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Comparison of adjoint and analytical Bayesian inversion methods for constraining Asian sources of carbon monoxide using satellite (MOPITT) measurements of CO columns

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[1] We apply the adjoint of an atmospheric chemical transport model (GEOS-Chem CTM) to constrain Asian sources of carbon monoxide (CO) with 2° × 2.5° spatial resolution using Measurement of Pollution in the Troposphere (MOPITT) satellite observations of CO columns in February–April 2001. Results are compared to the more common analytical method for solving the same Bayesian inverse problem and applied to the same data set. The analytical method is more exact but because of computational limitations it can only constrain emissions over coarse regions. We find that the correction factors to the a priori CO emission inventory from the adjoint inversion are generally consistent with those of the analytical inversion when averaged over the large regions of the latter. The adjoint solution reveals fine-scale variability (cities, political boundaries) that the analytical inversion cannot resolve, for example, in the Indian subcontinent or between Korea and Japan, and some of that variability is of opposite sign which points to large aggregation errors in the analytical solution. Upward correction factors to Chinese emissions from the prior inventory are largest in central and eastern China, consistent with a recent bottom-up revision of that inventory, although the revised inventory also sees the need for upward corrections in southern China where the adjoint and analytical inversions call for downward correction. Correction factors for biomass burning emissions derived from the adjoint and analytical inversions are consistent with a recent bottom-up inventory on the basis of MODIS satellite fire data.


1. Introduction

[2] Inverse modeling is a standard tool for combining observations of atmospheric composition with knowledge of atmospheric processes (transport, chemistry) to derive quantitative constraints on emissions to the atmosphere. A chemical transport model (CTM), known as the forward model for the inversion, solves the continuity equation to predict concentrations as a function of emissions. The inverse model then optimizes the emission estimates by fitting the CTM to the observed concentrations, subject to error weighting and a priori information on the emissions. We compare here analytical and adjoint methods for the inverse problem as applied to optimization of Asian emissions of carbon monoxide (CO) using Measurement of Pollution in the Troposphere (MOPITT) satellite observations of CO atmospheric columns [Deeter et al., 2002]. We demonstrate the ability of the adjoint approach to constrain emissions with high resolution when using a large satellite data set, revealing aggregation errors in the analytical method.

[3] Consider the general problem of estimating a set of emissions (assembled in a state vector \( x \)), given a set of observed atmospheric concentrations (observation vector \( y \)) and a CTM forward model \( y = F(x) \). One can define an optimal value of \( x \) as that which minimizes an error-weighted least squares (chi-square) scalar cost function \( J(x) \), derived from Bayes’ theorem with the assumption of Gaussian errors [Rodgers, 2000]. The cost function describes the error-weighted mismatch between the observed concentrations, \( y \), and those simulated with the forward model, \( F(x) \), as well as the error-weighted mismatch between the true state and the a priori estimate \( x_a \). The solution for \( \min(J(x)) \) with respect to \( x \) such that \( \nabla_x J(x) = 0 \) defines the Maximum A Posteriori (MAP) solution of the inverse problem [Rodgers, 2000].

[4] Most of the inverse modeling literature for atmospheric composition has used an analytical solution for \( \nabla_x J(x) = 0 \),
and we refer to this here as the “analytical method.” It has been applied extensively for example for inverse modeling of CO2 surface fluxes and CO emissions using observations from surface sites [Bousquet et al., 1999; Bergamaschi et al., 2000; Kasibhatla et al., 2002; Pétron et al., 2002] and aircraft [Palmer et al., 2003; Palmer et al., 2006]. Computing this analytical solution involves construction of the CTM Jacobian matrix (K = ∂y/∂x) and subsequent multiplication and inversion of matrices with dimensions of dim(x) and dim(y). This limits the practical size of x, i.e., the number of emission regions that can be optimized (limitations in the size of y can be overcome by partitioning the observations into independent packets assimilated by the inversion, i.e., “sequential updating” [Rodgers, 2000]). However, a large state vector x is desirable in applying the inverse method to satellite observations, where one would like to exploit the richness of the data to constrain emissions with high spatial and temporal resolution, limited only by the resolution of the CTM used as the forward model.

