



**HAL**  
open science

## Rice straw burning in Southeast Asia as a source of CO and COS to the atmosphere

Ba Cuong Nguyen, Nikolaos Mihalopoulos, Jean-Philippe Putaud, Ba Nguyen

► **To cite this version:**

Ba Cuong Nguyen, Nikolaos Mihalopoulos, Jean-Philippe Putaud, Ba Nguyen. Rice straw burning in Southeast Asia as a source of CO and COS to the atmosphere. *Journal of Geophysical Research*, 1994, 99 (D8), pp.16435. 10.1029/93JD03521 . hal-03612654

**HAL Id: hal-03612654**

**<https://hal.science/hal-03612654v1>**

Submitted on 18 Mar 2022

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

## Rice straw burning in Southeast Asia as a source of CO and COS to the atmosphere

Ba Cuong Nguyen, Nikolaos Mihalopoulos,<sup>1</sup> and Jean-Philippe Putaud

Centre des Faibles Radioactivités, Laboratoire mixte CNRS-CEA, Gif-sur-Yvette, France

**Abstract.** Atmospheric samples were collected during rice straw burning at four different locations in Viet Nam during the dry and wet seasons (March 1992, February 1993, and August 1992, respectively). The samples were analyzed for CO<sub>2</sub>, CO, and COS. The emission ratios relative to CO<sub>2</sub> for rice straw burning during the dry season were comparable to those observed on samples collected during burning of savanna in Africa or forest in the United States. On the contrary, during the wet season the emission ratios for CO and COS relative to CO<sub>2</sub> were 2 to 10 times higher. With these emission ratios and estimates of rice production from southeastern Asia, we estimated that burning of rice straw emits annually about 2.1 Tmol of CO (25 Tg C) and 0.6 Gmol of COS (0.02 Tg S) to the atmosphere. Taking into account these new results, CO and COS fluxes from biomass burning could be reevaluated by 5–21% and 10–67%, respectively, with regard to previous estimates of these gas emissions from all biomass burning activities.

### Introduction

As a substantial source of trace gases and aerosol particles to the atmosphere, biomass burning impacts both the troposphere's chemistry and its radiation balance [Andreae *et al.*, 1988; Crutzen and Andreae, 1990; Houghton, 1990; Lashof and Ahuja, 1990; Penner *et al.*, 1991]. Widespread, particularly in developing countries, burning of biomass is practiced as a means to convert forests to cultivated lands, to clean lands and to provide fuel for domestic heating. Also, many countries burn off agricultural wastes both as a means of disposal and as a mechanism of insect control. The single most important source of burned agricultural wastes is straw from rice, which makes up 31% of the agricultural waste in the developing world [Crutzen and Andreae, 1990].

One of the sulfur species emitted by biomass burning is carbonyl sulfide (COS), which is the predominant precursor of sulfate aerosols in the stratosphere during volcanic quiescent periods [Servant, 1989]. Emissions of this compound could therefore have a negative radiative effect. Up to now, very limited data regarding COS emission ratios from biomass burning have been reported. COS emission ratios have only been measured from forest fires in Colorado ( $n = 7$ ) and Brazil ( $n = 22$ ) by Crutzen *et al.* [1979, 1985], from African equatorial rainforest ( $n = 2$ ) by Bingemer *et al.* [1992], and from African savanna ( $n = 17$ ) by Nguyen *et al.* [1993a].

However, up to now, all data regarding trace gases and sulfur compound emissions from burning are originated from experiments performed on tropical forests and savanna fires. Measurements from other important biomass burning sources, such as agricultural waste which could account for about 17%–28% of the total carbon released [Crutzen and Andreae, 1990;

Chin and Davis, 1993], are not available. In this paper we present the first measurements of CO<sub>2</sub>, CO, and COS emissions from rice straw burning conducted in Viet Nam, situated in Southeast Asia. The most important crop in this country is rice, which is cultivated using both irrigated paddy and dry field culture on 75% of the total cropped land.

