

## **An Inventory of Gases and Particles Emissions for the 1860-2005 period**

Claire Granier

NOAA Earth Systems Science Laboratory, Boulder, CO, 80305 USA  
University of Colorado, Cooperative Institute for Research of the Environmental  
Sciences, Boulder, CO, 80305 USA  
UPMC, University Paris 06, and CNRS Service d'Aéronomie, UMR 7620, 75005  
Paris, France

[claire.granier@noaa.gov](mailto:claire.granier@noaa.gov)

Cathy Liousse, Bruno Guillaume

Laboratoire d'Aérodologie, 31400 Toulouse, France

[lioc@aero.obs-mip.fr](mailto:lioc@aero.obs-mip.fr), [guib@aero.obs-mip.fr](mailto:guib@aero.obs-mip.fr)

Aude Mieville

UPMC, University Paris 06, and CNRS Service d'Aéronomie, UMR 7620, 75005  
Paris, France

[Aude.mieville@aero.jussieu.fr](mailto:Aude.mieville@aero.jussieu.fr)

Jean-Marie Grégoire

Joint Research Center, Ispra, I-21020, Italy

[jean-marie.gregoire@jrc.it](mailto:jean-marie.gregoire@jrc.it)

Florent Mouillot

Centre d'Ecologie Fonctionnelle et évolutive, Montpellier, France

[mouillot@cefe.cnrs.fr](mailto:mouillot@cefe.cnrs.fr)

### **ABSTRACT**

Since the beginning of the 20th century, large changes in surface emissions of atmospheric chemical species have occurred. Previous inventories have been developed during the past few years which describe the changes in emissions, but they have focused either on gaseous species or on particles and their precursors. We have developed a new inventory of emissions, which covers the 1860-2005 period, and provides consistent emissions distributions of both gaseous species and aerosols. The species considered in our inventory are carbon dioxide (CO<sub>2</sub>), tropospheric ozone precursors, i.e. carbon monoxide (CO), nitrogen oxides (NO<sub>x</sub>) and volatile organic compounds (VOCs), and aerosols and their precursors, i.e. sulfur dioxide (SO<sub>2</sub>), and black and organic carbon. Our inventory provides emissions from both anthropogenic and biomass burning emissions at the global scale, at a 1x1 degree spatial resolution. The methodologies we have used to construct the inventory are discussed, as well as the statistical data and emissions factors that have been used. The distribution of emissions provided by the inventory are compared with other available inventories at the global or the regional scale.

## INTRODUCTION

Since the end of the 19<sup>th</sup> century, the development of industrial activities, of intensive agricultural practices and large scale deforestation have led to the emission of large amounts of chemical species into the atmosphere. The increase in the concentrations of greenhouse gases since the second part of the 19<sup>th</sup> century resulted in a climate forcing of  $1.6 \text{ Wm}^{-2}$  [0.6 to  $2.4 \text{ Wm}^{-2}$ ] (IPCC, 2007). The increase in the emissions of ozone precursors (methane, carbon monoxide, nitrogen oxides and volatile organic compounds) led to a significant increase in the concentrations of ozone, which resulted in a additional radiative forcing of  $0.35 \text{ Wm}^{-2}$  [0.25 à  $0.65 \text{ Wm}^{-2}$ ] (IPCC, 2007). The ozone forcing is however still uncertain: long-term changes in ozone and its precursors are still not well quantified, the main uncertainties being due to a poor quantification of the evolution of the emissions of ozone precursors. The climatic role of atmospheric particles from both natural and anthropogenic origin is still not well known: work conducted during the past few years has contributed to more accurate estimations of the aerosol climatic forcing since the preindustrial period. It is currently estimated to be  $-0.5 \text{ Wm}^{-2}$  [-0.9 to  $-0.1 \text{ Wm}^{-2}$ ]. Again, one of the main uncertainty is due to the lack of knowledge on surface emissions of particles and of their long-term changes.

