The Spatial and Source Type Distribution of Emissions of Selected Toxic Volatile Organic Compounds in the United States in 1990

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ABSTRACT

An improved interim toxic emission inventory for the purpose of screening-level regional dispersion and deposition modeling is estimated from a 1990 interim emission inventory of volatile organic compounds (VOCs) for the United States. The VOC emission inventory was derived by updating portions of the 1985 National Acid Precipitation Assessment Program emission inventory. The most current emission factors and speciation profiles available in late 1992 are used to derive emission estimates for specific toxic compounds. Emission factors are used in preference to speciation where possible. The annual anthropogenic emissions, principal contributing source types, and spatial distributions for four selected toxic VOCs, including acrylonitrile, benzene, perchloroethylene, and trichloroethylene, are presented for the United States. The resulting emission estimates are an improvement over an earlier application of this approach, despite necessary heavy reliance on general and default speciation profiles. Emission totals generally exceed national estimates based on partial or top-down inventory approaches by one to two orders of magnitude. The source type and geographical concentrations of toxic VOCs, as well as further emission data needs are discussed. For each toxic compound examined, annual emissions for dominant point, area, and mobile source categories are presented, with the emphasis on demonstrating any distinct geographic patterns. In general, point source toxic emissions are coincident with urban concentrations and chemical industry concentrations.

INTRODUCTION

The requirements of Title III of the Clean Air Act Amendments of 1990[1] have created a need for a variety of toxic air emission data, including numerous studies and regulatory activities for specified toxic substances. Title III also requires that the transport and deposition of toxic substances into the "Great Waters" (the Great Lakes, Lake Champlain, Chesapeake Bay, and other specified coastal waters) of the United States be investigated. This requires detailed toxic emission inventories for all sources on at least a national scale. Such an inventory based on traditional facility reporting does not exist.

To address the need for regional or national toxic emission inventories, EPA examined existing toxic emission inventory data in the absence of any single inventory with information suitable for regional air quality modeling, and began compilation of an estimated

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national interim toxic annual emission inventory.[2,3] The initial compilation effort focused on 26 compounds of interest in the Great Lakes area. Because of limited emission factors and activity data for toxic emissions, the emission estimates were based on speciation[4] of the base year 1985 total suspended particulate (TSP) and volatile organic compound (VOC) inventories gathered for the National Acid Precipitation Assessment Program (NAPAP).[5] The results were generally two or more orders of magnitude larger than existing toxic emission estimates, such as the emissions estimates prepared for the Province of Ontario.[6] This was due in part to old data used in NAPAP, in some cases dating to the 1970s; and to uniform application of speciation profiles of different quality to sources with variable characteristics. An effort to improve the interim toxic emission inventory is underway. This paper has three objectives with respect to the improvement effort. The first objective is to summarize the procedure for expansion and improvement of the interim toxic emission inventory using updated base inventories, the most current chemical and source-type-specific emission factors, and updated speciation profiles. The second objective is to examine the basis of the toxic emission estimates using four examples. Finally, the geographical distribution of the example toxic emissions are presented as examples of the magnitude and spatial variability of toxic emissions.

METHODOLOGY

The updated interim annual toxic emission inventory uses the 1990 EPA interim national emission inventory[7] as the basis for VOC-related toxic emissions. Because the 1990 inventory does not address particulate matter, the updated toxic emission inventory will use the 1985 NAPAP inventory for particulate-related toxic emissions. The 1990 interim inventory will be improved as 1990 emission data from the states are completed under the State Implementation Program for ozone precursor pollutants. There is generally insufficient information at this time to perform temporal allocation of the estimated emissions to seasonal, daily, or hourly values. Sixty compounds, including the 26 previously addressed, were tentatively selected for the updated toxic emission inventory, based on a draft ranking of Agency program needs and compound toxicity. Current information was examined to extract emission factors for each toxic pollutant-emission source category pair. Where there was no emission factor information, the best available speciation profile information was determined for speciation profile-emission source category pairs. Although the information available is improved from 1991, in most instances it was necessary to use a default profile. The default profiles are average representations of all profiles currently available, and although improved, are of low quality when applied to specific sources. Emission estimates attributable to default profiles were tracked to help define uncertainty in the interim toxic emission inventory. The estimated annual emissions of each toxic compound were calculated by following similar, but slightly different procedures for point, area and mobile sources.