### Experimental Procedure

Field experiments were conducted during March and August 1992 as well as February 1993 at four different locations near Ho Chi Minh City, Viet Nam (Figure 1) situated at 30 km, 50 km, 70 km, and 300 km to the north at Da Lat. Experiments were conducted during the day between 1000 and 1700 LT under full sunlight and temperatures between 25°C and 30°C. In the dry season, samples were taken during large-scale field burning of rice straws for land cleaning. In the wet season the samples were taken during small-scale field burning of already cut rice straws, previously spread on the ground and exposed to the Sun for few minutes before burning. Gas sampling was performed downwind, about 20–60 cm above the top of the flames of rice straw burning. Thus it is quite unlikely that unburnt gases or intermediate combustion products significantly affect the composition of the samples. Moreover, to obtain a more realistic picture of the "overall average" of fires, 2–4 samples were collected with an interval of few minutes during each experiment. The samples were stored in 2-L canisters of electropolished stainless steel, each fitted with a metal bellows valve. These canisters were previously conditioned by heating under vacuum. Tests conducted in our laboratory showed that no significant concentration change within the interval time of less than 1 month occurred between the sample collection and sample analysis [Mihalopoulos, 1989; Bonsang *et al.*, 1991].

COS was cryogenically trapped at -90°C on conditioned 60- to 80-mesh Tenax GC loaded into a FEP/Teflon U column. The quantitative analysis of the samples was performed with a Varian 3400 gas chromatograph, equipped with a double-flame photometric detector operating in the sulfur mode and calibrated by injection of small volumes (10–30  $\mu$ l) of a standard mixing of COS in helium at 85 ppm (Union Carbide).

<sup>1</sup>Now at Department of Chemistry, University of Crete, Iraklion, Greece.

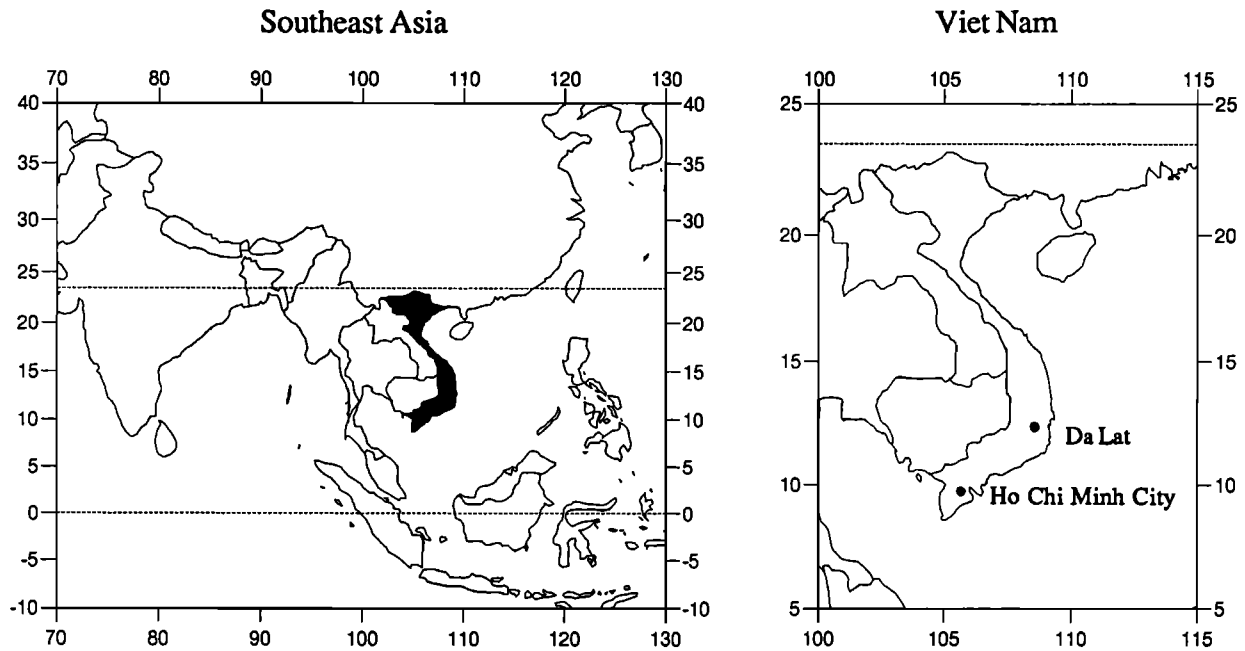


Figure 1. Location of the experimental stations in Viet Nam.

The overall precision of the method was about 7%. Further details of the analytical system have been described by *Belviso et al.* [1987].

