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**MOPITT-based
estimation of the CO
emissions in Africa**

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African CO emissions between years 2000 and 2006 as estimated from MOPITT observations

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Abstract

The space-time variations of the carbon budget at the Earth surface are highly variable and quantifying them represents a major scientific challenge. One strategy consists in inferring the carbon surface fluxes from the atmospheric concentrations. A variational scheme for the hydrocarbon oxidation chain, that includes CO and CH₄, is presented here with a focus on the African continent. The multi-tracer system has been built as an extension of a system initially developed for CO₂ and includes a new simplified non-linear chemistry module. Individual in situ measurements of methyl-chloroform and individual retrievals of CO concentrations from the Measurements Of Pollution In The Troposphere (MOPITT) space-born instrument have been processed by the new system for the period 2000–2006 to infer the time series of CO emissions at the resolution of 2.5°×3.75° (latitude, longitude). It is shown that the analysed concentrations improve the fit to five independent surface measurements located in or near Africa by up to 28% compared to standard inventories, which confirms that quantitative information about CO emissions can be obtained from MOPITT data. In practice, the inversion reduces the amplitude and the interannual variability of the seasonal cycle in the northern part of Africa, with a longer burning season. In the southern part, the inversion mainly shifts the emission peak by one month later in the season, consistent with previously-published inversion results.

1 Introduction

The prevailing climatic and economical conditions in Africa have hampered the observation of its carbon budget by conventional networks until now. The lack of measurements in turn makes the numerical models of the carbon cycle, mostly their soil and vegetation components, highly uncertain when applied to Africa since the models are usually developed and validated for different latitudes. Direct (i.e. concentrations of carbon compounds, Turner, 1993) and indirect (i.e. vegetation activity, Tucker, 1979)

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observations of the carbon cycle have been made from space for several decades. The retrievals of carbon compound concentrations (e.g., Deeter et al., 2003; Chédin et al., 2003) have poor vertical resolution and much higher uncertainties than surface measurements, but they circumvent the local constraints and provide large densities of data. Further, they are particularly suited for Africa because biomass burning is both a prominent component of the carbon budget there and a major signal in the satellite radiances, even for high-peaking channels (Chédin et al., 2005). The exploitation of these remotely-sensed concentrations in terms of carbon surface budget (for CO₂, CO and CH₄) has started only recently but the inversion systems are rapidly gaining in sophistication (Arellano et al., 2004; Pétron et al., 2004; Chevallier et al., 2005; Bergamaschi et al., 2007).

The present study focuses on the African CO budget over a seven-year period (2000–2006) by combining several sources of information within a Bayesian inversion system. The first information sources, which we will call here “a priori”, are version 2 of the Global Fire and Emission Database (GFEDv2) developed by Van der Werf et al. (2006) combined with the Emission Database for Global Atmospheric Research (EDGARv3) fossil fuel inventory of Olivier and Berdowski (2001). EDGARv3 is based on international statistics and GFEDv2 is a high-level product with contributions from different space-born observations of fires and of vegetation and from a numerical model of the terrestrial vegetation. The second information source, which we call “observations” in the inversion system, is the dataset of the CO concentrations retrieved from the MOPITT instrument (Deeter et al., 2003) that has been flying onboard NASA’s EOS Terra satellite since the end of 1999. In addition to these two information types, the inversion system gathers methyl-chloroform (MCF) surface measurements to constrain the concentrations of the hydroxyl radical OH, a global chemistry-transport model and the error statistics associated to each information piece. The code follows the Bayesian variational framework initiated by Chevallier et al. (2005) for CO₂. The method and the data are detailed in the next section. Section 3 presents the results and the last section concludes the study.

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2 Data and Method

2.1 Chemistry-transport model

The CO molecule represents one of the steps in the hydrocarbon oxidation chain with also a direct source from combustion processes. Its chemical consumption by the OH radical makes its average lifetime in the atmosphere about two months. Consequently CO is subjected to long range transport. A new chemistry-transport model has been developed to interpret the concentration measurements in terms of surface emissions. The model has been conceived as a simplification of LMDZ-INCA: LMDZ refers here to version 3 of the general circulation model of the Laboratoire de Météorologie Dynamique (Hourdin et al., 2007) and INCA stands for Interaction with Chemistry and Aerosols (Hauglustaine et al., 2004; Folberth et al., 2006). We will refer to the simplified model as LMDZ-Simplified Atmospheric Chemistry System (LMDZ-SACS). It is run here at horizontal resolution $3.75^\circ \times 2.5^\circ$ (longitude-latitude) and with 19 vertical levels between the surface and the mid-stratosphere (~30 km).

