An inventory of gaseous and primary aerosol emissions in Asia in the year 2000

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[1] An inventory of air pollutant emissions in Asia in the year 2000 is developed to support atmospheric modeling and analysis of observations taken during the TRACE-P experiment funded by the National Aeronautics and Space Administration (NASA) and the ACE-Asia experiment funded by the National Science Foundation (NSF) and the National Oceanic and Atmospheric Administration (NOAA). Emissions are estimated for all major anthropogenic sources, including biomass burning, in 64 regions of Asia. We estimate total Asian emissions as follows: 34.3 Tg SO_2, 26.8 Tg NO_x, 9870 Tg CO_2, 279 Tg CO, 107 Tg CH_4, 52.2 Tg NMVOC, 2.54 Tg black carbon (BC), 10.4 Tg organic carbon (OC), and 27.5 Tg NH_3. In addition, NMVOC are speciated into 19 subcategories according to functional groups and reactivity. Thus we are able to identify the major source regions and types for many of the significant gaseous and particle emissions that influence pollutant concentrations in the vicinity of the TRACE-P and ACE-Asia field measurements. Emissions in China dominate the signature of pollutant concentrations in this region, so special emphasis has been placed on the development of emission estimates for China. China's emissions are determined to be as follows: 20.4 Tg SO_2, 11.4 Tg NO_x, 3820 Tg CO_2, 116 Tg CO, 38.4 Tg CH_4, 17.4 Tg NMVOC, 1.05 Tg BC, 3.4 Tg OC, and 13.6 Tg NH_3. Emissions are gridded at a variety of spatial resolutions from $1° \times 1°$ to 30 s $\times$ 30 s, using the exact locations of large point sources and surrogate GIS distributions of urban and rural population, road networks, landcover, ship lanes, etc. The gridded emission estimates have been used as inputs to atmospheric simulation models and have proven to be generally robust in comparison with field observations, though there is reason to think that emissions of CO and possibly BC may be underestimated. Monthly emission estimates for China are developed for each species to aid TRACE-P and ACE-Asia data interpretation. During the observation period of March/April, emissions are roughly at their average values (one twelfth of annual). Uncertainties in the emission estimates, measured as 95% confidence intervals, range from a low of ±16% for SO_2 to a high of ±450% for OC. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; *KEYWORDS:* Asia, emissions, inventory, TRACE-P, aerosols

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1. Introduction

[2] A good understanding of air pollution concentrations and fluxes at regional scale requires the integration of observations from field campaigns with results from regional or global atmospheric models over the same domains of time and space. To exercise the atmospheric models requires a coordinated set of emission inputs of all necessary species. It is important that these emission fields correctly reflect the spatial and temporal emission profiles of sources that were operating during the time period of the field campaigns, otherwise good agreement between models and experiment cannot be expected. Knowledge of source strengths and locations is also a valuable aid for interpreting observations

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and model results and ultimately choosing appropriate mitigation strategies.

[3] For these reasons we have developed an emission inventory for Asia for the year 2000 to contribute to the NASA TRACE-P (Transport and Chemical Evolution over the Pacific) Mission [*Jacob et al.*, 2003] and ACE-Asia (Asian Pacific Regional Aerosol Characterization Experiment) funded by NSF and NOAA [*Huebert et al.*, 2003]. This is the first time that Asian emissions have been studied in a comprehensive and consistent way for a recent year. Though there are considerable uncertainties associated with some of the emission values, due to lack of national statistics in many countries and lack of knowledge about the performance of certain kinds of emitters, we nevertheless believe that the confidence level for many species is high and that the inventory will be valuable in gaining an understanding of the formation and fate of regional air pollutants in Asia.

[4] The domain stretches from Pakistan in the West to Japan in the East and from Indonesia in the South to Mongolia in the North. Figure 1 shows the countries and subregions that are part of this inventory. International shipping lanes are included, located according to the method of *Streets et al.* [2000b]. In order to focus on the needs of the two missions, the following priorities were given to emissions estimation: first, China; second, the rest of East Asia; third, Southeast Asia; and fourth, South Asia. Emissions are estimated for each country in the region and for each province of China. Often in this paper we emphasize emissions from China and the rest of East Asia because they dominate the pollution signature along the TRACE-P and ACE-Asia flight paths (in the South China Sea, East China Sea, Yellow Sea, the Sea of Japan, and the western Pacific Ocean).

[5] Figure 2 shows the general methodology for this activity. Emissions are estimated for nine major chemical species: SO_2, NO_x, CO_2, CO, CH_4, nonmethane volatile organic compounds (NMVOC), submicron black carbon aerosol (BC), submicron organic carbon aerosol (OC), and

Figure 1. Definition of the inventory domain, its constituent countries, international shipping lanes, and subregions (vertical lines, China; vertical lines with dark gray shading, East Asia; light gray shading, Southeast Asia; horizontal lines, South Asia).

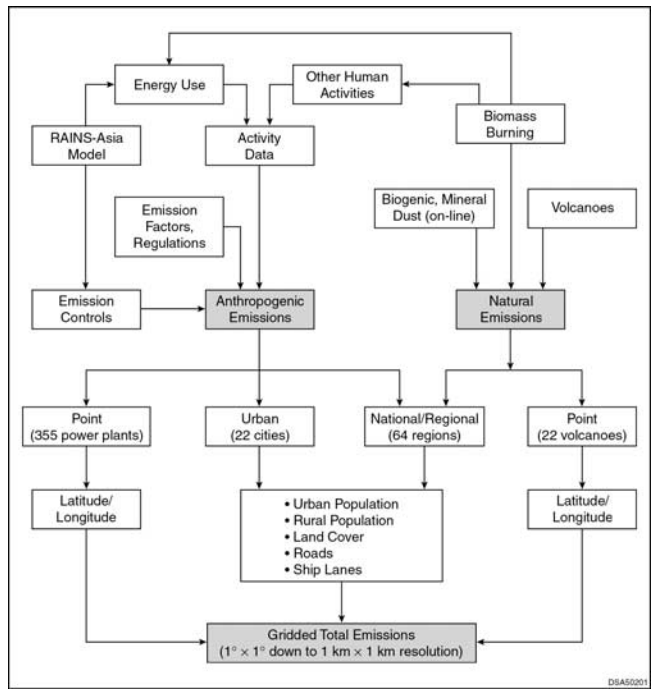

Figure 2. Schematic methodology for the development of Asian emission estimates.

NH_3. In addition, NMVOC emissions are speciated into 19 subcategories based on chemical reactivity and functional groups. Emissions are calculated for all source types believed to contribute significantly to the emissions of a particular chemical species. As examples, ten transportation categories are examined for all species and 82 emitting categories are compiled for NMVOC emissions. For energy use we often rely on the RAINS-Asia computer simulation model, as described below, supplemented with nonenergy activity data for such species as NMVOC, CH_4, and NH_3. Local regulations limiting the emissions of air pollutants are included in the model through the application of specific (required) abatement technologies.

[6] Emissions presented in this paper are for anthropogenic sources only. We do not include such natural sources as volcanic SO_2, biogenic NMVOC, CH_4 from wetlands, NO_x from lightning, or mineral dust; these are calculated online and added into the total gridded emissions as an integral part of the modeling process [*Carmichael et al.*, 2003a]. Neither are primary PM_{10} and $PM_{2.5}$ emissions reported in this paper, but they have been independently estimated by this team in coordination with the BC and OC emissions. However, in section 3.6, we summarize all natural and anthropogenic emissions estimates that were used in TRACE-P and ACE-Asia modeling. Biomass burning is included in this inventory because it is largely caused by human activity. (Even in remote areas it is believed that lightning is not the primary cause of fires.) Biomass burning is divided into three major categories: forest burning, savanna/grassland burning, and the burning of crop residues in the field. The reader should note that we distinguish carefully between this kind of ''open'' biomass burning and the combustion of biofuels (wood, crop residues, dung, etc.) in stoves, cookers, and heaters, which is

considered to be an energy source alongside fossil-fuel combustion. The detailed emission calculations are aggregated into seven primary source categories: industry, residential, transportation, power generation, agriculture, biomass burning, and other. It is at this aggregated level that summaries of emissions are presented later in this paper.

[7] Emission estimates are specifically for the year 2000 so as to incorporate many of the fundamental changes that occurred in Asia in the late 1990s. However, when this project began in late 2000, in order to provide premission modeling support to the field experiments that took place in March and April 2001, very little statistical data had been released for the year 2000 by Asian countries. Thus our approach consists of a mixture of actual year 2000 data, model forecasts for 2000 from a 1995 base, and trend extrapolations from emission estimates for the late 1990s.

[8] Section 3.1 presents the results for each species and compares the results with other literature estimates. Typically, there are no other estimates for a year as recent as 2000, so allowance for growth in the intervening years must be made. This section also compares and contrasts emission estimates among different species, sources, and countries. A separate discussion of the sources of individual organic chemical species and the differences among countries is presented in section 3.2.

[9] Emissions are initially calculated for an annual time period. However, it is recognized that there is considerable seasonal variation for some species, associated with such activities as the burning of fossil fuels for home heating in winter and the temperature dependence of releases of NH_3 from fertilizer application and CH_4 from manure. Biomass burning clearly has a very high degree of seasonality, determined by local agricultural practices and meteorological conditions. Such seasonality is important in preparing emissions for comparison with time-specific field experiments. For this reason we have apportioned annual emissions to daily and monthly emissions using a variety of techniques discussed in section 3.3. The techniques used to develop biomass burning estimates are elaborate and based on national surveys of fires, AVHRR satellite fire counts, and TOMS AI data; they are described in two separate papers [*Woo et al.*, 2003a; *Streets et al.*, 2003]. These efforts concentrated on realistically apportioning annual emissions to the months of March and April, which correspond to the timing of the TRACE-P and ACE-Asia field campaigns, and realistically distributing emissions spatially within the source regions.

[10] As Figure 2 shows, anthropogenic emissions are initially calculated for 64 regions of Asia. In this project we do not separately report emissions from the 22 major cities of Asia that are included in the model. Regional emissions are then gridded at a variety of spatial resolutions for input to the atmospheric simulation models, ranging from $1^\circ \times 1^\circ$ for most regional model applications down to 30 s $\times$ 30 s resolution for urban-scale studies. Each type of emission source is represented by a different spatially resolved surrogate parameter, using such data sets as urban and rural population, road networks, landcover, and ship lanes. Approximately 355 large Asian power plants are separately identified in the RAINS-Asia model and located according to their latitude/longitude coordinates. For input to the atmospheric models, gridded representations of anthropogenic area and point sources are developed separately and natural sources are added at the end. Thus all atmospheric releases are ultimately accounted for, such that comparison with observations can be meaningfully considered. The development of the gridded emission fields, the sources of surrogate GIS data that were used, and some examples are presented in section 3.4.

[11] An uncertainty analysis is presented, focusing on the combined uncertainties of activity levels and emission factors for each species in each of six prototypical regions. The uncertainty results are presented in section 3.5. Finally, in section 3.6, we present comparisons between modeled pollutant concentrations derived using this emission inventory and observed concentrations during the TRACE-P and ACE-Asia missions and at other ground stations. The findings are used to qualitatively assess the level of confidence that can be placed in the inventory values and, in some cases, their apparent shortcomings. All regional and gridded emissions data sets can be examined and downloaded from our emissions website at the University of Iowa (available at http://www.cgrer.uiowa.edu/EMISSION_DATA/index_16.htm).

2. Methodology

[12] The estimation of emissions from such a wide variety of source types, species, and regions cannot be described in complete detail due to space limitations. However, we can give a general overview of the methods, data, and data sources for the key elements of the emission calculations. The general approach used by us to estimate emissions has been described by *Klimont et al.* [2002]. The emissions of a particular species are estimated as a product of the activity rate, the unabated emission factor, and the removal efficiency of any applied emission abatement technologies, using the following equation:

$$E_{j,k} = \sum_{l} \sum_{m} \sum_{n} A_{j,k,l,m} ef_{j,k,l,m} \left(1 - \eta_{j,l,m,n} \alpha_{j,k,l,m,n}\right) X_{j,k,l,m,n} \quad (1)$$

where

- j, k, l, m, n species, region, sector, fuel/activity type, abatement technology;
- E emissions;
- A activity rate;
- ef unabated emission factor;
- η removal efficiency of abatement technology n;
- α maximum application rate of abatement technology n;
- X actual application rate of abatement technology n; note that the set of abatement technologies includes a "no-control" case, such that $\sum_n X = 1$.

[13] The parameter $\alpha_{j,k,l,m,n}$ is used only for NMVOC in this inventory to allow for the fact that emissions are generated by a large number of inhomogeneous sources, and consequently some control options apply only to a fraction of the total activity. Throughout most of developing Asia, there is little abatement technology in place, such that

Table 1. Fuel Use and Vegetation Burned in Asia in 2000

	Industry (PJ)			Domestic (PJ)			Transport (PJ)	Power (PJ)			Biomass Burned (Tg)		
Country	Coal	Oil	Other	Coal	Biofuel	Other	All Fuels	Coal	Oil	Other	Savanna/ Grassland	Forest	Crop Residue
China	5966	2523	2011	2417	7178	998	4246	9806	873	183	52	25	105
Japan	428	2490	1447	22	53	1506	3834	1783	2044	1757	0	1	2
Korea, Rep. of	232	1042	281	0	13	810	1175	752	373	230	0	0	2
Korea, DPR	651	12	21	0	294	24	57	199	0	0	0	1	1
Mongolia	32	14	22	0	22	59	9	51	4	0	23	9	0
Taiwan, China	239	585	186	0	6	136	485	466	316	113	0	0	0
Brunei	0	12	97	0	2	7	19	0	0	25	0	0	0
Cambodia	21	6	8	0	80	3	25	12	3	0	8	5	1
Indonesia	51	454	1059	0	1838	550	969	371	102	218	21	68	6
Laos	0	9	0	0	34	0	15	0	0	0	5	19	1
Malaysia	31	295	136	0	101	71	389	36	102	320	0	22	1
Myanmar	0	14	87	0	348	12	52	0	2	39	2	56	4
Philippines	21	219	183	0	250	109	302	152	105	0	0	17	7
Singapore	0	238	138	0	0	3	94	0	200	70	0	0	0
Thailand	183	243	648	0	319	70	908	320	206	341	12	36	8
Vietnam	70	82	42	19	850	26	101	63	21	23	12	15	6
Bangladesh	5	17	332	3	460	83	68	15	9	160	0	9	11
Bhutan	0	2	0	0	19	1	5	0	0	2	0	1	0
India	2085	893	1144	5	6740	841	2383	4597	133	350	9	37	84
Nepal	5	16	4	1	259	13	11	0	2	0	0	5	2
Pakistan	89	137	423	15	786	204	391	95	209	194	3	1	10
Sri Lanka	0	17	19	0	136	21	52	0	5	0	0	4	0
International Shipping	0	0	0	0	0	0	726	0	0	0	0	0	0
Asia Total	10109	9320	8289	2482	19788	5548	16316	18718	4710	4024	147	330	250

$(1 - \eta_{j,l,m,n}\alpha_{j,k,l,m,n})\, X_{j,k,l,m,n} = 1$, and emissions become simply a product of activity rates and emission factors. Abatement technologies must be considered for many species and sources in the developed countries of Asia (Japan in particular); however, in the rest of Asia it is generally only large sources of PM and SO_2 in the industry and power generation sectors for which abatement technologies must be considered. We discuss below the development of activity rates and emission factors. Additional detailed information on emissions by subsectors and emission factors can be found on the Web site.