CO and CO<sub>2</sub> were analyzed with a gas chromatograph (Intersmat), equipped with a 1/8", 2-m long 60- to 80-mesh carbosphere S-packed column held at 105°C, subsequently converted to CH<sub>4</sub> using a methanizer and quantified by a flame ionization detector (FID). Calibration for CO and CO<sub>2</sub> was performed using a diluted standard (Scotty). Precision for CO<sub>2</sub> and CO was better than 2%. Further details of the analytical system have been described by *Bonsang et al.* [1991].

Results and Discussion

CO and COS Emission Ratio Estimates From Fire Samples

COS concentrations reached values as high as 25 parts per million in volume (ppmv), exceeding the atmospheric background (500 parts per trillion in volume (pptv) [*Khalil and Rasmussen, 1984*]) by a factor of 5 x 10<sup>4</sup>. CO<sub>2</sub> concentrations varied from 1.2 x 10<sup>3</sup> to 2.6 x 10<sup>5</sup> ppmv and CO from 1.4 x 10<sup>2</sup> to 9.6 x 10<sup>4</sup> ppmv. Concentrations of CO<sub>2</sub> and CO in the fire samples also far exceed their background values

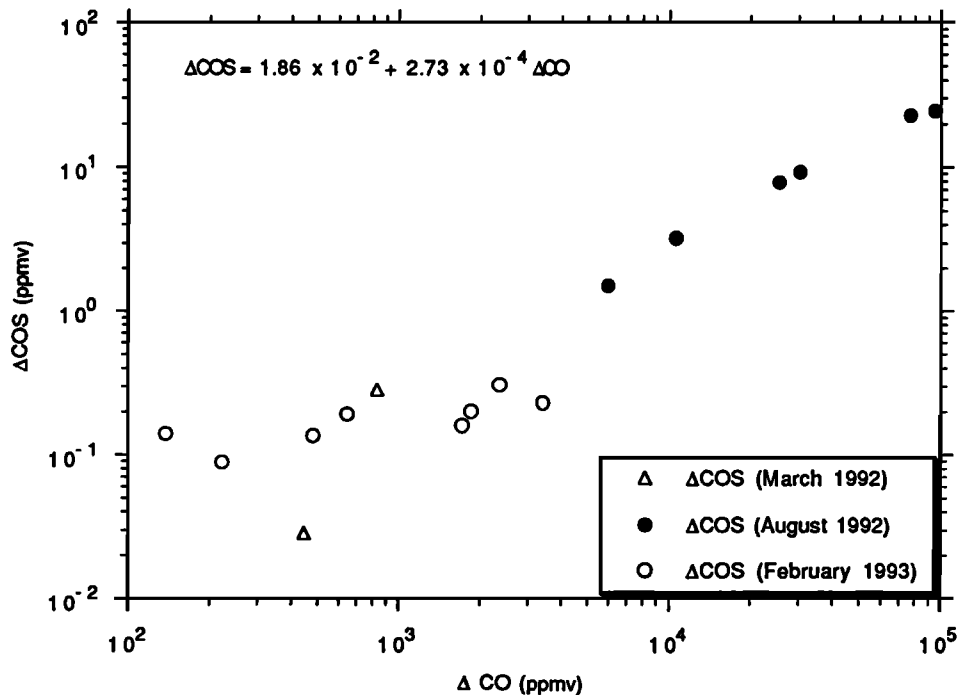


Figure 2. Relationship between ΔCO and ΔCOS during rice straw burning experiment in Viet Nam (solid circles, wet season; open circles and open triangles, dry season).

**Table 1.** CO and COS Emission Ratio ( $\Delta\text{CO}/\Delta\text{CO}_2$ ,  $\Delta\text{COS}/\Delta\text{CO}_2$ ) Estimates During Various Biomass Burning Experiments

