EMISSION ESTIMATES FOR AIR POLLUTION TRANSPORT MODELS

by

David G. Streets* and Stephanie T. Waldhoff
Decision and Information Sciences Division
Argonne National Laboratory
Argonne, IL 60439, U.S.A.

presented at

Workshop on Transport of Air Pollutants in Asia
International Institute for Applied Systems Analysis
Laxenburg, Austria

July 27-29, 1998

*Primary author to whom correspondence should be sent. Email: dstreets@anl.gov

EMISSION ESTIMATES FOR AIR POLLUTION TRANSPORT MODELS

DAVID G. STREETS and STEPHANIE T. WALDHOFF
Decision and Information Sciences Division, Argonne National Laboratory,
9700 South Cass Avenue, Argonne, IL 60439, USA

Abstract

The results of studies of energy consumption and emission inventories in Asia are discussed. These data primarily reflect emissions from fuel combustion (both biofuels and fossil fuels) and were collected to determine emissions of acid-deposition precursors (SO_2 and NO_x) and greenhouse gases (CO_2, CO, CH_4, and NMHC) appropriate to RAINS-Asia regions. Current work is focusing on black carbon (soot), volatile organic compounds, and ammonia.

Introduction

In order for transport and deposition models to be accurate, it is necessary to have reliable input data on emissions. Accuracy depends both on detailed fuel consumption data and the application of appropriate emission factors. In addition, in order for emissions data to be meaningful when gridded for input to atmospheric models, the fuel consumption data must be collected on a disaggregated geographic basis and apportioned using appropriate surrogate measures, i.e., population or production. For example, Chinese national data gridded by population are less accurate than Chinese provincial data gridded by population. To this end, energy consumption data have been gathered by RAINS-Asia regions, and, when possible, regional emission factors have been used.

Biofuel Combustion

In the RAINS-Asia Phase I Project, emissions from the use of commercial fuels were extensively documented (Arndt and others, 1997). Recent work has added the important component of biofuels (Streets and Waldhoff, 1998). Throughout the developing world, energy for cooking and heating is still often derived from the combustion of traditional biofuels: fuelwood, crop residues, dried animal waste, and charcoal. It has been estimated that approximately 15% of energy demand worldwide is supplied by these biofuels; in Asia the contribution is 25%. The *per capita* usage of traditional biofuels is declining (Figure 1), as they are supplanted by commercial fuels: kerosene, oil, natural gas, coal, and electricity; however, population is increasing in the developing countries, especially in the rural areas where biofuels are mainly used, such that total biofuel use is presently stable or only slowly declining. It was found that a significant correlation exists between *per capita* GDP and biofuel consumption as a percent of total energy use (Figure 2). This relationship, with a correlation coefficient (R^2) of 0.90, is expressed by the equation:

$$\% \text{ Biofuel Use} = 40{,}738\ (\text{GDP/cap})^{-1.17}$$

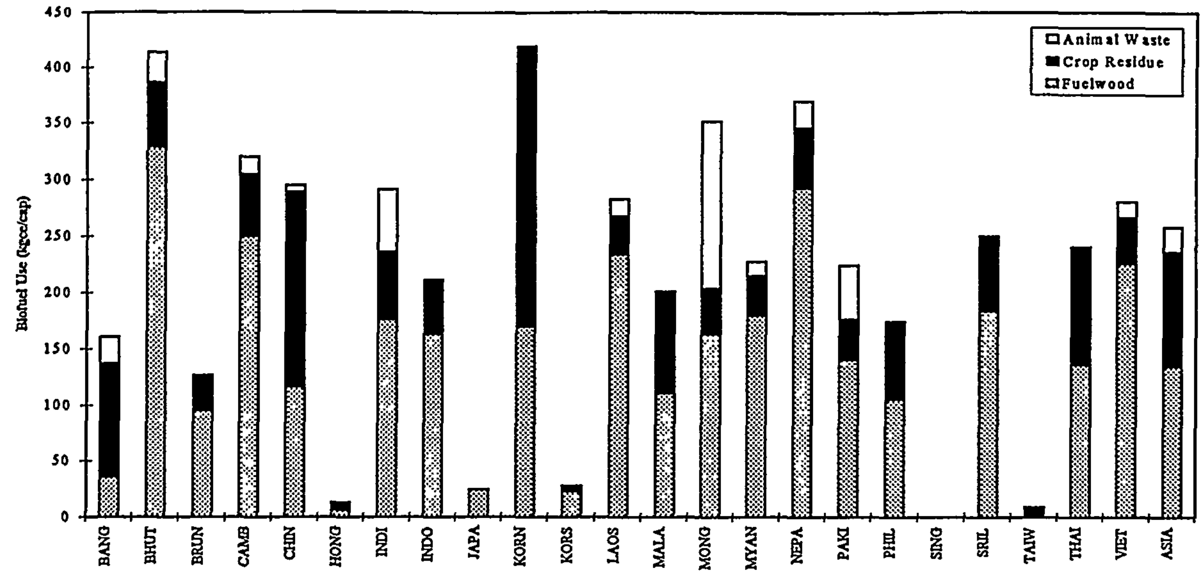


Figure 1 - Biofuel combustion *per capita* in Asian countries in 1990.

