Atmos. Chem. Phys. Discuss., 4, S3315–S3322, 2004
www.atmos-chem-phys.org/acpd/4/S3315/
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

Anonymous Referee #1

Received and published: 14 January 2005

The paper presents a coupled inversion of CO and NOx emissions using an adjoint model and iterative cost function minimization technique.

The application of this technique appears very promising for problems with very large numbers both of parameters to be optimized and observations. Furthermore, it allows to handle non-linear problems, such as chemical interaction.

In particular novel is the approach to simultaneously optimize two tracers (CO and NOx) and the paper nicely demonstrates the chemical feedbacks via the CO-NOx-NMHC-OH chemistry system but also the coupling via some common sources (in particular biomass burning).

A weak point of this study is the very simplified representation of transport in the IMAGES model, using monthly mean climatological wind fields. Therefore, the model cannot simulate synoptic variations (see general comment (3) below).

I would recommend the paper for publication in ACP after consideration of the following comments:

General comments

(1) Number of control parameters
As the authors point out, the adjoint technique allows to optimize large numbers of parameters. Nevertheless, in the presented study only a very small number (39) of control parameters is optimized, using 8 global continental regions only (+ ocean), and for some source categories (e.g. biogenic CO and VOC, oceanic CO) one global region only is used. It is not clear, why the study is restricted to such a small set of control parameters only. As pointed out e.g. by [Kaminski et al., 2001], the use of very large continental regions may lead to the "aggregation error", as no further optimization of the spatial emission pattern within these regions is performed. Using the the adjoint/variational approach, the use of much smaller regions should be possible without much higher computational costs ?

(2) Seasonal variations
Related to (1) is the question why no seasonal optimization has been performed (e.g. optimization of monthly mean emissions). Therefore, the study strongly relies on the temporal (and spatial) emission distribution of a priori bottom-up inventories. E.g. for biomass burning, different studies show different seasonalities. The authors should include some discussion about potential systematic errors arising from the use of fixed seasonalities.

(3) Model meteorology and data selection
A clear deficiency of the IMAGES model is the use of monthly averaged meteorol-

ogy (advection, convection, and turbulent mixing). Therefore, the model cannot reproduce synoptic variations. This appears especially critical for sites close to source regions (e.g. Mace Head or Hungary). In particular the use of data "representing non-background conditions" (as discussed in section 4.1 a) may lead to large systematic errors as such non-background conditions cannot be properly simulated by the IMAGES model. A further inconsistency arises from the fact that the meteorology applied is based on "mean climatological wind derived from a global analysis of ECMWF fields for the period 1985-1989", while observations are used for year 1997. It seems more appropriate to use also for the observations a climatological average over several years, taking into account their inter-annual variability. The preferred solution, however, would be fully consistent time periods both for meteorology and observations.

(4) Effectiveness of inversion in Southern Hemisphere

The authors state that the "inversion...is less efficient in the Southern Hemisphere" (Abstract, line 14 and sections 5 and 6). Clearly, there are less observations available in the Southern Hemisphere (SH) than in the Northern Hemisphere (NH). However, the authors further limit the use of SH data, by taking into account only Tierra Del Fuego from the high-latitude SH station, omitting Palmer, Syowa, Halley and South Pole ("to avoid redundancy" (page 8004, line 17)). These stations, however, are far distant from each other. The fact that their record look very similar is due to the well-mixed state of the high-latitude SH and the absence of sources, but does not mean that their information is redundant. The overall density of these stations (distribution over the global model grid) is not higher than in several other parts of the world. Previous studies which took into account all high-latitude SH stations showed an excellent agreement between inverse simulations and observations at these sites, e.g. [Bergamaschi et al., 2000; Kasibhatla et al., 2002; Petron et al., 2002].