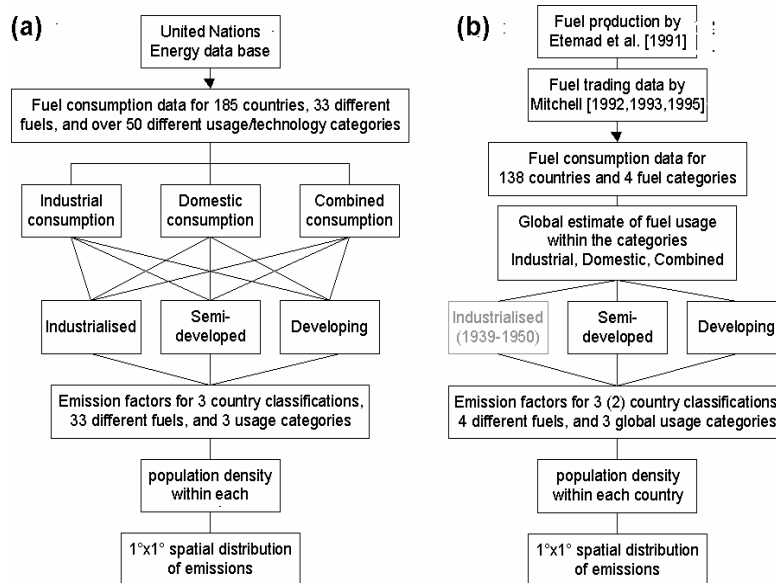
The main objective of the work discussed in this paper is to develop a methodology for the calculation of a consistent emission inventory of emissions of gaseous and particulate atmospheric compounds over the 1860-2005 period. Inventories for a similar period have been developed by other groups, but they take into account either gases or particles. The inventory we have developed considers gases and aerosols in a consistent way.

The paper focuses on the emissions resulting from anthropogenic activities and biomass burning for the following species: carbon dioxide (CO<sub>2</sub>), carbon monoxide (CO), nitrogen oxides (NO<sub>x</sub>), sulfur dioxide (SO<sub>2</sub>), black carbon (BC) and organic carbon (OC).

## ANTHROPOGENIC EMISSIONS

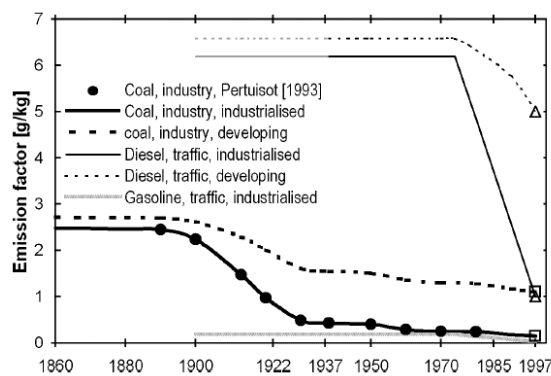
The method we have used is based on the algorithm carbonaceous aerosol emissions by Junker et Lioussé (2008). We use the statistical data provided by the United Nations Energy Statistics Database (UNSTAT : <http://unstats.un.org/unsd/>) together with estimations of emission factors.

A schematic view of the algorithm is shown in Figure 1. Since data for the 1860-1950 period are sparse, a simplified algorithm was used for this period (Figure 1.b). Three emission sectors are considered, « industrial », « domestic » and « combined », the « combined » category representing mostly traffic emissions.