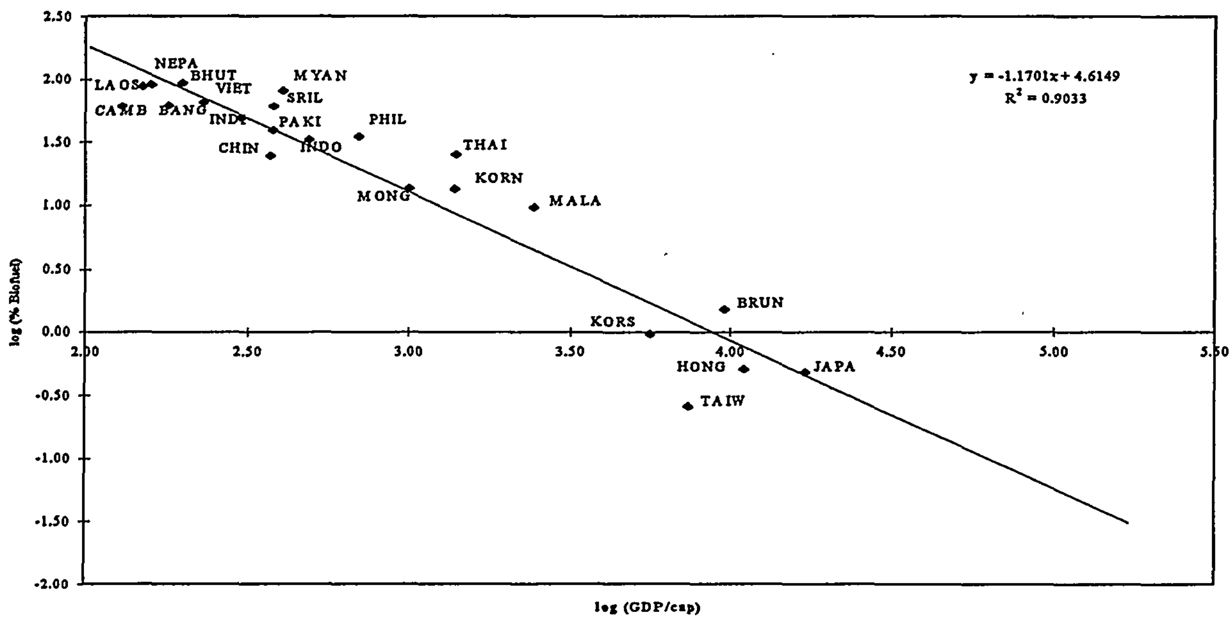


Figure 2 - Relationship between GDP *per capita* and biofuel use as a percent of total energy use in Asia in 1990.

Because the quantities of biofuels used in developing countries are not completely and consistently known and biofuel consumption data were largely absent in the RAINS-Asia model, it was decided that a detailed inventory of biofuel use should be created. One problem in this endeavor is that much of this fuel is noncommercial, that is to say, it is gathered by individual families for their own use on a subsistence basis. There are markets for firewood and charcoal near many of the larger towns and cities, but the fuel bought and sold in these establishments represents only a small fraction of total usage. National statistics on energy use are frequently limited to these commercial components. We report results of a detailed inventory of biofuel use at the regional level in Asia in 1990 (Streets and Waldhoff, 1998). The combustion products, SO_2, NO_X, and greenhouse gases are important for both indoor and local health effects, as well as regional and transboundary acidification and global warming. The results of this work are important for the development of both national and international policies for energy development and environmental protection.

Table 1 - 1990 SO_2 and NO_x emissions from biofuel combustion in Asia.

	Sulfur Dioxide					Nitrogen Oxides				
	Biofuel	Emissions (Gg)			% Biofuel	Biofuel	Emissions (Gg)			% Biofuel
Country	(g/capita)	Biofuel	Commer.	Total	Emissions	(g/capita)	Biofuel	Commer.	Total	Emissions
Bangladesh	474.0	50.3	44.3	94.6	53.2	379.8	40.3	50.4	90.7	44.4
Bhutan	769.2	1.1	0.2	1.4	82.5	629.4	0.9	0.4	1.3	69.2
Brunei	134.4	0.0	6.1	6.1	0.6	166.0	0.0	16.1	16.1	0.3
Cambodia	501.8	4.3	18.3	22.6	18.9	455.1	3.9	24.3	28.2	13.8
China	395.7	448.8	21908.2	22357.0	2.0	629.8	714.3	8272.7	8987.0	7.9
Hong Kong	17.2	0.1	136.4	136.5	0.1	34.5	0.2	245.3	245.5	0.1
India	1036.1	879.8	3480.6	4360.4	20.2	540.6	459.1	2642.6	3101.7	14.8
Indonesia	259.3	49.8	508.6	558.4	8.9	344.2	66.1	657.4	723.5	9.1
Japan	26.7	3.3	827.6	830.9	0.4	32.4	4.0	2468.5	2472.5	0.2
North Korea	459.3	10.0	343.1	353.2	2.8	891.0	19.4	518.3	537.7	3.6
South Korea	30.3	1.3	1663.6	1665.0	0.1	44.3	1.9	973.4	975.3	0.2
Laos	507.2	2.1	1.3	3.4	61.9	434.8	1.8	0.7	2.5	71.4
Malaysia	219.6	3.9	201.9	205.8	1.9	388.6	6.9	276.8	283.7	2.4
Mongolia	2302.6	4.9	76.0	80.8	6.0	704.9	1.5	29.0	30.5	4.9
Myanmar	404.6	16.5	2.8	19.3	85.7	365.3	14.9	17.0	31.9	46.8
Nepal	713.5	13.5	3.3	16.8	80.5	592.0	11.2	4.8	16.0	69.9
Pakistan	864.1	95.4	590.1	685.5	13.9	408.5	45.1	240.4	285.5	15.8
Philippines	190.8	11.6	399.7	411.3	2.8	322.4	19.6	165.7	185.3	10.6
Singapore	0.0	0.0	190.9	190.9	0.0	0.0	0.0	100.9	100.9	0.0
Sri Lanka	176.6	4.7	21.3	26.0	18.0	417.8	7.1	28.4	35.5	20.0
Taiwan	9.9	0.2	493.0	493.2	0.0	24.8	0.5	520.0	520.5	0.1
Thailand	264.6	14.9	973.4	988.3	1.5	458.3	25.8	442.1	467.9	5.5
Vietnam	489.4	31.9	81.1	113.0	28.2	444.9	29.0	78.1	107.1	27.1
Asia Total	**568.0**	**1648.5**	**31971.7**	**33620.2**	4.9	507.8	1473.5	17773.2	19246.7	7.7