2.1. Activity Rates

[14] Because of the need to generate a comprehensive picture of energy use by fuel type, sector, and Asian region, we often used the year 2000 forecasts of the RAINS-Asia simulation model from a 1995 base [*Foell et al.*, 1995; *Streets et al.*, 1999; *Shah et al.*, 2000]. One important exception is that of China, for which fundamental structural changes and a slowdown in the Chinese economy led to a reduction in coal use after 1996–97 [*Sinton and Fridley*, 2000; *Streets et al.*, 2001b], rather than the increase projected by the RAINS-Asia model; for China we therefore developed our own estimates of fuel use by sector and province based on the work of *Sinton and Fridley* [2000]. Table 1 presents the basic information on fossil fuel and biofuel energy use in the year 2000 by country, sector, and fuel type that was used to develop combustion-related emissions. The quantities of biomass burned are also included in Table 1, the derivation of which is described in a separate paper [*Streets et al.*, 2003]. Table 2 identifies the other, noncombustion source types that are included in the emission calculations, though we do not present the activity rates for these source types.

[15] A separate approach was taken for transportation emissions, because of the need to consider the variety of vehicles in use in Asia and their widely differing emission rates. Our approach was to estimate emissions as the product of numbers of vehicles in a country, annual vehicle km traveled, and emission rate in g km^{-1} of pollutant emitted. The numbers of vehicles and distances traveled were obtained where possible from World Road Statistics 2000 [*IRF*, 2000] and World Motor Vehicle Data [*AAMA*, 1998], inferred for some countries and vehicle types from national data sources and extrapolations. Table 3 shows the assumed numbers of each of ten types of vehicles in each country.

[16] The methods for determining activity levels for NMVOC and BC are the same as in previous analyses

Table 2. Noncombustion Activities Included in the Inventory

Species	Activities Included					
CO_2	cement production					
CO	steel production	pig iron production				
CH_4	rice cultivation	animal emissions	landfills	wastewater treatment	coal mining	leakage from oil and gas extraction and use
NMVOC[a]	vehicle refueling and evaporation	oil and gas extraction processes	solvent and paint use	chemical industry	miscellaneous industry	waste disposal
NH_3	cattle	pigs	other animals	fertilizer application	waste treatment and disposal	nitrogen fertilizer manufacturing

[a]Eighty-two source categories were considered for NMVOC. See *Klimont et al.* [2002].

Table 3. Numbers of Motor Vehicles by Country in Asia in 2000[a]

	Cars		Buses		Trucks					
Country	LDGV (Gas)	LDDV (Diesel)	HDGV (Gas)	HDDV (Diesel)	LDGT (Gas)	LDDT (Diesel)	HDGV (Gas)	HDDV (Diesel)	Tractors	Motorcycles
China	6205	773	368	217	9161	969	2075	2129	10124	13754
Japan	38976	15082	2	233	5089	4831	472	10019	144	13785
Korea, Rep. of	6926	2680	8	840	640	607	59	1259	161	3000
Korea, DPR	164	2	10	6	249	28	59	59	235	209
Mongolia	40	1	3	2	22	2	5	5	9	17
Taiwan, China	3764	6	0	23	177	168	16	349	169	11168
Brunei	166	0	0	2	4	6	0	4	3	5
Cambodia	68	0	0	1	5	7	0	4	1	568
Indonesia	2835	71	142	570	454	681	49	438	814	13635
Laos	11	0	0	1	2	3	0	2	20	356
Malaysia	3481	5	10	40	173	259	19	167	296	4928
Myanmar	24	58	116	466	12	18	1	11	181	3921
Philippines	778	147	293	1173	77	116	8	74	33	1200
Singapore	301	3	0	12	35	33	3	68	21	139
Thailand	2142	252	505	2019	1204	1807	129	1161	230	13980
Vietnam	1733	94	188	751	345	518	37	333	355	3620
Bangladesh	43	3	3	27	12	18	1	12	3	211
Bhutan	9	0	0	1	1	2	0	1	8	12
India	5300	54	54	483	614	922	66	593	4502	29486
Nepal	117	1	1	12	15	23	2	15	98	613
Pakistan	611	9	9	82	90	136	10	87	581	2131
Sri Lanka	276	6	6	58	96	143	10	92	118	839
Asia total	73970	19249	1722	7017	18478	11298	3024	16885	18106	117577

[a]Data are $\times 10^3$.

[*Klimont et al.*, 2001, 2002; *Streets et al.*, 2001c]. Activity levels for agricultural and related data needed for estimating CH_4 and NH_3 emissions were derived from national statistics wherever possible [e.g., *NBS*, 2000] and international statistics [e.g., *IFA*, 1998; *FAO*, 2000; *OECD*, 1995]. Often, data for 2000 were not available and were extrapolated from recent statistical information for 1998 and 1999 or derived from short-term forecasts [*OECD*, 1999].

2.2. Emission Factors

[17] Emission factors were developed from a wide range of sources. Table 4 summarizes the ranges of emission factors that were used for fossil fuel and biofuel combustion in different sectors. The ranges presented cover different countries or provinces; typically, the low end of the ranges applies to the most developed regions, like Japan or Hong Kong, and the upper end of the ranges applies to less-developed regions, like Laos or Xinjiang Province.

[18] Emission factors for vehicles were derived using the MOBILE5 model, as described for China in the work of *Fu et al.* [2001]. Emission factors were determined for each of the ten vehicle categories for six prototypical countries: China; India; Japan; Republic of Korea; Taiwan, China; and Thailand. Other countries were then assigned the same emission factors as the modeled country they most closely resembled. Emission factors for open biomass burning were taken from *Andreae and Merlet* [2001] and are shown in Table 5. Emission factors for CH_4 essentially followed IPCC methodology [*IPCC*, 1997], with special use of the China Country Study [*Tsinghua University et al.*, 1999]. Emission factors for NH_3 are based on *Bouwman et al.* [1997] but with livestock emissions adjusted for region-specific production efficiency of milk and meat and fertilizer emissions adjusted for differences in nitrogen losses between temperate and tropical regions [*Klimont et al.*, 2001]. For some source types that we believe are similar in Asia and the West (large power plants, for example), we used USEPA AP-42 emission factors [*USEPA*, 2002, available at http://www.epa.gov/ttn/chief/ap42/index.html]; these values were sometimes adjusted to reflect poorer performance of Asian technologies (see section 3.1.4). Emission factors and particulate collection efficiencies for BC are described by *Streets et al.* [2001c], and an overview of our assumptions about BC and OC emission factors will be reported elsewhere in the context of a global inventory (T. C. Bond et al., submitted manuscript, 2003).

[19] In selecting emission factors to be used in the development of this inventory, we were struck by the wide variation in literature estimates. One simple illustration of this variation can be obtained by comparing CO emissions from the stove tests of *Zhang et al.* [1999] and *Venkataraman and Rao* [2001]. The former, testing 15 wood-fired stove types in India and China, obtained a mean estimate of 78 g kg^{-1} CO, with values in the range of 42–170 g kg^{-1}. For four stoves fired with dung cake the values obtained by *Zhang et al.* [1999] were 43 (30–61) g kg^{-1} CO. *Venkataraman and Rao* [2001], on the other hand, report measured values of 11–12 g kg^{-1} CO for four Indian wood-fired stoves and 14–29 g kg^{-1} CO for four Indian stoves fired with dung cake. Such differences point out the very great and urgent need for comprehensive and rigorous testing of emission sources in the developing world and also partly explain why our uncertainty estimates (section 3.5) are so large for species like CO, BC, and OC, the emissions of which are dominated by small combustion sources.

3. Results

3.1. Asian and National Emission Estimates

[20] This section presents national and regional summaries of emissions of each of the nine major species studied and places them in the context of other emissions values

Table 4. Emission Factors for Fuel Combustion[a]

	Industry		Domestic		Transport			Power	
Country	Coal	Oil	Coal	Biofuel		Oil		Coal	Oil
					SO_2				
China	0.77 ~ 117	0.58 ~ 11.6	3.29 ~ 54.8	0.66 ~ 4.12		3.17 ~ 10.3		3.81 ~ 112.6	3.11 ~ 11.6
Japan	0.98 ~ 46.9	0.19 ~ 15.3	5.57 ~ 44.4	0.79 ~ 2.52		0.19 ~ 16.0		5.57 ~ 44.5	0.19 ~ 15.3
India	1.32 ~ 38.8	9.57 ~ 77.5	7.94 ~ 15.9	0.18 ~ 1.11		9.75 ~ 81.3		8.38 ~ 16.0	9.75 ~ 77.5
Other	5.57 ~ 121.7	3.24 ~ 72.0	5.28 ~ 121.7	0.65 ~ 1.53		3.95 ~ 25.3		11.7 ~ 121.7	19.31 ~ 72.0
					NO_x				
China	2.38 ~ 6.44	3.35 ~ 7.26	1.19 ~ 2.24	0.54 ~ 0.68		15.0 ~ 58.2		4.59 ~ 11.8	2.1 ~ 8.54
Japan	0.51 ~ 6.44	3.35 ~ 7.26	1.19 ~ 2.24	0.54 ~ 0.68		15.0 ~ 52.0		1.7 ~ 11.8	2.1 ~ 8.54
India	3.4 ~ 6.44	3.35 ~ 7.26	1.19 ~ 2.24	0.54 ~ 0.68		15.0 ~ 58.2		4.59 ~ 11.8	3.35 ~ 8.54
Other	3.4 ~ 6.44	3.35 ~ 7.26	1.19 ~ 2.24	0.54 ~ 0.68		15.0 ~ 58.2		4.59 ~ 11.8	3.35 ~ 8.54
					CO				
					LDV	HDV	MC		
China	37.6		74.0	43 ~ 77	(1.0 ~ 69.3)	(17.7 ~ 135)	(11.5 ~ 21.1)		
Japan	1.35		74.0	5.5	(2.1 ~ 6.8)	(6.8 ~ 40.8)	(10.0)		
India	12.6		74.0	43 ~ 77	(17.4 ~ 60.0)	(17.4 ~ 120)	(21.0)		
Other	37.6		74.0	43 ~ 77	(12.1 ~ 32.7)	(11.5 ~ 40.8)	(13.0)		
					BC				
China	0.056 ~ 0.6	0.25 ~ 0.36	0.12 ~ 3.7	1.00	(0.01 ~ 0.19)	(0.02 ~ 0.27)	(0.01)		0.36
Japan	0.0081 ~ 0.06	0.25 ~ 0.36	0.12 ~ 3.7	1.00	(0.003 ~ 0.11)	(0.01 ~ 0.16)	(0.001)		0.36
India	0.056 ~ 0.6	0.25 ~ 0.36	0.12 ~ 3.7	1.00	(0.01 ~ 0.60)	(0.04 ~ 0.87)	(0.01)		0.36
Other	0.030 ~ 0.37	0.25 ~ 0.36	0.12 ~ 3.7	1.00	(0.01 ~ 0.19)	(0.02 ~ 0.27)	(0.01)		0.36
					OC				
China	0.0081 ~ 0.056	0.19 ~ 0.27	0.12 ~ 3.00	5.00	(0.01 ~ 0.13)	(0.03 ~ 0.18)	(0.04)		0.27
Japan	0.003 ~ 0.0081	0.19 ~ 0.27	0.12 ~ 3.00	5.00	(0.006 ~ 0.13)	(0.02 ~ 0.18)	(0.001)		0.27
India	0.029 ~ 0.056	0.19 ~ 0.27	0.12 ~ 3.00	5.00	(0.05 ~ 0.41)	(0.14 ~ 0.59)	(0.07)		0.27
Other	0.018 ~ 0.030	0.19 ~ 0.27	0.12 ~ 3.00	5.00	(0.01 ~ 0.13)	(0.03 ~ 0.18)	(0.04)		0.27
					CO_2				
All	1462 ~ 2510	3011 ~ 3115	1869 ~ 2028	1018 ~ 1282		3252		2243 ~ 2313	3011 ~ 3115

[a]Emission factors are in g kg^{-1}, except for values in parenthesis, which are in g km^{-1}.

reported in the literature. It also identifies the major source types contributing to the emissions of each species. Table 6 summarizes national emissions of each species.

3.1.1. Sulfur Dioxide

[21] Of all the pollutants emitted in Asia, SO_2 is best known. Prompted by concerns over high ambient SO_2 concentrations in Asian cities and the threat of acid rain, Japanese researchers commenced studies of SO_2 emissions in Asia more than 10 years ago [*Fujita et al.*, 1991; *Kato and Akimoto*, 1992; *Akimoto and Narita*, 1994]. SO_2 emissions in Asia rose steadily to a peak of 39.9 Tg in 1996 [*Streets et al.*, 2000a], as ever-increasing quantities of fossil fuels were used to drive rapidly growing industrial economies. Since 1996, however, SO_2 emissions have slowly declined, due to a combination of economic slowdown, environmental regulations, and the fundamental restructuring of the Chinese industrial economy [*Sinton and Fridley*, 2000]. We estimate that Asian SO_2 emissions in 2000 were 34.3 Tg, which is in good agreement with the time trend of *Streets et al.* [2000a] through 1997. The power generation sector, dominated by coal-fired power plants in China, produced 45% of total Asian SO_2 emissions in 2000, and the industrial sector produced 36%.

