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Global emissions of hydrogen chloride and chloromethane from coal combustion, incineration and industrial activities: Reactive Chlorine Emissions Inventory

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Abstract. Much if not all of the chlorine present in fossil fuels is released into the atmosphere as hydrogen chloride (HCl) and chloromethane (CH_3Cl, methyl chloride). The chlorine content of oil-based fuels is so low that these sources can be neglected, but coal combustion provides significant releases. On the basis of national statistics for the quantity and quality of coal burned during 1990 in power and heat generation, industrial conversion and residential and commercial heating, coupled with information on the chlorine contents of coals, a global inventory of national HCl emissions from this source has been constructed. This was combined with an estimate of the national emissions of HCl from waste combustion (both large-scale incineration and trash burning) which was based on an estimate of the global quantity released from this source expressed per head of population. Account was taken of reduced emissions where flue gases were processed, for example to remove sulphur dioxide. The HCl emitted in 1990, comprising 4.6 ± 4.3 Tg Cl from fossil fuel and 2 ± 1.9 Tg Cl from waste burning, was spatially distributed using available information on point sources such as power generation utilities and population density by default. Also associated with these combustion sources are chloromethane emissions, calculated to be 0.075 ± 0.07 Tg as Cl (equivalent) from fossil fuels and 0.032 ± 0.023 Tg Cl (equivalent) from waste combustion. These were distributed spatially exactly as the HCl emissions, and a further 0.007 Tg Cl in chloromethane from industrial process activity was distributed by point sources.

1. Introduction

To help in the assessment of the global atmospheric chlorine cycle, the Reactive Chlorine Emissions Inventory (RCEI) has been organized within the Global Emissions Inventory Activity (GEIA) of the International Global Atmospheric Chemistry Program [*Graedel and Keene*, this issue]. The initial task of the RCEI is the development and verification of gridded global emission inventories for species that are important to the tropospheric burden of reactive chlorine or to the rate of chlorine cycling. These include hydrogen chloride (HCl), which has significant sources from sea-salt, biomass burning and fossil fuel combustion, and chloromethane (CH_3Cl, methyl chloride), for which the predominant sources are ocean and terrestrial biogenic processes, biomass burning, with a minor contribution from exclusively anthropogenic activities.

The anthropogenic sources described in this work represent minor but significant contributions to the atmospheric fluxes of hydrogen chloride (10% of the total) and chloromethane; the major fluxes from decomposition of sea-salt aerosol and biomass combustion are described in companion papers [*Erickson et al.*, this issue; *Lobert et al.*, this issue]. The various sources are likely to have very different geographical distributions, with sea-salt aerosol predominating over the oceans and the coast, and biomass burning being mainly tropical. Coal combustion, on the other hand, is a feature of industrial economies and so is mostly in temperate regions of the Northern Hemisphere; nevertheless, there are a few nations in the Southern Hemisphere where coal is a significant source of energy.

For hydrogen chloride, which is removed from the atmosphere principally by dissolution in water, a relatively rapid process with an average lifetime of 1.5 days [*Erickson et al.*, this issue] governed wholly by mixing, atmospheric concentrations are strongly influenced by local emissions. On the other hand, chloromethane has an atmospheric lifetime of 1.5 years, with oxidation by OH radicals being the main removal process [*Sanhueza*, 1994]. This means that substantial concentrations may exist far from local sources of emission. However, fossil fuel combustion sources could be important in regions such as Europe and some areas of North America and eastern Asia, particularly for modeling trajectories of air parcels.

To gain a measure of the size and global distribution of emissions from coal combustion, a global inventory of the coal burned during 1990 was developed; this was combined with information about the location of power stations to position the large releases from electricity generation. Population distribution was used to position industrial, commercial, and residential emissions. In much the same way as for other inventories in the RCEI activity [e.g., *Aucott et al.*,

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this issue] the quantity of coal burned in each location was combined with an emission function, which in this case was related to the quantity of chlorine estimated to be in the original coal, to estimate the hydrogen chloride and chloromethane release at that location.

Similarly, municipal and domestic waste contains chloride and could be expected to yield HCl and possibly chloromethane on combustion. Since it has not been possible to position municipal waste incinerators in the same way as coal-burning energy generators, the estimated emissions from this source have been distributed geographically on the basis of population.

Finally, there are direct chemical emissions during process operations when the materials are either manufactured or used to produce other materials. In the case of hydrogen chloride these emissions are not thought to be significant to atmospheric chemistry; they are generally strictly controlled because of the noxious nature of the material, and it is only catastrophic loss of containment (beyond the scope of this study) which leads to environmentally important concentrations. On the other hand, chloromethane is emitted from process operations, and an estimate of the magnitude and distribution of these releases is included in this work. The releases estimated from the various sources were combined into the end products: 1° latitude × 1° longitude grids of HCl and chloromethane emissions in 1990.

2. Global Combustion of Coal During 1990

2.1. Development of the Database

A globally consistent database for coal combustion is a prerequisite of this work. Several sources of information were identified [*International Energy Agency (IEA)*, 1992a,b; *Marland and Boden*, 1993; *United Nations (UN)*, 1995], and the data provided by the IEA proved to be the most comprehensive in terms of nations covered, consistency of reporting, and details of end uses. The database in *IEA* [1992a] describes the quantities of individual fuels burned in countries of the Organisation for Economic Cooperation and Development (OECD) in a number of categories: (1) transformation, including public and private generation of electricity, combined heat and power (CHP), and district heating plants; (2) industrial consumption, including iron and steel, nonferrous metals, and eight other categories expected to be large-scale combustion processes; and (3) small-scale combustion, categorized separately into agricultural, commerce, residential, and transport.

All solid, liquid, and gaseous fuels are tabulated for each country and category, and the coal statistics cover coking, steam, and subbituminous and brown coals. For non-OECD countries the data are presented in a much simpler format in *IEA* [1992b]: power and heat generation, other transformation, iron and steel industry, nonmetallic minerals, residential, and unspecified. The latter list represents the maximum categorization that could be accomplished globally; the usages in the much more detailed description of OECD countries were therefore aggregated to fit the non-OECD categories. In almost all of the non-OECD countries one category of consumption predominated, and so the values "unspecified" were added into that category.

Governed by the need to differentiate chemical emissions, which were presumed to be dependent on the composition of the coal and the way in which it is burned, the quantities in the IEA database were combined into broad categories in which the fuels were potentially of the same quality. The chlorine content of coal varies from a few parts per million (ppm) to thousands of ppm [*Sloss*, 1992] but there is very little possibility of obtaining accurate chemical compositions of each sort of coal burned, so some more general correlation was sought. From the work of *Emmel et al.* [1989], based on *Misenheimer et al.* [1985], the largest difference in emissions of HCl is between hard coals and lignite, the latter releasing 1/100th to 1/4 that of the former. Thus in this work the primary subdivision was into hard coals (including coking, steam, and subbituminous coals) and lignite (including brown coal and patent fuels manufactured from it). Quantities were calculated for consumption in large combustion processes (power and heat generation by utilities and industry) and conversion processes (essentially iron and steel manufacture, together with nonmetallic mineral processing). Small-scale combustion in residential and commercial premises was calculated separately. These categories were also designed to enable geographical discrimination of specific emissions from point sources within a nation, where information on the location of, for example, public electricity-generating utilities could be obtained.