[5] An alternative to the analytical method is to seek a numerical solution to ∇J(x) = 0 by using the CTM adjoint to efficiently compute ∇w(x) from successive estimates of x starting with the a priori, and applying an iterative optimization algorithm to converge to the solution. We refer to this here as the “adjoint method.” Pioneering studies applying the adjoint method to optimize emissions include the work of Elbernt et al. [1997], Eibern and Schmidt [1999], and Kaminski et al. [1999]. Recent studies have applied the method to constrain aerosol emissions [Hakami et al., 2005; Dubovik et al., 2008], global CO and NOx emissions using surface measurements of CO from the NOAA/CMDL network and NO2 columns from the GOME satellite instrument [Müller and Stavrakou, 2005], global CO emissions using MOPITT columns [Stavrakou and Müller, 2006], and East Asian CO sources using measurements from ground stations [Yumimoto and Uno, 2006].

[6] Inversion of CO sources is an attractive application of the adjoint method because of the availability of dense and high-quality satellite observations. The MOPITT instrument is a nadir viewing pressure modulator radiometer that measures broadband infrared radiation in thermal emission, from which CO column and profile concentrations are retrieved. It was launched onboard NASA’s EOS Terra in 1999 in a sun-synchronous polar orbit [Deeter et al., 2002], provides measurements with 1–2 pieces of information in the vertical and global coverage every 3 days. CO is emitted by incomplete combustion and is also produced in the atmosphere by oxidation of volatile organic compounds (VOCs). Its sink is oxidation by OH with a lifetime of about two months. Several recent inverse studies have used MOPITT CO data to constrain CO sources [Arellano et al., 2004; Heald et al., 2004; Pétron et al., 2004; Arellano et al., 2006; Stavrakou and Müller, 2006; Arellano et al., 2007]. All, except Stavrakou and Müller [2006], used the analytical method. Arellano et al. [2006] constrained a state vector of emissions including over 100 elements, much larger than previous studies, but still not commensurate to the density of data provided by MOPITT.

[7] Stavrakou and Müller [2006] applied an adjoint method to 2000–2001 MOPITT CO columns to constrain global CO sources, and compared “large region” and “grid-based” approaches to the inversion. This corresponded to CO source inversion at low (18 regions) versus high (5° × 5°) resolution, in both cases using an adjoint of the IMAGES CTM driven by monthly mean wind fields. They found that the grid-based inversion yielded better agreement with the observed CO columns and allowed for greater exploitation of the data. Here we also constrain CO sources at the native resolution of our CTM (2.5° × 2.5°), but rather than assess the impact of the resolution alone, we compare the adjoint to the analytical inversion method. Adjoint and analytical methods yield theoretically the same MAP solution but practically we may expect differences from the numerical approximation involved in the adjoint method. In addition, the analytical method provides exact covariance information on the solution which the adjoint method does not.

[8] We focus our analysis on Asian CO sources using MOPITT observations for the March–April 2001 period of the NASA/TRACE-P aircraft mission over the NW Pacific. This mission focused on characterizing the chemical outflow from the Asian continent and provided validation data for MOPITT [Jacob et al., 2003]. Palmer et al. [2003] and Wang et al. [2004] previously used the TRACE-P aircraft observations to invert for Asian CO sources, using as a priori a detailed Asian emissions inventory for 2000 [Street et al., 2003]. Heald et al. [2004] compared the information content from the TRACE-P aircraft and MOPITT satellite observations as constraints on Asian CO sources and concluded that the satellite observations were far richer. All these studies used the analytical method for the inversion. Additional studies for the TRACE-P period used simpler methods to constrain Asian CO sources from the aircraft and MOPITT observations [Carmichael et al., 2003; Allen et al., 2004], all with consistent results. March–April 2001 thus represents a well-studied period for Asian CO sources, and the study of Heald et al. [2004] is of particular value to us as a reference for the analytical solution to the inverse problem.