[22] Emissions of SO_2 in China in 2000 are estimated to be 20.4 Tg. This is in excellent agreement with the official Chinese estimate of 19.95 Tg [*SEPA*, 2000]. Though we do not know the details of SEPA's calculation, it is likely that it does not include some contributing sources that we do include, such as domestic biofuel combustion (450 Gg), biomass burning (83 Gg), and Hong Kong sources (76 Gg), so our estimate may actually be in even better agreement. An estimate for 1995 of 25.2 Tg [*Streets and Waldhoff*, 2000] is higher than the official SEPA value of 23.7 Tg, for reasons discussed in the text of that paper. We estimate emissions of 5.54 Tg in India in 2000. This can be compared with recent estimates of 4.35 Tg for 1996–1997 by *Reddy and Venkataraman* [2002a, 2002b] and 4.64 Tg for 1995 [*Garg et al.*, 2001b]. India's emissions of SO_2 have grown unabated throughout the 1990s, at an annual rate of about 5.5% according to *Garg et al.* [2001b], so our estimate is quite consistent. Our estimates for other countries in Asia, such as 801 Gg for Japan; 829 Gg for the Republic of Korea; 376 Gg for Taiwan, China; and 961 Gg

Table 5. Emission Factors for Open Biomass Burning[a]

Species	Savanna/ Grassland	Tropical Forest	Extra-tropical Forest	Crop Residue
SO_2	0.35	0.57	1.00	0.40
NO_x	3.90	1.60	3.00	2.50
CO_2	1613	1580	1569	1515
CO	65	104	107	92
CH_4	2.30	6.80	4.70	2.70
NMVOC	9.73	19.32	27.79	15.70
BC	0.48	0.66	0.56	0.69
OC	3.40	5.20	9.15	3.30
NH_3	1.05	1.30	1.40	1.30

[a]Emission factors are in g kg^{-1}. Source: *Andreae and Merlet* [2001].

Table 6. Summary of National Emissions of Each Species in Asia in 2000[a]

Country	SO_2	NO_x	CO_2	CO	CH_4	NMVOC	BC	OC	NH_3
China	20,385	11,347	3,817	115,749	38,356	17,432	1049	3385	13,570
Japan	801	2198	1,203	6806	1143	1920	53	74	352
Korea, Rep. of	829	1322	411	2824	1433	1161	22	28	172
Korea, DPR	227	273	120	3556	1345	234	22	106	98
Mongolia	101	221	69	2861	472	452	19	173	155
Taiwan, China	376	521	200	2127	560	510	8	10	152
Brunei	6	20	10	15	50	43	0	0	2
Cambodia	40	89	36	1707	708	305	14	89	86
Indonesia	884	1317	587	23,105	6443	6903	206	1138	1390
Laos	21	96	44	2547	387	486	18	129	58
Malaysia	273	494	144	5552	869	1424	26	151	146
Myanmar	65	226	145	8446	2691	1671	65	421	341
Philippines	713	326	152	4102	2563	1398	36	192	273
Singapore	163	185	56	138	85	81	3	2	4
Thailand	961	1086	351	10,815	3567	3052	72	364	388
Vietnam	193	283	169	9248	2907	1390	88	432	686
Bangladesh	140	220	123	4827	3608	819	52	268	763
Bhutan	6	8	4	172	42	36	2	12	10
India	5536	4591	1,886	63,340	32,851	10,844	600	2837	7399
Nepal	38	55	39	2,087	917	346	21	135	168
Pakistan	1416	539	221	7076	5415	1344	85	368	1214
Sri Lanka	58	57	28	1348	407	275	11	55	92
International shipping	1083	1292	51	117	1	27	68	51	0
Asia total	34,316	26,768	9,868	278,564	106,821	52,150	2541	10,420	27,519

[a]Data are in Gg (Tg for CO_2).

for Thailand, can be seen to be reasonable from both the long-term modeled trends and the official estimates through 1997 presented by *Streets et al.* [2000a]. Additional support for the validity of our SO_2 emission estimates is provided by the good agreement for SO_2 and SO_4 concentrations and wet sulfate deposition between values simulated by eight long-range transport models using our emission estimates and observations at 18 surface sites in East Asia [*Carmichael et al.*, 2002a]. A detailed treatment of long-term SO_2 emission and deposition trends can be found in the work of *Carmichael et al.* [2002b].

3.1.2. Nitrogen Oxides

[23] We estimate that total NO_x emissions in Asia were 26.8 Tg in 2000 (all values herein calculated as NO_2). There are no other estimates of recent NO_x emissions in Asia against which to compare. *Akimoto and Narita* [1994] estimated Asian emissions in 1987 to be 15.5 Tg, *van Aardenne et al.* [1999] estimated emissions in 1990 to be 19.2 Tg with a projection for the year 2000 of 31.9 Tg, and the 1990 value for Asia from the global inventories of *Olivier et al.* [1996] is 23.1 Tg. All researchers agree that the rapid growth of the transportation sector in Asia has caused NO_x emissions to climb steeply. Therefore our numbers seem reasonable continent-wide and consistent with historical NO_x emission trends and other earlier Asian estimates [*Streets et al.*, 2001a]. The transportation sector has the highest share of Asian NO_x emissions (37%), followed by power generation (27%) and industry (18%).

[24] For China, we calculate that emissions in 2000 were 11.3 Tg. Power generation has the largest contribution in China (39%), with industry and transportation both contributing about 25%. *Hao et al.* [2002] recently published estimates of NO_x emissions from commercial energy use in China in the period 1995–1998. A declining trend since 1996 is presented by *Hao et al.* [2002] (12.03 Tg in 1996, 11.66 Tg in 1997, and 11.18 Tg in 1998), consistent with the declines in emissions of other species in China reported elsewhere in this paper. Because *Hao et al.* [2002] only consider commercial energy, our value can be expected to be higher because of the inclusion of biofuel consumption and biomass burning. We also show a lower share of industrial emissions, consistent with reduced industrial coal use [*Sinton and Fridley*, 2000], and a higher share of transportation emissions, due to the rapid growth in vehicle ownership in China in the latter part of the 1990s. The value of 11.99 Tg for 1995 reported by *Streets and Waldhoff* [2000] is consistent with the value of *Hao et al.* [2002] for 1996 but higher than that reported by *Klimont et al.* [2001] for 1995, i.e., 9.7 Tg. *Wang et al.* [1996] reported that NO_x emissions in China in 1990 were 8.4 Tg, showing just how fast emissions grew during the early to mid-1990s.

[25] For much of the rest of East Asia (Japan; Republic of Korea; and Taiwan, China) our estimates are in good agreement with government statistical reports. For the rest of Asia, however, there are few estimates of NO_x emissions against which to compare. *Garg et al.* [2001b] report an estimate of 3.46 Tg for India's NO_x emissions in 1995, growing at an annual rate of about 5.5%. This value tallies well with our year-2000 estimate of 4.59 Tg. *Garg et al.* [2001b] calculate that in 1995 the power and transportation sectors both contributed about 28% to total NO_x emissions in India; our work suggests that by 2000 the share of the transportation sector had risen to 34%, while power generation had dropped to 26%.

3.1.3. Carbon Dioxide

[26] Though this study is not intended to support global climate change studies, atmospheric scientists are interested in local CO_2 concentrations and their relationships to concentrations of other carbon compounds like CO and CH_4. Our estimate for CO_2 emissions in Asia in the year 2000 is 9870 Tg. This estimate consists of all direct releases, including biomass burning, biofuel combustion, and releases from cement production, but it does not include any uptake from growing vegetation. Some researchers do

not include vegetation burning in their CO_2 accounting, arguing that the resource is renewable and the released CO_2 is taken up by regrowth of the vegetation in later time periods. We prefer to separate the accounting of CO_2 release from that of CO_2 uptake. We have shown [*Streets and Waldhoff*, 1999] that throughout Asia vegetation combustion is so inefficient that it releases large quantities of non-CO_2 gases and particles (such as CO, CH_4, NMVOC, and carbonaceous aerosols), which may not be returned to the vegetative ecosystem as usable carbon for some time. This can amount to 10–15% of the carbon in the fuel. In addition, much harvesting and burning of vegetation in Asia is nonsustainable. We leave it to others to balance this released CO_2 with carbon uptake in Asia.

[27] As part of its global emissions inventory, the IPCC estimates 7440 Tg for anthropogenic CO_2 releases in Asia in 2000 [*Nakicenovic et al.*, 2000], compared with our value of 8730 Tg (without the inclusion of open biomass burning but including biofuels). It is not clear if the suite of sources included in the two studies is the same. We estimate the distribution of Asian CO_2 emissions across sectors as 27% residential (including biofuels), 23% industry, 23% power generation, 12% transportation, and 12% biomass burning. This contrasts with the larger proportion of transportation and smaller share of residential emissions in the West.

[28] For China, we estimate CO_2 emissions to be 3820 Tg. This is the same as the value we reported in a separate study of recent trends in China's CO_2 emissions, allowing for the inclusion of carbon uptake by soils and growing vegetation in that study [*Streets et al.*, 2001b]. Other available estimates of China's CO_2 emissions [e.g., *Tsinghua University et al.*, 1999; *Wu and Wei*, 1997] should be compared with caution, because they are usually for an earlier time period like 1990 and omit biofuel combustion and biomass burning. For India, we estimate 1890 Tg. *Garg et al.* [2001a] report 778 Tg CO_2 in 1995 but apparently without the inclusion of emissions from biofuel use or biomass burning. As with continent-wide emissions, there is little reliable data for individual countries, and the data that do exist are inconsistent with regard to inclusion of various types of sources and sinks.

3.1.4. Carbon Monoxide

[29] Carbon monoxide emissions in Asia are not known with any great confidence because emission factors are highly dependent on the efficiency of the combustion process and the operation and maintenance of the equipment, none of which have ever been surveyed in a systematic manner. Throughout most of developing Asia, equipment performance is poor and CO emissions are high. We estimate that Asian emissions of CO were 279 Tg in 2000, of which 67 Tg, or 24%, comes from open biomass burning. The energy component is largely due to residential biofuel combustion (96 Tg or 34%) and transportation sources (78 Tg or 28%). *Olivier et al.* [1996] report 290 Tg for 1990, but this value is believed to be high due to overestimation of emissions from the burning of agricultural waste (see EDGAR emission inventory web site for a discussion of the differences between EDGAR Versions 2 and 3 at http://arch.rivm.nl/env/int/coredata/edgar/index.html.)

[30] Our estimate for China is 116 Tg. This value is high because vehicles in China are relatively high emitters of CO [*Fu et al.*, 2001] and because emission factors for small coal combustors are 2–3 times higher than comparable sources in the West [*U.S. EPA*, 2002]. There is a pressing need to undertake a program of field testing of sources in the region, particularly transportation and industrial, so that emissions of CO and other products of incomplete combustion can be quantified more reliably. The only other estimates of CO emissions in China are 87.4 Tg for 1994–1995 by *Tonooka et al.* [2001] and our earlier estimate of 113 Tg for 1995 [*Streets and Waldhoff*, 2000]. We believe that CO emissions in China are relatively stable today, as vehicles, industrial processes, and household energy sources with improved combustion efficiencies gradually replace outdated technology. We estimate CO emissions in India to be 63.3 Tg. Emissions in Southeast Asia are 65.7 Tg, of which almost 50% originates from biomass burning practices, especially slash-and-burn agriculture and deforestation.

[31] The CO estimates for India are very sensitive to the assumed emission factors for biofuel combustion in small stoves. As discussed earlier, *Zhang et al.* [1999] and *Venkataraman and Rao* [2001] agree that emission factors, especially for the burning of dung cake, are very high, but the magnitude is highly dependent on stove design and combustion conditions, both of which are poorly known for vast areas of developing South Asia. Emissions from Indian vehicles are also thought to be high, especially for two-wheelers and two-stroke engines in general [*Das et al.*, 2001]. *Bhattacharya et al.* [2000] have estimated the emissions of several species from biofuel use in Asian countries, and the results for CO emissions are informative, even though their results are characteristic of the early 1990s. The two sets of results are in good agreement for a number of countries, e.g., (our values first) China (36 Tg versus 31 Tg), Pakistan (4.1 Tg versus 2.8 Tg), Philippines (1.2 Tg versus 1.4 Tg), and Sri Lanka (610 Gg versus 740 Gg). However, for India, our value is considerably higher (29 Tg versus 9.9 Tg), as it is for Vietnam (4.3 Tg versus 1.9 Tg). The differences are partly due to the use of different emission factors and partly due to differences in estimates of the amounts of biofuel used, with the countries that disagree the most having the greatest differences in biofuel usage estimates. CO emissions are further discussed in section 3.6.

3.1.5. Methane

[32] Estimation of CH_4 emissions in Asia requires consideration of many varied source types not considered for other species. In this work we estimate emissions from rice cultivation, animals (both manure and enteric fermentation), landfills, wastewater treatment, coal mining and combustion, oil and gas extraction and combustion, biofuel combustion, and biomass burning. Emissions from wetlands are considered a natural source and are not included in this anthropogenic inventory. Our estimate of CH_4 emissions for all of Asia in 2000 is 107 Tg. The IPCC estimate is 125 Tg [*Nakicenovic et al.*, 2000], while the EDGAR inventory reports 118 Tg for 1990 [*Olivier et al.*, 1996]. China is the largest emitter in our inventory at 38.4 Tg, followed closely by India at 32.9 Tg. This estimate for China is higher than the value of 33.3 Tg that we reported in the work of *Streets et al.* [2001b] due to the inclusion of several additional source types and our own estimation of biomass burning emissions rather than that of *Olivier et al.* [1996]. The estimate from the China Country Study is 32.4 Tg for 1990 [*Tsinghua University et al.*, 1999].

[33] We find that emissions from manure and enteric fermentation in animals represent the largest source of CH_4 emissions in Asia at 36.2 Tg (34%), followed by rice cultivation (24.2 Tg, 23%), wastewater treatment (14.0 Tg, 13%), landfills (8.9 Tg, 8.3%), biofuel combustion (8.6 Tg, 8.1%), coal mining and combustion (8.4 Tg, 7.9%), oil and gas extraction and use (3.4 Tg, 3.1%), and biomass burning (3.1 Tg, 2.9%). The distribution of CH_4 emissions among source types varies considerably from country to country. In China, for example, rice cultivation is the largest source type (26% of national total), while in India animal emissions dominate (54%), and in Japan and the Republic of Korea landfills are the largest source type (31% and 39%, respectively). These balances of source types are continually shifting as national priorities change. For example, oil and gas extraction and use is rapidly increasing in China; municipal waste incineration is replacing landfills in developed countries with limited land area, like Japan, Singapore, and Taiwan, China; and biomass burning is now declining in Southeast Asian countries.