For the OECD countries the explicit data in *IEA* [1992a] on the quantites of hard coal and lignite used in each category were combined into the groups described above. For non-OECD countries, where the tabulated information was simply the quantity of all types of coal, a more circuitous process had to be used to estimate the qualities of the coals. The IEA database [*IEA*, 1992b] contains information on the quantities of hard coal and lignite produced by each country, together with statistics for electricity and thermal energy generated and the calorific values of the coals. By comparing the quantity inferred from these additional data with the total quantity of coal consumed, it was possible to identify the dominant quality of the coal used within each country, to which all usage of coal within that country was assigned.

The countries of the former USSR presented a special case; information for the four categories of use, in 1990, was provided only for the USSR as a whole. However, the statistics for power generation from all fuels were tabulated for all 15 of the independent states. Hard coal production statistics are available for seven states, lignite production for four, and energy balances (which included energy used in power generation, industrial applications, and small-scale combustion such as transport, commercial, and residential) for five states, plus the former USSR as a whole. The database also held calorific values of the coals for all 15 states, plus the average for the USSR. By assuming that missing values defaulted to the mean, it was possible to reconstruct the categorized coal consumption for all 15 independent states.

The results of these calculations, for all countries that have significant coal usage, are shown in Table 1. The remaining nations of the world either have no significant coal consumption or no significant energy use and are not considered further in this work. While it is not possible to assign rigorous uncertainty to these data, observations on the qualities of individual national statistics were included in the IEA tabulations. These show that the data with lowest uncertainty are those from the OECD tables, followed by those from the non-OECD tabulation and then those from centrally planned economies. The information is useful only in a qualitative sense, and an independent check on the data was sought.

2.2. Verification

The database developed here was compared with two other databases, apparently independent: (1) *UN* [1995], which contains a listing of the aggregate amounts of coal used in many of the nations under consideration; and (2) the national carbon dioxide emissions in the data compendium of the Carbon Dioxide Information Analysis Center (CDIAC) [*Marland and Boden*, 1993].

Figure 1 shows the direct comparison, between the aggregate coal consumptions in 1990 calculated here from the IEA tables [*IEA*, 1992a,b] and the similar data in *UN* [1995] for each country that appears in both databases. There is a systematic difference between

Table 1. Coal Consumption and Emissions of Hydrogen Chloride and Chloromethane, Country by Country

Country	Large Combustion Processes				Conversion Processes				Small Combustion Processes		Chlorine Content of Coal		FGD on Utilities, %	Cl Emission From Coal Combustion to Be Distributed		FGD on Incinerators, %	Cl From Combustion of Waste	Chloromethane Emission to Be Distributed		Notes
	Power and Heat Generation		Industry		Iron and Steel		Nonmetallic Minerals													
	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, %	Lignite, %		By Point Sources, Gg Cl	By population, Gg Cl		By Population, Gg Cl	By Point Sources, Mg Cl	By Population, Mg Cl	
Albania	0	477	0	0	0	62	0	0	0	1635	0.24	0.01		0	0.22		1.4	0	26	2, 6
Algeria	0	0	0	0	0	0	913	0	24	0	0.12	0.05		0	1.1		11	0	190	2, 5
Argentina	205	0	0	0	1060	0	0	0	112	0	0.12	0.05		0	1.6		14	0	250	2, 5
Australia	38,250	44,463	3621	493	502	0	995	66	321	99	0.2	0.3		210	13		7.2	3400	330	1, 9
Austria	1430	2175	21	197	1168	0	260	23	791	398	0.09	0.003	76	0.3	2	28	2.4	5	86	1, 10
Azerbaijan	10,510	0	0	0	2640	0	409	0	6176	0	0.12	0.05		13	11		3.0	200	230	4, 5
Bangladesh	0	0	0	0	0	0	540	0	18	0	0.12	0.05		0	0.67		48	0	790	2, 5
Belarus	723	0	0	0	521	0	81	0	1748	0	0.24	0.01		1.7	5.6		4.3	28	160	3, 6
Belgium	6637	0	522	2	4040	0	894	276	724	77	0.1	0.1		6.6	6.5	12	3.7	110	174	1, 9
Brazil	2758	0	804	0	10,336	0	1336	0	0	0	0.12	0.05		0	18		63	0	1300	2, 9, 10
Bulgaria	0	27,563	0	3996	0	1876	0	0	0	4314	0.12	0.05		0	19		3.8	0	370	3, 5
Canada	33,067	9117	880	231	1912	0	851	0	108	8	0.11	0.032		0	3		11	0	230	1, 5
Chile	2521	0	479	0	540	0	194	0	27	0	0.12	0.05		0	4.5		5.6	0	160	2, 5
Colombia	1398	0	1293	0	557	0	1929	0	280	0	0.12	0.05		0	6.5		14	0	330	2, 5
Cuba	0	0	0	0	140	0	0	0	0	0	0.12	0.05		0	0.17		4.5	0	76	2, 5
Cyprus	0	0	0	0	0	0	97	0	0	0	0.12	0.05		0	0.12		0.3	0	7	2, 5
Czech Republic	0	71,180	0	2289	0	8264	0	0	0	7130	0.1	0.015		11	2.6		4.4	170	110	4, 7
Denmark	9416	0	199	0	3	0	289	0	123	6	0.05	0.05	8	4.3	0.31		2.2	70	40	1, 10
Egypt	1	0	50	0	1289	0	0	0	0	0	0.12	0.05		0	1.6		22	0	390	2, 5
Estonia	7920	0	0	0	1990	0	308	0	4854	0	0.24	0.01		19	17		0.6	310	290	4, 6
Finland	3835	0	278	0	366	0	993	0	161	0	0.12	0.05	36	0	4.3		2.1	0	100	1, 5
France	11,642	1784	2654	214	5681	0	2050	0	1805	771	0.13	0.05	4	15	16		24	250	650	1, 5, 10
Georgia	1435	0	0	0	361	0	56	0	843	0	0.12	0.05		0	3.2		2.3	0	90	4, 5
Germany (former W.)	44,127	90,404	3571	2080	9954	39	1789	1704	1384	1892	0.1	0.015	28	42	18	59	14	670	829	1, 9
Germany (former E.)	0	11,211	0	18,934	0	33,483	0	11,704	0	10,976	0.1	0.015		1.7	11			27		9, 12
Greece	0	50,531	126	588	0	0	1293	17	3	115	0.12	0.05		25	2.1		4.3	410	100	1, 5
Hong Kong	8925	0	0	0	0	0	0	0	3	0	0.12	0.05		11	0.00		2.4	170	39	2, 5
Hungary	0	14,904	0	1830	0	993	0	234	0	2333	0.24	0.01		0	2		4.4	0	100	2, 6
Iceland	0	0	0	0	52	0	13	0	0	0	0.12	0.05		0	0.08		0.1	0	3	1, 5
India	123,880	0	54,834	0	30,910	0	12,140	0	5657	0	0.44			545	456		359	8800	13,000	2, 9

Table 1. (continued)

