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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Emergence of a linear tracer source from air concentration measurements" *by* J.-P. Issartel

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The paper describes mathematical developments with respect to the reconstruction of sources of atmospheric trace gases from concentration measurements. In the framework of inverse estimation, the presented method involves a suitable renormalization of space-time. The aim of this renormalization is to counteract the fact that the measurements at discrete locations and time instants offer a very inhomogenous information content, which is in contrast to the desire to infer the complete space-time field of the sources. The method is presented in a general theoretical form, and then applied to several illustrative examples.

The work is an original and interesting contribution. It addresses a topic essential for atmospheric inverse estimation as used in air pollution or biogeochemical research (what

information is 'seen' by atmospheric measurements, and how can we best retrieve it?). It should be published in Atmospheric Chemistry and Physics, after revisions as detailed below.

All of the *General comments*, *Specific comments*, and *Technical corrections* have been deliberately written *before* I had any look at the referee report by S. Houweling at the reply by J.-P. Issartel, in order to be able to present an independent view. It turns out, however, that a number of my comments confirm what has been said before by the other referee, and are partially addressed already by the author in the reply. I therefore added another section, *Interactive discussion*, to refer to this.

General comments

I explicitly welcome the submission of more theoretically inclined manuscripts to atmospheric journals, as the link to methods from different fields of science has the potential to give new insights into particular problems. Having said this, however, leads to my major point of criticism of the present paper. As the CO_2, CO, or CH_4 inversion problem is explicity mentioned as the motivation of the work, I feel that the method should be presented in a way more accessible to that community. This refers, on the one hand, to the terminology, as detailed in Specific comments below. On the other hand, it seems to me that the compact mathematical notation, and the invocation of the quantum mechanics formalism, could pose a high, and possibly unnecessary, obstacle to many readers of Atmospheric Chemistry and Physics. In the present form of the manuscript, it is quite hard to recognise the distinction between the core ideas necessary to solve the practical problem, and the interpretation meant to widen the view and to give the algorithm a solid foundation. For example, two full chapters (7 and 8) are spent to introduce yet another notation and to interpret the estimation problem as parallel to quantum entities, in order to then deal with measurement errors in terms of mixed states (chapter 9). However, could chapter 9 not just be replaced by simple error propagation, to arrive at the same result (as lines 10/11 on page 2642 seem to confirm)? If this was indeed true, then I would suggest to maybe move chapters 7 and

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8 into an appendix (it might also be worthwhile considering to present these interesting considerations as a separate paper in some other journal, the readers of which could appreciate it more fully). The same comment applies, maybe to lesser extend, to the presentation of the basic algorithm in the first chapters. For example, it is easy to get lost among the various different scalar products; I think it would help very much to at least back-translate them into integrals in key equations. A list of symbols, or maybe also a repetitive summary of the algorithm, could also be helpful.

Specific comments

Title:
In my opinion, the title is not very specific about the paper, and potentially even decieving.

It is said that the illumination diverges close to the measurements. In practice, samples are taken from air parcels which can be considered well-mixed within some space-time surrounding (i.e., they are non-infinitesimal space-time elements, or, in the model world, even grid cells). How relevant is the divergence then? That is, does some degree of spatial or temporal smoothing already remove the singularities? (Same question also for the 'excess entropy' discussed in Chapter 6.)

Equation (1), 'zero boundary conditions':
The mixing ratio is not generally zero at the boundaries. Rather, there are periodic boundary conditions, or zero *flux* boundary conditions, respectively, depending on the dimension.

As anounced in the General comments above, I'd suggest to provide explicit links between the terminology used in the paper, and that common among the biogeochemical community. For example, the identification of 'adjoint concentrations' with 'retroplumes' should be made explicit; you might also say that they represent lines from the 'transport matrix (operator)' usually introduced in the biogeochemical literature. Further, the 'background error matrix **B**' seems to correspond to what is called 'a-priori flux covari-

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ance matrix' elsewhere.

Equation (8):
Is there a short argument to see this?

page 2620, line 14, 'Dirac detectors':
Though it can be guessed what this means, I'd suggest to say it explicitly.

page 2621, line 5, 'contribution from this point':
Can you define explicitly what this means?

page 2622, line 19/20:
It is not clear to me what this actually means. Do you refer to different natures of measurement and model errors?

text after Equation (13):
If my assumption is correct that the 'background error matrix **B**' is what is called 'a-priori flux covariance matrix' elsewhere: It is suggested here that this matrix is just given, or following naturally from the problem. However, in the practice of e.g. biogeochemical tracers, there is major debate how this matrix looks like, and different choices lead to substantial differences in the results.

beginning of chapter 4:
It is stated that σ (itself) can be decomposed, but the equations refer to expectation values. Can you clarify this?