Point Sources

To estimate point source emissions, it was necessary to determine if an emission factor existed for each compound-source type combination. If so, emissions were estimated by multiplying the emission factor with the corresponding activity data for each source. If not, available information was investigated to determine if a source type-speciation profile pair was available for the toxic compound. If so, the speciation factor in the profile for the toxic compound was multiplied against the total VOC or TSP for the source, as appropriate.

If there was neither an emission factor nor directly applicable speciation profile, a general default profile was used.

Area Sources

Because of a lack of appropriate emission factors and area source activity data, area source emissions were estimated by speciation. If a source type-speciation profile pair was available that included the toxic compound, the speciation weight factor was multiplied with the VOC emission for the source. If no directly applicable source type-speciation profile wa available, a speciation weight factor from a general default profile was used.

Mobile Sources

Mobile source emissions for criteria pollutants are calculated for specific vehicle types, road use, and environmental conditions. This procedure is not available for direct determination of toxic emissions. Consequently, mobile source emissions were estimated by speciation. Speciation profiles specific to vehicle exhaust and portions of evaporative loss were used where available. If the specific profiles were not applicable, more general profile linked to mobile source types were used. In the absence of either kind of emission profile, a general default speciation profile was used.

BASIS OF INFORMATION

This paper presents example preliminary emission estimates for each state for the VOC-based toxic compounds acrylonitrile, benzene, perchloroethylene and trichloroethylene (Table 1). These compounds were selected because of their common use. Benzene is a constituent of fuels, while perchloroethylene and trichloroethylene are solvents often used in degreasing. Emission estimates for these examples are based on the VOC emissions in the 1990 EPA interim emission inventory. In each case, the point source emissions were estimated using a combination of emission factors and speciation. The results are of the same order of magnitude as the previous version of the interim toxic emission inventory. Decreases and increases varied from none to a factor of eight, reflecting changes in emissior factors and speciation profiles. The fraction of estimated emissions for point, area, and mobile sources for each compound derived from emission factors, speciation with source-type-specific profiles, and general default profiles is given in Tables 2 and 3.

Despite substantial new information, the frequency of use of general default speciatio profiles ranges between 90 and 100 percent of all sources for most toxic emissions. Benzen is an exception because more emission factors are available. The emission estimates in this paper for acrylonitrile and benzene are more than two orders of magnitude larger than emission estimates by the 1991 Toxic Chemical Release Inventory System (TRIS).[8] The trichloroethylene emission estimates are one order of magnitude larger than the TRIS estimate. The differences are attributable to several factors. The TRIS does not include are or mobile sources in estimating emissions, or many non-manufacturing point sources. Because area sources are extremely important for emissions of three of the four toxic air pollutants addressed in this paper, it is to be expected that the emission estimates will be larger than the TRIS estimates. There is evidence that, even when only point sources from a reported facility-level inventory are compared to TRIS, TRIS emissions are at least an order of magnitude too low.[9] Limited measured emission data makes it difficult to quantify the accuracy of the estimates presented in this paper. The estimates may be somewhat high,

despite the use of new emission factors and profiles, because of the continued need to use generalized speciation or default profiles for many source categories. Application of averaged speciation profiles to many small sources tends to increase the total emissions. Small sources can not be ignored in a comprehensive inventory. However, use of averaged or default speciation profiles tends to overestimate emissions because all existing speciation profiles are not complete (which may inflate the relative percentage weight of compounds that are in the profile), and averaged profiles are probably not applicable to some of the sources. Tables 2 and 3 illustrate the heavy reliance on speciation profiles, both in terms of the number of sources addressed and in terms of the amount of emissions. The basis for estimating overall emissions will improve as more air toxic emission factors are developed and used. In the meantime, estimates made using the approach described here provide a first order working estimate of toxic emissions from all sources and the means by which screening-level dispersion and deposition modeling can begin.

DISTRIBUTION OF EMISSIONS

As expected, the geographical distribution of emissions reflects population, manufacturing, and petrochemical industry concentrations. States with large chemical industries generally have the greatest point source emissions of the selected toxic compounds (Table 4). Benzene emissions are a factor of two to three larger than emissions of the other three chemicals because of the prevalence of benzene in fuels and chemical processes. Emissions from area sources (Table 5) and mobile sources (Table 6) reflect the relative populations and concomitant economic activity levels of the states, consistent with estimates of VOC for area and point sources.

Mobile and area sources of vehicle exhaust and evaporative loss, fuel transport and storage, degreasing, and cleaning are the predominant emission sources of acrylonitrile, benzene, and perchloroethylene (Table 7). Trichloroethylene emissions are principally from point sources of cleaning, degreasing and adhesive application. The manufacture of these compounds is also an important emission source. The locations of the dominant source types are consistent with the geographical distribution of the emissions mentioned previously.