Country	Large Combustion Processes				Conversion Processes				Small Combustion Processes		Chlorine Content of Coal		FGD on Utilities, %	Cl Emission From Coal Combustion to Be Distributed		FGD on Incinerators, %	Cl From Combustion of Waste	Chloromethane Emission to Be Distributed		Notes
	Power and Heat Generation		Industry		Iron and Steel		Nonmetallic Minerals													
	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, %	Lignite, %		By Point Sources, Gg Cl	By population, Gg Cl		By Population, Gg Cl	By Point Sources, Mg Cl	By Population, Mg Cl	
Indonesia	4572	0	0	0	0	0	1804	0	0	0	0.12	0.05		5.5	2.2		78	89	1300	2, 5
Iran	0	0	1250	0	450	0	0	0	0	0	0.12	0.05		0	2		25	0	430	2,5
Ireland	1977	0	480	64	43	0	0	0	777	375	0.25	0.05		4.9	3.5		1.5	80	80	1, 8
Israel	3702	0	18	0	0	0	0	0	0	0	0.12	0.05		0	4.5		2.9	0	120	2, 5
Italy	10,782	1056	378	2	4470	22	1460	0	144	0	0.1	0.05		11	6.5	15	21	180	500	1, 10
Japan	29,334	0	10,199	0	31,612	0	8417	0	74	123	0.046	0.12	29	9.6	23		52	160	1200	1, 11
Kazakstan	57,610	0	0	0	14,428	0	2236	0	33,749	0	0.12	0.25		69	60		7.1	1100	1100	4, 5
Kenya	0	0	0	0	0	0	151	0	0	0	0.12	0.12		0	0.18		10	0	170	2, 5
Kyrgyzstan	1145	0	0	0	324	0	50	0	759	0	0.12	0.05		0	2.7		1.9	0	75	4, 5
Latvia	990	0	0	0	249	0	39	0	582	0	0.24	0.01		0	4.5		1.1	0	91	4, 6
Lithuania	5051	0	0	0	1269	0	197	0	2968	0	0.24	0.01		12	11		1.6	200	200	4, 6
Luxembourg	0	0	0	9	943	0	158	0	1	11	0.12	0.05		0	1.3		0.1	0	24	1, 5
Malaysia	1322	0	0	0	0	0	834	0	0	0	0.12	0.05		1.6	1		7.6	26	140	2, 5
Malta	300	0	0	0	0	0	0	0	0	0	0.12	0.05		0	0.36		0.1	0	8	2, 5
Mexico	3078	0	0	0	2890	0	0	0	0	0	0.12	0.05		0	7.2		36	0	700	2, 5
Moldova	2439	0	0	0	607	0	94	0	1421	0	0.1	0.015		0	4.6		1.9	0	100	4, 7
Morocco	1200	0	604	0	0	0	0	0	0	0	0.12	0.05		0	2.2		11	0	210	2, 5
Myanmar	20	0	98	0	0	0	0	0	0	0	0.12	0.05		0	0.14		18	0	290	2, 5
Netherlands	7917	0	421	2	1775	0	77	56	58	17	0.09	0.05	73	1.9	2.1	5	6	31	137	1, 11
New Zealand	232	0	915	101	647	0	0	0	224	74	0.04	0.05		0	0.9		1.4	0	37	1, 9
Nigeria	9	0	0	0	0	0	43	0	3	0	0.01	0.007		0	0.00		46	0	750	2, 9
North Korea	11,000	0	37,950	0	7000	0	0	0	0	0	0.12	0.05		13	54		9.2	210	1000	3, 5
Norway	22	0	213	0	868	0	127	0	14	0	0.12	0.05		0	1.5		1.8	0	53	1, 5
Pakistan	40	0	0	0	1102	0	3096	0	7	0	0.12	0.05		0.05	5		50	1	890	2, 5
Peoples Rep. of China	301,998	0	209,765	0	183,510	0	94,251	0	225,20	0	0.083			251	592		480	4100	17,000	3, 9
Peru	0	0	68	0	49	0	0	0	39	0	0.12	0.05		0	0.19		9.2	0	150	2, 5
Philippines	971	0	410	0	12	0	664	0	0	0	0.12	0.05		1.2	1.3		27	19	450	2, 5
Poland	0	144,468	880	0	18,261	0	3207	0	20,507	0	0.24	0.01		14	103		16	230	1900	3, 9
Portugal	3259	0	33	0	153	0	811	0	2	0	0.12	0.05		3.9	1.2		4.2	63	87	1, 5
Rep. of South Africa	76,251	0	13,912	0	8337	0	2211	0	1797	0	0.02			15	5.3		16	250	350	9, 13
Romania	36,954	0	1536	0	5097	0	121	0	1815	0	0.24	0.01		0	109		9.9	0	1900	3, 6
Russia	203,328	0	0	0	91,393	0	14,167	0	98,431	0	0.12	0.05		244	245		63	3900	5000	3, 5

Table 1. (continued)

Country	Large Combustion Processes				Conversion Processes				Small Combustion Processes		Chlorine Content of Coal		FGD on Utilities, %	Cl Emission From Coal Combustion to Be Distributed		FGD on Incinerators, %	Cl From Combustion of Waste	Chloromethane Emission to Be Distributed		Notes
	Power and Heat Generation		Industry		Iron and Steel		Nonmetallic Minerals													
	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, Gg	Lignite, Gg	Hard Coal, %	Lignite, %		By Point Sources, Gg Cl	By population, Gg Cl		By Population, Gg Cl	By Point Sources, Mg Cl	By Population, Mg Cl	
Slovakia	0	7800	0	1311	0	4136	0	0	0	4086	0.12	0.05		0	8.7		2.3	0	180	4, 5
South Korea	7706	0	46	0	11,906	0	4418	0	18,779	0	0.12	0.05		9.2	42		18	150	980	2, 5
Spain	22,922	16,895	511	0	2500	0	1779	0	578	5	0.1	0.05		31	5.4		17	510	350	1, 9, 10
Sweden	853	0	659	0	925	0	456	0	75	0	0.1		100	0	2.1	2	3.6	0	93	1, 11
Switzerland	21	0	81	0	0	0	409	0	10	16	0.1	0.007		0.0	0.5	8	2.6	0	54	1, 9, 10
Taiwan	7964	0	2097	0	4281	0	2827	0	0	0	0.12	0.05		9.6	11		8.6	160	320	2, 5
Tajikistan	759	0	0	0	191	0	30	0	446	0	0.12	0.05		0	1.7		2.3	0	64	4, 5
Thailand	0	9875	0	631	0	0	2133	0	0	0	0.12	0.05		4.9	2.9		23	80	420	2, 5
Turkey	474	29,884	1671	5168	1753	0	7	0	1522	9170	0.12	0.05		0	29		24	0	850	1, 5
Turkmenistan	6407	0	0	0	1610	0	250	0	3765	0	0.12	0.05		7.7	6.8		1.6	120	140	4, 5
United Kingdom	84,378	0	4323	0	4382	0	1434	0	7062	664	0.25	0.05		211	43		25	3400	1100	1, 11
Ukraine	52,402	0	0	0	44,597	0	6913	0	2216	0	0.12	0.05		63	64		22	1000	1400	4, 5
United States	629,112	70,943	40,969	7812	17,160	0	11,966	2	13,913	0	0.1	0.03	27	475	86		106	7700	3100	1, 11
Uzbekistan	22,898	0	0	0	5753	0	892	0	13,457	0	0.12	0.05		27	24		8.8	440	530	4, 5
Venezuela	0	0	0	0	23	0	332	0	0	0	0.12	0.05		0	0.43		8.2	0	140	2, 5
Viet Nam	2765	0	1206	0	0	0	0	0	95	0	0.12	0.05		3.3	1.6		28	54	480	2, 5
Yugoslavia (former)	0	68,863	0	3497	0	3260	0	439	0	1223	0.1	0.015		0	12		10	0	350	2, 7
Zambia	15	0	327	0	30	0	0	0	0	0	0.12	0.05		0	0.45		3.4	0	63	2, 5
Zimbabwe	2669	0	913	0	474	0	0	0	911	0	0.12	0.05		0	6		4.2	0	170	2, 5
totals, Gg Cl														2406	2233		1974	39	68	

Data quality, source 1 has the highest quality, source 4 the lowest: 1, from energy statistics of OECD countries [*IEA*, 1992a]; 2, from energy statistics and balances of non-OECD countries [*IEA*, 1992b]; 3, data as from 2 but tagged by IEA as being of lower quality, generally from centrally planned economies; 4, data calculated by difference from data in 2 and 3. Chlorine contents: 5, at global average; 6, at east European average; 7, similar to that in eastern Germany; 8, similar to that in the United Kingdom; 9, from *Swaine* [1990]; 10, from *Lightowlers & Cape* [1988]; 11, from *Sloss* [1992]; 12, total derived from *UN* [1995], assumed to be all of lignite quality, power requirement from *IEA* [1992b], remainder apportioned as in rest of Germany; 13, conversion to synthetic petroleum has been discounted from these figures.