2. Analytical Versus Adjoint Solutions to the Inverse Problem

[9] Here we address the inverse problem of determining emissions x given observed atmospheric concentrations y and a CTM forward model:

\[ y = F(x) + \varepsilon \]  

where ε is the “observation error” including contributions from the measurements and from imperfection in the forward model (e.g., transport error). We apply an a priori constraint x0 on the emissions subject to error εa. Application of Bayes’ theorem with assumption of Gaussian errors leads to a MAP solution for x given y as the minimum of the cost function J(x) [Rodgers, 2000]:

\[ J(x) = (F(x) - y)^T S_\Sigma^{-1} (F(x) - y) + \gamma (x - x_0)^T S_a^{-1} (x - x_0) \]  

Here S_Σ and S_a are the observational and a priori error covariance matrices representing the error statistics of ε and ε_a, respectively. The regularization parameter γ controls the relative constraints applied by the observational and a priori parts of the cost function [Hakami et al., 2005; Müller and Stavrakou, 2005; Yumimoto and Uno, 2006; Henze et al., 2007]. Bayes’ theorem would have γ = 1, but this assumes that S_Σ and S_a are adequately characterized, which is difficult
Figure 1. Mean atmospheric CO columns over eastern Asia and downwind for the TRACE-P period (21 February to 10 April 2001). (top) MOPITT satellite observations and GEOS-Chem model values using (middle) a priori and (bottom) a posteriori CO sources (Table 1). The GEOS-Chem values are smoothed with the local MOPITT averaging kernels.

3. MOPITT Observations and a Priori Sources

We use here the same MOPITT observations, forward model (GEOS-Chem CTM), a priori emissions, and error covariance matrices as in the inverse analysis of Heald et al. [2004]. The MOPITT data are daytime CO columns (1030 local time overpass) during the TRACE-P period (21 February to 10 April 2001) over the Asian domain (10°S–55°N, 50°E–180°E), and averaged over the 2° × 2.5° GEOS-Chem grid. This amounts to 21,569 observations (Figure 1). The “best case” MAP solution reported by Heald et al. [2004] used a slightly narrower latitudinal domain (0°–55°N) and included chi-square filtering of outliers, reducing the total number of observations to
The effects of Case 3 for use assuming no seasonal variation in fuel sources and a seasonal variation in biomass burning as described by [2003b], based on the climatology of Streets et al. [2003]. Following Duncan et al. [2006], we augment these emissions by 19% (anthropogenic) and 16% (biomass burning) to account for rapid oxidation to CO of coemitted nonmethane volatile organic compounds (NMVOCs). Following Duncan et al. [2006], we augment these emissions by 19% (anthropogenic) and 16% (biomass burning) to account for rapid oxidation to CO of coemitted nonmethane volatile organic compounds (NMVOCs).

Figure 2 shows the a priori emissions for our simulation period (1 February to 10 April 2001), featuring maxima in the biomass burning regions of India and southeastern Asia as well as high values from fuel use in eastern China. Also following Heald et al. [2004], we include in the state vector a separate chemical source of CO lumping production from methane and from biogenic NMVOCs including isoprene.