CONCLUSIONS

This paper presents an estimation procedure and resulting national and state estimates of annual anthropogenic emissions of four example toxic compounds from the 60 compounds for which emission estimates are being prepared. Although heavily dependent upon default speciation profiles, these emission estimates represent an improved first order, possibly large estimate for a regional dispersion modeling inventory including most sources. The TRIS estimates are by definition less than the total emissions of any given toxic air emission. The uncertainty in the emission estimates provides an accuracy limitation on regional dispersion modeling efforts, and limits modeling results to relative transport and deposition patterns demonstrating ranges of quantitative results.

DISCLAIMER

REFERENCES

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Table 1. Selected annual state total toxic volatile organic compound emissions (tons per year) based on 1990 interim emissions inventory.

State	Acrylo-nitrile	Benzene	Perchloro-ethylene	Trichloro-ethylene
AL	3589	17949	3909	2666
AZ	1539	8242	1796	1141
AR	1381	8003	1620	1027
CA	11349	64284	13921	25527
CO	1317	7547	1580	2830
CT	1168	6473	1335	829
DE	678	3694	813	499
DC	144	832	173	106
FL	5121	27836	6165	5466
GA	3813	20966	4055	2817
ID	1017	5285	1227	754
IL	6135	39443	8196	62270
IN	3697	20898	1202	10896
IA	1370	7116	1608	1492
KS	1380	7811	1645	1177
KY	2143	12898	2566	1525
LA	4221	26191	4086	39333
ME	537	3295	645	397
MD	1692	9395	1980	1231
MA	2374	13237	2867	52725
MI	4493	26395	5577	4309
MN	2335	13541	2807	2115
MS	1863	9948	2422	1992
MO	3114	16422	3991	5614
MT	796	4911	958	588
NE	764	4061	928	592
NV	485	2802	537	360
NH	435	2552	531	11860
NJ	3551	21472	3734	2541
NM	813	4979	978	601
NY	7305	35116	7002	4376
NC	4787	26046	5498	4668
ND	457	2478	540	336
OH	5893	28352	6214	4231
OK	1723	9818	2441	4771
OR	1582	8711	1901	1179
PA	5871	31186	5956	4450
RI	418	2451	519	299
SC	4103	19668	2739	3308
SD	537	2815	648	399
TN	3673	21993	4290	4977
TX	34088	144786	24840	14811
UT	794	4602	932	580
VT	246	1468	296	211
VA	4726	19056	4247	2887
WA	2726	15965	3213	2008
WV	3873	22866	1976	36872
WI	2838	15736	3956	18329
WY	360	2624	431	265
TOTAL	159314	834216	165956	354237

Table 2. Frequency of emission estimation method use.

	Point sources	Area sources	Mobile sources
Acrylonitrile			
Emission factors			
Number of times used	7	0	0
Percent of total	0.1	0	0
Specified emission profiles			
Number of times used	4275	618	0
Percent of total	9.0	0.3	0
Default profile			
Number of times used	41132	207692	187744
Percent of total	90.9	99.7	100.0
Total number of sources	45414	208310	187744
Benzene			
Emission factors			
Number of times used	122	0	0
Percent of total	0.3	0	0
Specified emission profiles			
Number of times used	27242	5660	187744
Percent of total	60.0	3.0	100.0
Default profile			
Number of times used	18050	202650	0
Percent of total	39.7	97.0	0
Total number of sources	45414	208310	187744
Perchloroethylene			
Emission factors			
Number of times used	77	0	0
Percent of total	0.2	0	0
Specified emission profiles			
Number of times used	916	1886	0
Percent of total	2.0	1.0	0
Default profile			
Number of times used	44421	206424	187744
Percent of total	97.8	99.0	100.0
Total number of sources	45414	208310	187744
Trichloroethylene			
Emission factors			
Number of times used	230	0	0
Percent of total	1.0	0	0
Specified emission profiles			
Number of times used	853	0	0
Percent of total	2.0	0	0
Default profile			
Number of times used	44331	208310	187744
Percent of total	97.0	100.0	100.0
Total number of sources	45419	45419	45419

Table 3. Estimation basis of emission amounts (tons per year).