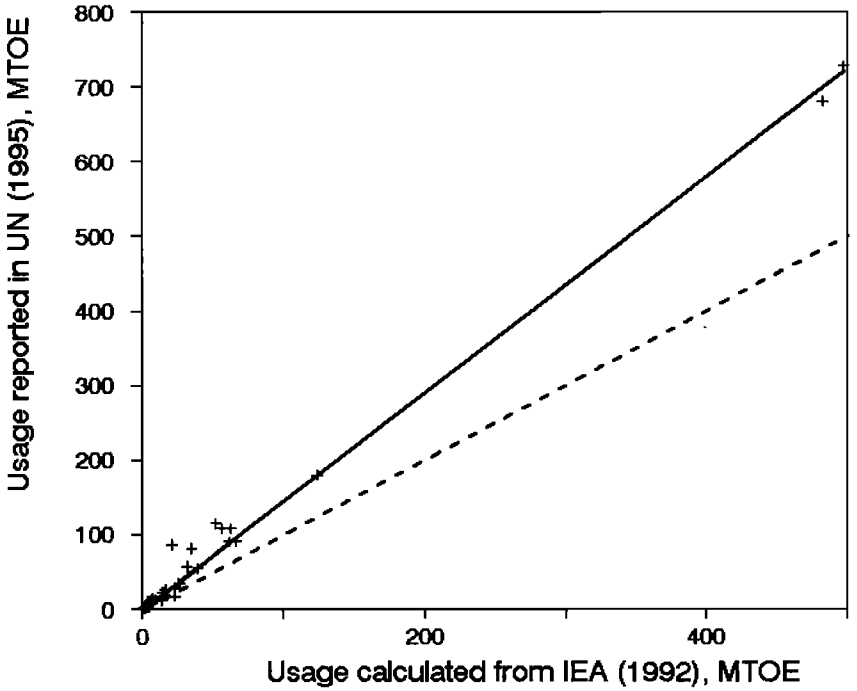

Figure 1. Comparison of total coal usage during 1990 in each country as reported in United Nations statistics [*UN*, 1995] with the values estimated in this work from the International Energy Agency databases [*IEA*, 1992a,b]. The solid line represents the best straight line fit to the points, and equivalence is shown by the dashed line.

the sets of data, with the values in this work consistently lower than those in the UN data by a factor of nearly 30%.

There are also some significant individual differences, and it is possible that there are problems with the consistency of units in the UN compilation; data for most countries seem to be in mass units, but there are some that seem to have been adjusted for calorific values. The adjustment factors are not given in the tabulations, so it is not possible to pursue the individual national differences further.

There is a much better agreement between the CDIAC database and the present work. To make the two databases comparable, the national values from this work were converted into national emissions of carbon dioxide from coal combustion. This was done on the basis of calorific value of the coal burned and a conversion factor, from calorific value to carbon dioxide emitted, which was developed from the data in *Jaques* [1992]. That work lists data for some 11 different varieties of coal burned in Canada. The empirical relationship between calorific value and carbon content, shown as the line fitted to the points in Figure 2, is

$$C = 87.16 \times T + 8.37 \qquad R^2 = 0.982 \qquad (1)$$

where C is the percentage of carbon in the coal, and T is the calorific value of the coal in tonnes of oil equivalent per tonne of coal (the units used in *IEA* [1992a,b]).

Using this relationship to estimate national carbon dioxide emissions from coal combustion, the data in this work compares very favorably with that for CO_2 from solids in the CDIAC compilation [*Marland and Boden*, 1993], as shown in Figure 3. There is a good fit over the 61 data points (R^2 is 0.996), and the CDIAC estimate is consistently 6% greater than that developed here, with the variability for an individual nation between the two data sets being approximately ±20% of scale. The fact that both databases show similar emissions of carbon dioxide may simply indicate that they both rely on the same basic information. Nevertheless, the fact that the data in the CDIAC compilation can be substantially reconstructed from those here shows that not only are the national coal usages consistent with other information but also the split between different grades of coal (with widely varying calorific values) must be reliable.

3. Chlorine Contents of Coals and Emission Factors

3.1. Chlorine Content

In one Australian study, the proportion of chlorine released from brown coal was shown to fall as the chlorine content of the coal increased [*Black and Fitzgerald*, 1986]. However, as a general rule it seems that most of the chlorine is actually released from burning coal and is emitted as hydrogen chloride; *Iapalucci et al.* [1969] estimated 94% emission and *Sloss* [1992] reported 99%. In this work it has been assumed that unless the flue gas is treated to remove ionic species, all of the chlorine contained in coal is emitted to the atmosphere as chloride ion (either on particulate material or as hydrogen chloride) or as chloromethane. The error that the assumption of total emission introduces is small compared to the uncertainties in the estimates of the chlorine contents of coal consumed.

Within a particular coalfield, deeper mined coal may have a higher chlorine content than shallow-mined material, but there does not seem to be a generally applicable correlation between type of hard coal and chlorine content; and the variation with depth has been assigned to local variation in the chloride content of the groundwater (the chloride concentration generally increases with depth below ground level) [*Sloss*, 1992]. According to *Hodges et al.* [1983] there are three principal forms of chlorine in coal: chloride ions dissolved in water in the coal matrix, solid inorganic chlorides, and organochlorine compounds in which the chlorine is chemically bonded to carbon. All are presumed to be released when the coal is burned, but the variation between the three types of chlorine, between coal measures and locally within each coalfield, precludes generalizations about chlorine content.