[34] It is interesting to compare our CH_4 values for China with other data sources. We estimate emissions of 9.8 Tg from rice cultivation. This is significantly lower than the estimate by *Verburg and Denier van der Gon* [2001] of 20.2 Tg for 1991. Our method is based on the detailed analysis by *Tsinghua University et al.* [1999], which yielded a value of 11.2 Tg. Even Verburg and Denier van der Gon acknowledge that "most recent estimates are . . . between 9 and 15 Tg," though they claim that rice harvested area is underreported in Chinese statistics. Other poorly known factors like the degree of organic amendment, irrigation, and flooding introduce considerable uncertainty into the estimate (see also *Lin et al.* [1997]). Our results for CH_4 emissions from rice cultivation for China and the rest of Asia are in good agreement with recent studies by *Yan et al.* [2003a, 2003b]. For CH_4 emissions from enteric fermentation and manure management in China, our value of 8.9 Tg for 2000 is in good agreement with the value of 8.8 Tg for 1991 from *Verburg and Denier van der Gon* [2001]. Our estimate for CH_4 emissions in India is 32.9 Tg, dominated by emissions from animals, particularly cattle. This is considerably higher than the value of 18 Tg reported by *Garg et al.* [2001a] for 1995. Resolution of differences must await a detailed side-by-side comparison of emission factors and activity levels.

3.1.6. Nonmethane Volatile Organic Compounds

[35] Emissions of nonmethane volatile organic compounds (NMVOC) in Asia are subject to a high degree of uncertainty due to the great variety of sources, many of which are poorly known in terms of their operating characteristics and emission controls, nor have many other attempts been made to estimate NMVOC emissions in Asia. We estimate anthropogenic emissions in all of Asia to be 52.2 Tg (measured as full molecular weights of the constituent compounds); this paper does not address biogenic emissions. Emissions are largest from the residential combustion of coal and biofuels (about 34%) and from transportation (27%). We believe that emissions of NMVOC are growing rapidly in Asia as countries extract and use petroleum products and natural gas in increasing amounts, expand the production of industrial organic chemicals, use paints and other surface coatings, and develop high-technology industries that use solvents for cleaning. *Olivier et al.* [1996] estimated 1990 emissions of NMVOC in Asia to be 53.3 Tg; however, this value is believed to be high because of over-estimation of emissions from agricultural waste burning (see section 3.1.3). A global inventory by *Piccot et al.* [1992] has a value of 20.4 Tg for a period characteristic of Asia in the mid-1980s.

[36] Our NMVOC methodology for Asia is fully described by *Klimont et al.* [2002], which addressed China only. The year 2000 value for China in that report is 15.6 Tg. The China estimate presented in this analysis is 17.4 Tg, the difference being due to the addition of emissions from savanna and forest burning. To the best of our knowledge there are no estimates of NMVOC emissions in China made by Chinese researchers. *Tonooka et al.* [2001] calculated China's NMVOC emissions in 1994/1995 to be 13.9 Tg, without open biomass burning, which is quite consistent with this work. For all of East Asia, we estimate 21.7 Tg for 2000, compared with 17.1 Tg for 1995 by *Klimont et al* [2001] and 17.7 Tg for 1994/1995 by *Tonooka et al.* [2001], both without full biomass burning emissions, so again agreement is reasonable. Outside East Asia there are no NMVOC emission estimates against which to compare, so we simply state that our estimate for all of Southeast Asia is 16.8 Tg and for India 10.8 Tg, both of which are quite uncertain due to lack of detailed information about industrial activities and evaporative sources. Section 3.3 discusses the speciation of total NMVOC emissions into its chemical components.

3.1.7. Black Carbon

[37] Submicron black carbon emissions in Asia are estimated to be 2.54 Tg, with a high degree of uncertainty, as discussed in section 3.5. The approach taken and emission factors chosen are consistent with *Streets et al.* [2001c] for China. The residential sector, consisting of the combustion of coal, kerosene, and biofuels in stoves, cookers, and heaters, generates the majority (64%) of these emissions. The other large source is biomass burning (18%), especially in Southeast Asia. Because BC emission factors have been revised significantly since the earlier work of *Cooke et al.* [1999], as discussed by *Streets et al.* [2001c], their estimate of 2.04 Tg of submicron BC in Asia for 1984 without biofuels or biomass burning is not directly comparable with our work.

[38] We estimate 1.05 Tg for China in 2000, significantly less than the value of 1.34 Tg we reported for 1995 [*Streets et al.*, 2001c]. This real reduction we attribute to a combination of several factors: economic downturn in the late 1990s; improved energy efficiency; the conversion of many residential users from raw coal to coal briquettes and cleaner fuels like LPG and electricity, especially in cities; and a concerted effort to clean up the air in China's more polluted cities through the closure of small coal-burning factories. As reported by *Streets et al.* [2001c], BC emissions in China are dominated by the residential sector (781 Gg, 74%), due to the continued direct combustion of coal and biofuels throughout much of China. Though emissions from individual diesel and gasoline vehicles tend to be high in China and the use of vehicles of all kinds grows rapidly, the transport sector is still a small part of the Chinese economy and contributes only 6% to national BC emissions. The value of *Cooke et al.* [1999] for China is 1.15 Tg (but see above).

[39] We estimate BC emissions in India to be 600 Gg, though this is even more uncertain than the estimate for China because of doubts about the emission factors of Indian vehicles, the degree of fuel adulteration in Indian vehicles, and emission factors for dung-burning cookstoves. *Reddy and Venkataraman* [2002a, 2002b] estimate Indian BC emissions to be 380 Gg for 1998–1999, using a similar approach and emission factors to ours, though the belief has been expressed in the literature that BC emissions could be considerably higher [*Dickerson et al.*, 2002], possibly as high as 2–3 Tg, based on atmospheric measurements during INDOEX. While the source strength of BC is indeed uncertain, comparison between models and ambient measurements is further confounded by inconsistencies between different BC measurement techniques. BC emissions are further discussed in section 3.6.

3.1.8. Organic Carbon

[40] Submicron organic carbon emissions in Asia are estimated to be 10.4 Tg C, with a high degree of uncertainty similar to BC. As for BC, residential fuel combustion represents the largest share of the Asian total (6.7 Tg, 65%), but for OC biomass burning is also large (3.3 Tg, 32%). All other sources of OC are relatively small. *Cooke et al.* [1999] report OC emissions of 2.44 Tg submicron OC for Asia in 1984, but the same caveats apply as for BC noted above; the values of *Cooke et al.* [1999] are 2–3 times lower than values obtained by *Liousse et al.* [1996], which included biofuel combustion and open biomass burning.

[41] For China we obtain an OC emission estimate of 3.38 Tg C, 2.57 Tg (76%) from residential fuel combustion and 0.73 Tg (22%) from biomass burning. These two source types are even more dominant for China. The value of *Cooke et al.* [1999] for China is 1.54 Tg. Our estimate for India is 2.84 Tg. This can be compared with 1.25 Tg for 1998–1999 from *Reddy and Venkataraman* [2002a, 2002b]. Differences between emission estimates for both BC and OC highlight the need for more source testing and intercomparison of measurement methods for primary aerosols.

3.1.9. Ammonia

[42] The major sources of ammonia are not energy-related, so they require a greater emphasis on agricultural activities. The large quantities of fertilizer, specifically urea and ammonium bicarbonate (ABC), used in China make this activity a big source of NH_3 emissions. We estimate that NH_3 emissions in Asia in 2000 were 27.5 Tg. *Zhao and Wang* [1994] estimated that annual Asian NH_3 emissions for the period 1989/1991 were 24.6 Tg, while the global inventory of *Olivier et al.* [1998] gives 22.5 Tg for 1990. Growth in agricultural production and fertilizer application in the decade of the 1990s can account for the increase. Similar growth rates for East Asian NH_3 emissions are reported by *Klimont et al.* [2001]. Our shares of Asian NH_3 emissions are: fertilizer application 45%, animals 38%, miscellaneous sources (humans, pets, waste disposal, fertilizer manufacture, etc.) 7.7%, biofuel combustion 6.1%, and biomass burning 3.3%.

[43] For China, we estimate NH_3 emissions in 2000 were 13.6 Tg, of which 50% comes from fertilizer application and fully 88% from the agriculture sector as a whole. This is in good agreement with *Zhou and Wang* [1994], though their China estimate of 13.6 Tg in 1989/1991 is larger than would be consistent with our approach. This can be largely explained by a discrepancy in estimates of fuel combustion; *Zhou and Wang* [1994] estimated significantly larger emissions from this source. *Bouwman et al.* [1997] agree with us that fuel combustion is a minor source of NH_3. Our estimate for NH_3 emissions from agriculture in Japan, 266 Gg, agrees better with the estimate for 1990 of *Murano et al.* [1995] of 200 Gg than the higher value for Japan of *Zhou and Wang* [1994] of 346 Gg for 1989/1991. For India our estimate is 7.4 Tg NH_3.

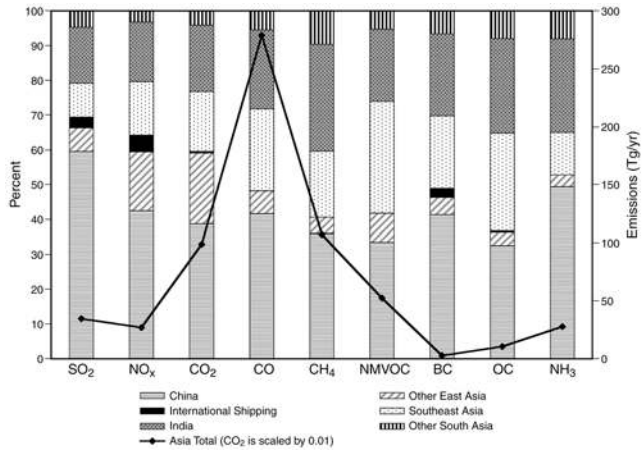

Figure 3. Species emissions by Asian region and absolute values of emissions.

[44] It needs to be stressed that estimates of NH_3 emissions carry a significant uncertainty linked to the use of European-based emission factors (especially for livestock). This applies to studies performed by both European and Asian researchers. Practices in the Asian livestock industry are different from those in Europe, but there is insufficient information to adjust European emission factors for the important variables since measurements in Asia are lacking. This observation can generally be applied to a number of sources and species in this inventory (particularly NMVOC), though not all.

3.1.10. Intercomparison of Emissions

[45] Table 6 summarizes the emissions of each species in each Asian country and shows that China dominates emissions of most species. China contributes the following percentages of total Asian emissions of each species: 59% SO_2, 42% NO_x, 39% CO_2, 42% CO, 36% CH_4, 33% NMVOC, 41% BC, 32% OC, and 49% NH_3. India also contributes significantly to the totals, as follows: 16% SO_2, 17% NO_x, 19% CO_2, 23% CO, 31% CH_4, 21% NMVOC, 24% BC, 27% OC, and 27% NH_3. Other countries contribute much smaller individual shares, though Japan becomes important for CO_2 (12%).

[46] Figure 3 shows the shares of Asian emissions among different source regions. Southeast Asia contributes significantly to emissions of CO, NMVOC, and OC, due to the strong component of biomass burning. South Asia, including India, is a major contributor to CH_4 and NH_3 emissions because of the agrarian component, especially livestock and rice production. Other East Asian countries, being at a higher level of economic development and industrialization, contribute most to NO_x and CO_2 emissions.

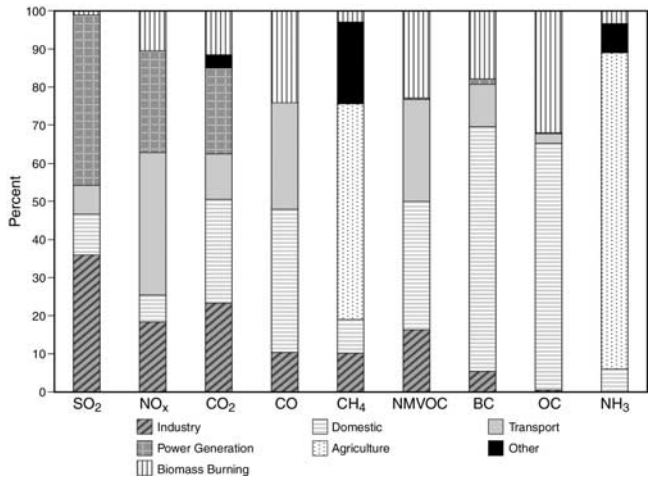

Figure 4. Species emissions by emitting sector.

[47] Figure 4 is particularly informative in its depiction of the source types that contribute most to the emissions of each species. It can be seen that the traditional air pollution sources, namely industry, power generation, and transportation, indeed contribute most to the traditional air pollutant species that have been most studied in the West, SO_2 and NO_x. However, for carbonaceous species, there is much more involvement of other source types in Asia. Emissions of the primary aerosol species BC and OC, for example, are dominated by the residential and biomass burning sectors, and emissions of CH_4 and NH_3 are dominated by agriculture. This means that much greater attention has to be paid to these source types in Asia than has been devoted to them in the West.

3.2. NMVOC Speciation

[48] A detailed analysis of the chemical speciation of NMVOC emissions has been performed. As described in the methodology section, we have speciated the NMVOC emissions into 19 categories based on chemical reactivity and functional groups. This representation was performed for each geographical area in the region for each of 82 emitting source types [*Klimont et al.*, 2002]. The speciation profiles for each source type were drawn from four major information sources: the USEPA SPECIATE database [*USEPA*, 2000, available at http://www.epa.gov/ttn/chief/software/speciate], the CORINAIR inventory [*EEA*, 2000], *Smith et al.* [1992] for the special case of Asian biofuel stoves, and *Andreae and Merlet* [2001] for open biomass burning. For the first two information sources, we recognize the implicit assumption that the species profile for a given source type in Asia is the same as for the same source type in the West; this may or may not be a reasonable assumption, but with a complete lack of VOC emissions test data for most sources in the Asian setting we have no other choice.

[49] Table 7 presents the distribution of aggregated chemical species groups by country. In summary, for all of Asia we estimate that 27% of NMVOC emissions are alkanes, 22% alkenes, 5% ethyne, 17% aromatics, 9% aldehydes and ketones, and 20% halocarbons and other organic compounds. This kind of distribution is typical of all of Asia except the developed or industrializing parts of East Asia (such as Japan; the Republic of Korea; and Taiwan, China).

