Monthly CO surface sources inventory based on the 2000–2001 MOPITT satellite data

Gabrielle Pétron[1]

Advanced Study Program/Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, Colorado, USA

Claire Granier[2,3]

Service d'Aéronomie, Institut Pierre-Simon Laplace, Université Paris 6/Centre National de la Recherche Scientifique, Paris, France

Boris Khattatov, Valery Yudin, Jean-François Lamarque, Louisa Emmons, John Gille, and David P. Edwards

Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, Colorado, USA

Received 19 May 2004; revised 6 October 2004; accepted 14 October 2004; published 10 November 2004.

[1] This paper presents results of the inverse modeling of carbon monoxide surface sources on a monthly and regional basis using the MOPITT (Measurement Of the Pollution In The Troposphere) CO retrievals. The targeted time period is from April 2000 to March 2001. A sequential and time-dependent inversion scheme is implemented to correct an a priori set of monthly mean CO sources. The a posteriori estimates for the total anthropogenic (fossil fuel + biofuel + biomass burning) surface sources of CO in TgCO/yr are 509 in Asia, 267 in Africa, 140 in North America, 90 in Europe and 84 in Central and South America. Inverting on a monthly scale allows one to assess a corrected seasonality specific to each source type and each region. Forward CTM simulations with the a posteriori emissions show a substantial improvement of the agreement between modeled CO and independent in situ observations. *INDEX TERMS:* 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 3337 Meteorology and Atmospheric Dynamics: Numerical modeling and data assimilation. **Citation:** Pétron, G., C. Granier, B. Khattotov, V. Yudin, J.-F. Lamarque, L. Emmons, J. Gille, and D. P. Edwards (2004), Monthly CO surface sources inventory based on the 2000–2001 MOPITT satellite data, *Geophys. Res. Lett.*, *31*, L21107, doi:10.1029/2004GL020560.

1. Introduction

[2] Carbon monoxide plays a key role in the composition of the troposphere: it is the main sink for OH radicals and it is a precursor of ozone. The two largest surface sources of CO are the combustion of fossil fuel and the combustion of biomass (forest and savanna fires, biofuel use, and waste burning). Substantial amounts of CO are also produced in the atmosphere through the incomplete oxidation of methane and other hydrocarbons. The global average lifetime of CO is about two months. Its main sink is its reaction with OH. Uncertainties in the CO budget are still fairly large due to the difficulty in quantifying the variability of CO sources and sinks and due to a lack of measurements and emissions statistics used to derive emissions inventories.

[3] Several numerical studies with chemistry and transport models (CTMs) have shown a progressive convergence in the global estimates of the sources of CO [*Intergovernmental Panel on Climate Change*, 2001]. The direct source of CO (1000–1400 Tg/yr) due to biomass, biofuel and fossil fuel burning contributes close to 50% of the total CO source. The chemical production of CO is estimated between 880 TgCO/yr and 1400 TgCO/yr. The natural direct emission of CO from the vegetation, soils and the ocean surface represents a minor source in the CO budget.

[4] Recently, Bayesian synthesis inversion approaches have been implemented for the inverse modeling of the CO global budget, using the NOAA Climate and Monitoring Diagnostic Laboratory (CMDL) surface CO measurements and a global 3D tropospheric CTM [*Bergamaschi et al.*, 2000; *Pétron et al.*, 2002; *Kasibhatla et al.*, 2002]. The major result in *Pétron et al.* [2002] and *Kasibhatla et al.* [2002] was that the CO source in Asia had been previously substantially underestimated. Since March 2000, the MOPITT instrument has been providing a continuous and quasi global monitoring of CO in the lower atmosphere. Several groups have been using this new global dataset to constrain CO emissions regionally [*Allen et al.*, 2004] or globally [*Arellano et al.*, 2004].

[5] In this paper, we present the results of the first time-dependent inversion of CO global surface fluxes based on the MOPITT CO retrievals. In the next section, we briefly describe the selected observations. Section 3 presents our methodology as well as the CTM and the a priori emissions inventory used for the inversion. Major results including a

[1]Was also at Service d'Aéronomie, L'Institut Pierre-Simon Laplace, Université Paris 6/Centre National de la Recherche Scientifique, Paris, France, when this work was done.

[2]Also at Aeronomy Laboratory, Cooperative Institute for Research in Environmental Studies, National Oceanic and Atmospheric Administration, Boulder, Colorado, USA.

[3]Also at Max Planck Institut für Meteorologie, Hamburg, Germany.