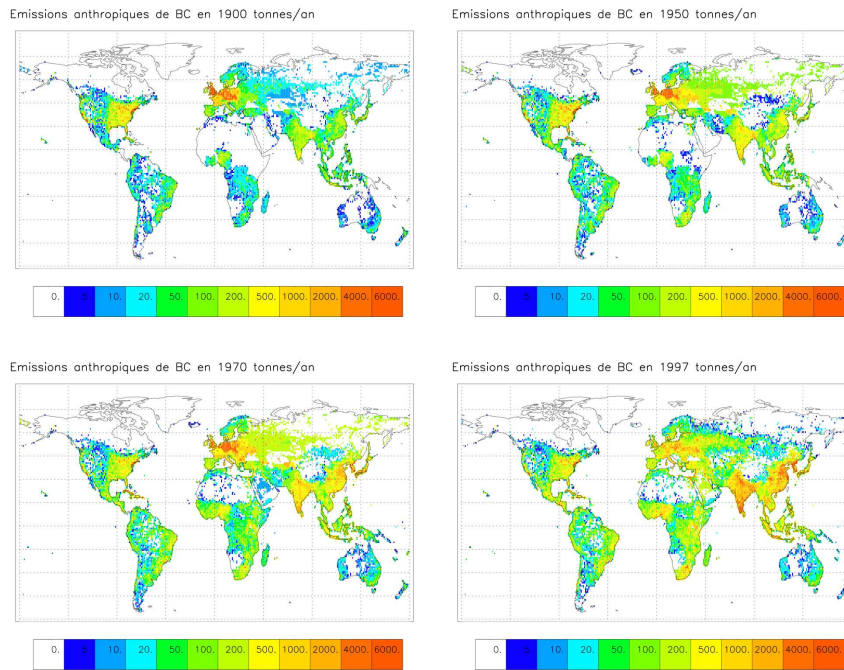
**Figure 1** Algorithms used to calculate anthropogenic emissions for the (a) 1950-1997 and (b) 1860-1949 periods.

We have considered two scenarios for the evolution of the emissions factors: a scenario where emission factors are kept constant during the full period, and one scenario where the impact of changes in technologies on the emission factors are taken into account. Figure 2 shows the evolution of emission factors for coal, diesel and fuel use in developed and developing countries for black carbon. Similar evolution of the emission factors for the other species considered in the inventory were also implemented.

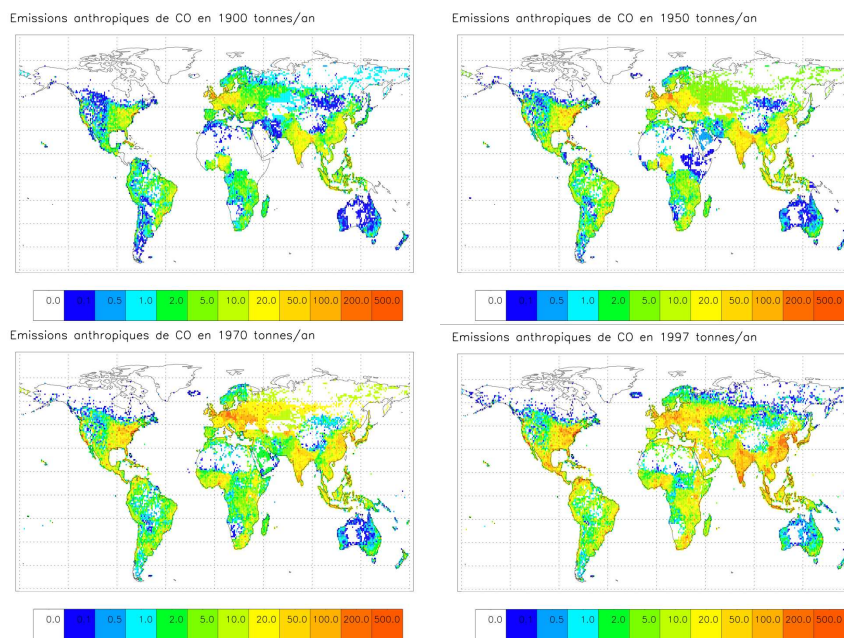


**Figure 2** : BC Emission factors (g/kg) for coal and fuel use. Comparison with values from Pertuisot [1993], values from Cooke et al. (1999).

Figure 3 and 4 show the emissions of BC and CO in 1900, 1950, 1970 and 1997. Large differences are obtained in industrialized area and in developing countries such as India and China, mostly over the past two decades. BC and CO emissions have decreased since the 1970s in Europe and North America, as a result of the implementation of new technologies.



**Figure 3 :** BC anthropogenic emissions in 1900, 1950, 1970 et 1997 : emissions are given in Tg C/year.



**Figure 4 :** CO anthropogenic emissions in 1900, 1950, 1970 et 1997 : emissions are given in Tg C/year.

## BIOMASS BURNING EMISSIONS

Biomass burning emissions since 1900 are determined in two steps: emissions were first quantified using satellite data for the past decade. Emissions for the 1900-1997 period are then determined using historical data.