Table 7. Speciation of NMVOC by Country in Asia in 2000[a]

Country	Alkanes	Alkenes	Ethyne	Aromatics	Ald./ Ket.	Other	Total[b]
China	5067	3629	892	3435	1324	3094	17,440
Japan	611	141	20	301	83	770	1925
Korea, Rep. of	507	130	25	201	48	253	1164
Korea, DPR	87	27	6	55	15	44	234
Mongolia	36	64	10	29	108	205	452
Taiwan, China	257	39	7	88	21	100	512
Brunei	38	1	0	2	0	3	43
Cambodia	48	67	12	34	53	91	305
Indonesia	1577	1745	338	1250	614	1395	6918
Laos	58	95	13	34	111	176	486
Malaysia	485	239	39	218	141	304	1426
Myanmar	256	354	54	174	318	516	1672
Philippines	279	292	59	225	157	393	1404
Singapore	24	9	1	10	9	28	81
Thailand	783	618	108	565	312	670	3056
Vietnam	275	375	72	214	167	287	1391
Bangladesh	164	208	39	98	113	197	820
Bhutan	7	8	1	5	5	10	36
India	2826	2811	588	1796	955	1874	10,850
Nepal	53	96	19	49	45	85	346
Pakistan	416	331	71	214	101	211	1344
Sri Lanka	68	67	12	45	28	55	275
International shipping	1	20	3	2	0	0	27
Asia total	13,920	11,365	2390	9044	4729	10,759	52,206

[a]Data are in Gg.
[b]Rounding errors in the speciation calculations cause these numbers to be slightly different to those in Table 6.

In this region, the share of alkanes is higher (about 35%) and the share of alkenes is much lower (about 9%). This is indicative of greater use of petroleum products, particularly for transportation, and far less vegetation burning. *Piccot et al.* [1992] calculated the following shares for Asian NMVOC in the mid-1980s: alkanes 46%, alkenes 30%, and aromatics 22% (others not matching our categories). This higher share of hydrocarbons might be due to relatively greater combustion activities (fossil fuels and biofuels) in the mid-1980s and less use of industrial chemicals and solvents [*Klimont et al.*, 2002]. Figure 5 shows the species profiles of emissions from four different countries. China

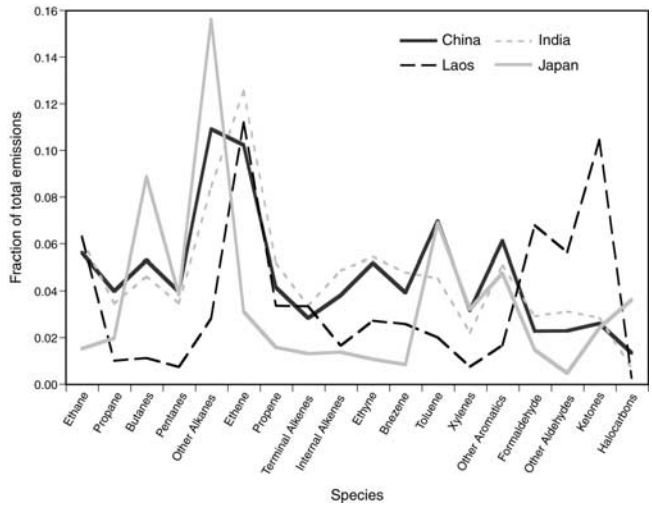

Figure 5. Fractional emissions of NMVOC species in four selected countries.

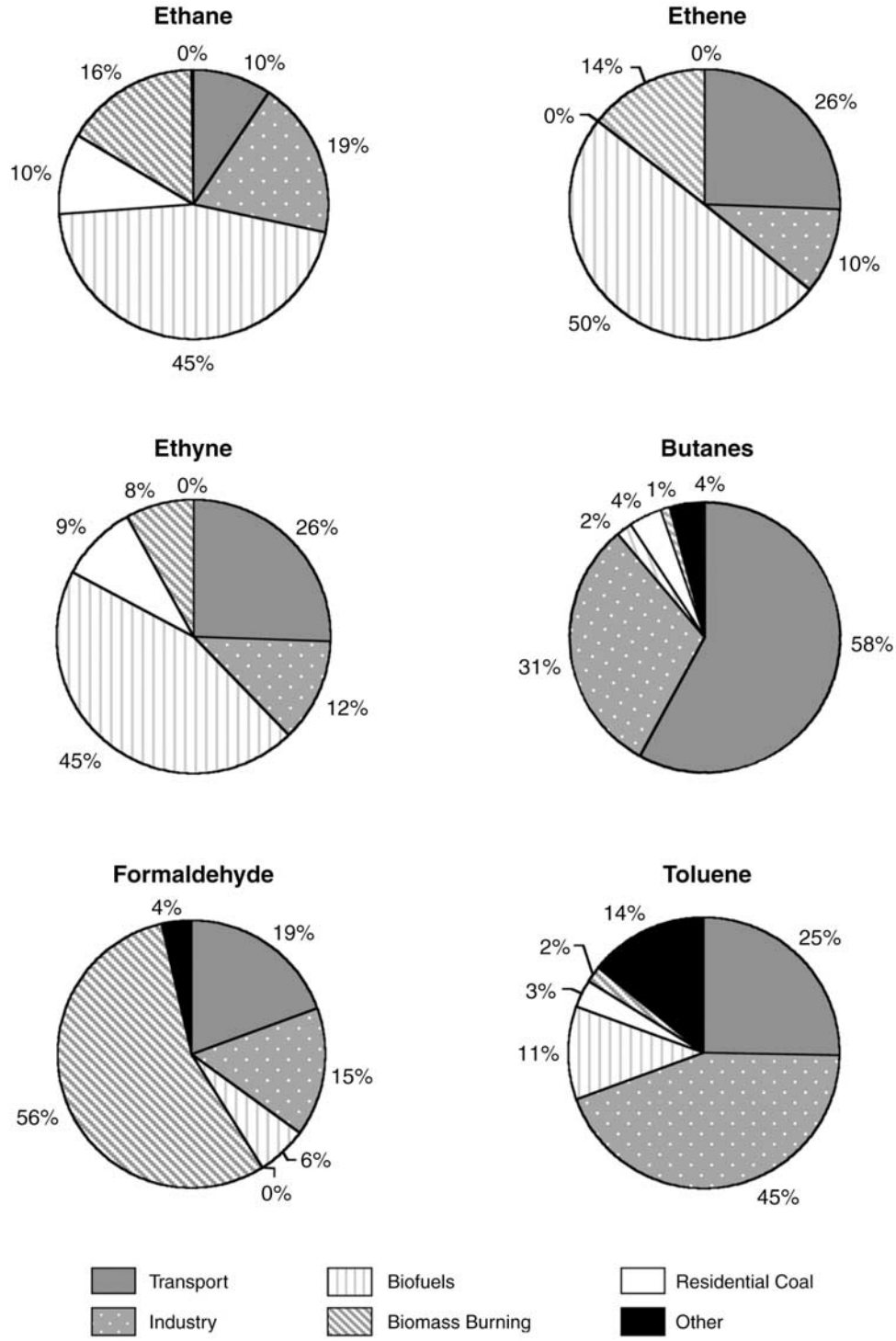

Figure 6. Source contributions to emissions of six selected organic species in East Asia.

and India have remarkably similar profiles, reflecting similarities in the state of development and resource use. Japan, on the other hand, shows a much larger fraction of alkanes, particularly butanes and $>C_5$ alkanes, indicative of a petroleum-rich, developed economy with heavy transportation needs; emissions of alkenes, aldehydes, and ketones are much lower. The case of Japan can be contrasted with that of impoverished Laos, where alkane emissions are very low and emissions of alkenes, aldehydes, and ketones are high.

[50] Figure 6 shows the major sources of some important compounds for East Asia. Biofuel use dominates the emission profiles of ethane (45%), ethene (50%), and ethyne (45%). Emissions of butanes are primarily from transportation (58%). Formaldehyde emissions are mainly from biomass burning (56%), and toluene emissions are mainly from industry (45%). Such data are important for identifying the major source regions that contribute to observed concentrations of particular species.

3.3. Seasonality of Emissions

[51] There is undoubtedly seasonal dependence of emissions, though this is not easy to determine. Generally, we can expect that the space-heating component of residential energy use will depend on outdoor temperature, though the cooking component will not. For the purposes of illustrating the seasonal variation of fuel-combustion emissions (no open biomass burning included), we have examined the potential monthly variation of residential energy use in China, while assuming that other components remain constant. (Of course, this is a big assumption because we can expect that power generation and industrial energy use will also have some seasonal variation; however, there is no information available to estimate these changes; China does not report provincial fuel use by sector and month.) We developed monthly mean temperatures for each province of China from *Lin* [1995]. We then assumed the following dependence of stove operation on provincial monthly mean temperature: <0°C, 16 hr/d; 0–5°C, 12 hr/d; 5–10°C, 6 hr/d; >10°C, 3 hr/d. The last value in this list approximates cooking-only stove usage, based on the work of *Ravindranath and Ramakrishna* [1997]. Annual emissions at provincial level were shared out by month according to this residential stove usage scheme.

[52] Figure 7 shows the results for the primarily combustion-related species SO_2, NO_x, CO, NMVOC, and BC. The strongest seasonal dependence is observed for BC and CO, because residential fuel use constitutes the largest contributor to total emissions for these species. We find a peak in January, with a significant shoulder in the period February–March. The ratio of monthly emissions in January to emissions in June, the minimum, is approximately 1.9. We find that SO_2 and NO_x emissions have very similar seasonal dependence, with a ratio of 1.2 between maxima and minima. Emissions during the TRACE-P and ACE-Asia months of March and April are almost exactly at the monthly average fraction of 0.083 of annual emissions, so detailed seasonal analysis is not necessary for these campaigns; however we would expect that observations should be higher in March than in April by perhaps 10%. Figure 7 also shows the latitudinal dependence of monthly CO emissions for northern China (defined as provinces north of the Huang He or Yellow River), southern China (provinces south of the Chang Jiang or Yangtze River), and central China (provinces in between these two rivers). Perhaps surprisingly, the greatest relative seasonal difference is observed for the central region, because of the sharper contrast between summer and winter. In the other two regions there are longer cold and warm seasons which flatten out the seasonality of emissions.

[53] Seasonality of CH_4 and NH_3 emissions in China has also been determined. In these cases we examined the temperature dependence of emissions of CH_4 from animal waste and rice cultivation and of NH_3 from animal waste and fertilizer application. For rice cultivation we considered the planting cycles of various rice crops by province, and for fertilizer application we considered the application of fertilizer prior to the planting of wheat, corn, and rice by province. The seasonality of CH_4 emissions from domestic fuel combustion was also incorporated. The results are shown in Figure 8. The profile generally is the opposite

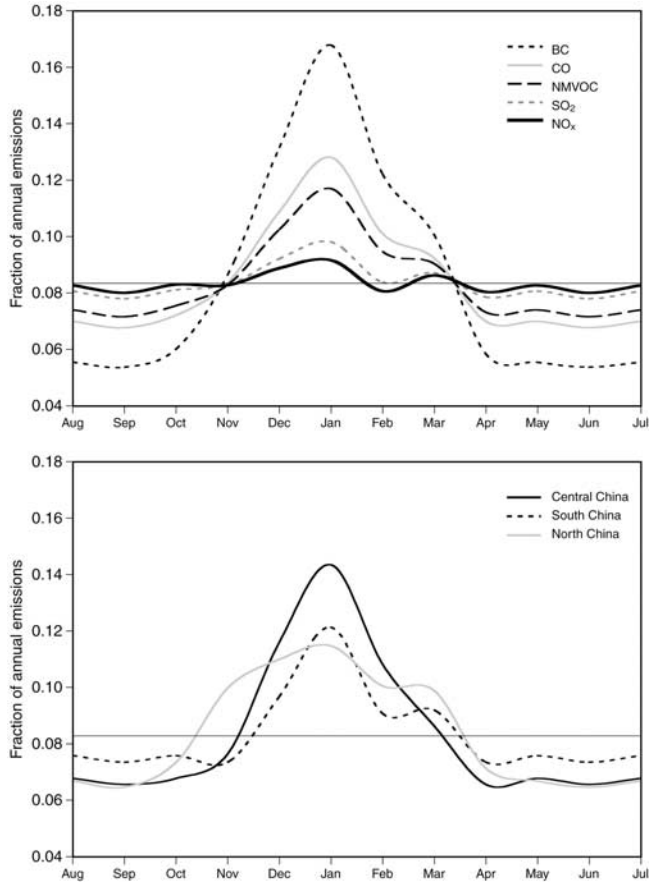

Figure 7. Seasonality of (top) five primarily combustion-related species in China and (bottom) CO emissions by latitude in China. Open biomass burning is not included.

of the profiles of combustion-related species (Figure 7). NH_3 emissions peak in June, and CH_4 emissions peak in August. The greatest seasonal variation in NH_3 emissions (Figure 8) is observed in northern China, but there is no great difference by latitude.

3.4. Gridded Emissions

[54] Spatial allocation (gridding) is a process to transform large and irregularly shaped emission data to relatively small and uniform (usually rectangular) data. Since air pollution is not a spatially uniform problem, spatial allocation with different resolutions is important for different levels of model application and analysis, i.e., at global, regional, urban, and local scales.

[55] For this inventory, we developed a multiresolution, spatial allocation technique using Geographical Information System (GIS) and Remote Sensing (RS) technology to support the various scales of research needs for TRACE-P and ACE-Asia. Basically, we use a "top down" approach to break regional- or country-based emission data down to their various gridded forms. This method is adapted from previous research, but further advanced in terms of calculation efficiency and flexibility [*Woo et al.*, 2003b]. Since the "top" emissions data are different for each administrative region and sector, the allocation factors are separately developed for each category so that emissions can be

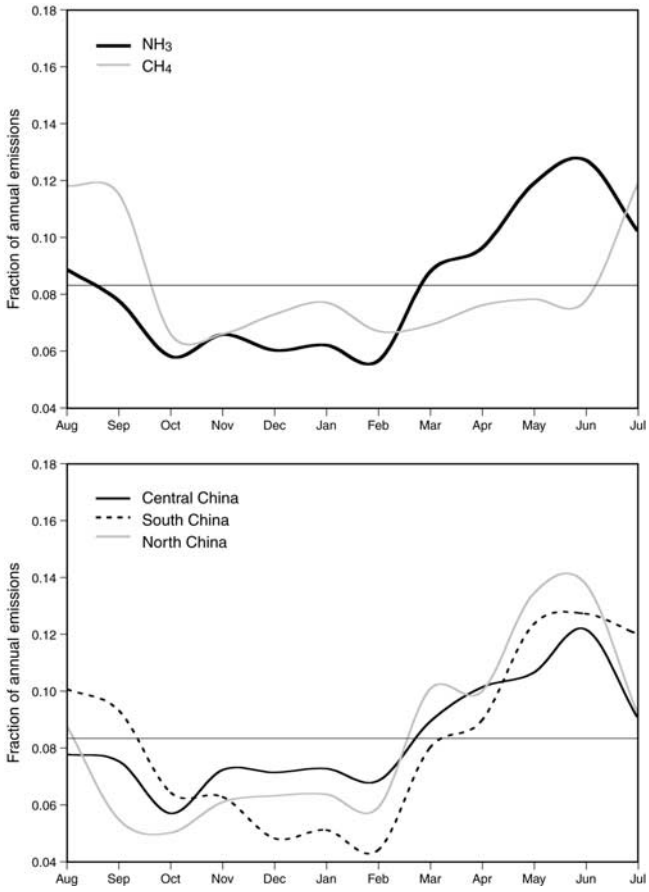

Figure 8. Seasonality of (top) two primarily noncombustion-related species in China and (bottom) NH_3 emissions by latitude in China.

faithfully allocated "down" to each grid cell. To generate the allocation factors, various types of GIS/RS datasets are used: (1) for common use, administrative boundaries are extracted from the RAINS-Asia model [*Foell et al.*, 1995; *Shah et al.*, 2000] and the Digital Chart of the World (DCW) [*Defense Mapping Agency*, 1989]; (2) for large point sources, exact geographical coordinates are taken from the RAINS-Asia model and the GEIA inventory [*Graedel et al.*, 1993]; (3) for line sources, road networks and ship lanes are extracted from the Digital Chart of the World and previous research by *Streets et al.* [2000b]; (4) for other area sources, high-resolution (30 s × 30 s) population and landcover data are extracted from the LandScan Global Population database developed by Oak Ridge National Laboratory [*ORNL*, 1999].