Chapter 6:
If I understood right, the matrix **H** is the covariance of the measurements arising from the fact that they depend on the same (stochastic) source field, which is distinct from the covariance matrix of the measurement *errors*. To avoid confusion here, it may be good to say this once more.

page 2642, line 11:
Uncorrelated measurement errors are a common assumption, but definitely not true

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Interactive Comment

(e.g., due to common standard gases and other systematic influences).

page 2645, line 22, ‘the known domain is finite’:
This was surprising to me, and I would welcome more discussion there. A point of confusion for me is that atmospheric diffusion leads, theoretically, to immediate spread of information throughout space. Thus a tiny (but non-zero) sensitivity of any measurement would exist with respect to any point in space throughout all the past.

page 2648, lines 5ff:
It did not become clear to me whether or not the transport model POLAIR is a state-of-the-art “realistic” transport model including the usual parametrizations of transport phenomena, or a simplified scheme.

page 2650, lines 17ff:
It is implied that the calculation of the renormalization function takes orders of magnitude more time than a single estimation run. This may pose a serious obstacle in using the algorithm for practical problems.

page 2650, last paragraph:
The example case described here (source written as a decomposition into Gaussian elements centered around the measurements) is labelled a ‘more traditional inversion’. However, I do not see why such a decomposition would be an obvious choice for the considered case. If I knew a-priori that the source is shaped like a Gaussian in space and time, and wanted to infer its location and characteristic width, I would probably rather choose some homogeneous source decomposition (e.g. a regular array of Gaussians, not at the measurement locations). Can you comment on this?

page 2651, line 12/13, ‘Contrary to our expectation’:
This was indeed also contrary to my expectation. Could you comment a bit more on this? For example, could it have to do with the empirical choice of $f(x)$ involving $E_{max}/1000$? Or could it be related to the gridded numerical representation?

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page 2651, line 14/15, 'The quality of the renormalized estimates is clearly improved.': This is only true for the estimates with positivity constraint, while without, the result considerably deteriorates. On the other hand, comparing for example panels a3 and b3 of Fig. 2, to a lesser extent also c3 and d3, the spatial structure is actually quite similar (suggesting that the main improvement by the renormalization is in the total amount). In all panels, the source peak estimated near Monterfil is still 1 to 2 orders of magnitude larger than the spurious sources at the detectors (as far as it can be read off from the colors). Can you comment on this?

page 2653, lines 22ff:
In the case of the 'Gaussian' source, how do the retrievals compare between using/not using the renormalization?

Technical corrections

Equations (6), (7), and (11):
The first factors in the dot products should wear the transposition sign as used later.

page 2622, line 7, 'altitude':
Replace by 'surface altitude'.

page 2629, line 12:
(x,t) should be (x).

page 2655, line 20:
The method used in Rödenbeck et al. (2003) is not actually a (sequential) Kalman filter, but just a one-step Bayesian estimation.

Fig. 1:
It seems that the detectors were operated according to the expected movement of the tracer cloud. Is there a way to make this obvious in the Figure (e.g., by choosing the colours in a West-East progression)? This would help understanding Figure 3 later. Further, what do the symbols at the bottom line of the right panels mean?

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Interactive Comment

About the 'background error matrix **B**':
It seems that the other referee also identified **B** with an a-priori covariance, but according to item 7) of the Author's reply this is not what is meant (this makes one of my comments above obsolete). Rather, if I now understand right, **B** is *purely* a property of transport and of the set of locations/times of the observations (in terms of notations used in biogeochemical inversion literature, **B** is a unique function of the transport matrix, which in turn is nothing else than the set of the **r**'s, at least before renormalization). Yet, in the cost function, **B** stands at the place where the 'traditional' Bayesian inversion puts the a-priori covariance matrix which represents additional outside information. I am interested to understand this better, and feel that this warrants more discussion, to which I'd like to invite even after the closing of the public discussion phase.

Concerning item 5) of the Author's reply:
From the paper, I had meant to understand basically how the algorithm works, but this item again causes confusion. What exactly is the source of randomness in the anticipation? If it is the randomness in the fluxes as described by **B**, isn't the consideration circular?

Concerning item 9) of the Author's reply:
Like the other referee, I have expressed above the feeling that the comparison to quantum mechanics was an interesting line of thought, but more in the sense of broadening the view, and that it should therefore be much less prominent in the manuscript. In contrast, I realise that it is meant by the author as an essential part of the work. I agree with the author that it is valuable to raise new concepts, and see scope to do so in Atmospheric Chemistry and Physics. My hesitation against the corresponding chapters in the manuscript comes from the impression that, up to now, it is mainly the compact notation and terminology that has been used, but that it did not yet provide substantial new insights. If the author agrees to this, and if it was the intention to present the link to quantum formalism as a *potentially valuable* tool (as item 9) indicates), then I suggest

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to at least state this intention more clearly, in order to reduce the risk to expel potential readers.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 2615, 2004.