Acid-Deposition Precursors

Although fossil fuels are commonly thought to be the most significant sources of acid-precursors, in areas where biofuels are heavily used, their contribution to SO_2 and NO_x emissions can be very significant (Table 1). In some countries biofuel use represents more than 80% of SO_2

emissions (Bhutan, Myanmar, and Nepal) and more than 60% of NO_x emissions (Bhutan, Laos, and Nepal). SO_2 emissions *per capita* from biofuel combustion were highest in countries that burn large amounts of animal waste (with a high emission factor) while NO_x emissions were highest in countries that used primarily woody-type biofuels (Figure 3).

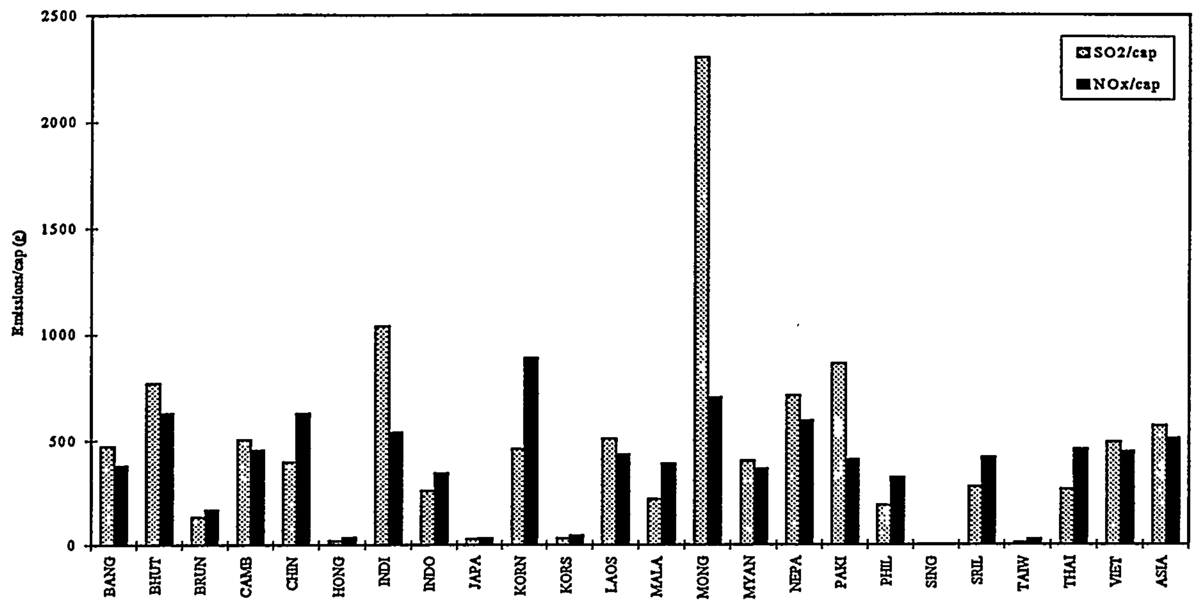


Figure 3 - 1990 SO_2 and NO_x emissions *per capita,* by country

These studies found that total SO_2 emissions from biofuel use in Asia in 1990 are 1.65 Tg, or 4.9% of total SO_2 emissions from fuel combustion in Asia. The greatest amount of SO_2 emissions came from India, 880 Gg, the figure being especially high because of the large amounts of animal waste burned, which has a high sulfur content. Though China consumes the largest amount of biofuels, its SO_2 emissions are lower, at 449 Gg, because the predominant biofuels used are woody products (fuelwood and crop residue) that have lower sulfur contents. Together, these two countries account for nearly 81% of total SO_2 emissions from biofuels.

Total 1990 NO_x emissions from the combustion of biofuels in Asia are estimated to be 1.47 Tg. Again, the two countries with the highest absolute emissions are China (714 Gg) and India (459 Gg). Biofuels represent 7.7% of total NO_x emissions from all sources in Asia (Table 1). In the rural economies that have few industrial sources and minimal fossil-fuel-based transportation systems, biofuel NO_x can represent a high proportion of total NO_x emissions (>50% in Laos, Nepal, and Bhutan).

In addition, a need was identified within another project, the NASA China-MAP study, to develop emission profiles for China for the years 1992/93 and 1995, for the purpose of comparing calculated deposition with observed data for sulfate and nitrate deposition in Chinese monitoring data sets. The RAINS-Asia model generates sulfur dioxide emission estimates for the years 1990 and

2000 (nitrogen oxide emissions estimates were determined by multiplying energy data from the RAINS-Asia model by appropriate emissions factors). It was decided that interpolating between the RAINS-Asia emission values for 1990 and 2000 would provide results consistent with other parts of the China-MAP project. An improved methodology is currently being developed for the RAINS-Asia Phase II project, which involves extrapolation from 1990 to the period 1985-1995 using fuel-use proxies from IEA statistics (IEA, 1997). For the present, however, interpolation is considered a reasonable method that relies on real data for 1990 and anticipated growth rates by region and sector for the period 1990-2000.