The simplifications of LMDZ-SACS compared to LMDZ-INCA have been introduced to minimize the computational burden in the context of a variational (iterative) inversion system. For the transport part, the simplification consists in using a frozen archive of 3-hourly transport mass fluxes rather than running the full general circulation model LMDZ (Hourdin et al., 2006). The archive has been obtained from a previous simulation of LMDZ for the same dates, guided by the horizontal winds analysed by the European center for Medium-Range Weather Forecasts (ECMWF). The simplification for the chemistry consists in explicitly solving the main reactions of a limited set of four species only: CH₄, CO, formaldehyde (HCHO) and MCF. The reactions considered are:

- the oxidation of CH₄ by OH, with HCHO as a product,
- the oxidation of HCHO by OH, with CO as a product,

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- the oxidation of CO by OH,
- the oxidation of MCF by OH,
- the reaction between CH₄ and excited atomic oxygen (O^{1D}),
- the photolysis of HCHO.

5 Intermediate reactions are considered as very fast compared to the reactions above. Their yields are pre-computed using the full LMDZ-INCA that considers the NMHC/CH₄/O_x/NO_x chemistry as described in Folberth et al. (2006). The surface emissions of CH₄, CO, HCHO and MCF are included in LMDZ-SACS. Biomass burning emissions are emitted at the surface in the model since fire emissions are usually not directly injected in the free troposphere (Labonne et al., 2007; Mazzoni et al., 2007). An additional source of HCHO arises from the degradation of volatile organic compounds (VOC) and is prescribed in the simplified model based on a previous simulation with the full LMDZ-INCA model. OH concentrations and constants of reactions are prescribed in the same way. Comparisons between LMDZ-SACS and LMDZ-INCA indicate that the simplified model differs from the reference by less than 15% for CO individual concentrations with negligible bias (Pison et al., manuscript in preparation).

15 The adjoint code of LMDZ-SACS has been developed for an efficient minimization of the cost function J described in the next section.

2.2 Inversion scheme

20 The inversion system described by Chevallier et al. (2005, 2007) aims at optimizing a series of variables gathered in a state vector \mathbf{x} , based on some observations \mathbf{y} and on some prior guess \mathbf{x}_b . The merging of information exploits the error statistics of each information source, assumed to be Gaussian-distributed and unbiased, within a Bayesian methodology. Technically, the optimal solution (in a statistical sense) is found by iteratively minimizing the following cost function:

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$$J(\mathbf{x}) = (\mathbf{x} - \mathbf{x}_b)^T \mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_b) + (H(\mathbf{x}) - \mathbf{y})^T \mathbf{R}^{-1}(H(\mathbf{x}) - \mathbf{y}) \quad (1)$$

B and **R** are the error covariance matrices of the prior and of the observations, respectively. *H* is the operator that computes the equivalent of **y** from **x**, i.e. the combination of LMDZ-SACS and of the MOPITT averaging kernels. This strategy follows the data assimilation systems that the numerical weather prediction community has been developing for several decades (Le Dimet and Talagrand, 1986).

Consistent with LMDZ-SACS, the vector **x** is made of:

- global 2-D maps of CO emissions at an 8-day and 3.75° × 2.5° (longitude-latitude) resolution over a chosen period of time (here 13 months at a time),
- global 2-D maps of CH₄ emissions at the same resolution,
- global 2-D maps of factors, at the same resolution, to scale at once the surface emissions of HCHO and its 3-D atmospheric production by VOC,
- scaling factors for the 4-D OH concentrations fields every 8 days for four latitude bands (90 S–30 S, 30 S–0, 0–30 N, 30 N–90 N). Note that more regions could be introduced if there were more stations (Bousquet et al., 2005),
- global 2-D maps of factors, at 3.75° × 2.5° resolution, that scale the 3-D concentrations of CO, CH₄ and HCHO at the initial time step of the inversion window.