Table 1. Optimization of CO Sources by Analytical and Adjoint Inverse Methods

<table>
<thead>
<tr>
<th>Source Region</th>
<th>A Priori Streets et al. [2006]</th>
<th>Best Estimate</th>
<th>Ensemble Range</th>
<th>Case 1</th>
<th>Case 2</th>
<th>Case 3</th>
<th>Adjoint Inversion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Japan</td>
<td>7.7</td>
<td>2.67</td>
<td>(0.47–1.83)</td>
<td>1.69</td>
<td>1.87</td>
<td>1.88</td>
<td>1.11</td>
</tr>
<tr>
<td>Korea</td>
<td>6.3</td>
<td>2.67</td>
<td>(0.47–1.83)</td>
<td>1.69</td>
<td>1.87</td>
<td>1.88</td>
<td>0.94</td>
</tr>
<tr>
<td>N. China</td>
<td>9.6</td>
<td>0.32</td>
<td>(0.47–1.83)</td>
<td>0.87</td>
<td>0.72</td>
<td>0.76</td>
<td>1.13</td>
</tr>
<tr>
<td>C. China</td>
<td>52.9</td>
<td>1.48</td>
<td>(1.29–1.75)</td>
<td>1.56</td>
<td>1.59</td>
<td>1.83</td>
<td>1.34</td>
</tr>
<tr>
<td>W. China</td>
<td>33.0</td>
<td>2.12</td>
<td>(1.29–1.75)</td>
<td>2.30</td>
<td>2.13</td>
<td>2.38</td>
<td>1.10</td>
</tr>
<tr>
<td>S. China</td>
<td>25.0</td>
<td>0.87</td>
<td>(0.63–1.37)</td>
<td>0.42</td>
<td>0.50</td>
<td>0.31</td>
<td>0.94</td>
</tr>
<tr>
<td>SE Asia</td>
<td>69.2</td>
<td>0.61</td>
<td>(0.37–0.90)</td>
<td>0.64</td>
<td>0.68</td>
<td>0.63</td>
<td>0.78</td>
</tr>
<tr>
<td>Philippines</td>
<td>5.6</td>
<td>0.48</td>
<td>(0.48–1.11)</td>
<td>0.62</td>
<td>0.87</td>
<td>0.89</td>
<td>0.90</td>
</tr>
<tr>
<td>Indonesia</td>
<td>55.6</td>
<td>1.41</td>
<td>(1.01–1.14)</td>
<td>1.44</td>
<td>0.94</td>
<td>0.96</td>
<td>0.97</td>
</tr>
<tr>
<td>India</td>
<td>89.9</td>
<td>0.51</td>
<td>(1.01–1.14)</td>
<td>0.56</td>
<td>0.61</td>
<td>0.50</td>
<td>0.37</td>
</tr>
<tr>
<td>Europe</td>
<td>145</td>
<td>0.73</td>
<td>(1.01–1.14)</td>
<td>0.77</td>
<td>0.80</td>
<td>0.75</td>
<td>1.07</td>
</tr>
<tr>
<td>Rest of world</td>
<td>596</td>
<td>1.15</td>
<td>(1.01–1.14)</td>
<td>1.15</td>
<td>1.14</td>
<td>1.16</td>
<td>1.16</td>
</tr>
<tr>
<td>Methane and biogenic NMVOCs</td>
<td>1205</td>
<td>1.15</td>
<td>(1.01–1.14)</td>
<td>1.15</td>
<td>1.14</td>
<td>1.16</td>
<td>0.99</td>
</tr>
<tr>
<td>Number of MOPITT observations</td>
<td>18,295</td>
<td>18,295</td>
<td>18,295</td>
<td>20,542</td>
<td>21,569</td>
<td>21,569</td>
<td></td>
</tr>
</tbody>
</table>

*Sources for the TRACE-P period (February–April 2001), converted to equivalent Tg a⁻¹ assuming no seasonal variation in fuel sources and a seasonal variation in biomass burning as described by Duncan et al. [2003]. The regional sources include direct emissions from fossil fuel, biofuel, and biomass burning, as well as chemical production from anthropogenic nonmethane volatile organic compounds (NMVOCs) coemitted with CO [Duncan et al., 2006].

*Source regions are those of Heald et al. [2004] and follow the same numbering as in that paper. See Figure 3a for region boundaries. Japan and Korea were treated as one single source region in the Heald et al. [2004] analytical inversion, and so were the sources from oxidation of methane, biogenic NMVOCs, and emissions outside Eurasia (rest of world).

*Best case inverse solution from Heald et al. [2004] constraining an 11-element state vector using the analytical method. Relative to this best case from Heald et al. [2004], our adjoint solution presented here assumes uncorrelated observational error (no off-diagonal terms in the observational covariance matrix), uses an extended latitudinal domain (10°S–55°N versus 0°–55°N), and does not remove outliers in the MOPITT data (χ² filter). The effects of these successive modifications on the Heald et al. [2004] analytical inversion are shown in the Table as Cases 1–3.

*Range of source constraints obtained by Heald et al. [2004] in an ensemble of inversions with varying inversion parameters, corresponds to Heald et al. [2004, Figure 10].

*No observational error covariance.

*No observational error covariance, extended domain.

*No observational error covariance, extended domain, no χ² filter. These conditions reproduce exactly those used in the adjoint inversion (results in the next column).

*The adjoint inversion optimizes the CO source on the 2° × 2.5° grid of the GEOS-Chem CTM, and the solution is averaged here over the 11 regions of the analytical inversion for purpose of comparison. See Figure 5 for the fine structure of the adjoint solution.