	Point sources	Area sources	Mobile sources
Acrylonitrile			
Emission factors			
Amount due to emission factors	53	0	0
Percent of total	0.1	0	0
Specified emission profiles			
Amount due to profiles	27603	2737	0
Percent of total	58.0	4.0	0
Default profile			
Amount due to default profile	19583	70160	39315
Percent of total	41.9	96.0	100.0
Total amount	47240	72897	39315
Benzene			
Emission factors			
Amount due to emission factors	2313	0	0
Percent of total	1.0	0	0
Specified emission profiles			
Amount due to profiles	150488	84198	272965
Percent of total	70.0	24.0	100.0
Default profile			
Amount due to default profile	61602	264303	0
Percent of total	29.0	76.0	0
Total amount	214403	348501	272965
Perchloroethylene			
Emission factors			
Amount due to emission factors	3776	0	0
Percent of total	9.0	0	0
Specified emission profiles			
Amount due to profiles	9830	1940	0
Percent of total	23.0	3.0	0
Default profile			
Amount due to default profile	29207	73921	47450
Percent of total	97.8	99.0	100.0
Total amount	42813	75861	47450
Trichloroethylene			
Emission factors			
Amount due to emission factors	246023	0	0
Percent of total	91.0	0	0
Specified emission profiles			
Amount due to profiles	7287	0	0
Percent of total	3.0	0	0
Default profile			
Amount due to default profile	17857	53749	29148
Percent of total	6.0	100.0	100.0
Total amount	271167	53749	29148

Table 4. Annual estimated point source emissions of selected toxic volatile organic compounds (tons per year) based on the 1990 interim emissions inventory.

State	Acrylo-nitrile	Benzene	Perchloro-ethylene	Trichloro-ethylene
AL	1285	6077	1489	962
AZ	13	274	16	10
AR	238	2247	269	182
CA	720	3168	1673	17687
CO	35	167	42	1607
CT	85	399	57	27
DE	78	461	93	57
DC	4	21	5	3
FL	130	1189	156	1770
GA	292	2069	347	213
ID	4	97	5	3
IL	2081	15092	3429	59286
IN	940	5231	2515	8860
IA	99	336	79	551
KS	177	1029	211	289
KY	599	4071	732	384
LA	999	7985	998	36971
ME	32	317	39	24
MD	175	670	167	109
MA	347	1478	438	51225
MI	606	3330	1050	1432
MN	335	1943	405	637
MS	342	1737	631	873
MO	867	3977	1300	3952
MT	35	735	42	26
NE	27	110	42	47
NV	4	14	4	3
NH	28	157	41	11558
NJ	672	5718	644	416
NM	47	361	57	35
NY	2355	6497	1105	714
NC	1299	8029	1367	2089
ND	11	63	11	7
OH	1511	2982	1250	995
OK	144	560	632	3609
OR	257	1394	312	198
PA	1236	4815	882	1033
RI	69	465	101	42
SC	177	949	214	398
SD	45	227	55	34
TN	1273	9110	1428	3203
TX	23060	87164	14690	6672
UT	63	245	65	40
VT	6	37	7	33
VA	2095	4614	1092	940
WA	314	1836	339	228
WV	1220	10496	748	34906
WI	711	3430	1421	16756
WY	98	1030	118	72
TOTAL	47240	214403	42812	271167

Table 5. Annual estimated area source emissions of selected volatile organic compounds (tons per year) based on the 1990 interim emission inventory.

State	Acrylo-nitrile	Benzene	Perchloro-ethylene	Trichloro-ethylene
AL	1394	6398	1322	1030
AZ	858	4017	974	636
AR	732	3193	856	541
CA	6402	32305	7146	4706
CO	793	3745	948	587
CT	694	3235	809	514
DE	489	2420	586	360
DC	82	396	98	60
FL	2731	12807	3282	2021
GA	2189	10171	2100	1616
ID	828	3862	999	614
IL	2532	13009	2930	1855
IN	1761	8310	1951	1298
IA	841	3723	1010	622
KS	785	3783	930	578
KY	942	4604	1107	694
LA	2424	13238	2125	1770
ME	289	1300	346	213
MD	837	3925	992	618
MA	1271	6062	1516	939
MI	2279	10736	2586	1685
MN	1277	6012	1529	942
MS	1031	5278	1200	756
MO	1323	5857	1576	977
MT	604	2974	726	446
NE	485	2116	581	358
NV	280	1296	291	208
NH	234	1082	282	174
NJ	1943	9144	1960	1431
NM	403	1911	483	297
NY	3040	14179	3591	2246
NC	2261	10177	2650	1669
ND	318	1453	374	234
OH	2715	13343	2952	2000
OK	966	5109	1069	707
OR	866	4042	1035	640
PA	3024	14633	3130	2223
RI	224	1045	267	165
SC	3264	14744	1725	2419
SD	352	1552	424	261
TN	1487	7051	1760	1097
TX	7846	37744	6310	5780
UT	428	2135	501	315
VT	135	612	162	100
VA	1682	7779	2010	1243
WA	1578	8059	1867	1161
WV	2386	10529	906	1768
WI	1302	5993	1540	962
WY	153	758	182	112
TOTAL	72760	347847	75696	53648

Table 6. Annual estimated mobile source emissions of selected volatile organic compounds (tons per year) based on 1990 interim emissions inventory.