Another factor contributing to the weak weighting of the high-latitude SH stations is the application of a constant percentage of 10 % of mean mixing ratio for the representativity error (equation 20). Due to low spatial and synoptic variability at the high-latitude

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SH stations the representativity error should be very small at these sites, but very high at sites close to source regions. Therefore, more appropriate than a constant percentage seem approaches which take into account the spatial and temporal gradients [Rödenbeck et al., 2003].

Specific comments

page 7987, line 25-27 ("It is implicitly assumed... well predicted by the model") and section 2 ("The IMAGES model")
It would be useful to include some information about validation of the IMAGES model (e.g. using standard tracers such as 222Rn and SF6 and intercomparison with other models).

page 7988-7989 (Introduction)
It should be mentioned that the adjoint model can be used to explicitly calculate inverse base functions for a given set of observations (as applied e.g. by [Houweling et al., 1999; Kaminski et al., 1999a; Kaminski et al., 1999b]) with subsequent analytical solution of the optimization problem or alternatively, as in this study, in an iterative approach. Also it would be helpful to explain the similarity of this iterative approach to 3DVAR and 4DVAR techniques.

page 7996, equation 2
The use of exponentiation of control parameters is certainly a very interesting approach to "ensure positiveness of optimized fluxes". However it leads to a non-Gaussian probability density function. This should be explicitly mentioned and discussed. The probability density function p is related to the cost function J as:
J = -ln p
For Table 15 it should be explained how the a priori and a posteriori uncertainties of emissions are derived from the Δ f values.

page 7997, line 4:

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I assume that ti refer to monthly mean values. Would be helpful, to explicitly explain.

page 7997, equation 4 / 5: Both equations are identical (?)

page 7999/8000 (section "3.2 Adjoint code generation and minimizer")
The first part of this section (page 7999, line 8 to page 8000, line 14) could be left out, as similar descriptions of the adjoint technique can be found in standard references (e.g. [Giering and Kaminski, 1998]).

page 8001, line 4 ("15 minutes for a complete simulation")
Does this refer to a 1-year simulation with full chemistry ?

page 8001, line 11 ("convergence criterion")
How many ierations are typically required ? Does the optimization include also the spin-up time, or is the spin-up period 09/1996-12/1996 kept fixed and only the target period 01-12/1997 optimized ?

page 8004, line 26 ("more than three times greater than the mean value")
Does "mean" value refer to the monthly value ? Please specify.

page 8005, line 5 ("actual errors")
I assume that measurement errors are meant.

page 8005, line 8-10 ("It is usually small when the number of measurements is sufficiently large")
This relationship is not clear. Large number of measurements gives better statistics, but not smaller standard deviation. Only the error of the mean (sigma / sqrt(n)) would decrease with increasing n (in case of random errors only).

page 8015, line 19-21 ("increasing the number of control variables... leads...to higher uncertainties")
However such higher uncertainties are probably more realistic. Small number of control variables (e.g.. large regions) imply the assumption of a perfect correlation between

emissions of different grid cells within one region (and in this study also in time, as no seasonal optimization is performed). See e.g. discussion by [Houweling et al., 1999].

page 8016, line 7-8 ("a posteriori are smaller than the a priori error estimates, except for the African anthropogenic CO sources")
A posteriori errors cannot exceed the a priori errors. I assume that in this case it is due to the approximations by the finite differences calculations.

page 8017/8018 (discussion of correlations)
It could be interesting to provide a figure with the correlation matix.

page 8022, line 20 ("...are significantly higher")
I assume "lower" is meant instead of "higher" ?

page 8039, Table 4
give value of a priori flux for CO deposition

page 8040, Table 4
give value of a priori flux for NOx deposition

page 8041, Table 5
would be interesting to include the emission factors in this table

page 8059, Fig 8
would be helpful to include the names of the regions in this figure (as the region names are used in Fig. 14 and 15)

page 8066, Fig 15
I assume that model values are extracted for the corresponding month of observations ? If so, this information should be added in the figure

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