[56] The spatial allocation of large point sources is a two-step process. First, the regional emission total is subdivided into the sum of large point source emissions and small area source emissions in the appropriate sector. Second, the sum of large point source emissions is allocated to each facility on the basis of plant size or plant production. In the present scheme, large point sources are primarily power plants, except that large iron and steel plants are included for CO emissions. Line and area source emissions allocation is conducted in one simple step, by multiplying pregenerated allocation factors by regional or national emission totals. The allocation factors for line and area sources are created by combination and transformation of georegistered information, such as road network type (e.g., connector, dual-lane highway, primary or secondary roads, and tracks, trails or footpaths), ship-lane information (e.g., major/minor ship lanes and harbors), population distribution (total, urban, and rural), landcover classes (urban/suburban, crop fields, forest, savanna, grassland, and wetlands). For example, CO emissions from biofuel combustion are allocated by rural population distribution, since most biofuels are consumed in rural households. After the spatial allocation of all emissions by chemical species and sectors is completed, sectoral aggregation is conducted to sum emissions up to two emission types, point and area, for the practical purpose of reducing the number of unnecessary files. Special emission allocation schemes can be developed for special research needs, as necessary.

[57] Figure 9 shows the area source emission distribution of four of the gaseous species: SO_2, NO_x, NMVOC, and NH_3, at 30 min × 30 min resolution. These figures are presented at the same scale, so comparative observations can be made. A number of features stand out: the source strengths of NH_3 in the agricultural areas of central China and northern India, the relatively strong signature of NO_x from transportation sources in Japan, significant emissions of NMVOC from biomass burning in Southeast Asia, and large SO_2 emissions in the Sichuan Basin and industrialized eastern China. In Figure 10, BC and OC emissions are compared, also at 30 min × 30 min resolution. This figure shows how much stronger the OC signal is than BC throughout the region, as well as the broad distribution of both species across rural areas of China, where domestic coal and biofuel combustion are prevalent. Figure 11 presents the distributions of four gaseous organic chemical species: total alkanes (except CH_4), ethene, toluene, and formaldehyde. Note that the two upper maps are at the same scale, but the two lower maps are at a different scale. These graphs conform to Figure 6, in revealing the dominant source associations of these species: alkanes and ethene with biofuel combustion, toluene with industry, and formaldehyde with biomass burning. These maps are specifically for the month of January, and therefore they emphasize emissions from domestic heating in China and biomass burning in Southeast Asia.

[58] In terms of spatial resolution, the initial allocation procedures are conducted at the highest possible resolution (30 s × 30 s, about 0.7 km × 0.7 km) then aggregated to lower resolution to meet the needs of modeling and analysis. In theory, any resolution is possible that is lower than or equal to 30 s × 30 s. Figure 12 shows SO_2 and CO emission distributions at the highest resolution in the Pearl River Delta of China as an example. In this figure, the effects of regional and sectoral differences can be clearly seen for the two different chemical species. This level of detail makes focused studies of large metropolitan air-sheds feasible with this inventory, e.g., the contribution of emissions in Guangzhou to air quality in Hong Kong, in the example presented.

3.5. Uncertainty in Emission Estimates

[59] We acknowledge that for some types of sources in some countries very little is known about actual activity levels and emission factors, and our choices for this inven-

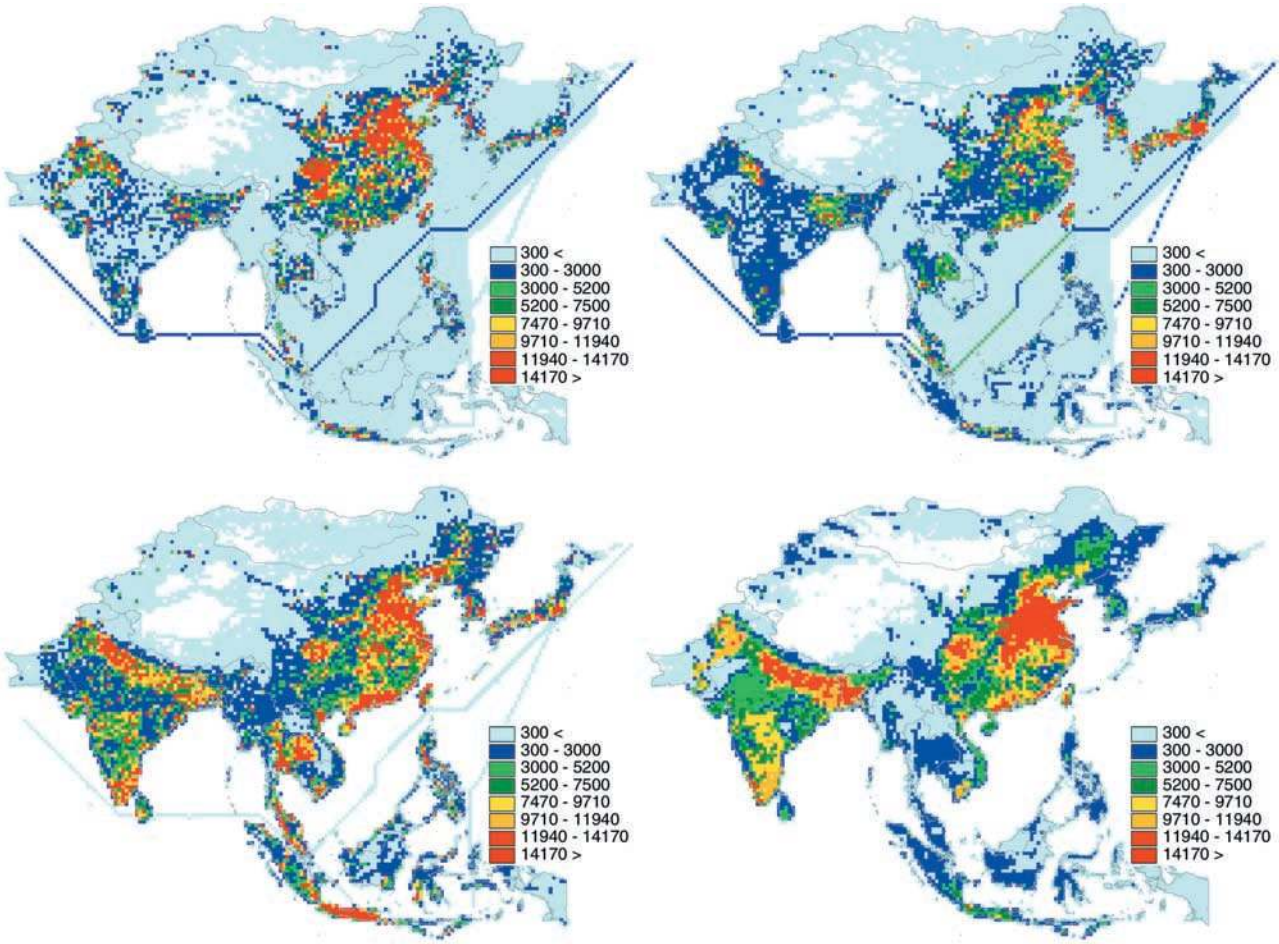

Figure 9. Area source emission distributions at 30 min × 30 min resolution of selected gaseous species: (top left) SO_2, (top right) NO_x, (bottom left) NMVOC, and (bottom right) NH_3. The scale is the same for each species in units of Mg yr^{-1} per grid cell.

tory in such cases rely heavily on inferences of activity levels from quite limited and uncertain statistical information and extrapolations of emission factors from western or other Asian source types. Several factors influence the estimation of emissions, including emission factors, activity levels, fuel quality, and removal efficiency of abatement technology. We estimate the error for each emitting subsector by combining the coefficients of variation (CV, or the standard deviation divided by the mean) of the contributing factors. We then combine these uncertainties to estimate the uncertainty of emission estimates for each country. We tried wherever possible to use harmonized data across different source types and regions, even though the reporting practices of countries are not always consistent and available.

[60] Since there is no way to judge the accuracy of activity estimates, expert judgment was used, classified according to a ten-point scale of CV, which varied for each group of provinces or countries based on the level of economic development and perceived statistical quality. For example, coal consumption estimates for Japan and Hong Kong are believed to be highly reliable and are assigned the lowest value of ±5%; at the other extreme, the quantity of grassland burned in Myanmar is extremely difficult to estimate reliably and is assigned the highest value of ±1000%. For the CV of emission factors, the available literature sources that reported emission factors based on laboratory tests usually included standard deviations that could be used. In some cases no uncertainty estimate was provided with the average value, so expert judgment was again used. Whenever emission factors were taken from the USEPA's AP-42 compilation [*USEPA*, 2002], the letter-grade emission factor ratings of A ∼ E were transferred to CV of ±5% ∼ 500% in our estimations. We have assumed that the underlying emission factor measurements are normally distributed [*Suutari et al.*, 2001], but a case can be made for a lognormal distribution of combustion emission measurements (T. C. Bond et al., submitted manuscript, 2003).

[61] Combining uncertainties requires assumptions about the dependence or independence of parameters. To combine multiplicative, independent random variables such as activities and emission factors, we used Goodman's formula for the product of variables. The relative 95% confidence intervals for emissions are calculated as 1.96 times CV. However, combining uncertainties across emitting sectors or provinces within a country is more problematic. When uncertainties are independent they can be combined in quadrature; this can be done for different sectors such as industry, residential, agriculture, biomass burning, etc. However, since the same emission factors may be used to

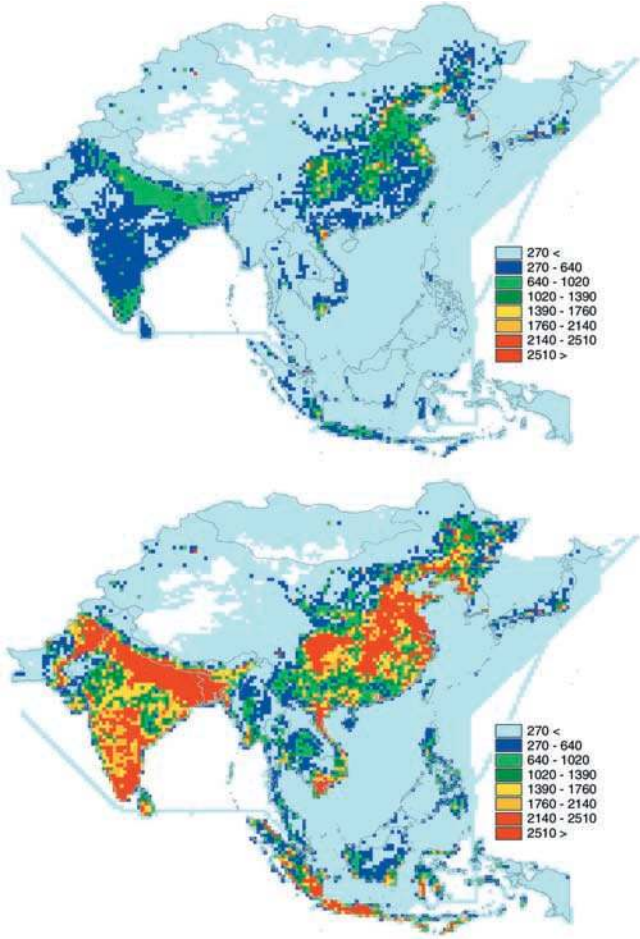

Figure 10. Area source emission distributions at 30 min × 30 min resolution of primary aerosol species: (top) BC and (bottom) OC. The scale is the same for each species in units of Mg yr^{-1} per grid cell.

calculate the emissions from two provinces, for example, these uncertainties are not independent and should not be added in quadrature. The same is true of two different sources for which an identical emission factor is used. For each sector, then, uncertainties are added linearly when the same emission factor is used, and uncertainties in emissions from different provinces are also added linearly. These aggregated uncertainties are then independent of each other and are combined in quadrature.

[62] Table 8 and Figure 13 show the results for each species for seven prototypical Asian regions. Our general findings are that emissions are known least well in India, the rest of South Asia, and Southeast Asia. This has much to do with the large contributions of biomass burning and uncertainties about liquid fuel consumption in vehicles. Emissions are best known in Japan and the other East Asian countries. The overall uncertainty in emissions for all of Asia is as follows, ranked in increasing order of uncertainty and measured as 95% confidence intervals: ±16% (SO_2), ±31% (CO_2), ±37% (NO_x), ±65% (CH_4), ±72% (NH_3), ±130% (NMVOC), ±185% (CO), ±360% (BC), and ±450% (OC). So, for example, we are 95% confident that Asian emissions of SO_2 are within ±16% of the stated value. As the confidence intervals are frequently greater than the mean, our presentation of relative confidence intervals >±100% might suggest that the lower confidence interval is negative. However, the true confidence interval is not symmetric about the mean because some of the underlying variables are lognormally distributed. A better interpretation of "±400%", for example, might be "within a factor of five" so that the confidence interval would be 20–500% of the mean given.

[63] These values for Asian emission uncertainties are similar to recent estimates made for some European countries [*Suutari et al.*, 2001]. In that study, the following ranges of 95% confidence intervals were obtained for emissions in European countries: ±6–23% for SO_2; ±9–26% for NO_x; and ±9–23% for NH_3. We obtain similar values for Japan, where meticulous record keeping and source testing compares with European countries. However, in the rest of Asia uncertainties are higher (Table 8).