The eight-day temporal resolution materializes the idea that the prior information likely has large temporal error correlations at the scale of days and small error correlations at the scale of weeks. This subjective guess has been confirmed for CO₂ fluxes by Chevallier et al. (2006) and similar studies should be conducted in the future for the species considered here. Emissions within any of the eight-day periods are here interpolated in time from the control variables.

The observations **y** are the individual MCF measurements and the MOPITT retrievals of CO, sampled per orbit at the 3.75° × 2.5° model resolution. The processed

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datasets cover the globe, and the results outside of Africa will be presented in a future paper rather than in this special issue.

The presence of OH among the optimized variables makes the H operator diverge from linearity and makes the cost function J diverge from quadraticity. In this context, our minimisation strategy relies here on the M1QN3 limited memory quasi-Newton software (Gilbert and Lemaréchal, 1989) applied to the classical preconditioned variables $\mathbf{z} = \mathbf{B}^{-1/2} (\mathbf{x} - \mathbf{x}_b)$.

For technical reasons, the seven-year period considered here is processed in consecutive 13-month chunks with a 1-month overlap from one chunk to the next. The overlap was found large enough not to introduce significant artificial discontinuity. Each chunk involves about 950 000 observations and 1 870 000 variables to optimize. Five iterations are performed that reduce the norm of the gradient of J with respect to the control variables by about 95%.

The Bayesian theory provides the errors of the optimal solution to the inversion problem (e.g., Rodgers, 2000). Such a theoretical estimation is an interesting diagnostic, but poses numerical problems when both the state vector and the observation vector are large. The Monte Carlo method of Chevallier et al. (2007) is used here for this computation. We recall its successive steps:

- run the LMDZ-SACS chemistry-transport model with a climatology of surface emissions to generate a set of pseudo MOPITT and MCF observations at the same location and time than the actual measurements,
- perturb the pseudo-observations consistently with assumed observation error statistics (described later in the section),
- perturb the state vector (that includes the surface flux climatology) consistently with assumed error statistics (described later in the section),
- perform a Bayesian inversion of the surface fluxes using the perturbed pseudo-observations as data and the state vector as the prior field,

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- compare the estimate of the inversion to the flux climatology to get the errors in the estimate.

The method is applied several times with different perturbations each time, in order to compute the posterior error statistics. When using a statistical ensemble of increasing size, the method converges towards the usual Bayesian theoretical formula. In the present study, an ensemble of two one-year global inversions of eight-day surface fluxes is used.

2.3 satellite CO retrievals

The MOPITT instrument has been operated since March 2000 on the polar-orbiting sun-synchronous Terra platform, which crosses the equator at 1030 and 2230 local time approximately. Two to three days worth of MOPITT radiances offer a near global coverage with a $22 \times 22 \text{ km}^2$ horizontal resolution for the nadir fields of view. The official MOPITT CO concentration product (level 2 version 5) is available in the form of profiles of mixing ratios at seven nominal levels between the surface and 150 hPa (Deeter et al., 2003). The retrieval algorithm is based on optimal estimation, like our emission inversion system. It evolved between Phase 1 (March 2000–May 2001) and Phase 2 (from August 2001) of the MOPITT operations due to the failure of four channels since May 2001 (Emmons et al., 2004). An averaging kernel is associated to each profile. It describes how the measurements and the prior concentration profile influence the retrieval at each level and quantifies the retrieval uncertainty. Since the retrieval is based on measurements in the thermal infrared spectral range, the actual vertical resolution of a retrieved profile depends on the thermal contrast between the surface and the atmosphere and on the thermal gradients within the troposphere. Even in the most favourable observation conditions, the measurements provide less than two pieces of information in the vertical with larger information content for tropical daytime scenes (Deeter et al., 2004): the upper levels in the retrieved CO profile are primarily sensitive to upper tropospheric CO and the lower levels in the retrieved profile are primarily

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sensitive to CO between 300 and 800 mbar (Deeter et al., 2003). As a compromise between data volume, closeness to the surface and retrieval noise, only the 700 hPa-level retrievals, together with their respective averaging kernels, are used here. Following the recommendations associated with the product (<http://web.eos.ucar.edu/mopitt/data/>, access: 28 May 2008), data within 25° from the poles, or corresponding to nighttime, or for which the so-called “prior contribution” exceeds 50% have been left out. The data have then been sampled at the resolution of the chemistry-transport model to avoid possibly large correlations for measurement and model errors within a model grid box. This screening keeps between 2500 and 5000 MOPITT spots per day in the inversion system.