0094-8276/04/2004GL020560$05.00

comparison with other CO emissions inventories are discussed in Section 4.

2. MOPITT CO Retrievals

[6] Since March 2000, the MOPITT instrument, a downward viewing sun-synchronous gas correlation radiometer on board the NASA/EOS Terra platform, has been monitoring the CO content in the troposphere. The retrieval algorithm for CO uses a single a priori profile for all regions and all seasons [*Deeter et al.*, 2003]. A cooler failure in May 2001 marks the limit between two phases in the MOPITT CO retrievals. Validation results for both phases have been described in *Emmons et al.* [2004]. In this study we use the MOPITT CO retrievals at the 700 hPa level, which has the highest sensitivity to the measured radiances in the troposphere. The MOPITT CO profiles and averaging kernels are binned onto the 2.8° × 2.8° horizontal grid of the CTM employed for the inversion and monthly averages are calculated from April 2000 to March 2001. We use data between 65°N and 65°S, as the weight of the a priori CO profile in the MOPITT retrievals increases towards the pole. The number of selected observations after quality control is between 4000 and 6000 each month.

3. Methodology

[7] For the optimization of CO emissions based on the MOPITT CO retrievals, we use an a priori set of sources and a global CTM to relate perturbations in the CO surface emissions to perturbations in the CO tropospheric amounts. This relationship, called observation matrix, needs to be "inverted" to transform the differences between the observed and the modeled CO distributions into corrections of the specified a priori CO fluxes. The magnitude of these corrections depends on the relative uncertainties assigned to the observations and to the a priori emissions. The inversion is designed to produce the best linear unbiased estimate of the emissions by solving a weighted least squares problem. The technique has been described in detail in *Pétron et al.* [2002].

[8] The global 3D CTM used in this study is MOZART (Model for OZone And Related chemical Tracers) [*Horowitz et al.*, 2003]. The model simulates the evolution of the distributions of 63 trace gases in the lower atmosphere with a horizontal resolution of 2.8° × 2.8° and with 28 vertical levels between the surface and 4 hPa. The meteorological fields are taken from the NCEP (National Center for Environmental Prediction) reanalysis [*Kalnay et al.*, 1996]. The emissions in the model are prescribed on a monthly basis. For CO emitted through technological activities and biofuel use, the a priori emissions have no seasonality and are based on annual estimates from the EDGAR-3 inventory (Emission Database for Global Atmospheric Research) [*Olivier and Berdowski*, 2001] extrapolated to 2000. The a priori biomass burning emissions have been derived on a monthly basis from the ATSR (Along the Track Scanning Radiometer) fire counts products for the period 1996–2001 (C. Granier and J.-F. Lamarque, personal communication, 2002). The modeled CO distributions are transformed using the MOPITT a priori CO profile and averaging kernels to allow the comparison with the satellite observations.

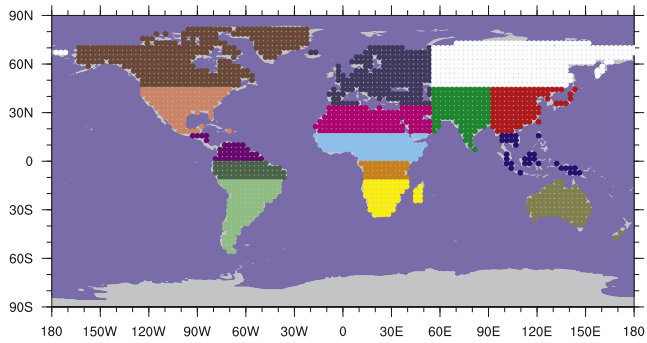

Figure 1. Partition of the globe into 15 regions for the anthropogenic emissions: Europe, North Asia, South Asia, East Asia, South East Asia, Oceania, Northern Africa (>10°N), North Equatorial Africa (10°N–0°), South Equatorial Africa (0°–10°S), South Africa (<10°S), Canada-Alaska, USA, Northern Central America, Southern Central America, South America (<10°S).

[9] A tracer version of MOZART has been run to compute the observation matrices used in the inversion. It uses the OH field (daily averages) archived from a simulation with the full chemistry and the a priori surface sources. The globe has been divided into 15 continental regions (see Figure 1) for fossil fuel use, biofuel use, as well as biomass burning emissions and into four latitudinal bands for the continental biogenic and for the oceanic net sources. The total number of fluxes to be optimized each month is 53. The geographical distribution of the CO flux inside each region is fixed. The a priori errors on the various emissions types are assumed to be uncorrelated in space and time. The a priori relative uncertainty assigned to the emissions is 100%, except for the sources due to fossil fuel use which are supposed to be better quantified and were therefore given a 50% uncertainty.