The method used to determine 1992/93 and 1995 values is based on the simple formula for fixed annual growth in this period:

$$E_{2000} = E_{1990} (1 + r)^{10}$$

where E represents the emissions, in Gg, of pollutant and r is the annual rate of growth. By manipulating this function algebraically, we find that:

$$E_{1995} = (E_{1990} * E_{2000})^{1/2}$$

Values for 1992/93 were calculated in a similar manner:

$$E_{1992/93} = (E_{1990} * E_{1995})^{1/2}$$

For three of the sectors–industry, domestic, and transportation–1992/93 and 1995 values were interpolated by applying this formula. Slightly different methods were used for the power and non-commercial sectors.

It was found that the industrial sector was the largest contributor to SO_2 emissions in China in 1990, followed by the power sector, the domestic sector (fossil-fuel emissions), non-commercial sector (biofuel), and transportation (Figure 4). It is expected that emissions from these sectors will grow through 2000, while keeping their relative rankings. 1990 NO_x emissions in China were dominated by the power and industrial sectors with relatively similar amounts coming from the domestic (fossil-fuel), transportation, and non-commercial (biofuel) sectors (Figure 5). The biggest change in NO_x emissions between 1990 and 2000 is expected to take place in the transportation sector. As cars become more commonplace in China, this sector is expected to grow relatively quickly, with NO_x emissions growing similarly.

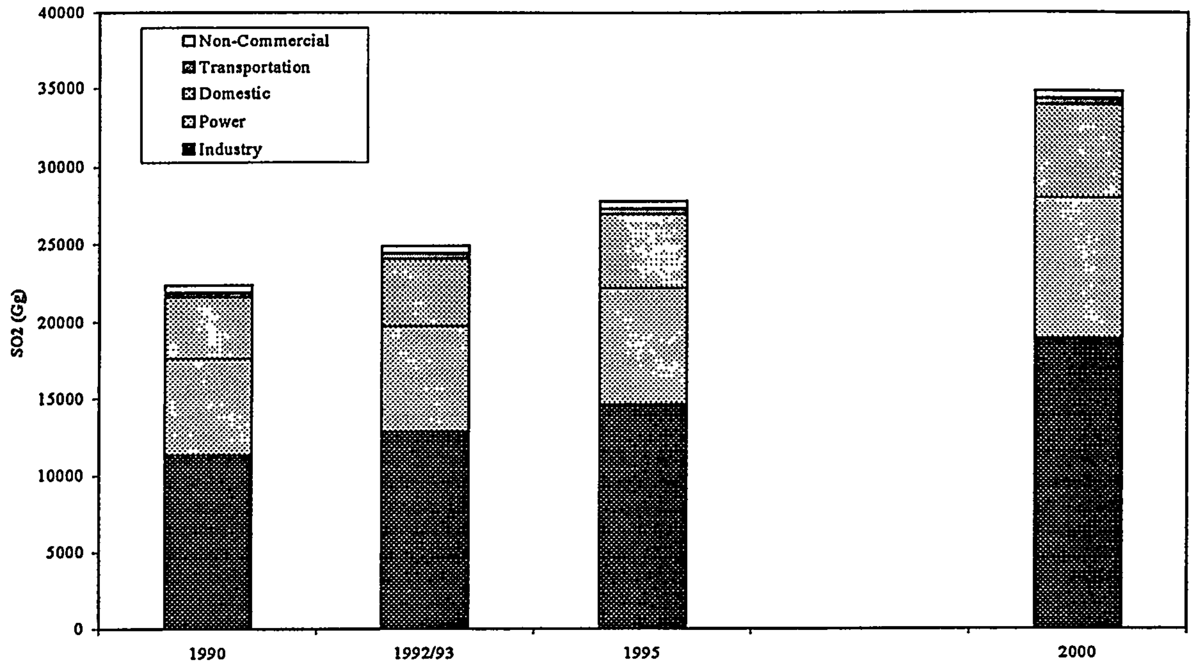


Figure 4 - 1990 Chinese SO_2 emissions by sector.

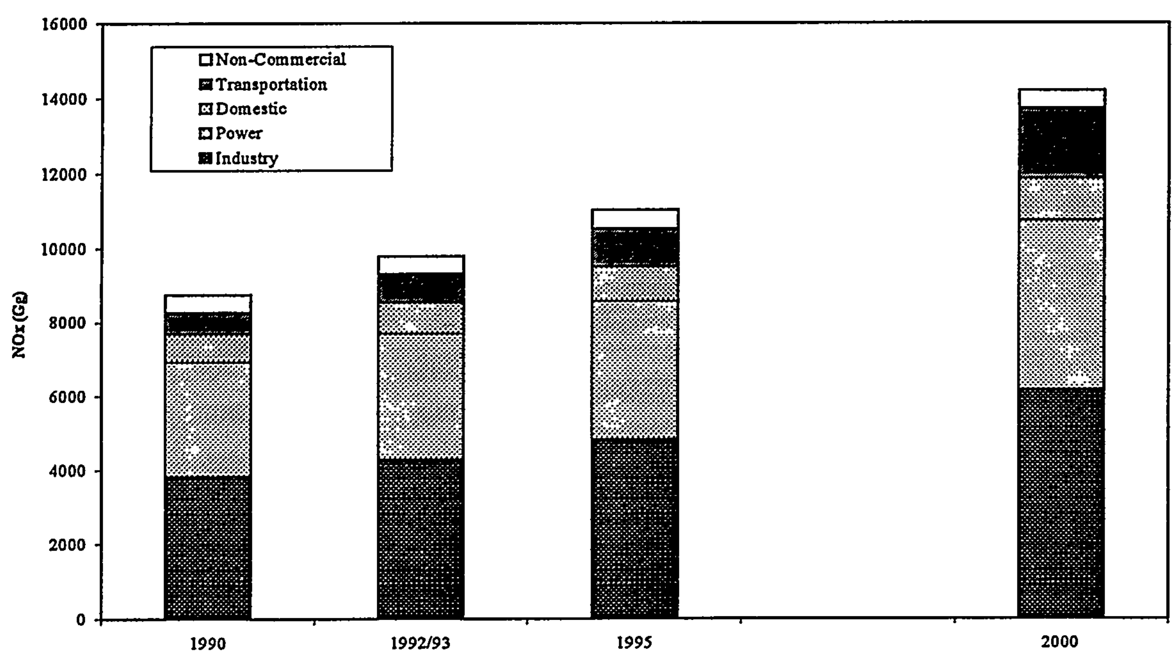


Figure 5 - 1990 Chinese NO_x emissions by sector.