Type of Vegetation	$\Delta\text{CO}/\Delta\text{CO}_2$ ( $\times 10^{-6}$ )	$\Delta\text{COS}/\Delta\text{CO}_2$ ( $\times 10^{-4}$ )	$\Delta\text{COS}/\Delta\text{CO}$	References
Wild basin fire, Colorado	0.15	15.8	1.05	<i>Crutzen et al.</i> [1979]
Tropical forest, Brazil	$0.15 \pm 0.10$	8.2	0.55	<i>Crutzen et al.</i> [1985]
Equatorial rainforest, Congo		6 - 42		<i>Bingemer et al.</i> [1992]
Savanna, Ivory Coast	0.11	$11.4 \pm 15.1$	1.03	<i>Nguyen et al.</i> [1993a]
Rice straw, Viet Nam (dry season)	$0.13 \pm 0.10$	$37.4 \pm 41.1$	2.87	this work
Rice straw, Viet Nam (wet season)	$0.28 \pm 0.11$	$78.2 \pm 31.4$	2.80	this work

in tropical areas of 350 ppmv and 0.1 ppmv, respectively [Marenco et al., 1989; Rudolph et al., 1991]

Figure 2 shows the relationship between  $\Delta\text{COS}$  and  $\Delta\text{CO}$ , which represent the increase of CO and COS mixing ratios above their background levels for both the wet (solid circles) and the dry seasons (open circles, open triangles). A significant regression is observed ( $r^2=0.99$ , slope  $2.7 \times 10^{-4}$ ) in agreement with our previous conclusions that COS and CO are both mainly produced during the smoldering stage [Nguyen et al., 1993a].

Emission ratios relative to  $\text{CO}_2$  ( $\Delta X/\Delta\text{CO}_2$ ), defined as

$$\Delta X/\Delta\text{CO}_2 = ([X]_{\text{sample}} - [X]_{\text{background}}) / ([\text{CO}_2]_{\text{sample}} - [\text{CO}_2]_{\text{background}})$$

for  $X = \text{CO}$  and  $\text{COS}$ , were calculated for both the wet and the dry seasons (Table 1). Two different emission ratios relative to  $\text{CO}_2$  are observed for both seasons and both compounds. The COS emission ratio falls from  $78 \times 10^{-6}$  during the wet season to  $27 \times 10^{-6}$  during the dry season. The CO emission ratio also falls from 0.28 during the wet season to 0.13 during the dry season. It should be noted that emission ratios relative to  $\text{CO}_2$  for both seasons and both compounds present a high variability (up to 110%) also in agreement with our previous observations and confirm our conclusion that most  $\text{CO}_2$  emission occurs during a different combustion stage (flaming) than CO and COS, mainly produced during the smoldering stage [Nguyen et al., 1993a].

Table 1 also compiles the recent estimates on the emission ratios of CO and COS relative to  $\text{CO}_2$  from biomass burning. Even during the dry season the CO and COS emission ratios from rice straw burning are close to the highest emission ratios already published. The  $\text{COS}/\text{CO}_2$  emission ratio is, for instance, more than twice higher than those observed during burning savanna in Africa [Nguyen et al., 1993a] or burning of forest in the United States [Crutzen et al., 1979] (Table 1). During the wet season both CO and COS emission ratios relative to  $\text{CO}_2$  increase by a factor of 2 with respect to the dry season. During this period,  $\Delta\text{COS}/\Delta\text{CO}_2$  and  $\Delta\text{CO}/\Delta\text{CO}_2$  are thus almost twice higher than the highest emission ratios observed in the plumes of savanna burning [Nguyen et al., 1993a; Bingemer et al., 1992] or forest burning in the United States [Crutzen et al., 1979] (Table 1). These results indicate that the combustion of rice straw produces much more CO and COS with regard to  $\text{CO}_2$  than the other known sources of

biomass burning. Enhanced emission ratios have also been observed for  $\text{CH}_4$  [Nguyen et al., 1993b] and could be explained by differences in the conditions of combustion among forest, savanna, and rice straw. In the same way the greater emission ratio for CO and COS relative to  $\text{CO}_2$  during the wet season could be due to the higher humidity in rice straw during this time period. Indeed, the presence of water in the burning material reduces the flame temperature, which induces changes in the thermodynamical conditions of combustion producing higher CO and  $\text{CH}_4$  concentrations [Lobert and Warnatz, 1993]. It should be noted that the  $\text{COS}/\text{CO}$  ratio is constant all over the year (Table 1). This suggests that CO and COS emissions from rice straw burning are controlled by the same combustion factors and that COS emission can be accurately estimated from CO emission and the  $\Delta\text{COS}/\Delta\text{CO}$  ratio.