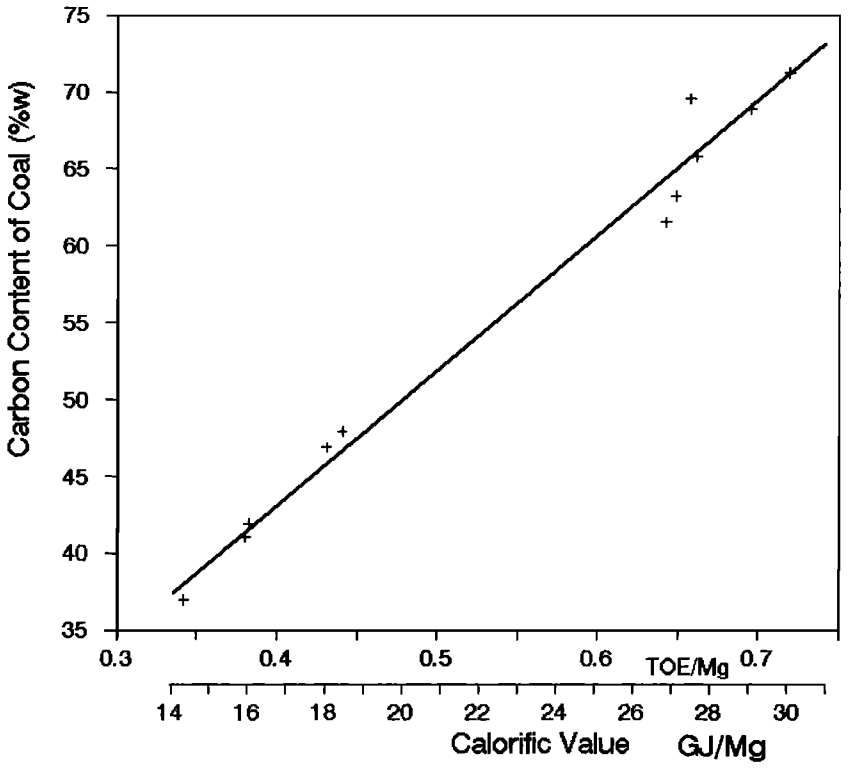

Figure 2. Relationship between carbon content of coals and their calorific values from the data in *Jaques* [1992]. The solid line is the best straight line fit to the points.

Chlorine contents of coals vary from a few tens of parts per million up to over 1%, and although there is no systematic variation of hard coals for the reasons shown above, the chlorine content of brown coal is generally less than that of hard coals, with the notable exception of some Australian lignites [*Swaine*, 1990]. On the basis of the *Lightowlers and Cape* [1988], *Swaine* [1990], and *Sloss* [1992] analyses, which describe chlorine in 54% of the coal used globally, mean values were developed for the chlorine contents of

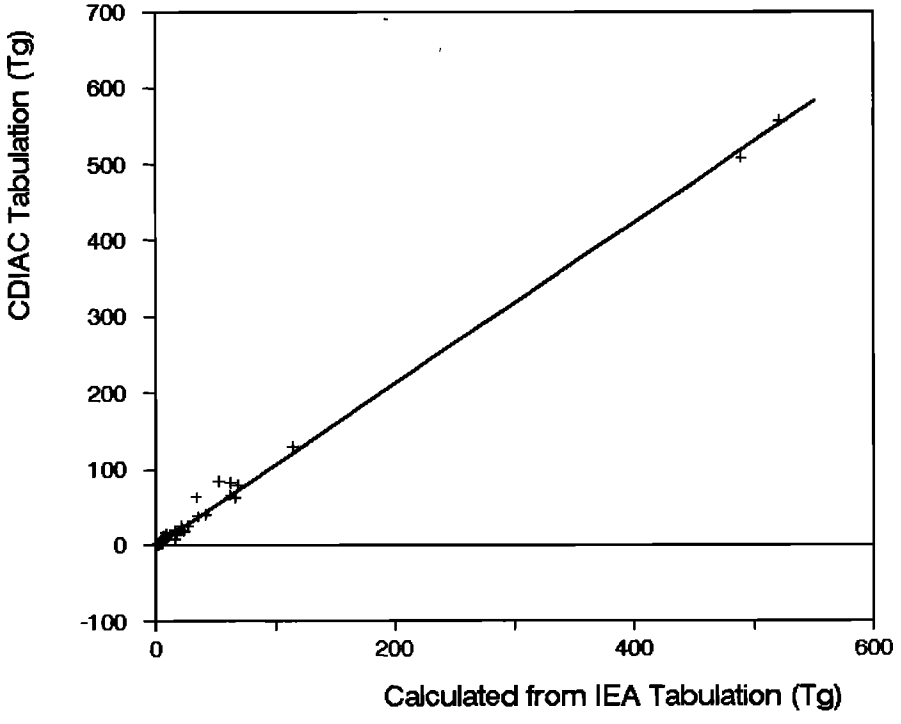

Figure 3. Comparison of national emissions of carbon dioxide from coal combustion calculated from the database developed in this work with that from the Carbon Dioxide Information Analysis Center database [*Marland and Boden*, 1993]. The solid line is the best fit to the points.

hard coal and lignite in eastern Europe (0.24% and 0.01%, respectively) and in the rest of the world (0.12% and 0.05%). These were used to calculate emissions from countries for which there were no specific data on coal chlorine contents. The complete list is shown in Table 1.

3.2. HCl Emissions

In calculating the mass fluxes of emissions, no account was taken of any dependency on combustion process, although there is some evidence that hydrogen chloride emissions change with the type of furnace; residential boilers have been shown, in some cases, to release 3 times as much chloride from anthracite as industrial furnaces and 25 times as much from lignite [*Misenheimer et al.*; 1985]. It is possible that this difference was due to the form in which the chloride was released, particulate versus hydrogen chloride, and the present work does not discriminate between these. Consequently, the maximum case was assumed in which all of the chlorine is released, 98.4% as HCl and 1.6% as chloromethane.

Allowance was made, however, for flue gas desulphurization (FGD). In electricity utilities this has been shown to reduce atmospheric chloride emissions substantially (by 90% or more [*Meij and Anderliesten*, 1989]). FGD plants generally convert sulphur dioxide into calcium sulphate using lime or limestone [*Maibodi*, 1991], so most of the chloride is sequestered into the aqueous effluent stream as $CaCl_2.xH_2O$ in wet processes and into the precipitated dust as $CaCl_2$ in dry processes [*Hammerschmid et al.*, 1990]. From a number of original references, *Sloss* [1992] lists the FGD capacity in the late 1980s in a number of countries. The chloride emission from electricity utilities in these countries was reduced pro rata to the fraction that this capacity represented of the total generating capacity, as reported in *IEA* [1992a]. The reduction factors are given in Table 1.

3.3. Chloromethane Emissions

While most of the chlorine content of burning coal is emitted as chloride ion in one or another form, a small fraction is converted into chloromethane and released as such. M.A.K. Khalil (personal communication, 1997) has analyzed emissions from burning Chinese and U.S. coals and found significant differences in emission rates expressed relative to CO_2 (2.2×10^{-6} and 4.6×10^{-6} g/g CO_2, respectively). However, the carbon content of coal, and hence CO_2 emitted, depends on calorific value as described in equation (1). U.S. coal has a higher average calorific value and a higher typical chlorine content, and so when the average results are adjusted for these, the emission functions become closer at 1.9×10^{-2} g/g Cl for China and 2.6×10^{-2} g/g for the United States. An average of 2.3×10^{-2} g chloromethane/g Cl was used to estimate national emissions from coal combustion; this is equivalent to 1.6% of the chlorine in coal being liberated as chlorine in chloromethane. Since there is no evidence that flue gas desulphurization affects other than ionic species, the chloromethane emissions were not adjusted for FGD processing. The values shown in Table 1 include the releases from waste incineration (see below) and are expressed as the chlorine content of chloromethane. Note also that since the numbers are 50 times smaller than the ionic chlorine emissions, they are expressed in the table as Mg.

4. Hydrogen Chloride and Chloromethane From Waste Incineration

In much the same way as chlorine in coal and biomass is liberated as hydrogen chloride on combustion, so the chlorine in municipal and domestic wastes may also be released when they are burned, and the global emission of chlorine from this source has been estimated to be in the region of 2 Tg yr^{-1} [*Graedel and Keene*, 1995; *Aucott*, 1997].