Carbonaceous Greenhouse Gases

The biofuel consumption inventory used to calculate SO_2 and NO_X emissions was also used to calculate 1990 greenhouse gas emissions (CO_2, CO, CH_4, and NMHC) from biofuel combustion in Asia (Table 2), which were then compared with emissions from fossil-fuel combustion. Data were examined from two perspectives: total carbon released and total global warming potential (GWP) of the gases (Tables 3-5). Two time frames, 20 years and 100 years, were used to compare the total global warming potentials of these emissions, as GWP decreases over time as the products of incomplete combustion (PIC)—CO, CH_4, and NMHC—are removed from the atmosphere. It is estimated that biofuels contributed 28% of total carbon emissions resulting from energy-related combustion in Asia in 1990 (Table 3). Because of the high rate of incomplete combustion in typical biofuel stoves and the high GWP coefficients of the PIC, biofuels comprise an even larger share in terms of global warming potential (CO_2 equivalents): 38% over 20 years and 31% over 100 years (Tables 4 and 5). It was found that even when biofuel is assumed to be harvested on a completely sustainable basis (all CO_2 emissions are reabsorbed in the following growing season), PIC emissions from biofuel combustion account for almost 5% of total carbon emissions and nearly 25% of CO_2 equivalents in terms of total short-term (20-year) GWP.

Table 2 - 1990 Greenhouse gas emissions from biofuel combustion, by fuel source and species (Gg-C)

Country	Fuelwood				Crop Residue				Animal Waste			
	CO_2	CO	CH_4	NMHC	CO_2	CO	CH_4	NMHC	CO_2	CO	CH_4	NMHC
Bangladesh	2508.5	242.4	19.9	18.8	7512.9	962.5	88.6	60.2	1503.5	202.9	75.3	87.5
Bhutan	312.1	30.2	2.5	2.3	57.0	7.3	0.7	0.5	24.0	3.2	1.2	1.4
Brunei	15.9	1.5	0.1	0.1	5.6	0.7	0.1	0.0	0.0	0.0	0.0	0.0
Cambodia	1425.9	137.8	11.3	10.7	321.3	41.2	3.8	2.6	79.3	10.7	4.0	4.6
China	87733.5	8479.1	696.9	658.2	136309.5	17462.3	1608.4	1091.4	3862.7	521.3	193.4	224.7
Hong Kong	22.6	2.2	0.2	0.2	28.5	3.7	0.3	0.2	0.0	0.0	0.0	0.0
India	99359.8	9602.7	789.3	745.4	35430.0	4538.9	418.1	283.7	28860.1	3894.8	1445.1	1679.0
Indonesia	20828.6	2013.0	165.5	156.3	6155.0	788.5	72.6	49.3	243.4	32.8	12.2	14.2
Japan	1995.7	192.9	15.9	15.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
North Korea	2451.8	237.0	19.5	18.4	3784.8	484.9	44.7	30.3	0.0	0.0	0.0	0.0
South Korea	636.2	61.5	5.1	4.8	166.2	21.3	2.0	1.3	0.0	0.0	0.0	0.0
Laos	642.9	62.1	5.1	4.8	98.1	12.6	1.2	0.8	38.1	5.1	1.9	2.2
Malaysia	1296.4	125.3	10.3	9.7	1124.6	144.1	13.3	9.0	0.0	0.0	0.0	0.0
Mongolia	230.4	22.3	1.8	1.7	60.5	7.8	0.7	0.5	193.0	26.0	9.7	11.2
Myanmar	4858.0	469.5	38.6	36.4	1019.6	130.6	12.0	8.2	300.4	40.5	15.0	17.5
Nepal	3677.9	355.5	29.2	27.6	699.7	89.6	8.3	5.6	276.0	37.2	13.8	16.1
Pakistan	10293.8	994.9	81.8	77.2	2747.1	351.9	32.4	22.0	3238.9	437.1	162.2	188.4
Philippines	4228.4	408.7	33.6	31.7	2909.3	372.7	34.3	23.3	0.0	0.0	0.0	0.0
Singapore	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Sri Lanka	2067.4	199.8	16.4	15.5	792.8	101.6	9.4	6.3	0.0	0.0	0.0	0.0
Taiwan	4.6	0.4	0.0	0.0	125.9	16.1	1.5	1.0	0.0	0.0	0.0	0.0
Thailand	5082.6	491.2	40.4	38.1	4088.7	523.8	48.2	32.7	0.0	0.0	0.0	0.0
Vietnam	9763.2	943.6	77.6	73.2	1874.3	240.1	22.1	15.0	560.5	75.6	28.1	32.6
Asia	259436.3	25073	2060.8	1946.3	205311.4	26302.0	2422.6	1643.9	39179.8	5287.5	1961.8	2279.4

Table 3 - Contribution of biofuels and fossil fuels to carbon emissions in 1990 (Gg-C).