#### Estimates of Regional and Global Emissions of COS from Biomass Burning

With data from the Vietnamese Ministry of Agriculture [1991] we estimated that about 20 Tg of rice straw are exposed to fire annually in Viet Nam. From that we calculated from our measured mean dry / total matter ratio of 0.9 and the carbon / dry matter ratio of 0.45 [Crutzen and Andreae, 1990] that 8.1 Tg of C (680 Gmol) from rice straw are burned each year in Viet Nam. About 95% of the carbon exposed to fire is released in the atmosphere in the gaseous form [Crutzen and Andreae, 1990], so that each year, 640 Gmol of C must be delivered to the atmosphere as  $\text{CO}_2$ , CO,  $\text{CH}_4$ , non-methane-hydrocarbons (NMHC), and COS:

$$N = n(\text{CO}_2) + n(\text{CO}) + n(\text{CH}_4) + n(\text{NMHC}) + n(\text{COS}) \quad (1)$$

where  $N$  is the total number of gaseous C moles annually released to the atmosphere (=640 Gmol) and  $n(X)$  is the number of moles of each compound.

The contributions of each of the above mentioned carbon species to the gaseous C released to the atmosphere can be calculated for both seasons using the following formulas:

$$n(\text{CO}_2)/N = (1 + n\text{CO}/n\text{CO}_2 + n\text{CH}_4/n\text{CO}_2 + n\text{NMHC}/n\text{CO}_2 + n\text{COS}/n\text{CO}_2)^{-1} \quad (2)$$

$$n(X)/N = n(X)/n(\text{CO}_2) \times n(\text{CO}_2)/N \quad (3)$$

**Table 2.** Percent Contributions of CO<sub>2</sub>, CO, CH<sub>4</sub>, NMHC and COS to the Gaseous C Released to the Atmosphere for Both Wet and Dry Season

Season	CO <sub>2</sub>	CO	CH <sub>4</sub>	NMHC
Dry	87	11	1	1
Wet	74	20	3	3

NMHC, nonmethane hydrocarbons

As  $n(X) = \Delta X \times V$  where  $V$  is the "volume" of the plume:

$$n(X)/n(\text{CO}_2) = (\Delta X \times V) / (\Delta \text{CO}_2 \times V) = \Delta X / \Delta \text{CO}_2 \quad (4)$$

Therefore we obtain

$$n(\text{CO}_2)/N = (1 + \Delta \text{CO} / \Delta \text{CO}_2 + \Delta \text{CH}_4 / \Delta \text{CO}_2 + \Delta \text{NMHC} / \Delta \text{CO}_2 + \Delta \text{COS} / \Delta \text{CO}_2)^{-1} \quad (5)$$

$$n(\text{CO})/N = (\Delta \text{CO} / \Delta \text{CO}_2) (n(\text{CO}_2)/N) \quad (6)$$

As emission ratios relative to CO<sub>2</sub> vary as a function of the season, we have performed the calculations for both wet and dry seasons. For CO we have taken emission ratios relative to CO<sub>2</sub> from Table 1. For CH<sub>4</sub> we have taken emission ratio relative to CO<sub>2</sub> equal to  $4.7 \times 10^{-2}$  and  $7.2 \times 10^{-3}$  for wet and dry seasons, respectively [Nguyen *et al.*, 1993b]. For NMHC we have assumed an emission ratio equal to that of CH<sub>4</sub>, as measured in Africa by Bonsang *et al.* [1991]. There appear significant changes in the gaseous composition of the plume of rice straw burning between the wet and the dry season (Table 2).

From field observations it was estimated that about 80% of the rice straw is burned during the dry season, whereas the remaining 20% is burned during the wet season [D. Canh, personal communication, 1993]. The annual weighted mean contributions to the gaseous C released to the atmosphere have thus been estimated to 84% for CO<sub>2</sub>, 13% for CO, and 1.1% for each of CH<sub>4</sub> and NMHC. Therefore we calculated that each year, rice straw burning in Viet Nam releases  $640 \times 0.13 = 83$  Gmol CO (1 Tg C). From the CO flux and the COS/CO ratio of  $2.7 \times 10^{-4}$ , the annual COS flux from rice straw burning can be estimated to 24 Mmol (0.8 Gg S).