The inversion system does not distinguish between the representativeness errors, the errors of the observation operator and the measurement errors. The MOPITT official product includes an estimation of the measurement random error for each individual retrieval. It is about 10% standard deviation for each retrieval, with regional biases of a few particles per billion (ppb) (Emmons et al., 2004). The errors of the observation operator (i.e. LMDZ-SACS) and those of representativeness (i.e. the mismatch between observation and model resolutions) are difficult to quantify. LMDZ-SACS accumulates the errors of the reference model LMDZ-INCA (which are bounded but not quantified) and those of the simplifications (about 15% as mentioned in Sect. 2.1). As a consequence, we arbitrarily defined the variance of the individual observation errors in \mathbf{R} by summing the square of the MOPITT error standard deviations with the variance corresponding to 50% of the measured values (to account for representativeness random errors, model random errors and likely measurement and model biases). Error correlations between different observations were ignored. Doing so, the free chemistry-transport model already fits the observations within their assigned uncertainty. The minimization further improves the fit.

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2.4 MCF measurements

CO is the main species reacting with OH in the troposphere: OH affects both its atmospheric production and its removal. Therefore OH is a key component for the analysis of CO concentrations. However, its high reactivity (inducing a very short lifetime) prevents observations of OH with a sufficient spatio-temporal representativity to deduce its global climatology. Several authors have used MCF measurements to infer OH fields because MCF has rather well known sources and interacts only with OH (e.g., Prinn et al., 1995; Bousquet et al., 2005). The present study innovates because it exploits MCF in synergy with other reactive species rather than as a separate preliminary step: CO concentrations complement MCF to infer the OH concentrations and inferred CO emissions benefit from the full statistical information about MCF without any filter.

A series of 12 stations measuring MCF (CH_3CCl_3) nearly continuously for the 2000–2005 period has been selected from the ALE/GAGE/AGAGE and NOAA/ESRL networks (e.g., Prinn et al., 2000; Montzka et al., 2000). The stations are: Alert (Canada), Barrow (US-AK), Cape Grim (Australia), Cape Kumukahi (US-HI), Park Falls (US-WI), Cape Matalula (American Samoa), Mace Head (Ireland), Mauna Loa (US-HI), Niwot Ridge (USA-CO), Palmer Station (Antartica), Ragged Point (Barabados), South Pole. Errors are defined at each station from Bousquet et al. (2005) and range between 1 and 2 ppt (particles per trillion).

2.5 Prior emissions

Prior emissions are taken from standard databases for biomass burning (GFEDv2, Van der Werf et al. (2006)) and fossil fuel (EDGARv3, Olivier and Berdowski (2001)).

Standard deviations of the eight-day prior emission errors are arbitrarily set to 100% of the maximum value of the emission time series during the year for each grid point, except for MCF where it is set to 1% as its emissions are supposed to be better known than for CO (Bousquet et al., 2005). Figure 1 displays the map of CO prior error standard deviations over Africa for year 2004 as an example, with negligible values

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over the deserts and the oceans and large ones over biomass-burning areas. The prior scale factors are set to 1 with arbitrary error standard deviations of 100% and 10% for HCHO and OH, respectively.

The assigned correlations of the prior errors have been standardised for all variables following what had been done for CO₂ by Chevallier et al. (2007): temporal correlations are neglected and spatial correlation are defined by a short e-folding length (500 km, i.e. slightly coarser than the chemistry-transport model spatial resolution at the Equator) over land and a larger one (1000 km) over the ocean.