18,295 (Table 1). For the purposes of our comparison we repeated the Heald et al. [2004] analytical solution for our extended data set with no data filters (see Table 1), and results will be discussed in section 5. Although we do not correct MOPITT observations by the known 6% high bias [Emmons et al., 2007], Heald et al. [2004] showed that the effect of the correction on the a posteriori solution was minimal. The observation error covariance matrix Sₑ for use in the inversion is dominated by the forward model error with a spatial covariance structure found by Heald et al. [2004] to decay over a second-order autoregressive length scale of 150 km. Since this length scale is less than our grid resolution we treat Sₑ as diagonal. Observational errors are derived with the relative residual error (RRE) method of Palmer et al. [2003] and are in the range 5–26%, as shown by Heald et al. [2004, Figure 4].

Our a priori CO sources, following Heald et al. [2004], include monthly Asian anthropogenic emissions (fossil fuel and biofuel combustion) from Streets et al. [2003] and daily biomass burning emissions from Heald et al. [2003b], based on the climatology of Duncan et al. [2003]. Following Duncan et al. [2006], we augment these emissions by 19% (anthropogenic) and 16% (biomass burning) to account for rapid oxidation to CO of coemitted nonmethane volatile organic compounds (NMVOCs). Figure 2 shows the a priori emissions for our simulation period (1 February to 10 April 2001). The sources are in units of kilograms per 2° × 2.5° grid square.

Figure 2. A priori CO source from fossil fuel, biofuel, and biomass burning during the TRACE-P period (1 February to 10 April 2001). The sources are in units of kilograms per 2° × 2.5° grid square.
monoterpenes, acetone, and methanol. This chemical source
is as described by Duncan et al. [2006].

Following Heald et al. [2004], errors on the a priori
fuel CO sources are taken from Streets et al. [2003] and
capped at 100%. Errors on the biomass burning sources are
assumed to be 50%, and these two errors are added in
quadrature and again capped at 100% to obtain the regional
source error, which ranges from 17% in Japan to 100%
in southeastern Asia, India, Philippines, and Indonesia. We
assume errors to be spatially uncorrelated so that $S_a$ is
diagonal.

4. GEOS-Chem Forward Model and Its Adjoint

[17] The GEOS-Chem CTM used as forward model in the
inversion is driven here by assimilated meteorological data
from the Goddard Earth Observing System (GEOS-3) of the
NASA Global Modeling and Assimilation Office (GMAO). We
use a linear CO simulation (GEOS-Chem version 6-02-05;
http://www-as.harvard.edu/chemistry/trop/geo) with stored
monthly mean OH concentration fields from a previous
O$_3$-NO$_x$-NMVOC simulation [Fiore et al., 2003]. Our global
annual mean pressure-weighted OH concentration below
200 hPa is 1.00 × 10$^6$ cm$^{-3}$. The resolution is 2° × 2.5°
in the horizontal, with 30 vertical levels and 15 min transport
steps. We apply local MOPITT averaging kernels [Deeter et al.,
2002] to the GEOS-Chem vertical profiles of CO and obtain column values for comparison to
MOPITT (Figure 1). The MOPITT averaging kernels have
greatest sensitivity in the middle troposphere. Our linear CO
simulation is the same as that used in previous applications
of GEOS-Chem to interpret CO observations from the
TRACE-P period [Heald et al., 2003a; Jones et al., 2003;
Palmer et al., 2003; Heald et al., 2004; Wang et al., 2004].

[18] Our forward and adjoint model simulations cover the
February–April 2001 period, starting from observed fields
on 1 February. This initialization is done by spinning up
GEOS-Chem from January 2000 to February 2001 and then
adjusting the 1 February 2001, model CO concentrations by
the ratio of mean model CO columns in each 2° zonal band
to the corresponding MOPITT observations on that day. The
adjustment factors range from +20% in the tropics to +4–8%
at northern midlatitudes and −2% in the Arctic. Such an
adjustment was not done in the work by Heald et al. [2004],
where instead the inversion optimized for year-round emis-
sions assuming known temporal variability. This would
cause some difference with our results if the correction
factors for the emissions after 1 February are different from
those before, although there is no particular reason why that
should be so. The 1 February adjustment in our work also
corrects the latitudinal background so that our observational
error derived from the RRE method is slightly lower (by up
to 3%) than in the work by Heald et al. [2004]. We did not
implement MOPITT scaling and 1 February start date in
order to preserve as much of the analytical inversion setup
as possible.