	CO2	CO	CH4	NMHC/ VOC	Total
Biofuels	503,927.5	56,663.0	6,445.2	5,869.6	572,905.3
Fossil Fuels	1,455,634.5	17,749.7	1,280.0	1,201.1	1,475,865.3
Total	1,959,562.0	74,412.7	7,725.2	7,070.7	2,048,770.6
% Biofuel	25.7	76.1	83.4	83.0	28.0

Table 4 - Contribution of biofuels and fossil fuels to overall 20-year GWP in Asia (CO_2 Equivalent)

	CO2	CO	CH4	NMHC/ VOC	Total
Biofuels	503,927.5	254,983.5	141,803.2	70,432.8	971,147.0
Fossil Fuels	1,455,634.5	79,873.7	28,160.4	25,212.7	1,588,881.3
Total	1,959,562.0	334,857.2	169,963.6	95,645.5	2,560,028.3
% Biofuel	25.7	76.1	83.4	73.6	37.9

Table 5 - Contribution of biofuels and fossil fuels to overall 100-year GWP in Asia (CO_2 Equivalent)

	CO2	CO	CH4	NMHC/ VOC	Total
Biofuels	503,927.5	107,659.7	48,342.0	24,064.5	683,993.7
Fossil Fuels	1,455,634.5	33,724.4	9,600.1	8,614.3	1,507,573.3
Total	1,959,562.0	141,384.1	57,942.1	32,678.8	2,191,567.0
% Biofuel	25.7	76.1	83.4	73.6	31.2

Carbon emissions (CO_2, CO, CH_4, NMHC) *per capita* from biofuel combustion were proportional to SO_2 and NO_x emissions (Figure 6). Total emissions were again dominated by India and China (together contributing more than 75% of biofuel emissions for each gas), but there were significant differences between the specific gases. CO_2 and CO emissions were dominated by China (Figure 7a-b), with Indonesia's (a country that burns primarily woody biofuels) emissions ranking third. However, India's role was much more significant for CH_4 and NMHC (gases associated with animal waste burning) and Pakistan was the third largest emitter for these two gases (Figure 7c-d).

Chinese emissions of carbon monoxide were examined in more detail for 1995. Five sectors were determined to be the most significant contributors to CO emissions in China. These are domestic (both fossil-fuel and biofuel combustion), industry (combustion of fossil fuels and emissions from processes such as iron and steel manufacture), transportation (primarily passenger and freight highway traffic), field combustion (disposal of crop residues by direct combustion in fields), and electric power generation (Figure 8). Though they are thought to be significant, savannah burning and spontaneous coal mine combustion are not included in this estimate, due to difficulties in determining the magnitude and location of these activities. It was estimated that approximately 120 mt of CO were released in China in 1995. This figure is more than 40% greater than the CO emission estimate of 83.5 mt for the United States in 1995. Sectoral distribution is also significantly different, with the majority of U.S. emissions coming from the transportation sector, while in China the transportation sector is responsible for less than 20% of total CO in 1995. The domestic sector is by

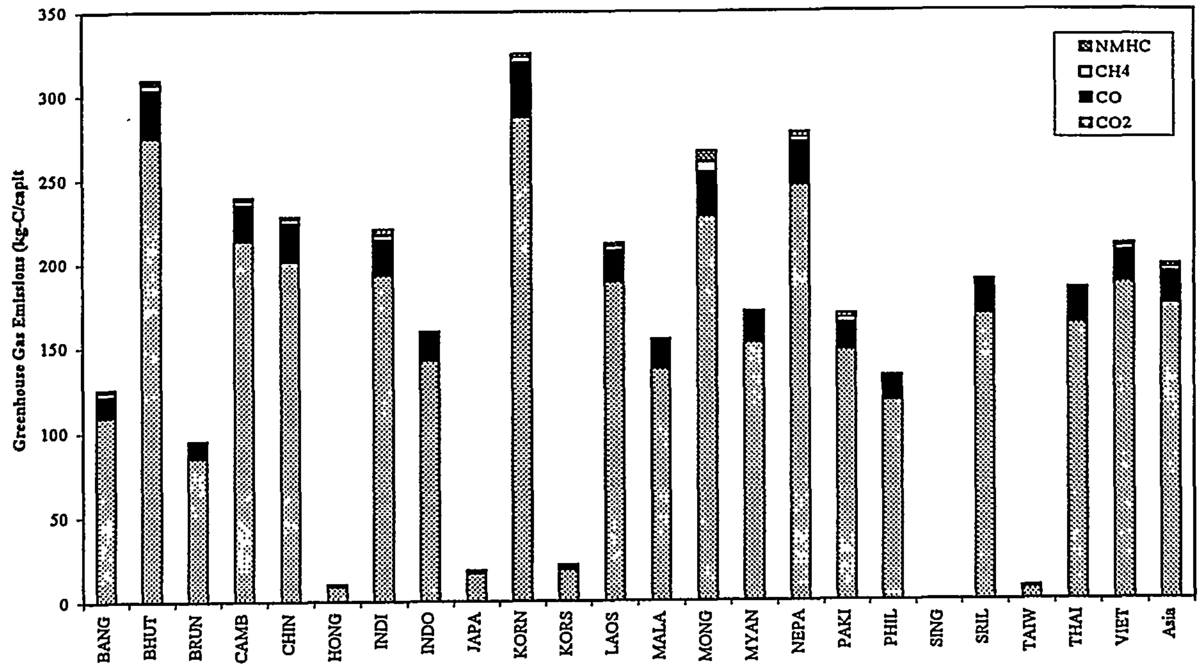
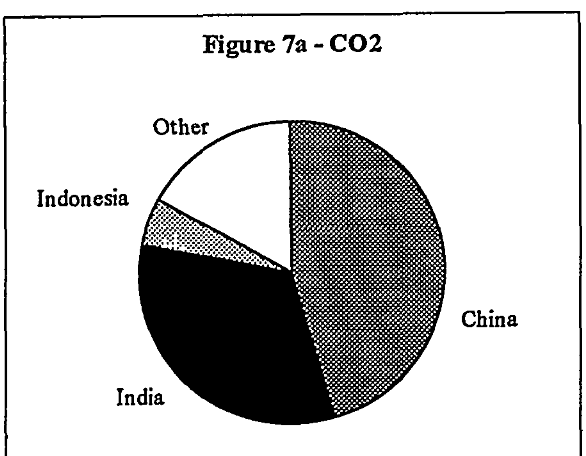