### *Emissions for the 1997-2005 period*

Two fire satellite products are used for estimating biomass burning emissions :

- GBA2000 (Global Burnt Area 2000, Tansey et al., 2004) : this product provides burnt area data for year 2000 on a global scale.
- ATSR WFA, World Fire Atlas (Arino et al., 1999) : this product provides daily fire spots data for the 1995-2007 period, derived from the ATSR instruments (Arino et al., 2001)

A vegetation map is used to determine which type of vegetation is burning, when a fire is detected at a given location of the globe. The map used in this study is GLC2000 (Global Land Cover 2000, Bartholomé and Belward, 2005). CO<sub>2</sub> emissions related to each GLC2000 vegetation type are first estimated from GBA2000 burnt areas. These quantities are then used to calibrate ATSR fire pixels, in order to derive the spatial and temporal distribution of the emissions for the 1997-2005 period.

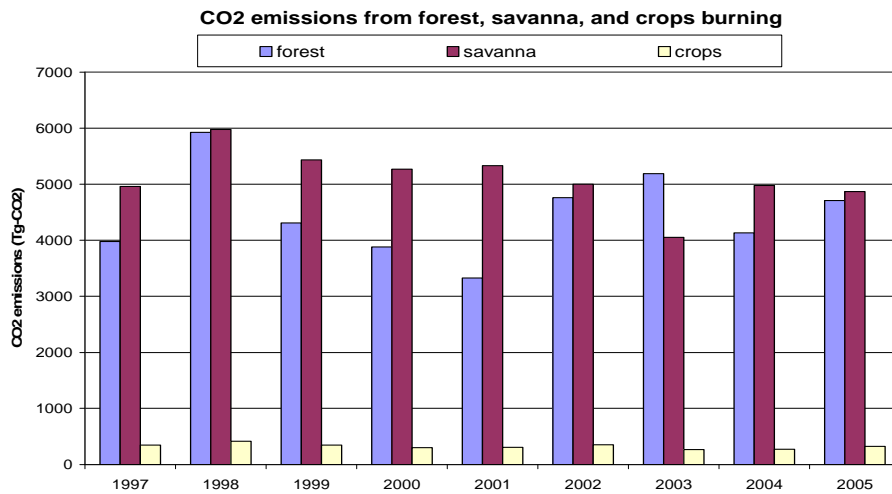
For each vegetation class provided by the vegetation map, the emissions of species X is calculated as the product of burnt area, biomass density, burning efficiency and CO<sub>2</sub> emission factor (Seiler and Crutzen, 1980) :

$$E = BA \times BD \times BE \times EF(X)$$

where BA represents the burnt area (m<sup>2</sup>), BD is the biomass density (g/m<sup>2</sup>), BE represents the burning efficiency, and EF(X) is the emission factor for species X (in kg(X)/kg dry matter) for each vegetation class.

Since the GBA-2000 product is only available for year 2000, fire pixel observations are used for the rest of the period. Fire pixels give precious information on the location of fires, but it is not straightforward to estimate the amount of biomass burnt from a fire pixel. The emissions derived previously for 2000 using GBA2000 are used to calibrate the amount of biomass burned in each fire pixel. Emissions factors are taken from the values compiled by Andreae and Merlet (2001), and for BC and OC from Lioussé et al. (2004), and Michel et al. (2005).

Figure 5 displays the CO<sub>2</sub> emissions from forest fires, from savanna burning and agricultural waste burning from 1997 to 2005. A large year-to-year variation is obtained, which reflects the variability in meteorological conditions as well as in deforestation.

