shape of the points alongside the colour. It is also not clear to me what these points represent? Do they stand for the results of the performed experiments and the solid lines are splines connecting those points? Could the authors also state in the caption for what values of the correlation lengths these symbol stand?

3 Technical corrections

1. The coordinates of the Ochsenkopf station are specified four times in the paper: Abstract (expressed in minutes), Introduction (in hundredths of a degree), Section 2.2 (in minutes) and Summary (in minutes). Could you please make them consistent?

2. Introduction, p. 26074, l.25, last sentence 'Second, it plays ...'
To me there is an 'it' missing in this sentence. Moreover, I would also suggest splitting it into two sentences: 'Second ... (). In the 21st century it ...'

3. Introduction, p.26075, l.16
Please, spell radon with a lower case 'r'.

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4. Introduction, p.26076, l.27, 'For example ...'
   Please consider replacing 'do not need' with 'does not need'. I would remove 'as long as it (is) taken into account'. To me it is self-explanatory from the previous sentence.

5. Atmospheric transport model, p.26079, l.7-9
   I would suggest to go with this problem to somebody speaking English as their mother tongue but I would rewrite the last sentence: 'The model domain has been chosen to be Europe extending ...' or, better: 'Europe, extending from ... in the centre, has been chosen for a model domain.'

6. Bayesian inversion set-up, p.26082, l.2
   It is actually only F which is a forward operator, or model if you prefer. f should rather be seen as model parameters.

7. Bayesian inversion set-up, p.26082, l.5-6
   This sentence is just difficult to grasp. I would propose rewriting it. The least the authors could do is to replace 'each' with 'any'.

8. Bayesian inversion set-up, p.26082, l.27-28
   I would propose to add a word to this sentence '... this is achieved by specifying a probability ...'

9. Bayesian inversion set-up, p.26084, l.3
   I am convinced it should be 'with regard to', without 's'.

10. Bayesian inversion set-up, p.26084, l.4-7
    Please check it but to me it looks like you are having 'the pattern' in singular and 'do not' and 'those' in plural in this sentence.

11. Synthetic data generation, p.26086, l.23

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I would insert here, say after 'prior fluxes' a reference to Eg. 6 to make understanding easier.

12. Synthetic data generation, p.26086, l.2
It should be 'have' instead of 'has'.

13. Reference inversion, p.26087, l.4
Be careful here as you have not introduced this cost function in your paper.

14. Sensitivity to temporal ..., p.26087, l.20
I would suggest replacing 'a similar set-up as was used' with 'a similar set-up to the one used'.

15. Sensitivity to temporal ..., p.26088, l.5
I believe the authors want to say 'have' and not 'has'

16. Sensitivity to temporal ..., p.26089, l.27
Please consider replacing 'accounted' with 'accounted for'.

17. Temporal resolution ..., p.26090, l.1-5
Please consider splitting this sentence into two ones with a period after 'this error'. You could start the next sentence with 'The algorithm is ...'

18. Temporal resolution ..., p.26090, l.14
What is this temporal resolution? I can read from Fig.4 that it is most likely 7,10,14,30 days but I do not find it in the text.

19. Averaging ..., p.26091, l.2
I believe the authors mean 'effect' and not 'affect'.

20. Unless ACP does not allow using diacritics, please consider spelling: 'Frédéric' and 'Białystok'.

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