Figure 6 - 1990 carbon emissions *per capita* by species and country.

far the largest contributor in China, emitting approximately 73.3 mt, or 61% of all the CO emissions in China for 1995. Within the domestic sector, direct burning of biofuels (wood, crop residues, and animal waste) was the primary contributor (85% of domestic emissions), with coal use contributing an additional 10.6 mt. The industrial, agricultural, and transportation sectors were also large emitters, releasing 13.7 mt, 9.8 mt, and 23.0 mt of CO, respectively. These results are presently undergoing internal review. Initial estimates for black carbon (aerosol) emissions in China are also included (Figure 9); these estimates are also in the process of internal review.



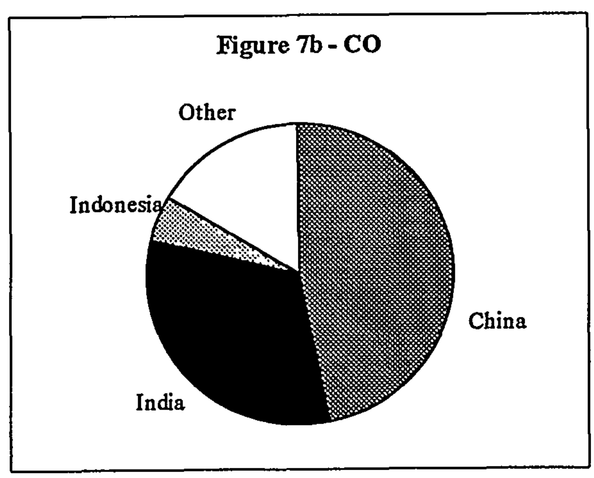
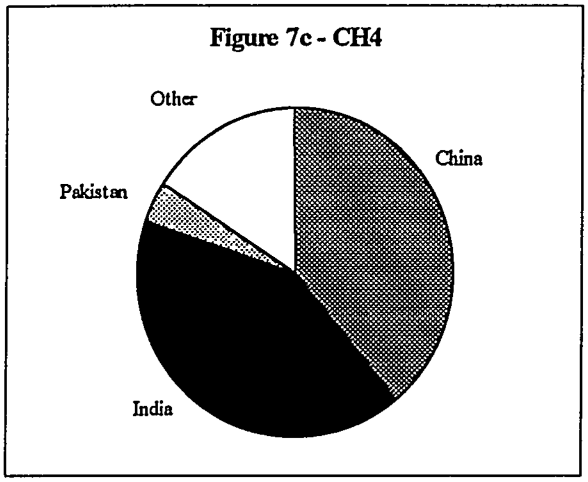
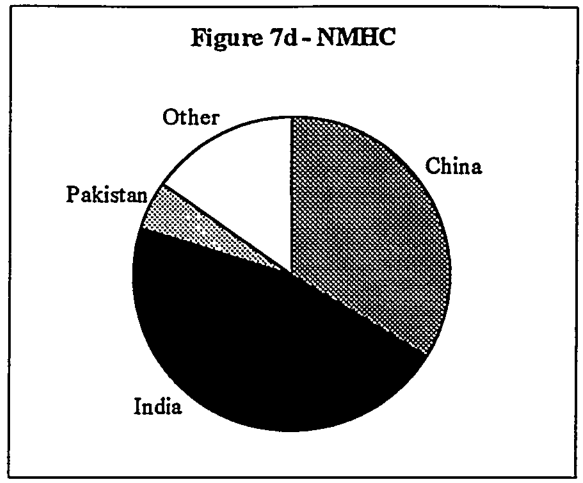


Figure 7 - 1990 Greenhouse gas emissions from biofuel combustion in Asia, by species.