[10] Due to the average CO lifetime of two months, and to give more importance to large scale features in the observations such as plumes extending over and downwind of major source-regions, the lag of the inversion has been fixed to 2 months. For every month of observations, the sequential inversion algorithm computes updates of both the emissions of the past 2 months and the associated errors. The inversion produces a new set of CO emissions taken as the last updated estimates for each month. However, the number of fluxes to be optimized is too large and prevents any significant decrease in the a posteriori emissions uncertainties. In another inversion exercise, the anthropogenic emissions have been added up over the 15 continental regions to infer the a posteriori uncertainties for regional total anthropogenic sources.

[11] To test and ensure the a posteriori consistency between the CO sources and sinks, as well as to take into account the lasting impact of the CO sources (beyond the fixed 2-month lag in the inversion itself), the inversion is iterated three times. Updated OH and CO fields are computed with the full-chemistry CTM using the a posteriori CO emissions from the previous inversion. The new OH field is then read in the tracer model to compute updated observation matrices. These matrices, as well as the updated CO field are used to iterate the inversion.

Table 1. A Priori CO Global Budget Sources and Results of the Three Inversion Iterations (Unit TgCO/yr)

		A Posteriori		
	A Priori	*xa*1	*xa*2	*xa*3
Fossil fuel use	306	382	365	365
Biofuel use	246	328	310	318
Vegetation fires	322	435	373	408
Biogenic	160	239	180	183
Total	1034	1384	1228	1274
Total sink due to OH	2360	2710	2590	2630
Total chemical production	1580	1640	1650	1650

[12] The "observation" errors used for the inversion have to take into account the retrieval errors reported in the MOPITT dataset (10–15%), the representation errors due to the monthly averaging (~15%) and to the smoothing (10–20%, [*Pan et al.*, 1998]) and the model errors (assumed at 30%). The model errors describe the uncertainties in the forward model which links the observables to the surface emissions. They are for example errors in the boundary layer ventilation, the location of the emissions, the production and loss of CO, the meteorological fields, as well as aggregation errors. Practically, the forward model errors are difficult to characterize [*Kaminski et al.*, 2001]. For the monthly inversion exercise discussed in this letter, the various errors described above are supposed to be uncorrelated. More work has to be undertaken however in the near future to properly assess –spatial and temporal- covariances of the errors on satellite data retrievals. The total relative observation errors assigned here vary between 50 and 100%, depending on the location and season.

4. Results and Discussion

[13] The sequential inversion has been iterated to test the impact of the CO-OH chemistry non-linearity. CO budgets derived from MOZART simulations using either the emissions from the a priori inventory or from three successive iterations of the inversion have been compared (Table 1). The first inversion tends to overestimate the correction to be applied to the sources. The relative changes in the regional estimates derived from the second and the third iteration of the inversion are within 5% for emissions due to fossil fuel use, 10% for emissions due to biofuel use, and 25% for biomass burning emissions. It is possible that the a posteriori estimates oscillate around the solution of the inverse problem, due to the non-linear feedbacks in the system. In North America and Europe, the estimates for the CO sources due to both fossil fuel and biofuel use are 121 Tg/yr and 77 Tg/yr respectively, while the Asian estimate is as high as 352 Tg/yr. East Asia is the largest regional emitter of CO, representing 20% of the anthropogenic global emission. The a posteriori estimates for the East Asian emission of CO due to fossil fuel use, biofuel use and vegetation burning are 95, 95 and 21 Tg/yr, respectively. The biogenic and oceanic emissions of CO after the second and third inversion are within 12% of the a priori estimates. In *Arellano et al.* [2004], the a posteriori global CO source due to fossil fuel and biofuel use is between 782 and 960Tg/yr and the source due to vegetation fires is estimated between 486 and 633 Tg/yr depending on which MOPITT data are used.

[14] The implementation of the sequential inversion with a monthly frequency enables the derivation of a noticeable seasonality for emissions due fossil fuel and biofuel. These sources are respectively 30% and 100% higher during the winter months in the northern hemisphere compared to the summer months. The time-dependent inversion also derives several emission peaks lasting 2 to 3 months due to vegetation fires in Africa (December to February north of the Equator, July to September from the Equator to 10°S, and September–October south of 10°S). The a posteriori emissions for the South American region peak in October 2000.