State	Acrylo-nitrile	Benzene	Perchloro-ethylene	Trichloro-ethylene
AL	910	5474	1098	674
AZ	668	3951	806	495
AR	411	2563	495	304
CA	4227	28811	5102	3134
CO	489	3635	590	363
CT	389	2839	469	288
DE	111	813	134	82
DC	58	415	70	43
FL	2260	13840	2727	1675
GA	1332	8726	1608	988
ID	185	1326	223	137
IL	1522	11342	1837	1129
IN	996	7357	1202	738
IA	430	3057	519	319
KS	418	2999	504	310
KY	602	4223	727	447
LA	798	4968	963	592
ME	216	1678	260	160
MD	680	4800	821	504
MA	756	5697	913	561
MI	1608	12329	1941	1192
MN	723	5586	873	536
MS	490	2933	591	363
MO	924	6588	1115	685
MT	157	1202	190	117
NE	252	1835	305	187
NV	201	1492	242	149
NH	173	1313	208	128
NJ	936	6610	1130	694
NM	363	2707	438	269
NY	1910	14440	2306	1416
NC	1227	7840	1481	910
ND	128	962	155	95
OH	1667	12027	2012	1236
OK	613	4149	740	455
OR	459	3275	554	341
PA	1611	11738	1944	1194
RI	125	941	151	92
SC	662	3975	800	491
SD	140	1036	169	104
TN	913	5832	1102	677
TX	3182	19878	3840	2359
UT	303	2222	366	225
VT	105	819	127	78
VA	949	6663	1145	704
WA	834	6070	1007	619
WV	267	1841	322	198
WI	825	6313	995	611
WY	109	836	131	81
TOTAL	39314	271966	47448	29149

Table 7. Source category codes* associated with the greater portion of emissions from selected toxic volatile organic compounds.

Percent of Estimated Annual Total Emissions in the United States

ACRYLONITRILE		BENZENE		PERCHLOROETHYLENE		TRICHLOROETHYLEN	
SCC	%	SCC	%	SCC	%	SCC	%
30	11	30	14	30	13	40100306	23
109	7	28	6	95	8	40200701	21
95	7	95	6	28	6	30101801	17
30125405	6	109	6	39	5	40100205	8
28	5	34	5	40100203	5	30	4
39	4	39	4	34	4	109	2
34	4	30199999	4	54	3	95	2
30190099	3	104	3	93	3	28	2
30101899	3	32	3	78	3	39	1
54	3	54	2	103	2	34	1
Subtotal	53		53		52		81
Others	47		47		48		19
Total	100		100		100		100
Percent of total emissions by general source group							
Point	30		26		26		77
Area	46		42		46		15
Mobile	24		32		28		8

* Source Category Code (SCC Code) key. Two and three-digit codes are area or mobile sources, seven-digit codes are point sources.

28 - Lgt. duty gas vehicles, rural roads
30 - Lgt. duty gas vehicles, urban roads
32 - Lgt. duty gas trucks, rural roads
34 - Lgt. duty gas trucks, urban roads
39 - Off-highway gasoline vehicles
54 - Gasoline marketing
78 - Degreasing
93 - Misc. industrial manufacturing
95 - Misc. nonindustrial solvent use
103 - Bulk terminals and plants
104 - Fugitive emiss., petrol.refineries
109 - Hazard. waste treatment and storage
30100801 - Chloro-alkali production
30101801 - Polyvinyl chloride production
30101899 - Misc. general plastics production
30125405 - Acrylonitrile production
30190099 - Waste gas flares
30199999 - Misc. chemical manufacturing
40100203 - Perchloroethylene degreasing
40100205 - Trichloroethylene degreasing
40100306 - Trichloroethylene cleaning
40200701 - General adhesive application

KEY WORDS

emission inventory
emission factors
acrylonitrile
benzene
perchloroethylene
trichloroethylene