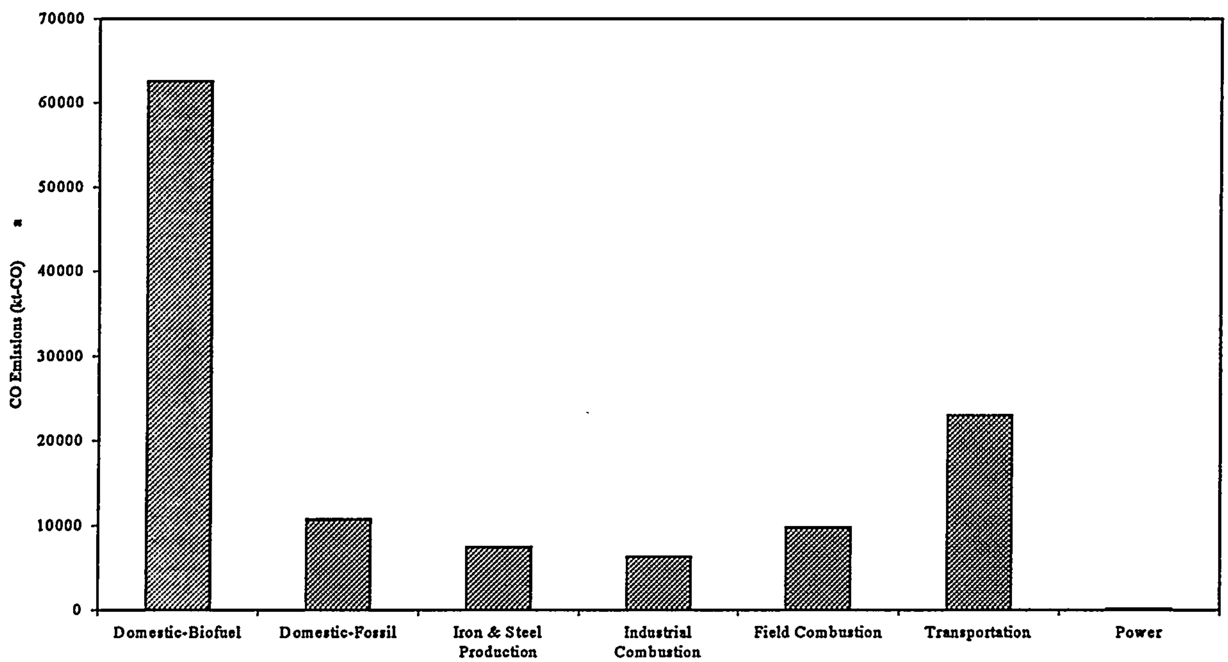


Figure 8 - 1995 preliminary emissions of CO in China, by source.

Conclusions

Emission inventories are the base upon which atmospheric transport and deposition models are built. If the emissions data are faulty, the results of future modeling will reflect and possibly amplify these problems, so it is extremely important to estimate emissions as accurately as possible. As the state of the art of atmospheric modeling for Asia advances, an increasingly wide range of species becomes important. For example, the prediction of elevated ozone concentrations—which is a critical, but presently unmet need—requires knowledge of an array of emitted species, including NO_x, CO, and NMHC. In order to be most effective, sophisticated climate models need accurate inputs of concentrations of aerosol species, including SO_4, NO_3, black carbon, etc. Even acidification models need base cation and ammonia emissions to produce an accurate picture of the physical and chemical processes that occur. Because many of these species are driven by the same anthropogenic and natural processes, it is efficient to integrate the approaches used to estimate their emissions. This is a goal of much of the emissions work presently being undertaken at Argonne.

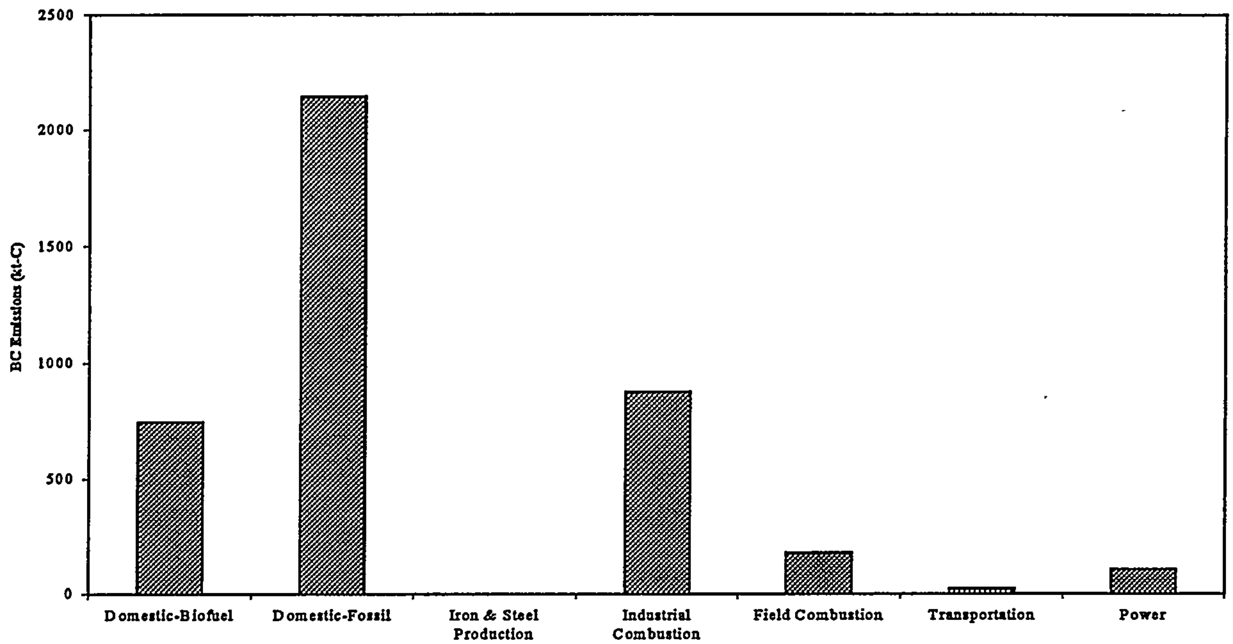


Figure 9 - 1995 preliminary emissions of BC in China, by source.

Acknowledgments

This work was funded by the National Aeronautics and Space Administration, as part of the China-MAP program, under Interagency Agreement No. W-19,313. The opinions presented here are those of the authors themselves and are not necessarily the opinions of Argonne National Laboratory or NASA.

References

Arndt, R.L., Carmichael, G.R., Streets, D.G. and Bhatti, N., *Atmospheric Environment*, 1997, **31**, 1553.

IEA, "Energy Statistics and Balances," IEA and OECD, Paris, 1997.

Streets, D.G. and Waldhoff, S.T., *Energy–The International Journal*, 1998, in press.