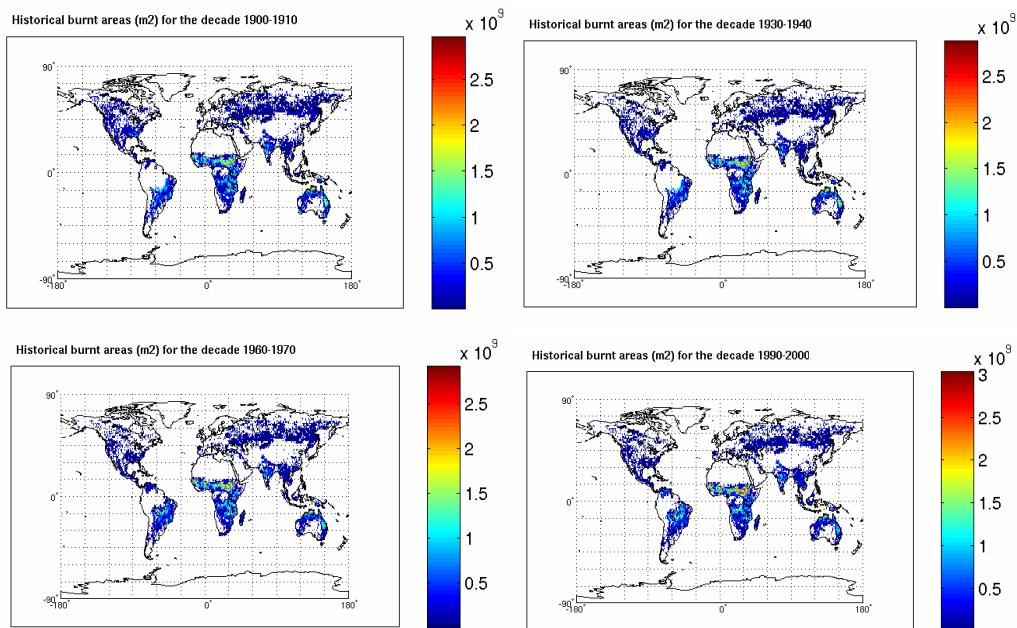


**Figure 5:** CO2 annual emission estimations from fires in forests, savannas, and cultivated areas, for the 1997-2005 period.

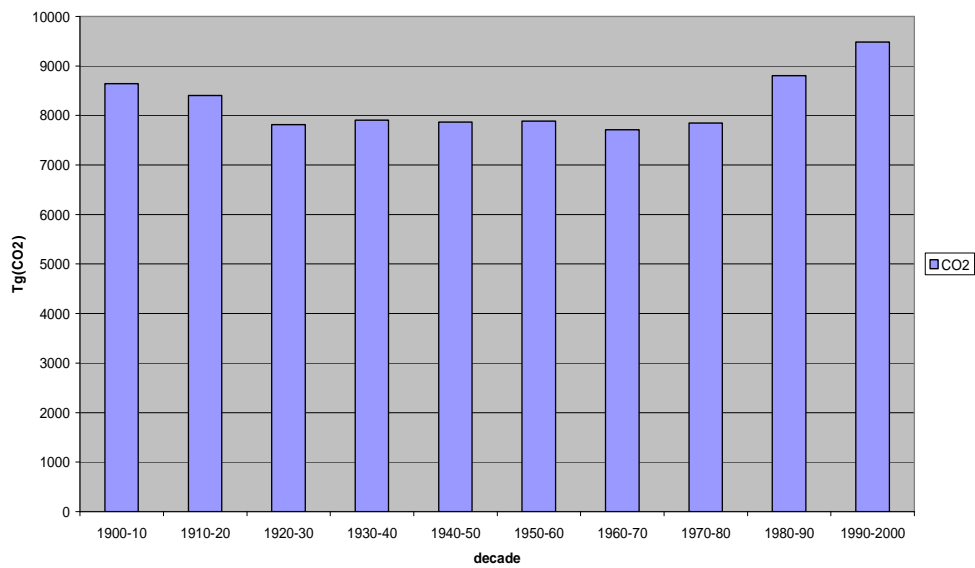
#### *Emissions for the 1900-1997 period*

Historical emissions are estimated from historical burnt areas data determined by Mouillot and Field (2005). This dataset provides the 1x1 degree distribution of burned areas for each decade since 1900. We used the GLC2000 vegetation map for calculating the emissions resulting from biomass burning. It should be noted that this current vegetation map might not be representative of the vegetation distribution for the beginning of the 20<sup>th</sup> century. However, evolutive vegetation maps are not readily available. In order to quantify the emissions for the past decades of the 20<sup>th</sup> century, it is assumed that the emissions for the 1990-2000 decade in the historical inventory provides the same emissions as the one determined using satellite observations during the 1997-2005 period.