[19] The construction and theoretical validation of the
GEOS-Chem adjoint is presented by Henze et al. [2007] in
the context of an aerosol source inversion. The adjoint code
was derived from the forward code using a (discrete) adjoint
of algorithms approach [Giering and Kaminski, 1998]. The
exception was the adjoint of the advection operator, for

which the continuous approach was adopted, wherein the
adjoint model equations are solved using the same Lin and
Rood [1996] advection scheme as in the forward model but
with reverse winds. Although the advective component of
the adjoint leads to sensitivities (gradients) that differ from
forward model sensitivities, these discrepancies are not
inaccuracies in either discrete or continuous approach, as
has been discussed extensively in previous work [e.g.,
Sirkes and Tziperman, 1997; Thuburn and Haine, 2001;
Vukicevic et al., 2001; Hakami et al., 2007; Henze et al.,
2007]. In fact, a continuous approach to an adjoint of
advective component has been shown to successfully con-
strain even a point source [Davoine and Bocquet, 2007]. We
added here self-adjoint modules for CO emissions and
chemical loss (these operator matrices are diagonal and
thus are not modified by transposition), as well as the
adjoint (transpose) of the MOPITT averaging kernel matri-
ces. The cost function gradient computed by the adjoint
model is used with a bounded quasi-Newtonian limited-
memory BFGS optimization [Liu and Nocedal, 1989] to
obtain the MAP solution for CO sources. In addition to
testing of the adjoint model conducted and previously
reported by Henze et al. [2007], we performed a series of
Observation System Simulation Experiments (OSSSes),
wherein we ascertained the ability of the optimization
system to successfully constrain CO sources given a varied
density of data.

5. Implementation of the Adjoint Method

[20] Our adjoint solution optimizes the combustion sour-
ces of CO (treated as surface fluxes) at the global 2° × 2.5°
horizontal resolution of the forward model. The state vector
consists of time-invariant correction factors with a zero
lower bound imposed by the optimization algorithm, applied
to the a priori inventory for the model grid squares where
these emissions are nonzero (3013 out of 13,104 surface
grid squares). Temporal variability of emissions is assumed
to be adequately constrained by the a priori emission
inventories of Streets et al. [2003] and Heald et al. [2003b]
(monthly for fuel, daily for biomass burning) and so is not
optimized here. We add a correction factor to
optimize the background source from oxidation of methane
and biogenic NMVOCs, so that the state vector has 3014
elements. The inversion is conducted for the TRACE-P
period (1 February to 10 April or 69 days).

[21] We initialized the GEOS-Chem CO field on 1 February
with MOPITT observations as described in section 4. As a
result, the sensitivity of the cost function to emissions
before 1 February is negligibly small; the norm of the
corresponding cost function gradient (0.01) is much smaller
than the norms of the a priori (7.20) and a posteriori (0.35)
cost function gradients. We determined an optimal value $\gamma = 0.01$ for the regularization parameter as that which yields
the lowest a posteriori value of the cost function (Figure 3),
however all values of $\gamma$ that are smaller than 1 yield an a
posteriori cost function within 1%, essentially eliminating
the influence of a priori constraint. When $\gamma > 1$, influence of
the a priori limits the optimization of the solution. A likely
reason for our need to reduce the weight of the a priori
information through $\gamma$ is that we did not account for spatial
correlation of a priori sources, whereas, in fact, CO source
errors within a given geopolitical region are expected to be correlated [Stavrakou and Müller, 2006].

[22] With \( \gamma = 0.01 \), the a posteriori cost function is 22,575 (starting from an a priori value of 29,191). This is comparable to the number of observations used in the inversion (21,569). Figure 4 shows the evolution of the cost function as a function of the iteration number in the inversion. The analytical method using the same data set as the adjoint method has a higher a priori cost function (37,686, see case 3 of Table 1), reflecting the GEOS-Chem initialization in the adjoint method with MOPITT observations on 1 February. The a posteriori cost function from the analytical method (28,762) is higher than from the adjoint method, indicating that the adjoint method provides a better fit to the observations. The a priori source constraint does not contribute significantly to the a posteriori cost function in either the adjoint or the analytical solution.