This estimate is consistent with 40% of municipal and domestic waste being burned. *Pirrone et al.* [1996] estimated the U.S. generation of solid waste to be 1 to 1.8 kg $person^{-1}$ d^{-1} and that people in developing countries produced 0.6 kg $person^{-1}$ d^{-1}. Taking the lower end of the U.S. range as representative of the rest of the developed world, the global rate of generation of waste is estimated to be 1.5 Pg yr^{-1}. For this exercise, waste is assumed to be burned only by the 89% of the global population in the coal combustion inventory. *Lightowlers and Cape* [1988] report that United Kingdom waste contains 0.32% insoluble chlorine (assumed to be present in the form of polyvinylchloride, PVC) and 0.27% soluble chloride ion. *Bouscaren et al.* [1985] found that 65% of the chlorine in PVC was mobilized into the gas phase as hydrogen chloride during combustion in a municipal incinerator, and Lightowlers and Cape suggest that 50% of the soluble chloride is emitted as HCl during combustion. Thus the total potential emission of HCl could be in the region of 5 Tg yr^{-1}, if all waste were burned.

In the developed world the proportion of domestic waste that is burned in municipal incinerators varies from zero in Ireland to 77% in Switzerland [*Lightowlers and Cape*, 1988]. A comparable amount could be burned in relatively uncontrolled trash fires in areas where central incineration is not practiced [*Lemieux*, 1997]. For the total emission from this source to be 2 Tg yr^{-1}, the extent of combustion would have to be 40% of all waste, averaged over developed and developing countries.

This is probably a high estimate; in a survey conducted in India, only 15% of the vegetable waste, 11% of paper waste, and 5% of plastic waste was burned by the first generators of the waste [*Chapman et al.*, 1997]; 74% of the vegetable waste was composted or reused, and 83% of paper and 89% of plastics in the waste were reused. In addition, trials of combustion of household waste in barrels (apparently a common method of disposal used in rural areas of the United States) showed that very little of the chlorine in PVC was liberated as HCl at this scale of burning [*Lemieux*, 1997]. Liberation of chlorine, in all forms, into the atmosphere was 6% in one trial and 13% in another. However, these observations may not reflect the situation in the majority of countries, and so to calculate emission functions, the global release of chlorine (as HCl and chloromethane) from waste combustion was set at 2 Tg yr^{-1}.

In developing the estimate of the spatial distribution of HCl releases, allowance was made for flue gas treatment on incineration units, which may significantly reduce hydrogen chloride emissions from the point source [*Lightowlers and Cape*, 1988]. Although such reductions do not actually apply to the uncontrolled and local trash burning which has been assumed to be practiced along with controlled incineration, the national hydrogen chloride emissions from all waste combustion were reduced pro rata to the extent of flue gas treatment on controlled plants. The reduction in emission resulting from this assumption is consistent with the expectation that a state that encourages the cleaning of incinerator gases would also discourage uncontrolled trash burning.

To minimize costs, municipal incinerators tend to be sited close to the centers of population that they service; for example, those in the United Kingdom are located in London and the industrial regions of the Midlands and north of England [*Environmental Business Magazine (EBM)*, 1997]. Thus the spatial distribution of controlled incineration as well as domestic trash burning may be described by population. Given all of the uncertainties, a single per capita function was assigned to the HCl emission from waste burning and used globally. This was calculated as the chlorine release (98.4% of 2Tg yr^{-1}) divided by the world population in the combustion inventory, resulting in a value of 0.42 kg Cl $person^{-1}$ yr^{-1}. National emissions

were then the product of population and this function; these emissions are shown separately in Table 1. The emissions were geographically distributed within each country using the population distribution of *Li* [1996] and, for each grid square, were added to the emissions from coal combustion.

As described above, some of the chlorine in coal is emitted as chloromethane during combustion, and it is to be expected that some of the chlorine in municipal waste would follow the same chemical pathway. The same emission function has been used in both cases, and so the chlorine content of chloromethane releases was calculated as 1.6% of the sum of the chlorine present in coal and the chlorine present in domestic and municipal waste in each country.

5. Distribution of Coal Combustion Emissions

5.1. Power Plants

The fraction of HCl and CH_3Cl emissions from coal combustion for power and heat generation have been distributed as point sources where power plant information was available. Information on coal-fired power plants was derived mainly from the work of *Benkovitz et al.*, [1996] for the GEIA 1985 inventories of anthropogenic emissions of SO_2 and NO_x. The 1985 information was supplemented with data obtained to update the GEIA inventories to base year 1990; this work is currently in progress. In total, seven sets of data covering 43 counties have been used in the work described here. Information in these data accounts for 90% of the 1990 global consumption of coal used for power and heat generation. Countries for which no point source data were available, such as those in Latin America and most of Africa, have had their HCl and CH_3Cl emissions for power and heat generation distributed on the basis of population.

The region covered, contributor, and information included are listed in Table 2. Most information included location, generating capacity, SO_2 emissions, NO_x emissions, and other plant information; however, some comprised only plant information and location. Where data on emissions were included, the values were used as surrogates to distribute the country total HCl and CH_3Cl emissions due to power and heat generation. The fractional contribution of each point source to the country SO_2 or NO_x emissions was used to distribute national HCl and CH_3Cl emissions, which were assigned to the grid location of each point source. In cases where no suitable surrogate parameter was available, country emissions due to power and heat generation were divided evenly between all the point sources listed for that country.

5.1.1. Eastern Europe, Former USSR (FSU), South Africa, and Australia. For the power plants in Australia and South Africa, SO_2 emissions were used as the surrogate to allocate HCl and CH_3Cl emissions. For eastern Europe and the former USSR countries, industry codes were used to select only power plants and NO_x emissions were used as the surrogate. For the former USSR each power plant was allocated to the appropriate independent state to match the allocation of the coal consumption data [*IEA*, 1992b]. No point sources were listed for Georgia, Kyrgyzstan, Moldavia, and Tajikistan, and coal in Latvia was consumed in processes other than power and heat generation.

5.1.2. Western Europe. Because of confidentiality restrictions, the European "CORINAIR" data do not list individual power plants but

Table 2. Databases of Spatial Distributions of Power Utilities Point Sources

Region	Contributor	Creator/Developer	Included
Asia	Markus Amann[a]	RAINS-ASIA[b]	plant name, country, region, longitude, latitude
United States	Dave Mobley[c]	Suzy Rothchild[d]	plant name, FIPS codes, latitude, longitude, capacity (MW)
Eastern Europe	Arthur Li[e]	Jozef Pacyna[f]	plant name, latitude, longitude, industry code, NO_x (1985), country
Former Soviet Union (FSU) or USSR	Arthur Li[e]	Jozef Pacyna[f]	plant name, latitude, longitude, industry code, NO_x
Western Europe	Trevor Sholtz[g]	CORINAIR[h]	latitude and longitude of GEIA grid square with SO_x and NO_x emissions due to power plants
South Africa	Trevor Sholtz[g]	S.M. Lloyd[i]	latitude, longitude, SO_2, NO_2, CO_2, and total gas volume
Australia	Frank Carnovale[j]	Frank Carnovale[j]	plant name, type, latitude, longitude, SO_2, NO_x

[a] International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria.
[b] Acid rain and emission reductions in Asia project.
[c] U.S. Environmental Protection Agency, Research Triangle Park, North Carolina.
[d] E.H. Pechan and Associates, Durham, North Carolina.
[e] Atmospheric Environment Service, Downsview, Ontario, Canada.
[f] Norsk Institut for Luftforskning (NILU), Kjeller, Norway.
[g] ORTECH International, Mississauga, Ontario, Canada.
[h] [*Bouscaren and Fontelle*, 1992].
[i] South African Department of National Health and Population Development, Pretoria.
[j] Coffey Partners International, Victoria, Australia.

allocate their emissions to the corresponding cell in the GEIA 1° × 1° grid at the appropriate vertical level (above and below 100 m). Grid cells with elevated releases were identified as locations of major point sources; these cells were then associated with individual countries [*Rand McNally*, 1993]. The CORINAIR SO_2 emissions from coal-burning power plants were used as the surrogate to allocate country HCl and CH_3Cl emissions.

