Atmos. Chem. Phys. Discuss., 4, S3367–S3371, 2004
www.atmos-chem-phys.org/acpd/4/S3367/
European Geosciences Union

Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on “Inversion of CO and NO$_x$ emissions using the adjoint of the IMAGES model” *by* J.-F. Müller and T. Stavrakou

S. Houweling (Referee)

s.houweling@phys.uu.nl

Received and published: 19 January 2005

This paper presents a novel method for top-down quantification of CO and NOx emissions. These emissions constitute an important class of processes that largely influence tropospheric chemistry. Yet the uncertainties are rather large and therefore methods, like the one presented in this manuscript, that provide additional constraints are very welcome and important. The approach is new in that it accounts for the chemical coupling of CO and NOx, which is an important methodological step forward. Besides the usefulness of the presented emission estimates, the synthesis approach - if followed by others - might make the scientific field more quantitative in the sense that it requires an uncertainty estimate for every piece of information that is taken into account. In the end this will likely help to identify the most urgent gaps in our understanding.

The approach that is used is thorough and sound, and - as a modeller - I admire the programming effort that went into this work. The manuscript is well organized and well written. I believe that some methodological information that could in principle be found in the literature is not redundant but makes it better accessible to interested readers who are not so specialized in this field. There is one point of criticism (major comment 1), which might hamper publication if not addressed appropriately. The other points should help to further improve the manuscript or trigger discussion.

MAJOR COMMENTS

1. Base year 1997

Chapter 8 emphasizes the general agreement between the results of this study and previous studies. This would be fine and confirm the validity of the approach were it not that 1997 was a very exceptional year. Because of this the agreement with studies that aim at different years should actually be considered highly surprising. The NOAA/ CMDL data archive for CO shows that the global CO growthrate exceeded 10% in 1997, while on a decadal time scale there doesn't seem to be a significant trend. Previous studies have identified enhanced biomass burning in response to El-Nino drought as the most plausible explanation. This study seems to almost fully ignore this significant anomaly. A priori biomass burning emissions are scaled to the climatological estimates by Hao et al 1994, while - most surprisingly - the inversion even reduces the biomass burning parameters. This is a significant and unexpected result that should be discussed. It is also surprising that a large effort goes into comparing the inversion results with previous studies, except those studies that specifically address 1997 (see e.g. van der Werf et al, Science, 2004).

MAJOR SUGGESTION

As mentioned in the abstract this is the first global inversion solving for parameters that are coupled through chemistry. It remains unclear, however, if the results yield new insights confirming that coupled chemistry inversions are worth the effort and can be considered a promising direction. Right now the main message seems to be that there is a broad agreement with other methods, suggesting that there isn't much gain. As often, disagreements are more interesting than agreements. This is certainly true if it can be shown that disagreements point to improvements. Maybe potentially interested scientists like me should be more patient, but this paper offers a nice opportunity to discuss the potentials and remaining stumbling blocks of the approach.

MINOR POINTS

p7987 'It is implicitly ... the emission inventories' What is the evidence that this assumption is satisfied? In inverse modelling of quasi passive tracers like CH4 and CO2 there is a serious concern about transport model errors. Coupled chemistry inversions add another dimension to this, which are the uncertainties in the chemistry parameterizations. One way to deal with error sources like these, which are difficult to formally include in the inversion, is to perform sensitivity calculations. The ones that are performed in this study are only a small subset of those that one would ideally test. At this stage, however, it wouldn't be fair to require a comprehensive senstivity assessment. It the same time, however, it should be acknowledged that relationships between CO, NOx, OH are uncertain and that emission updates may compensate for errors in the chemistry. In this regard, comparing the results to independent measurements of CO and NOx is useful, but more interesting would be a comparison of prior and posterior model skills in reproducing for example O3.

P7998 'Its value is initially taken equal to 5' An important difference between inverse modelling and model tuning is that objective and tracable criteria are used. In this inversion the relative weight of the priors, the weight of the CH4 lifetime and the weight of representation errors are difficult to trace. It doesn't seem like the results are tuned by the settings of these parameters. Still an explanation is required of the method that led to the specific numbers that were used. Most likely these parameters are needed because of some statistical inconsistency. A good diagnostic for this is the chi-square test. My guess is that this test would indicate that model errors have been underestimated. This would justify increasing representation errors (i.e. relaxing the data constraint) rather than tightening the prior constraint. In this way the 'tuning parameters' would become tracable and a more objective methods can be chosen to - attempt to - quantify them. Another hidden example of statistical conflicts is the omission of 'redundant' stations. In a sparse measurement network it seems to me that none of the stations of high quality data can be considered redundant. Even if stations were located so closely together that to the coarse resolution model they are actually in the same location you'd end up with approximately twice the number of measurements, which would justify a reduction in data uncertainty. The real argument for omitting the station, however, may be to make the network more homogeneous, thereby reducing biases caused by the use of large regions. This should be made clearer.

p8011 'The reasons for this poor representation of the seasonal cycle of NO2 columns at mid-latitudes are unclear' It is questionable whether the inversion should be expected to reproduce seasonality since it has hardly any degrees of freedom to adjust it. Errors in the assumed seasonalities of fossil CO and NOx emissions might partly explain these discrepancies. More generally, it remains unclear how sensitive the results are to the temporal and spatial discretization of the parameters. I take the argument that computational limitations prevent a larger number of parameters, but it should have been made clearer 1) what motivates the current descretization 2) how sensitive the outcome is to the particular choice of regions.

P8001 'Using 8 processors ... for the adjoint one' It would be interesting to know how many iterations are needed to satisfy the convergence criterion. A related question is whether the same solution is obtained when starting from different parameter values. Since the problem is non-linear there is the risk of ending up in a local minimum. In this particular inversion non-linearity may be mild enough to avoid this. Nevertheless, I assume this has been tested and in that case it is worth mentioning the outcome.

A prior CH4 lifetime I just wanted to make sure that this number is mentioned somewhere (it may be there, but I was unable to find it)

8016 'In all cases, the a posteriori errors are smaller than the a priori error estimates, except for the African anthropogenic CO sources' This is a bit surprizing because the equation for posterior uncertainty does not allow an increase of uncertainty. I assume this has to do with the approximation of the Hessian and is therefore an artifact of the method. If this is true it should be mentioned. Looking for an explanation of why the GOME NOx measurements couldn't provide significant constraints in tropical biomass burning, it struck me that there are large data gaps in Figure 6 and 7 over the tropics that I didn't quite understand (e.g. March over Amazonia and Australia; September over the Sahara and South-East Asia).

Fig 15 & 16 Although it is true that BGFS is less accurate than DFP it has the advantage that the approximations are always on the conservative side. DFP might justify invalid claims.

TECHNICAL CORRECTIONS

P7997 What is the difference between equation 4 and 5?

A reference in the text is missing for Figure 11 and Table 2 and 3.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 7985, 2004.

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

EGU