3.6. Discussion

[64] This paper presents an inventory of air pollutant emissions from Asian sources. It uses available information on source activity rates and emission factors and is compared with other inventories and official national estimates where they are available. All of this information is consistent. However, we would really like to know how robust the inventory is in representing actual source strengths in the real world. The inventory implicitly consists of thousands, in some cases millions, of individual pollution sources, most of which have never been tested, scattered across a very wide area. Is there any evidence from observations and computer simulations that can substantiate the inventory?

[65] One verification method that is being pursued in TRACE-P, ACE-Asia, and similar experiments is to input the emissions data into atmospheric models and compare the modeled concentration values with field observations at the same times and places. Consistency may increase confidence in the reasonableness of the emission values; inconsistency may cast doubt on the inventory. However, in the latter case one should not automatically assign problems to the emission estimates because the models have their own shortcomings and the measurements themselves are not always reliable or directly comparable with model predictions.

[66] Table 9 summarizes all the emission estimates that were used in the TRACE-P and ACE-Asia modeling of *Carmichael et al.* [2003a, 2003b] and several other groups. These estimates are specific to the 2-month period of March–April, 2001, when the TRACE-P and ACE-Asia measurements were taken. From this table, a comparison of the relative source strengths of anthropogenic and natural sources can be made. Also presented in Table 9 are the overall PM_{10} and $PM_{2.5}$ emission estimates. For some natural sources, biogenic NMVOC and mineral dust in particular, the levels of uncertainty are higher even than for anthropogenic emissions and can contribute to discrepancies between model and observation.

[67] Initial results from the TRACE-P and ACE-Asia experiments are encouraging. Table 10 presents a comparison between observed mean concentrations of ten selected species and concentrations simulated with the STEM model [*Carmichael et al.*, 2003a] using this emission inventory. In Table 10, measurements from TRACE-P DC-8 flights 6 through 17 are included. Values are limited to altitudes

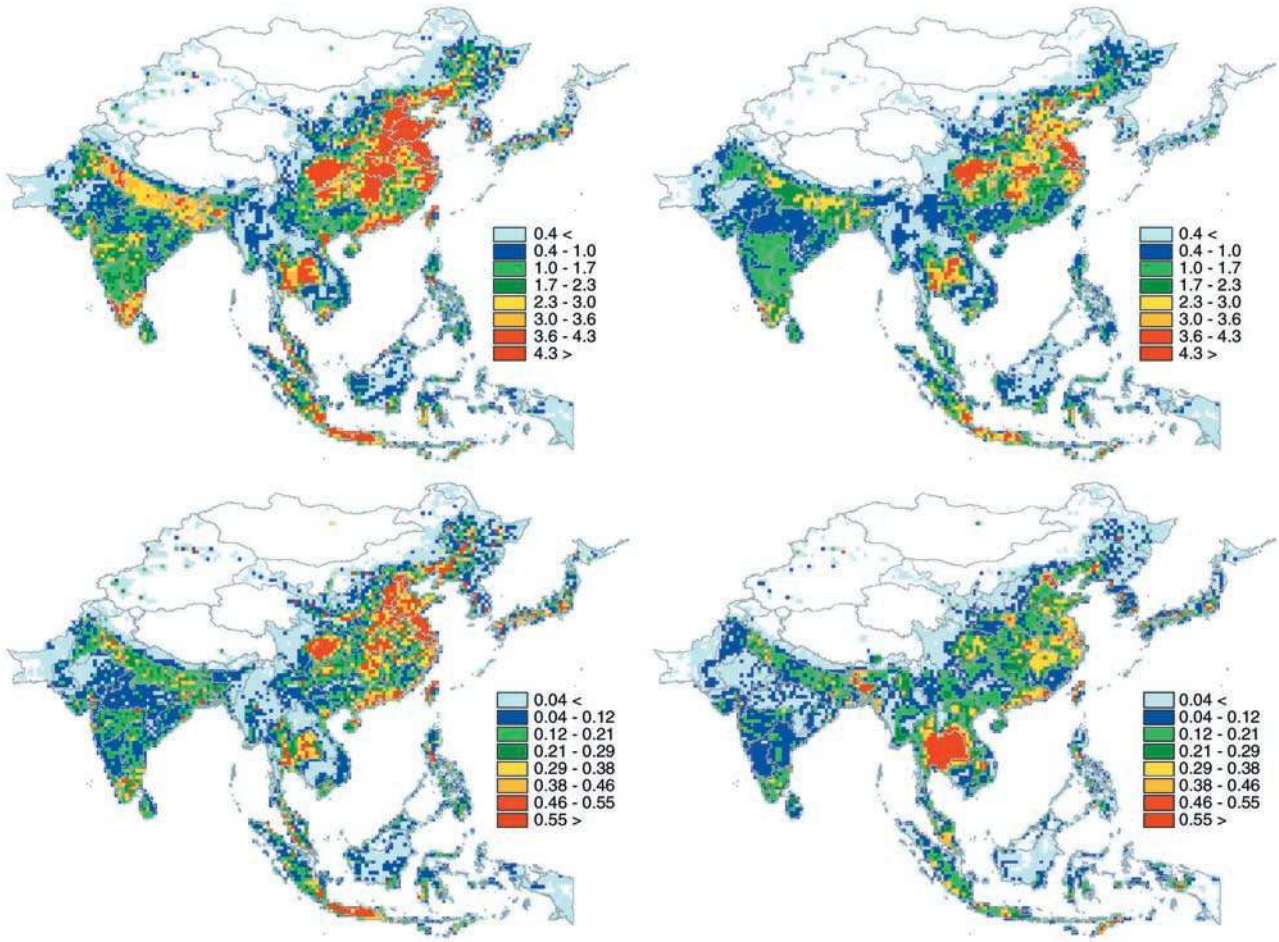

Figure 11. Area source emission distributions at 30 min × 30 min resolution of selected organic species: total (top left) alkanes, (top right) ethene, (bottom left) toluene, and (bottom right) formaldehyde. Scale is in units of moles s^{-1} per grid cell, different for the upper and lower pairs.

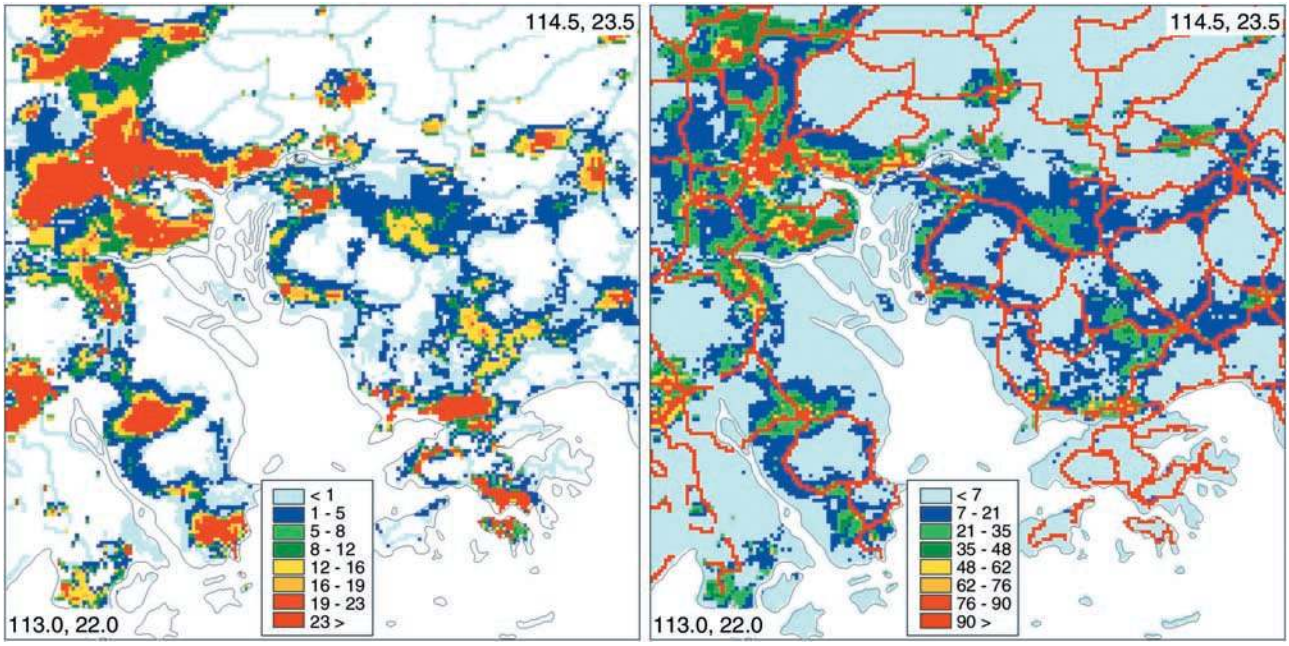

Figure 12. Illustration of sector-specific detail in the high-resolution (30 s × 30 s) inventory for the Pearl River Delta (at the bottom right in each frame is Hong Kong; near the upper left is Guangzhou). Left frame is SO_2; right frame is CO.

Table 8. Uncertainty in Pollutant Emission Estimates (±95% Confidence Intervals)[a]

Region	SO_2	NO_x	CO_2	CO	CH_4	NMVOC	BC	OC	NH_3
China	13	23	16	156	71	59	484	495	53
Japan	9	19	7	34	52	35	83	181	29
Other East Asia	12	24	13	84	101	49	160	233	31
Southeast Asia	27	92	91	214	95	218	257	345	87
India	26	48	33	238	67	149	359	544	101
Other South Asia	35	63	44	291	109	148	379	531	101
International shipping	44	56	40	72	72	204	402	402	–
All Asia	16	37	31	185	65	130	364	450	72

[a]Uncertainty in percent.

below 1 km because the concentrations at these altitudes often reflect sampling directly downwind of large sources (in the cases of flights in the Yellow Sea) and are not as influenced by complicating factors such as vertical transport by convection. The general agreement in Table 10 between observed and modeled values for a range of organic and inorganic species is reassuring. A more detailed table with more species and more comparison criteria can be found in the work of *Carmichael et al.* [2003a]. Similarly, *Zhang et al.* [2003] found that the CMAQ model using this inventory reproduces well the latitudinal and vertical distribution of CO, O_3, and SO_4^{2-} in the Asian outflow.

[68] One issue concerns the SO_2 and SO_4^{2-} observations. The SO_2 concentrations measured in the Yellow Sea were very high during TRACE-P, and significant increases were noted between PEM-West B (1994) and TRACE-P, even though SO_2 emissions have declined during the intervening time period in East Asia [*Streets et al.*, 2000a]. This is not believed to be a problem with the anthropogenic emissions, however, but more likely due to different precipitation regimes affecting the conversion of SO_2 to SO_4^{2-} in the two campaigns and/or the influence of the Miyakejima volcano that was erupting during TRACE-P. To further assist researchers in interpreting measurements in Asian field campaigns separated by a number of years, Table 11 is presented. This table shows trends for seven major species in the 1990–2000 period, with emphasis on differences between 1994 (PEM-West B) and 2000. A brief explanation of the reasons for the trends is included. Emissions of two species (SO_2 and BC) have declined in the latter half of the 1990s, two have changed little (CO_2 and CH_4), and three have increased significantly (NO_x, NMVOC, and NH_3). Though we do not have trend information for CO, we believe that CO emissions have been stable in recent years [*Streets and Waldhoff*, 2000].

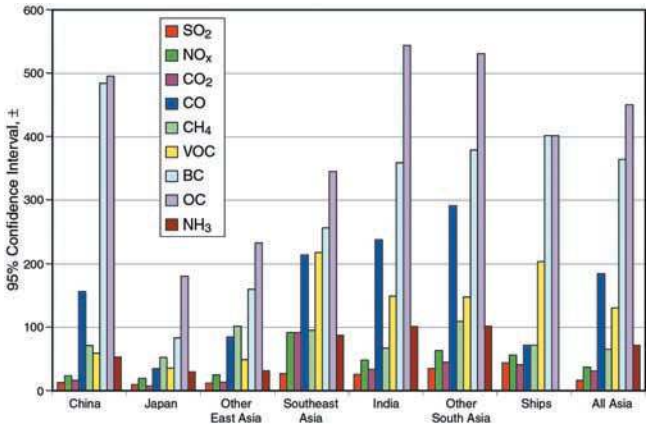

Figure 13. Uncertainty (%) in pollutant emission estimates (±95% confidence intervals).

Table 9. Emissions From All Source Types Included in TRACE-P Modeling for March–April 2001[a]

Species	Natural Sources	Anthropogenic (Other Work) Combustion	Anthropogenic (Other Work) Open Burning	Anthropogenic (This Work) Combustion	Anthropogenic (This Work) Open Burning	All Sources
SO_2	2.0[b]	0.002[c]	-	5.6	0.2	7.8
NO_x	0.005[d]	0.029[c]	-	4.0	1.1	5.1
CO_2	-	-	-	1453.0	459.6	1912.6
CO	-	0.013[c]	-	34.7	27.8	62.5
CH_4	-	-	-	16.5	1.3	17.8
NMVOC	18.3[e]	0.008[c]	-	6.7	5.0	30.0
BC	-	-	-	0.3	0.2	0.5
OC	-	-	-	1.2	1.4	2.6
NH_3	-	-	-	4.7	0.4	5.1
PM_{10}	107.7[f]	4.9[g]	3.4[g]	-	-	116.0
$PM_{2.5}$	26.2[f]	2.9[g]	2.1[g]	-	-	31.2
Sea Salt	41.6[h]	-	-	-	-	41.6

[a]Emissions are in Tg/2 mo.
[b]Volcanic emissions, based on *Fujita* [1992], *Andres and Kasgnoc* [1998], and S. Fujita et al. (manuscript in preparation, 2003).
[c]Aviation emissions, based on *Olivier et al.* [1996].
[d]Lightning emissions, based on *Carmichael et al.* [2003a].
[e]Biogenic emissions, based on *Guenther et al.* [1995]; this value is the sum of isoprene and monoterpene emissions for March and April.
[f]Wind-blown dust, based on *Uno et al.* [2003b].
[g]Estimate for all anthropogenic emissions, based on J. Dorwart et al., manuscript in preparation, 2003. Note that PM_{10} includes $PM_{2.5}$, and $PM_{2.5}$ includes BC and OC.
[h]Based on *Uno et al.* [2003b].