The uncertainty about emissions, when aggregated at the scale of regions and months, results both from the grid-point error standard deviation and from the correlations. For instance, in 2004, the assigned uncertainty amounts to about 14 Tg of CO per month for the regions of Africa North of the equator and to about 9 Tg per month in 2004 South of the Equator. Those two domains are referred to as “North Africa” (NAfrica) and “southern Africa” (SAfrica), respectively, in the following.

3 Results

3.1 Expected error reduction

As a preliminary step, the impact of the MOPITT data on the estimation of the CO fluxes is quantified using the Monte Carlo method described Sect. 2.2 and applied to the data of 2004. The map of the error reduction is shown in Fig. 2. As expected from a Bayesian system with rather uniform data coverage (Chevallier et al., 2007), the map is very similar to that of the prior error standard deviations shown in Fig. 1. In biomass burning areas the values are about 20%, reaching up to 45% for some grid points. By construction, those figures are yearly averages and should be larger in biomass burning seasons. Estimating the seasonal cycle of the error reduction would require a larger statistical ensemble of inversions.

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The aggregated posterior uncertainty amounts to about 11 Tg of CO per month of 2004 for NAfrica and 8 Tg per month of 2004 for SAfrica. The error reduction reaches about 20% for NAfrica and 10% for SAfrica.

3.2 CO Concentrations and emissions

5 Figure 3 illustrates the behaviour of the inversion system with some results for the month of July 2003. The MOPITT mean concentrations and the concentrations before and after the inversion are displayed. By construction, the inversion brings the concentrations closer to the observations than the prior, with mean increments of up to 25% of the prior concentrations. The values in NAfrica and South of 20 S are increased, while
10 those between the Equator and 20 S, associated to biomass burning in this season, are decreased. The model does not exactly fit the (uncertain) observations for two reasons. First, the finite prior uncertainty on the emissions pulls the analysis toward the prior values. Second, the chemistry-transport model is used as a hard constraint in the inversion and therefore smoothes out the space-time variations of the concentration fields.
15

Figure 4 presents the results for CO concentrations and emissions over the seven year period and for the two African regions. The series mainly reflect the seasonal cycle of biomass burning with maximum values in boreal winter for NAfrica and late austral winter in SAfrica. The inversion appears to smooth out the concentration time series
20 in the North, with a longer burning season and a smaller interannual variability. In the South, the inversion mainly shifts the emission peak by one month later in the season. The Southern emissions are still smaller than the Northern ones on an average. The annual budgets are given in Table 1. We performed a series of sensitivity studies to evaluate the robustness of the inverted fluxes by varying the OH assigned prior errors (from 10 to 35%), by varying the OH prior field (the alternate prior field had been obtained from a simulation of LMDZ-INCA with different emission inventories), by removing the MCF observations, by varying the CO assigned prior errors (from 100
25 to 50% of the annual maximum emission value), by switching to release number 4

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of LMDZ (we use version 3 here) and by adding offsets to MOPITT data (the offsets amount to a few ppb). In contrast to other parts of the globe (not shown here), the results over Africa appear to be very robust and the range of inverted fluxes lie within the theoretical posterior uncertainty of Sect. 3.1. Among the tests that we performed, varying the OH assigned prior errors from 10 to 35% had the largest impact. The two main inversion features (i.e. smoother series in the North and a shifted burning season in the South) were already noted by Pétron et al. (2004) and by Arellano et al. (2006) who independently processed the April 2000–March 2001 MOPITT data with different inversion systems.

The multi-tracer chemistry LMDZ-SACS couples the species. As a consequence, the CO observations induce increments for all species. HCHO increments over Africa are about a few per cent. Their geographical distribution is similar to the CO increments (not shown), which may indicate that the system is not able to separate the atmospheric production of CO from its surface emissions. CH₄ increments are negligible. OH is mainly constrained by MCF in our system. Its spatial distribution is hardly affected, but the amplitude of the seasonal cycle is reduced by about 2%. The phase of the seasonal cycle remains unchanged.