Considering that agricultural customs are the same all over Asia and based on data provided by the International Rice

Research Institute (IRRI) that rice production from Viet Nam (19.2 Tg/yr) accounts for the 4% of the total Asian rice production (476.8 Tg/yr), we calculated that rice straw burning in Asia releases each year (Table 3) 2.1 Tmol CO (25 Tg C) and 0.6 Gmol COS (0.02 Tg S).

For these gases, Crutzen and Andreae [1990] and Nguyen *et al.* [1993a] estimated that all biomass burning releases each year 120-510 Tg C as CO and 0.04-0.20 Tg S as COS. Our new estimates from rice straw burning in Asia represent substantial portions of earlier estimates, which until now had never been adequately considered (Table 3). Taking into account these emissions from Asia where 92% of the mundial rice is produced [IRRI, 1993], former fluxes of CO and COS from biomass burning could be reevaluated by 5-21% and 10-67%, respectively, reaching 145 to 535 Tg C/yr for CO and 0.05 to 0.22 Tg S/yr for COS (Table 3).

## Conclusion

Our field rice straw burning experiments conducted in Viet Nam show a seasonality in the CO and COS emission ratios relative to CO<sub>2</sub> with twice higher values during the wet season.

With the annual weighted mean emission ratios from rice straw burning and using data of rice production from southeastern Asia, we estimated that burning of rice straw emits annually about 2.1 Tmol of CO (25 Tg C) and 0.6 Gmol of COS (0.02 Tg S) to the atmosphere. Taking into account these new results, the global CO and COS fluxes from biomass burning could be reevaluated by 5-21% and 10-67%, respectively, in regard to previous estimates of these gas emissions from all biomass burning activities.

Based on data provided by the Vietnamese Ministry of Agriculture, the amount of rice straw burned each year increases by about 6%. Therefore rice straw burning should have an increasing impact on atmospheric chemistry and could become

**Table 3.** CO and COS Emissions From Biomass Burning and All Known Sources

	CO, Tg C / yr	COS, Tg S / yr	References
All known sources	600-1300	0.6-1.5 0.3-1.8	Crutzen and Andreae [1990] Nguyen <i>et al.</i> [1993a]
Biomass burning sources	120-510	0.04-0.20 0.03-0.11	Crutzen and Andreae [1990] Nguyen <i>et al.</i> [1993a]
Rice straw burning	25	0.02	this work
Biomass burning of all sources	145-535	0.05-0.22	this work

a significant source of CO and COS at the global scale. Our estimates are based on the assumption that the seasonal variations of  $\Delta\text{CO}/\Delta\text{CO}_2$  and  $\Delta\text{COS}/\Delta\text{CO}_2$  are the same all over Asia. The uncertainty introduced by linearly extrapolating the seasonal variations observed in Viet Nam to the whole Asia is less than 20%. However, since some parts of Asia are not submitted to a monsoon climate, detailed data concerning agricultural customs and additional measurements in other areas of Asia would be needed to more accurately determine gaseous flux from agricultural waste burning.

**Acknowledgments.** This work was supported by the French Centre National de la Recherche Scientifique and Commissariat à l'Energie Atomique. The authors thank the authorities of Viet Nam, J. Fontan, Doan Canh and J. Goldammer for helpful field assistance, J. Orr and the two anonymous reviewers for helpful comments. This is CFR contribution No 1576.