[69] Observations of BC during TRACE-P and ACE-Asia seem to yield good agreement with modeled concentrations using this inventory when concentrations are low and sampled air masses originate in Southeast Asia or southern China. For example, BC concentrations modeled using the CFORS model have been compared with observed BC concentrations measured during the ACE-Asia period at five Japanese ground stations: Rishiri, Sado, Hachijo, Chichijima, and Amami-Oshima [*Uno et al.*, 2003a]. Good agreement is obtained for both absolute concentration levels and the time variation of the concentrations. Also, analysis of BC aircraft measurements during TRACE-P (A.D. Clarke, University of Hawaii, manuscript in preparation, 2003, hereinafter referred to as Clarke, manuscript in preparation, 2003) produced reasonable agreement between observed absorp-

Table 10. Comparison of Observed and STEM-Simulated Mean Concentrations for TRACE-P DC-8 Flights 6 Through 17 at Altitudes Below 1 km

Species	Observed	Modeled
CO, ppbv	219	203
Ethane, ppbv	2.0	1.6
Propane, ppbv	0.62	0.46
Ethene, ppbv	0.18	0.20
Ethyne, ppbv	0.78	0.63
SO_2, ppbv	1.55	1.04
NO_2, ppbv	0.27	0.25
NO, ppbv	0.035	0.041
Benzene + toluene, ppbv	0.33	0.19
BC, μg m^{-3}	0.84	0.67

Table 11. Asian Emission Trends From Anthropogenic Sources[a]

Species	Region	1990	1994	1995	2000	94–00, %	Brief Explanation
SO_2 (Tg)	China	22.2[b]	24.2[b]	25.7[b]	20.3[c]	−16.1	reduction in coal use; switch to lower-S coals
	East Asia[d]	25.9[b]	27.3[b]	27.9[b]	22.6[c]	−17.2	China dominates S; lower-S oil in Taiwan and ROK
NO_x (Tg)	China	7.2[e]	9.2[f]	9.7[e]	10.3[c]	13.0	growth in transport offsets decline in coal use
	East Asia[d]	10.8[e]	13.3[f]	13.9[e]	14.8[c]	12.0	transport growing slower in developed East Asia
NMVOC (Tg)	China	11.4[e]	12.8[f]	13.1[e]	14.6[c]	14.1	unabated growth in chemicals, solvents, oil use
	East Asia[d]	15.2[e]	16.7[f]	17.1[e]	18.5[c]	10.8	emissions better controlled in developed East Asia
NH_3 (Tg)	China	9.7[e]	11.3[f]	11.7[e]	13.3[c]	17.7	increase in fertilizer use in recent years
	East Asia[d]	10.6[e]	12.2[f]	12.6[e]	14.2[c]	16.4	China dominates NH_3 emissions in East Asia
CO_2[g] (Pg)	China	30.7[h]	35.9[h]	37.1[h]	36.3[h]	1.1	reduction in coal use, improved energy efficiency
CH_4 (Tg)	China	29.1[h]	29.8[h]	30.6[h]	31.7[h]	6.4	reduced coal mining has slowed increase
BC (Tg)	China	N.A.	N.A.	1.27[i]	0.94[c]	−26.0	reduction in coal use by households and industry

[a]Emission trends are in Gg.
[b]*Streets et al.* [2000a].
[c]This work.
[d]Includes China; Japan; DPRK; ROK; Mongolia; and Taiwan, China.
[e]*Klimont et al.* [2001].
[f]Linearly interpolated between 1990 and 1995 values (growth was approximately linear in this period.)
[g]Direct emissions only; does not include take-up by growing vegetation.
[h]*Streets et al.* [2001b].
[i]*Streets et al.* [2001c]; the trend shown for BC is from 1995–2000 (no 1994 value available).

tion and modeled BC concentrations. Analysis of ACE-Asia observations yielded similar agreement for air masses that originated in Southeast Asia and had a predominantly biomass burning signature. However, for air masses with a signature of fossil-fuel emissions off the central China coast, comparison between measured and modeled BC concentrations suggests a significant underestimation of BC emissions (Clarke, manuscript in preparation, 2003).

[70] The major pollutant of contention is CO. Table 10 showed that mean CO concentrations simulated with the STEM model are in good agreement with the observational data set. Figure 14 elaborates on this simulation by showing the STEM model/observation intercomparison of all 5-min values for DC-8 flights 6 through 17 and P-3 flights 8 through 19. The results show that approximately 80% of the predicted values fall within ±30% of the observed values. However, there is a bias towards underprediction, especially at high CO concentrations (300–1000 ppbv). Approximately 8% of values are underpredicted by more than 30% and 1.5% of values are underpredicted by more than 50%. In particular, it seems that some of the very high concentrations observed during flights close to the coast of China are not well reproduced. It is possible that these measurements are of essentially urban plumes and that the limited model resolution is unable to capture them. For example, *de Laat et al.* [2001] noted that during the INDOEX program their high-resolution model (1.9° × 1.9°) was much better able than their low-resolution model (3.75° × 3.74°) to reproduce measured CO mixing ratios. They further state that the underestimation of CO in the model for air masses from the Bay of Bengal is resolution-related because discrepancies disappear at the higher resolution. Nevertheless, it does pose the question of whether emissions of CO are underestimated in this inventory.

[71] Analysis by TRACE-P global modelers suggests that CO emission estimates for China are low. The Asian emission inventory for CO described here was used in the GEOS-CHEM and other models to simulate observations of CO during the TRACE-P campaign [*Palmer et al.*, 2003; *Kiley et al.*, 2003]. A comparison of the modeled and observed distribution of CO showed that the model had a significant negative bias, especially in the boundary layer. An inverse model analysis of these data using GEOS-CHEM suggests that a 54% increase in China's anthropogenic CO emissions is called for [*Palmer et al.*, 2003]. *Kasibhatla et al.* [2002] used an inverse modeling methodology to study Asian CO emissions from fuel combustion and inferred that emissions in 1994 were 350–380 Tg, which is 110–140 Tg higher than derived using the bottom-up inventory of *Olivier et al.* [1996]. Palmer et al. conjecture that the high-density aircraft observations of continental outflow directly downwind of sources during TRACE-P are likely to yield more reliable inversion analyses than the sparse network of NOAA/CMDL surface observations relied on by *Kasibhatla et al.* [2002] and a related paper by *Petron et al.* [2002], which also implied a severe underestimation of CO emissions in Asia.

[72] Additional evidence is provided by ground-station measurements in China. *Wang et al.* [2001, 2002] have measured concentrations of CO, NO_x, and SO_2 at an essentially rural site in eastern China, Linan in Zhejiang

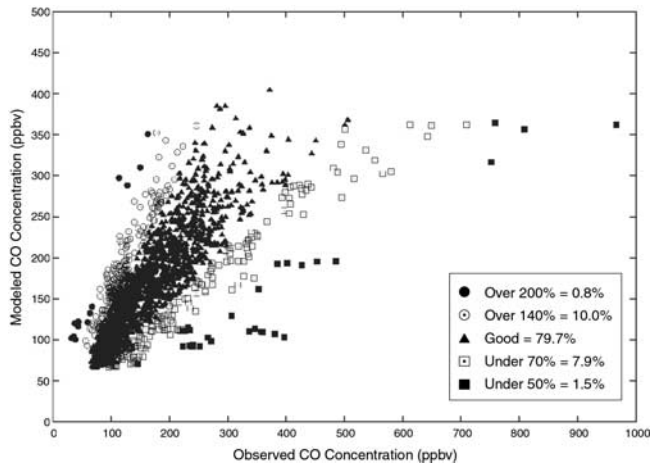

Figure 14. Comparison of modeled to observed CO mixing ratios for each 5 min along the flight paths for DC-8 flights 6 through 17 and P-3 flights 8 through 19.

Province, for more than 1 year. They have also compared their observations with emission ratios from our inventory. The very high levels of CO concentrations they observed led the authors to speculate that biofuel combustion or open biomass burning might be underestimated in our inventory.

[73] While it is true that biofuel consumption is not well known, it is tracked by Chinese energy experts and presented in energy statistics [*SSB*, 1998]. We have examined the question of biofuel usage for the province in which Linan is located. The amount of CO released from biofuel combustion in Zhejiang Province according to our inventory is 1370 Gg, representing 49% of total CO emissions in the province. This estimate is obtained using the high emission factor estimate of 77.5 g kg^{-1} for biofuel combustion [*Zhang et al.*, 1999], combined with Chinese energy statistics. As a check, we performed our own analysis of the amount of biofuel used in Zhejiang Province under a variety of assumptions about the number of biofuel stoves in the province, daily stove usage for cooking, and amount of biofuel consumed per day. Using the same emission factor, we obtained a range of emissions from biofuel combustion of 1110–1900 Gg CO. This analysis suggests that biofuel use is probably captured reasonably well in Chinese statistics.

[74] Is it possible that open biomass burning is underestimated? We can likely rule out the extensive forest and agricultural burning that occurs in Southeast Asia because these plumes are easily recognizable in the aircraft observations; also, they generally occur at high altitudes over eastern China and would not explain elevated ground-level CO concentrations at Linan. It is true that we know very little about farmers' practices as regards the burning of crop residues in the fields after harvest. Our calculations assume that 17% of crop residues are burned in the field in China [*Hao and Liu*, 1994], somewhat less than the estimate of 25% by *Crutzen and Andreae* [1990], though to our knowledge neither value is based on surveys of agricultural practice. Actually, it is our understanding that crop burning in the fields is now banned in many (all?) parts of China but persists nonetheless. Visual observation around Linan has indeed revealed burning fields at some times of the year (M. Bergin, Georgia Institute of Technology, personal communication, 2002); and it would be consistent with the high levels of OC (mean value of 44 $\mu g\ m^{-3}$) observed in November in the Yangtze delta [*Xu et al.*, 2002]. However, if this were the cause, one would expect seasonality to the CO concentration profile that would mirror the growing and harvesting seasons (around Linan, say). The CO data of *Wang et al.* [2001], though, show no strong seasonality, merely a small winter peak that is likely associated with a modest amount of additional home heating in Zhejiang and Jiangsu Provinces and more northerly areas. On the other hand, a good correlation between CO and CH_3Cl concentrations does suggest a biomass-burning source [*Wang et al.*, 2002]; cross-sectional analysis of K^+ concentrations and BC/OC ratios could elucidate whether biomass burning is the cause. Only an honest survey of farmers' practices can reveal if more than 17% of crop residues are burned in the field and when the burning occurs.

[75] Recent analysis of the Linan data has compared the measurements of *Wang et al.* [2001, 2002] with modeled concentrations using the STEM model and this inventory [*Carmichael et al.*, 2003b]. Significant underestimation of not only CO but also SO_2 and NO_y is found. This would suggest that the underprediction is more likely to reflect an underestimation of local coal use rather than biofuel use. In addition, back-trajectory analysis of the observed CO concentrations during TRACE-P [*Carmichael et al.*, 2003b] seems to identify the central heartland of China as being the source of many of the air masses in which underprediction is manifested. In combination, this evidence suggests an underestimation of domestic coal use in rural China.

[76] This is an intriguing question. Official Chinese energy statistics, on which our emission estimates are based, show a dramatic decrease in household use of coal in the late 1990s. From 135 million tons (mt) in 1995, it dropped to 122 mt in 1997 [*Sinton and Fridley*, 2000], to 89 mt in 1998 [*NBS*, 2000], and to 75 mt in 2000 (Jiang Kejun, Energy Research Institute of China, personal communication, 2001). According to the statistics, this decline was partially offset by an increase in the use of LPG and electricity. While such a trend is verifiably true in the developed coastal and urban areas of China, it is difficult to believe that it could be true in the central heartland of China, where rural population continues to rise and the penetration of clean residential fuels is slow (J. Sinton, Lawrence Berkeley National Laboratory, personal communication, 2002). In addition, though coal production has declined in this region according to official statistics, illicit mining is thought to be widespread, and much of that coal could find its way into rural households [*Sinton*, 2001]. So there certainly is the possibility for rural coal use to be underreported in this region. The same argument can be applied to the use of coal to fuel small industrial operations, such as kilns and ovens. In addition, the higher temperatures of these applications can generate more NO_x, which seems to be indicated by analysis of the Linan data. It should be noted here that this problem of underreported domestic coal use may be restricted to the central, less-developed region of China where small sources are the primary polluters. In other regions (coastal and northern), emissions are larger and dominated by big industrial and power generation sources. This could explain why we do not always see discrepancies between models and observations, only when air masses originate in the rural heartland of China.

[77] Another possibility is that there might be significant sources of emissions that are omitted from this inventory. Possibilities include coal mine fires, small coke ovens, and rural vehicles. We considered carefully whether open waste burning (of garbage) is significant in China. We believe that it is not. The amounts of garbage generated in China, especially rural China, are low compared with western countries; garbage tends to have higher moisture content than in the West and be more difficult to burn; and land disposal is a preferred option [*Wang and Nie*, 2001]. Though coal mine fires have been touted as a major environmental catastrophe in China, Chinese and other western authorities believe that the amount of coal consumed in out-of-control burning is probably about 14 million tons annually, yielding 490 Gg CO or 0.4% of China's total CO emissions in this inventory (A. Prakash, University of Alaska, personal communication, 2002, based on unpublished work by the Aerophotogrammetry and Remote Sensing Bureau of China). Also, many of the fires are in locations quite remote from Linan. Another possibil-

ity relates to the use of small coke ovens in eastern and central China, particularly in Shanxi Province, which are known to be high emitters of CO [*Polanske and McMichael*, 2002]. Unfortunately, we have no information on the number or emission rates of such facilities. However, this could be a way to increase CO emissions without increasing emissions of other species. A further possibility is that the numbers of vehicles in China might be underreported; anecdotal information suggests that there are many unregistered vehicles in rural China that are not included in the official statistics used for this inventory.

[78] Finally, is it possible that the high CO is a product of some unusual chemical reaction in the atmosphere? Perhaps the oxidation of biogenic hydrocarbons emitted from the extensive southern forest and woodland areas of China is greater than anticipated? However, then a summer peak in CO concentrations might be expected, which is not observed. Alternatively, it has been noted by other TRACE-P researchers that peroxyl and hydroxyl radicals over China are lower than expected, perhaps leading to less CO-destruction capability. More study of these possibilities is warranted.

[79] We conclude with the following observations. This emission inventory for Asia is the first to treat many species in many countries with a consistent, detailed methodology for a recent year. Our methodology is internally consistent and agrees well with the few previous, reliable estimates that have been made. The inventory performs well for most species when input into atmospheric models and compared with observations. Exceptions to this are CO and possibly BC, for which observations indicate that our estimates are low in the central heartland of China. Taken together, the CO and BC findings suggest that there are missing sources with poor combustion efficiency, possibly in rural areas. There are a number of possible explanations for this: that open burning of agricultural residues in the field or of household garbage is underestimated, that Chinese energy statistics underreport coal use in residential and small industrial applications, and/or that Chinese vehicle statistics are missing a substantial number of rural vehicles. Further analysis of data from the field campaigns in Asia may unravel this mystery. However, field testing of sources and surveys of source types and operating characteristics are the best ways to develop a better understanding of emissions in the region.

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