3.3 Fit to independent surface measurements

In order to validate the inversion, the posterior concentrations have been compared to independent surface flask measurements from the NOAA ESRL measurement network for the 2000–2006 period. We used five measurements stations located in the African continent or nearby: ASC (Ascension Island, 7.92 S, 14.42 W), ASK (Assekrem, Algeria, 23.18 N, 5.42 E), IZO (Tenerife, Canary Islands, 28.30 N, 16.48 W), SEY (Mahe Island, Seychelles, 4.67 S, 55.17 E) and WIS (Sede Boker, Israel, 31.13 N, 34.88 E). The departure statistics before and after the MOPITT inversion appear in Table 2. The analysis significantly improves the fit to the individual observations at all stations with respect to the prior. Over the six-year period, the root mean square departures are reduced by up to 28%, while the bias is also significantly reduced.

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This paper represents a first step in extending the variational inversion system of Chevallier et al. (2005) to reactive gases. Compared to the 2005 paper, a simplified chemistry module for hydrocarbon oxidation chain has been developed, the M1QN3 minimizer has been implemented, and the system has been interfaced to the individual MOPITT data, including their averaging kernels. The MOPITT archive over the 2000–2006 period has been processed and the results analysed for the African continent. The error budget indicates that the MOPITT data should in principle reduce the uncertainty about the African emissions significantly, even at the grid point level. This is confirmed on real data by the comparison of the prior (i.e. before the MOPITT-based inversion) and of the posterior (i.e. after the MOPITT-based inversion) model simulation at five independent stations. It will be interesting to extend the validation to aircraft measurements, like those of the MOZAIC program (Measurements of Ozone, Water Vapour, Carbon Monoxide and Nitrogen Oxides by In-Service Airbus Aircraft, Marengo et al. (1998)). The results for the rest of the globe are still being analysed.

A major advantage of the new system relies on its flexibility to accommodate new observations related to the hydrocarbon oxidation chain: CO, CH₄, HCHO and MCF observations can be analysed in synergy, with the observations about any one of the species helping constraining the other species. For the inversion of CO emissions, the major improvement is expected from the contribution of HCHO retrievals, like those of Global Ozone Monitoring Experiment (GOME) or from the SCanning Imaging Absorption SpectroMeter for Atmospheric CHartography (SCIAMACHY) (e.g., De Smedt et al., 2008). Additionally, introducing other CO retrieval products together with MOPITT in the system, like those from the Infrared Atmospheric Sounding Interferometer (IASI) (Turquety et al., 2004), should reduce the impact of possible biases in individual products.

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Table 1. Annual budget of CO emissions, in Tg of CO per month, for the two African regions, in the prior (FG) and in the posterior (AN) inventories. The top row corresponds to Phase 1 of the MOPITT instrument. The uncertainty figures for AN make use of the prior uncertainty and of the error reduction obtained for year 2004 by the randomization method.

Year	FG NAfrica	AN NAfrica	FG SAfrica	AN SAfrica
April 2000–March 2001	177±73	188±58	96±36	135±32
2002	151±48	121±38	96±33	100±29
2003	137±52	124±41	98±35	107±31
2004	142±45	127±36	95±31	104±29
2005	149±62	130±49	103±35	121±31
2006	135±52	129±41	90±29	103±26

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Table 2. Statistics of the model-minus-observation departures, in ppb, at five stations of the NOAA ESRL network. The model uses either the prior (FG) and the posterior (AN) CO emissions. The flask data have not been used in the inversion system.

Station	FG Bias	AN Bias	FG RMS	AN RMS
ASC	5	1	16	12
ASK	20	13	24	17
IZO	23	16	27	22
SEY	8	4	14	13
WIS	29	20	36	30

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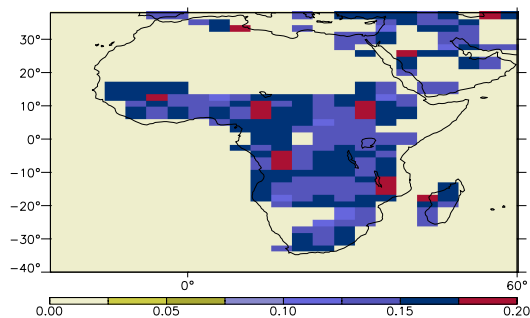


Fig. 1. Prior error standard deviations, in g.m^{-2} per day, for the eight-day mean grid point CO surface emissions in 2004.