5.1.3. United States. The data for the United States included power plant information, fuel type, and generating capacity. HCl and CH_3Cl emissions were distributed among the coal-fired power plants using generating capacity as the surrogate.

5.1.4. Asia. The data for Asia included power plant name, province, and country but did not include any appropriate surrogate to coal consumption. For these countries, HCl and CH_3Cl emissions have been distributed uniformly among all the power plants listed. While this introduces some error by treating all power plants within a country as equal and assuming that all power plants listed are coal fired, it will not affect a country's total HCl or CH_3Cl emissions but rather just the distribution of those emissions within an individual country and is preferable to distributing the power and heat generation emissions by population.

5.1.5. Emissions from residential and commercial activities. The quantities of coal burned in residential and commercial fires is a simple function of population. The population distribution sometimes follows the distribution of industrial activity, although the link is less distinct in mixed industrial economies that do not rely exclusively on heavy industries. *Tremolada et al.* [1996] found that the spatial distribution of the deposition of polycyclic aromatic hydrocarbon (PAH) combustion products in the United Kingdom was linked to population density. Although coal combustion is by no means the sole source of PAHs, this study establishes a connection between population and combustion emissions. Therefore the emissions of HCl and chloromethane from coal combustion in the residential and commercial categories, from industrial processes burning coal, and from coal-fired utilities (in nations where the actual locations of the power plants was not known) were distributed as described by *McCulloch et al.* [this issue], using the population density grid of *Li* [1996].

5.2. Chloromethane From Industrial Activities

5.2.1. National emissions and development of an emission function. Chloromethane is a widely used industrial chemical, predominantly as a feedstock for the manufacture of other materials. Approximately 80% of chloromethane production is converted into methyl chlorosilanes from which silicone fluid, elastomers, and resins are manufactured. Chloromethane is also converted into cellulose ethers and quaternary ammonium compounds, and in some countries, it is still used to make the gasoline antiknock additive, tetramethyl lead [*Linak and Yau*, 1995]. From all of the foregoing uses, emissions are adventitious and minor. However, when chloromethane is used as a solvent, as in the production of some butyl elastomers, significant quantities may be lost to atmosphere and can be an important point source [*National Pollutant Release Inventory (NPRI)*, 1995].

It was estimated that in 1990, some 251,000 Mg were used in the United States, comparable figures were 3000 Mg in Mexico, 1500 Mg in South Korea, 252,000 Mg in western Europe and 97,000 in Japan [*Linak and Yau*, 1995]. There are no data for the centrally planned economies of Europe and Asia, and in this work, it has been presumed that this indicated no use there.

Emissions from U.S. sources are recorded in the Toxics Release Inventory (TRI) [*USEPA*, 1994]. For this, enterprises in the United States employing more than 10 people are required to report their emissions of pollutants, inter alia chloromethane. Those manufacturing or processing less than 25,000 lbs/yr or otherwise using less than 10,000 lbs/yr are exempt. However, chloromethane is used in generally large-scale chemical manufacturing processes, and so it is reasonable to assume that the TRI captures most, if not all, of the releases. These values were then used to develop an emission function of 1.44×10^{-2} g emitted/g used, based on the total quantity recorded in the TRI and the estimate for the U.S. use of chloromethane.

5.2.2. Distribution of industrial emissions. The industrial emissions are described in terms of mass of chloromethane; all were distributed as point sources and, when placed in the 1° × 1° grid, were converted into the equivalent chlorine emission. In the United States and Canada, distribution was done directly from the regulatory databases: 3600 Mg U.S. emissions were distributed as in TRI [*USEPA*, 1994], and the 1100 Mg emitted from a single point source in Canada was assigned as in *NPRI* [1995]. In Europe and Japan the distribution of producers' facilities, as recorded by *Linak and Yau* [1995], was used as a surrogate for consumers. The rationale for this is that since chloromethane is used in large-scale chemical production, the likelihood is that production and consumption facilities will be in the same 1° x 1° grid square. Thus the 3600 Mg of European chloromethane emissions were distributed 20% from two locations in France, 45% from five locations in Germany, 2% from one location in Italy, 5% from one in Spain, 1% from one in Switzerland and 28% from two in the United Kingdom. Similarly, the 1400 Mg of Japanese emissions were distributed over seven localities covering six grid squares. Mexican (43 Mg) and South Korean (21 Mg) emissions were placed in single grid squares. The combined total of industrial process emissions, expressed as chlorine, amounts to 7 Gg in 1990.

6. Results and Discussion

For some 15 of the nations listed in Table 1, data for HCl emissions from coal combustion are also listed in the work of *Lightowlers and Cape* [1988] (with additional information from *Sloss* [1992]). They comprise some 22% of the global HCl emission from this source. These referenced data and the equivalent values developed in this work were shown to be not significantly different (using Student's paired *t*-statistic). The same is true for the HCl emissions from waste burning, although in this instance there are 14 countries in the data sets, covering 9% of global emissions from this source. For both coal and waste combustion the average of the literature values was a little larger than that of those developed in this work, but the difference was not statistically significant.

In section 2.2. on verification above we showed that the coal combustion database developed here gave results consistent with other databases, so consistency of chlorine emissions as hydrogen chloride between subsets of the databases is to be expected. However, the waste combustion databases were constructed in wholly different ways, the referenced data being based on national statistics (built from the bottom up) rather than by subdivision of a global total, as in this work. The fact that the data sets are consistent for Europe and Japan indicates not only that these techniques converge but also that the global fraction of the total release from Europe and Japan calculated in this work is of the right size. This builds confidence that it is valid to base emissions of HCl and chloromethane from waste burning on population statistics.

The distributions of hydrogen chloride and chloromethane from all industrial sources, expressed as chlorine, are shown in Plates 1 and 2. Clearly, emissions of both compounds from industrial activity and waste combustion are features of the Northern Hemisphere but not just the industrially developed regions; Asian developing countries also have significant emissions. Because of the factors controlling