6. Comparison of Adjoint and Analytical Solutions

[23] Figure 5 shows the a posteriori emission correction factors for the adjoint and analytical solutions. The correction factors for the adjoint solution range from the lower bound of zero (a few grid squares in biomass burning dominated regions in India), to 2.85 (northern China). Average model bias relative to the MOPITT observations decreases from a mean of +2.9% with a priori emissions to −2.0% with a posteriori emissions; however, on a regional scale, the a posteriori model bias is negligible in India, southeastern Asia and southern China, indicating vast improvement. The global source from oxidation of methane and biogenic NMVOCs decreases by only 1%, which provides some confidence in the model concentrations of OH. This confidence justifies in turn focusing the optimization on CO sources, rather than on the sink from oxidation by OH.

[24] The analytical solution in Figure 5 is for case 3 in Table 1, which has exactly the same setup as the adjoint solution. It departs from the best case analytical solution by Heald et al. [2004] in that it includes only diagonal terms of the observational error covariance, it uses an expanded data
domain, and it does not filter outliers. The effects of these successive changes in the analytical solution relative to the Heald et al. [2004] best case are shown as cases 1–3 in Table 1. Also shown in Table 1 are the ranges of results from the ensemble of analytical solutions presented by Heald et al. [2004] with varying choices of inversion parameters and MOPITT data processing, representing estimated uncertainty ranges in the inverse analysis. The correction factors obtained from our adjoint solution generally fall well within these ranges but tend to be smaller than for the analytical solution. Error in the adjoint solution would need to be estimated with a similar ensemble approach as in the work by Heald et al. [2004], and is likely narrower owing to reduction in the aggregation error.

Aircraft observations from the TRACE-P campaign over the NW Pacific in spring 2001 [Jacob et al., 2003] offer an independent evaluation of our adjoint solution. Figure 6 shows the latitudinal gradients of CO concentrations measured by the aircraft in two altitude ranges (0–3 and 3–12 km) and simulated by the model using a priori and a posteriori sources. The observations are averaged over the model grid squares, and the model is sampled at the location and time of the measurements. Palmer et al. [2003] previously pointed out that the model with a priori sources overestimates the TRACE-P observations in the free troposphere south of 25°N (excessive biomass burning emissions in SE Asia) and underestimates observations north of 30°N (insufficient Chinese anthropogenic emissions). Our a posteriori solution from the adjoint method affords significant improvement in the simulation of the TRACE-P data, as shown in Figure 6.

7. Fine Structure of the Adjoint Solution and Interpretation

The high-resolution adjoint inversion provides source constraints that distinguish individual cities, for example Tokyo and Mumbai in Figure 5. The adjoint inversion takes advantage of MOPITT’s ability to detect urban centers [Clerbaux et al., 2008]. It also distinguishes political boundaries as between China, Korea, and Japan, although these are not implemented as a priori constraints. It reveals compensating patterns of underestimate and overestimate within the same previously aggregated region of the analytical solution. This is most manifested in previously aggregated regions of India and Indonesia. In the Indian region from Heald et al. [2004], we find an underestimate of emissions in Mumbai, Sri Lanka and northern India, and an overestimate in the rest of India where
biomass burning emissions dominate. In the Indonesia region from Heald et al. [2004], emissions from Singapore and northern Indonesia are underestimated, but this is compensated on the regional scale by an overestimate of mostly biofuel emissions from Java (including Jakarta and other major cities. This points to the danger of aggregating large regions to afford analytical solution to the inverse problem.

[28] Streets et al. [2006] recently revisited their previous Chinese CO emissions inventory for the TRACE-P period [Streets et al., 2003] in light of evidence from inverse analyses of the TRACE-P and MOPITT data that their anthropogenic emission estimates from China (116 Tg CO a\(^{-1}\)) were too low by 40–55\% [Palmer et al., 2003; Heald et al., 2004; Wang et al., 2004]. We find the Chinese CO emissions underestimate to be only 15\% (Table 1). The updated Streets et al. [2006] inventory for China in 2001 is 157 Tg CO a\(^{-1}\), 36\% higher than that of Streets et al. [2003] due to inclusion of previously neglected sources (e.g., power plants, small coke ovens, synthetic ammonia production, unregistered rural vehicles, and coal mine fires), an update with 2001 data on activity rates, and corrections to emission factors (e.g., cement kilns, iron and steel industries, and vehicles).