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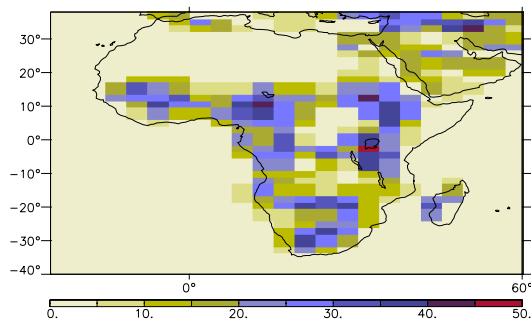


Fig. 2. Error reduction, in %, of the eight-day mean grid point CO surface emissions. The error reduction is defined as $(1 - \sigma_a/\sigma_b) \times 100$, with σ_b the prior error standard deviation shown in Fig. 1 and σ_a the posterior error standard deviation.

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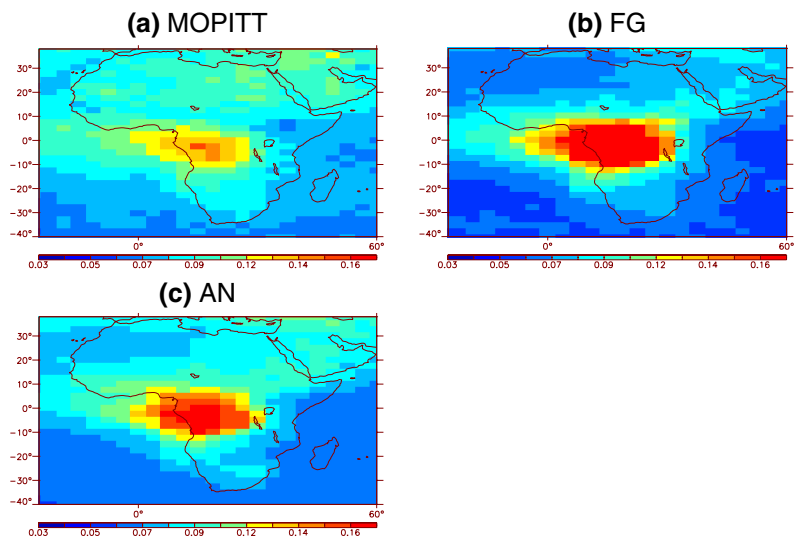


Fig. 3. Mean CO concentrations, in particles per million, for July 2003 in **(a)** the MOPITT observations, **(b)** the prior simulation (FG) and **(c)** the posterior simulation (AN).

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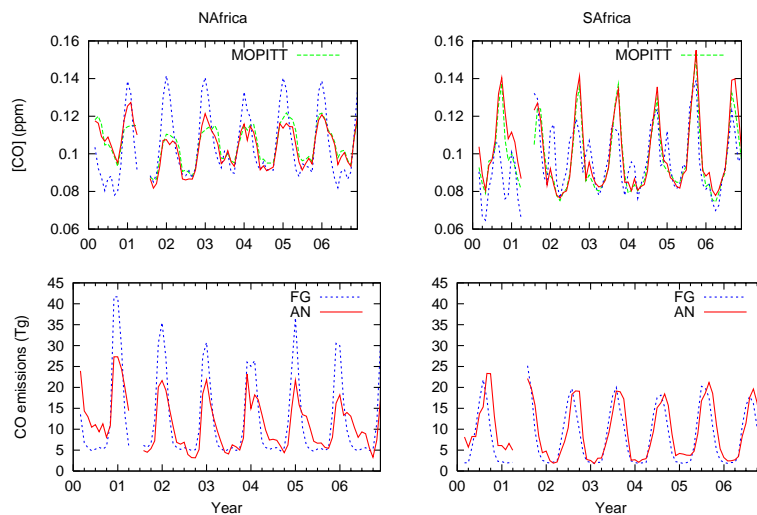


Fig. 4. Time series of the CO concentrations (top row) and of the CO emissions (bottom row) for regions NAfrica (left column) and SAfrica (right column) in the prior simulation (blue curve) and in the posterior simulation (red curve). CO concentrations from MOPITT (in green) are reported in the concentration figures (top row).

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