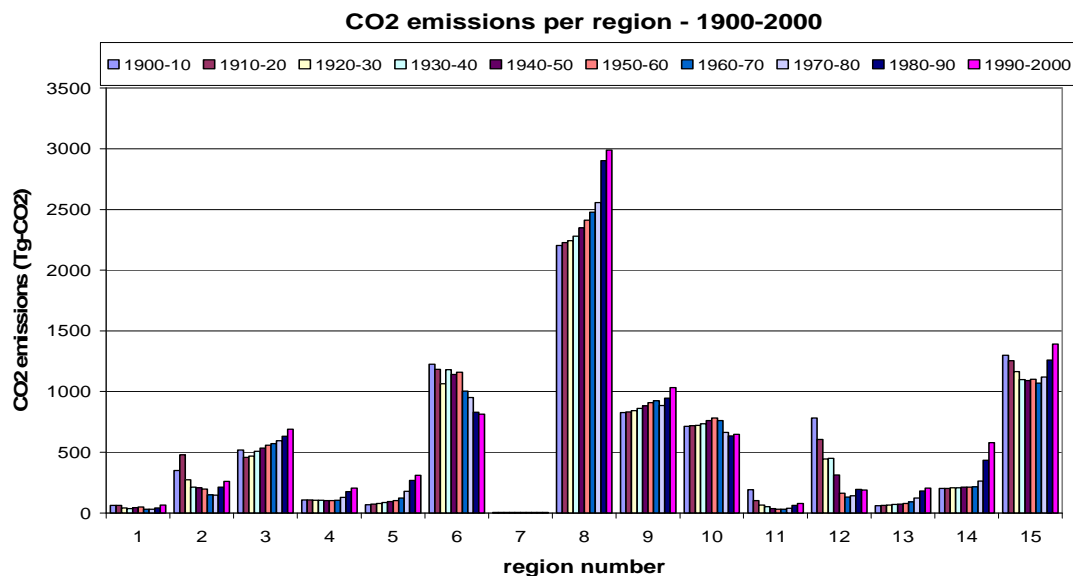
The evolution of burnt areas for selected decades from 1900 to 2000 is shown in Figure 6. Global CO2 emissions for each decade are shown in figure 7, and Figure 8 displays the emissions for different areas of the world. Biomass burning emissions decrease from the beginning of the century to the 1920s. The emissions remain stable then until the 1970s. Since the 1980s, emissions have increased at a very fast rate, as a result of intense deforestation in the tropics. In boreal regions however, burnt surfaces and emissions have decreased during the last century, as a result of fire suppression policy, and fire fighting systems.



**Figure 6:** Burned area distributions (in  $m^2$ ) de (Mouillot et Field, 2005), for the 1900-1910, 1930-1940, 1960-1970, 1990-2000 decades.



**Figure 7 :** Evolution of global CO<sub>2</sub> emissions between 1900 and 2000.



**Figure 8** : Emissions of CO2 for different areas of the world.

## CONCLUSIONS

A global inventory of emissions resulting from anthropogenic activities and biomass burning for the 1860-2005 period has been developed. This inventory takes into account in a consistent way the emissions of gases and aerosols, i.e. carbon dioxide, carbon monoxide, nitrogen oxides, black and organic carbon. Comparisons with existing inventories, which take into account either only gaseous species or only particulate species are under way. First results will be shown in the oral presentation.

These emissions are currently been used as boundary conditions in the MOZART-4 chemistry-transport model (Horowitz et al., 2003; Emmons et al., personal communication). In the oral presentation, first results of the model simulation will be discussed.

## REFERENCES

Andreae, M. O. and P. Merlet, Emission of trace gases and aerosols from biomass burning. *Global Biogeochemical Cycles*, 15: 955-966, 2000GB001382, 2001.