[15] The a posteriori estimates for CO emissions due to vegetation fires for the time period April 2000 to March 2001 are respectively 26, 13 and 19 TgCO/yr for three regions of South America (10°N-Equator, Equator-10°S, south of 10°S) and 66, 51 and 43 TgCO/yr for the same three latitudinal bands in Africa. Regional biomass burning sources have been compiled by *van der Werf et al.* [2004] based on satellite observations of fire activity and on the CASA terrestrial biogeochemical model. For the same time period, their source estimates for South America are respectively 24, 44 and 46 TgCO/yr. For Africa, *van der Werf et al.* [2004] regional estimates are respectively 89, 45 and 56 TgCO/yr. Figure 2 shows a comparison of our a priori and a posteriori monthly biomass burning sources in Africa with *van der Werf et al.* [2004] inventory. There are substantial differences in the timing and intensity of the

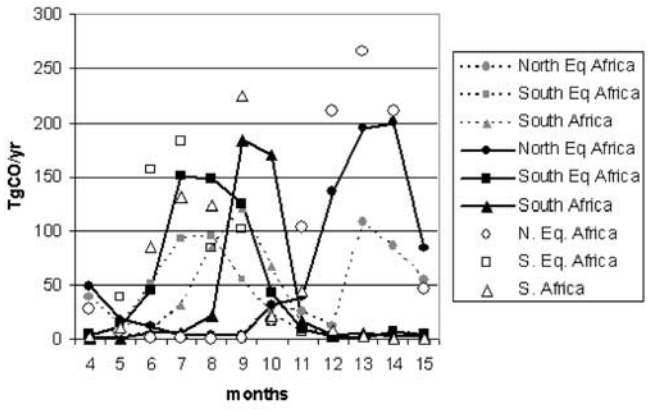

Figure 2. Comparison of our a priori (grey symbols) and a posteriori (last iteration, black symbols) monthly biomass burning sources in Africa with *van der Werf et al.* [2004] inventory (white symbols).

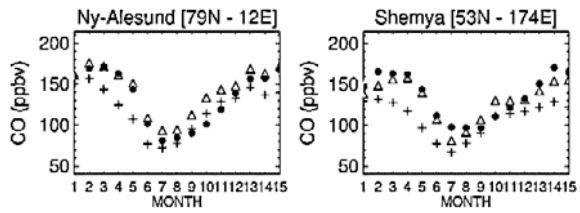

Figure 3. Comparison between CO weekly measurements (black dots) at two surface stations from the CMDL network and the a priori (crosses) and a posteriori (triangles) modeled CO mixing ratios.

Table 2. Regional A Priori (xb) and A Posteriori Total Anthropogenic CO Source Estimates (Unit TgCO/yr)[a]

Region	xb	sigb_ag	xa2	xa_ag	siga_ag
Europe	77.7	21	90.3	90	1
North Asia	20.3	17	20.2	20	2
South Asia	99.4	64	121.0	121	4
East Asia	141.6	117	207.1	203	37
South-East Asia	49.1	30	44.5	46	18
Oceania	72.9	112	89.6	89	57
Northern Africa	24.0	11	25.6	25	11
North Eq. Africa	77.1	125	119.1	128	50
South Eq. Africa	42.7	86	60.3	53	58
South Africa	41.8	91	48.7	57	52
Canada	8.4	10	8.6	9	10
USA	102.2	48	131.6	119	37

[a]The a posteriori fluxes have been derived from the sequential inversion (xa2) and from the non-sequential inversion of aggregated fluxes (xa_ag). For the non-sequential inversion of net fluxes, the a priori and a posteriori uncertainties are given as sigb_ag and siga_ag.

emissions peaks. There is also a 1 to 2-month delay between the peak in the MODIS fire counts and the peak in the MOPITT CO retrievals for most regions in the southern hemisphere (not shown).

[16] *Arellano et al.* [2004] a posteriori biomass burning emissions for the year 2000 are 30% larger than our estimates. The disagreement between the two inventories is quite large for South America and South-East Asia. More work is needed to improve and validate the derivation of biomass burning emissions from the combination of biosphere-atmosphere coupled models and a suite of in situ and remote sensing measurements.