## References

- Andreae, M. O., E. V. Browell, and M. Garstang, Biomass burning emissions and associated haze layers over Amazonia, *J. Geophys. Res.*, **93**, 1509-1527, 1988.
- Belviso, S., N. Mihalopoulos, and B. C. Nguyen, The supersaturation of carbonyl sulfide (OCS) in rain waters, *Atmos. Environ.*, **21**, 1363-1367, 1987.
- Bingemer, H. G., M. O. Andreae, T. M. Andreae, P. Artaxo, G. Helas, D. J. Jacob, N. Mihalopoulos, and B. C. Nguyen, Sulfur gases and aerosols in and above the equatorial African rainforest, *J. Geophys. Res.*, **97**, 6207-6217, 1992.
- Bonsang, B., G. Lambert, and C. Boissard, Light hydrocarbons emissions from African savanna burnings, in *Global Biomass Burning*, edited by J.S. Levine, pp. 155-161, MIT Press, Cambridge, Mass., 1991.
- Chin, M., and D. D. Davis, Global sources and sinks of COS and CS<sub>2</sub> and their distributions, *Global Biogeochem. Cycles.*, **7**, 321-337, 1993.
- Crutzen, P., and M. O. Andreae, Biomass burning in the tropics: Impact on atmospheric chemistry and biogeochemical cycles, *Science*, **250**, 1669-1678, 1990.
- Crutzen, P., L. E. Heidt, J. P. Krasnec, W. H. Pollock, and W. Seiler, Biomass burning as a source of atmospheric trace gases, *Nature*, **282**, 253-256, 1979.
- Crutzen, P., A. C. Delany, J. Greenberg, P. Haagenson, L. E. Heidt, R. Lueb, W. H. Pollock, W. Seiler, A. Wartburg, and P. Zimmerman, Tropospheric chemical composition measurements in Brazil during the dry season, *J. Atmos. Chem.*, **2**, 233-256, 1985.
- Houghton, R. A., The global effects of tropical deforestation, *Environ. Sci. Technol.*, **24**, 414-422, 1990.
- International Rice Research Institute (IRRI), *Rice Facts*, Manila, March 1993.
- Khalil, M. A. K., and R. A. Rasmussen, Global sources, lifetimes and mass balances of carbonyl sulfide (OCS) and carbon disulfide (CS<sub>2</sub>) in the earth's atmosphere, *Atmos. Environ.*, **18**, 1805-1813, 1984.
- Lashof, D. A., and D. R. Ahuja, Relative contribution of greenhouse gas emission to the global warming, *Nature*, **344**, 529-531, 1990.
- Lobert, J., and J. Wamatz, Emissions from the combustion process in *Fire in the Environment: The Ecological, Climatic and Atmospheric Chemical Importance of Vegetation Fires*, edited by P.J. Crutzen and J.G. Goldammer, pp 15-37, John Wiley, New York, 1993.
- Marengo, A., M. Macaigne, and S. Prieur, Meridional and vertical CO and CH<sub>4</sub> distributions in the background troposphere (70°N-60°S; 0-12 km altitude) from scientific aircraft measurements during the STRATOZ III experiment (June 1984), *Atmos. Environ.*, **23**, 185-200, 1989.
- Mihalopoulos, N., Contribution à l'étude du cycle biogéochimique de l'oxy-sulfure de carbone dans l'atmosphère, Ph.D thesis, Univ. of Paris, 1989.
- Ministry of Agriculture of Viet Nam, *35 Year (1956-1990) Agricultural Statistic Data Report*, Publishing House "Statistics", Hanoi, 1991.
- Nguyen, B. C., N. Mihalopoulos, J. P. Putaud, and B. Bonsang, Carbonyl sulfide emissions from biomass burning in tropics, *J. Atmos. Chem.*, in press, 1993a.
- Nguyen, B. C., J. P. Putaud, N. Mihalopoulos, and B. Bonsang, CH<sub>4</sub> and CO emissions from rice straw burning in tropics, paper presented at the international Symposium on Non-CO<sub>2</sub> greenhouse gases, Commission of the European Communities, Maastricht, Netherlands, December 13-15, 1993b.
- Penner, J. E., J. S. Ghan, and J. J. Walton, The role of biomass burning in the budget and cycle of carbonaceous soot aerosols and their climate impact, in *Global Biomass Burning*, edited by J.S. Levine, pp. 387-393, MIT Press, Cambridge, Mass., 1991.
- Rudolph, J., R. Koppman, and B. Bonsang, Measurements of light hydrocarbons and carbon monoxide over an equatorial forest in Africa, paper presented at the AGU Fall Meeting, Am. Geophys. Union., San Francisco, Calif., Dec. 5-9, 1991.
- Servant, J., The burden of the sulfate layer of the stratosphere during volcanic quiescent periods, *Tellus*, **38B**, 74-79, 1989.

B.C. Nguyen and J.-P. Putaud, Centre des Faibles Radioactivités, Laboratoire mixte CNRS-CEA, F-91198 Gif-sur-Yvette Cedex, France. (e-mail: putaud@asterix.saclay.cea.fr)

N. Mihalopoulos, Department of Chemistry, University of Crete, P.O.Box 1470, 71409 Iraklion, Greece. (e-mail: mihalo@hercules.cc.uh.gr)

(Received August 6, 1993; revised December 9, 1993; accepted December 10, 1993.)