Arino, O. and J.M. Rosaz, 1997 and 1998 world ATSR fire atlas using ERS-2 ATSR-2 data., *Proceedings of the Joint Fire Science Conference*, Boise, Idaho, 15–17 June 1999, edited by Neuenschwander, L. F., Ryan, K. C., & Golberg, G. E. (Eds), Boise: University of Idaho and the International Association of Wildland Fire, pp. 177–182, 1999.



Arino, O., Simon, M., Piccolini, I. and J.M. Rosaz, The ERS-2 ATSR-2 World Fire Atlas and the ERS-2 ATSR-2 World Burnt Surface Atlas projects. *Proceedings of the 8th ISPRS conference on Physical Measurement and Signatures in Remote Sensing*, Aussois, 8-12 January 2001.

Bartholomé, E. and Belward, A.S., GLC2000 : a new approach to global land cover mapping from Earth observation data. *International Journal of Remote Sensing*, vol. 26(9), 1959-1977, 2005.

Cooke, W. F., C. Liousse, H. Cachier and J. Feichter, *J. Geophys. Res.* 104 (D18), 22137-22162, 1999.

Horowitz, L., S. Walters, D.L. Mauzerall, L.K. Emmons, P.J. Rasch, C. Granier, X. Tie, J-F. Lamarque, M.G. Schultz, G.S. Tyndall, J.J. Orlando, and G.P. Brasseur, A global simulation of tropospheric ozone and related tracers: Description and evaluation of MOZART, version 2, *J. Geophys. Res.*, 108(D24), 4784, doi:10.1029/2002JD002853, 2003.

IPCC, 2007: Climate Change 2007: The Physical Science Basis, Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M. Tignor, and H.L. Miller eds, Cambridge University Press, United Kingdom and New York, NY, USA, 2007.

Junker, C., and Liousse, C.: A global emission inventory of carbonaceous aerosol from historic records of fossil fuel and biofuel consumption for the period 1860-1997, *Atmos. Chem. Phys.*, 8, 1-13, 2008.

Liousse, C., M. O. Andreae, P. Artaxo, P. Barbose, H. Cachier, J.-M. Grégoire, P. Hobbs, D. Lavoué, F. Mouillot, J. Penner, M. Scholes and M. Schultz, Deriving Global Quantitative Estimates for Spatial and Temporal Distributions of Biomass Burning Emissions, in *Emissions of Atmospheric Trace Compounds*, C. Granier, P. Artaxo, and C. Reeves (Eds.), 560 pp, 2004.

Michel, C., C. Liousse, J.-M. Grégoire, K. Tansey, G.R. Carmichael and J.-H. Woo, Biomass burning emissions inventory from burnt area data given by SPOT-Vegetation satellite for TRACE-P and ACE-Asia case studies, *J. of Geophys. Res.*, 110, D09304, doi:10.1029/2004JD005461, 2005.

Mouillot, F., and C. B. Field, Fire history and the global carbon budget: A 1° × 1° fire history reconstruction for the 20th century, *Global Change Biology*, 11, 398-420, 2005.

Pertuisot, M. H.: Transfert du carbone atmosphérique dans les neiges et les pluies, dissertation Université Paris VII, UFR de Chimie, 1993.

Seiler, W. and P.J. Crutzen, Estimates of gross and net fluxes of carbon between the biosphere and the atmosphere from biomass burning, *Climatic Change*, 2, 207-247, 1980.

Tansey, K., J.-M. Grégoire, E. Binaghi, L. Boschetti, P.A. Brivio, D. Ershov, S. Flasse, R. Fraser, D. Graetz, M. Maggi, P. Peduzzi, J. Pereira, J. Silva, A. Sousa, and D. Stroppiana, A global inventory of burned areas at 1 km resolution for the year 2000 derived from SPOT VEGETATION data, *Climatic Change*, **67**, 345-377, doi:10.1007/s10584-004-2800-3, 2004.

**KEY WORDS**

Anthropogenic emissions

Biomass burning

Historical emissions

Ozone precursors

Black carbon

Global scale