[30] Figure 7 compares the geographical distribution of the corrections to the Streets et al. [2003] inventory from our adjoint inversion and from the revised bottom-up inventory of Streets et al. [2006]. Both show large and consistent upward corrections in central and eastern China. They agree on upward correction in northern China (where the “best estimate” Heald et al. [2004] inversion pointed to a downward correction) and in western China. There are however large discrepancies in southern China, where both the adjoint and analytical inversions find the need for downward correction but Streets et al. [2006] only find decreases in the southern edge of the country near the Myanmar border (where emissions are mostly from biomass burning). Streets et al. [2006] find large upward corrections in coastal southern China that are not seen in the adjoint inversion.

A recent biomass burning inventory for 2001 by van der Werf et al. [2006] using MODIS satellite fire counts finds much smaller CO emissions than the Heald et al. [2003b] inventory for India and southeastern Asia used here as a priori. They find biomass burning CO emissions to be largely absent in India and about 36\% below Heald et al. [2003b] in southeastern Asia, which agrees closely with the adjoint solution (Table 1 and Figure 5).

8. Conclusions

[32] We presented a comparison of adjoint and analytical methods for inverting Asian CO sources on the basis of MOPITT satellite observations of CO columns over Asia and the North Pacific during the TRACE-P aircraft campaign (February–April 2001). The adjoint method provides a powerful tool for exploiting high-density observations of atmospheric composition from space to constrain emissions with high resolution. The standard analytical method, requiring construction and operations of the Jacobian matrix of the forward model, is severely limited in terms of the source information that it can resolve. Our motivation was to illustrate the capability of the adjoint method through comparison to the analytical method. Several previous studies had applied the analytical method to invert Asian CO sources on the basis of MOPITT and TRACE-P data for the same period.

[33] We started from the previous analytical inversion by Heald et al. [2004] which used MOPITT CO column observations for the TRACE-P period, and the GEOS-Chem chemical transport model (CTM) as forward model, to constrain CO sources from 9 Asian regions. We used the same MOPITT observations, forward model, a priori source information, and error characterization as Heald et al. [2004], but optimized the Asian CO sources at the 2° × 2.5° horizontal grid resolution of the CTM rather than for the 9 coarse regions of Heald et al. [2004]. The resulting a posteriori cost function is lower in the adjoint solution than in the analytical solution, indicating a better fit to the observations.
We compared the results from the adjoint and analytical inversions by averaging over the 9 coarse Asian regions of the latter. The large-scale features of the a posteriori source constraints are very similar in both solutions; a priori Chinese fossil fuel and biofuel emissions are underestimated in the Streets et al. (2003) inventory, and emissions from biomass burning regions in India and southeastern Asia are overestimated in the Heald et al. (2003b) inventory. However, correction factors in the adjoint solution tend to be smaller than in the analytical solution. Also, the analytical solution finds the need for large increases in emissions in Korea-Japan, whereas the adjoint solution finds only a small increase in Japan and a decrease in Korea.

The high resolution of the adjoint solution provides constraints on emissions are the scale of individual cities, and reveals particularly large underestimation of CO emissions from Tokyo and Mumbai. The adjoint solution can also resolve geographical boundaries between China, Korea, and Japan even though these are not in the a priori constraints. Comparison of the adjoint and analytical solutions warns of large aggregation errors when optimizing sources averaged over coarse regions in the analytical solution. For example, in the Indian subcontinent, the adjoint solution is able to separate increases in Mumbai, northern India, and Sri Lanka from decreases in the rest of the region where the seasonal source is mostly from biomass burning.

Studies et al. (2006) recently revised their prior 2001 anthropogenic emission inventory for China [Streets et al., 2003] to address the underestimates found in previous inverse model analyses. They added previously neglected sources, updated information on activity rates, and corrected emission factors. Their updated inventory finds a 36% increase over the prior, as compared to 41–55% found in previous inverse studies and 15% in our work. Our adjoint solution agrees with Streets et al. [2006] in attributing most of the increase to central and eastern China where emissions are highest, but there are inconsistencies in the fine structure, and Streets et al. [2006] find the need for significant upward corrections in southern China that are not apparent in our adjoint or analytical inversions. The recent biomass burning inventory of van der Werf et al. [2006] yields estimates for eastern India and southeastern Asia that are much lower than Heald et al. [2003b] and consistent with our results. Independent comparison with in situ aircraft CO data from TRACE-P campaign shows improved model-data agreement when a posteriori sources instead of a priori sources are used.

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