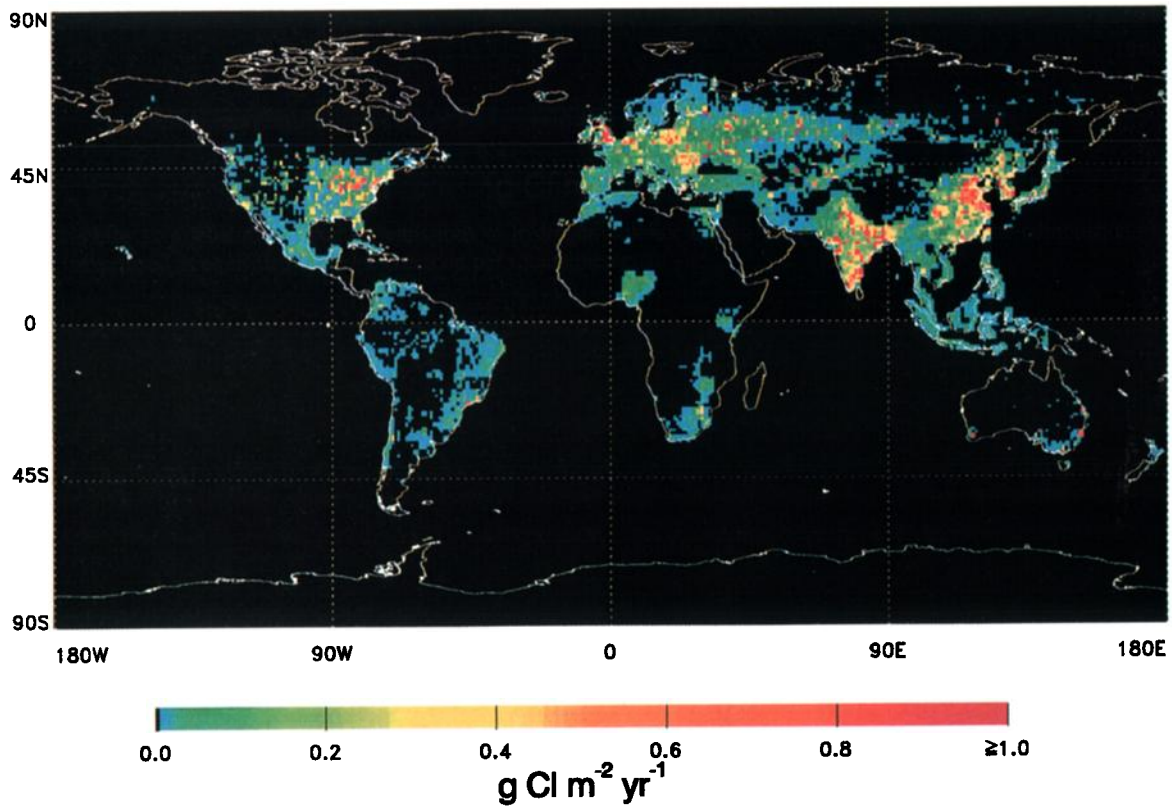

Plate 1. Global distribution of emissions of hydrogen chloride during 1990 from coal and waste combustion, expressed as equivalent chlorine emission per square meter.

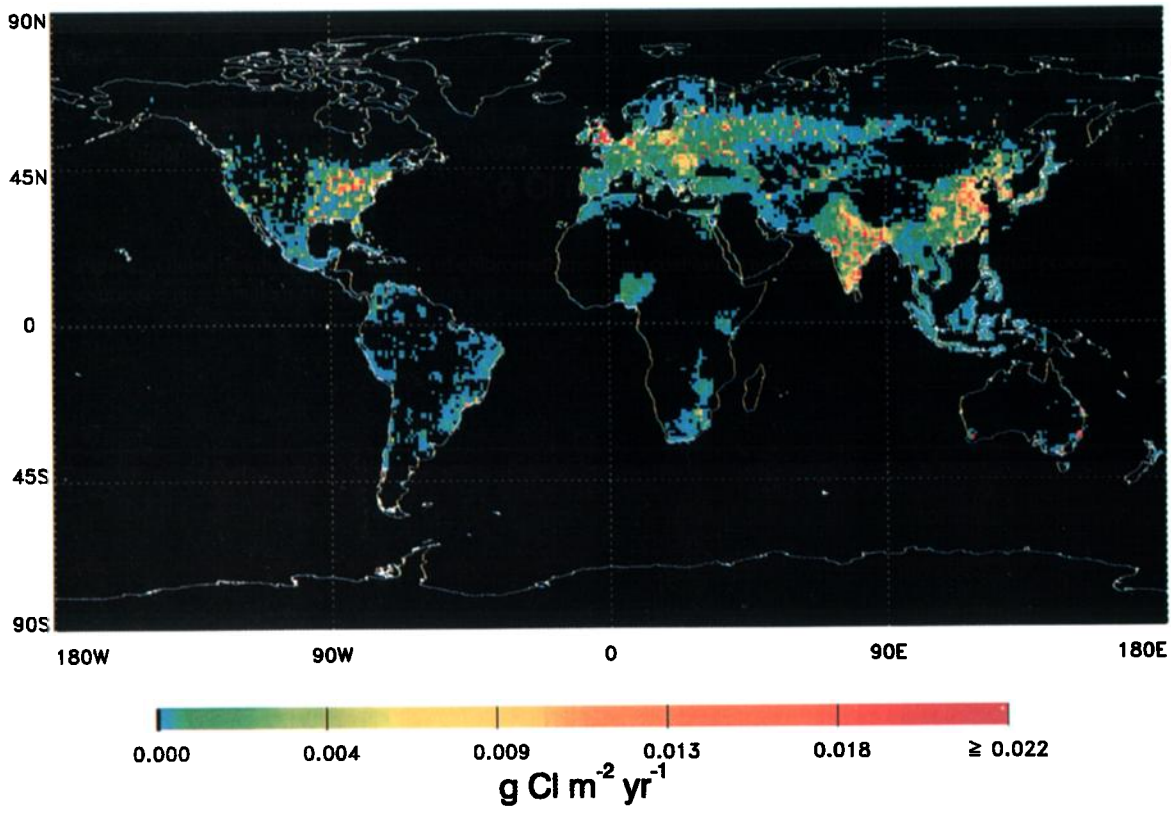

Plate 2. Global distribution of emissions of chloromethane from coal and waste combustion and industrial processes, expressed as equivalent chlorine emission per square meter.

releases being both industrial activity and population density, the emissions in these regions arise from a combination of point sources and a high background. By contrast, those southern hemispherical emissions, which are significant, are from point sources. This is made the more obvious by our decision not to assign waste combustion emissions to those countries that did not have emissions from coal burning, so much of Africa remains blank. The releases here are at the bottom of the scale and are not shown.

The HCl emissions for 1990 distributed as in Plate 1 total some 6.6 ± 6.2 Tg, 4.6 ± 4.3 Tg from all forms of coal combustion, and 2 ± 1.9 Tg from waste burning. The relatively large, 1σ, uncertainty in the coal release is due to the variability of the chlorine content. It is not possible to formalize uncertainty in the same way for waste incineration, and it is likely to be at least as large as that for coal. These emissions are anthropogenic and are, with the exception of trash burning, industrial; they contribute about 10% of the global source strength shown by *Keene et al.* [this issue]. They are distinct from the emissions that fall within the broader definition of anthropogenic, which include HCl liberated from sea-salt aerosol by acidification due to anthropogenic SO_2 and NO_x releases into the atmosphere.

Chloromethane emissions distributed as in Plate 2 total some 107 ± 93 Gg (as chlorine), most of which (100 Gg) arises from combustion. The rest is due to losses from industrial manufacturing processes that use chloromethane. As was the case with HCl, the uncertainty arises from variability in the chlorine content of coals. These industrial anthropogenic emissions are a minor component, less than 4%, of the global flux of 2.8 Tg yr^{-1} estimated from the known sinks by *Keene et al.* [this issue]. Although the anthropogenic emissions of neither compound are important for the global flux, anthropogenic HCl in particular could influence atmospheric chemistry locally in regions remote from the influence of oceanic air. This is particularly true in central Asia where only northerly airstreams are free of the immediate influence of industrial activity.

7. Access to Data

The inventory grids generated from this work are available on-line from the GEIA website at http://groundhog.sprl.umich.edu/geia/rcei. It is planned to update these inventories as new information becomes available.

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