[17] The MOZART CTM was next run with the last a posteriori CO emissions (xa3) from December 1999 to March 2001. We have assumed that the emissions from December 1999 to March 2000 were identical to the optimized emissions from December 2000 to March 2001. The resulting CO distribution was then compared with the MOPITT data. The RMS of the differences (RMSD) between the modeled CO and the observations at 700, 500 and 350 hPa respectively are on average reduced by a factor of 2–3 from December to March, by a factor of 2 during spring and fall and by a factor of 1.5 from June to August. The a posteriori RMSD is less than 11 ppbv at 700 hPa.

[18] Using regional top-down approaches and in situ CO observations from the NASA/TRACE-P (TRAnsport and Chemical Evolution over the Pacific) campaign, the estimates for the Chinese emissions due to fossil fuel and biofuel use range from 145 Tg/yr [*Allen et al.*, 2004] to 165 Tg/yr [*Palmer et al.*, 2003]. These values are lower than our estimate of the East Asian source (186Tg/yr) and *Arellano et al.* [2004] result (196–214 Tg/yr), both based on the MOPITT data. This could be related to a positive bias in the MOPITT CO retrievals (10–20 ppbv at 700 hPa) reported by *Emmons et al.* [2004].

[19] Our a posteriori CO emissions have been tested by comparing the a priori and a posteriori modeled CO distributions with CO mixing ratio profiles measured during the TRACE-P and the TOPSE (Tropospheric Ozone Production about the Spring Equinox, North America, spring 2000) campaigns. The a priori and a posteriori RMSD between the observed and the modeled CO profiles below 2 km are 81 and 20 ppbv for TRACE-P and 29 and 17 ppbv for TOPSE. The underestimation of CO in the lower layers of the model with the a priori emissions during spring at the mid and high northern latitudes is corrected when using the a posteriori emissions. The model performance is also improved at 26 of the 31 CMDL stations used for the validation. Figure 3 shows an example of this improvement at two stations.

[20] To evaluate the uncertainty on the emissions a posteriori estimates, we have added up the anthropogenic sources over all 15 regions considered earlier. The inversion technique has also been modified to be non-sequential, as in *Pétron et al.* [2002]. The a priori monthly emissions (xb_net) were taken as constant annual averages derived from a previous sequential inversion (xa2) and the attached variances (sigb_ag^2) were set (arbitrarily) as 100 (Tg/yr)^2 plus twice the square differences between the estimates xa2 and xb_net. Table 2 shows the resulting a posteriori regional sources and their uncertainties. Only in South America, for which the results stay very close to the prior xb_net, is this inversion unable to retrieve a seasonal cycle and amplitude close to the xa2 inventory. For the other regions, the optimized total fluxes are quite close to the xa2 and xa3 estimates regardless of the statistics on the a priori emissions and on the observations.

[21] Part of the differences between the sources estimates obtained in the inversion of the MOPITT retrievals can be related to differences in the inversion techniques: time-dependent versus time-independent inversion. Other differences in the optimization procedures are: 1) the number and specification of the regions, 2) the errors assigned to the emissions and to the observations, 3) the selection, filtering and binning of the observations and 4) the CTM (transport, OH fields). To conclude, further comparison studies are needed to evaluate and improve the inversion techniques including the adequate characterization of the model and observations random errors and biases.

[22] An extended summary of this study can also be found at the URL: http://acd.ucar.edu/~boris/inversion.htm.

[23] **Acknowledgments.** The authors acknowledge Peter Hess and David Baker and two anonymous reviewers for their useful and constructive comments. The authors would like to thank the MOPITT groups at the University of Toronto and at NCAR, P. Novelli, G. Sachse and D. Blake for providing CO data-sets from respectively MOPITT, the TRACE-P and the TOPSE campaigns. Work of G. P., B. K., V. Y. and J. F. L was in part supported by the EOS Interdisciplinary Science Program (EOS/IDS) via grant proposal "A Study of Tropospheric Budgets of CO and CH_4 Using Data Assimilation". Work of C. G. and G. P. was in part supported by the European Commission under the POET project (contract EVK2-1999-00011), by the Program Gestion et Impacts du Changement Climatique of the French Ministry of Environment and by the French National Program for Atmospheric Chemistry (PNCA). The National Center for Atmospheric Research is sponsored by the National Science Foundation.

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D. P. Edwards, L. Emmons, J. Gille, B. Khattatov, J.-F. Lamarque, G. Pétron, and V. Yudin, Atmospheric Chemistry Division, National Center for Atmospheric Research, P. O. Box 3000, Boulder, CO 80307, USA. (gap@ucar.edu)

C. Granier, Service d'Aéronomie, Université Pierre et Marie Curie, 4 place Jussieu, F-75252 Paris Cedex 05, France.