Preliminary Results of the 2005 NATA Model-to-Monitor Comparison

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ABSTRACT

Acute and chronic exposure to specific hazardous air pollutants (HAPs) can lead to cancer and/or noncancer effects. Since the passage of the 1990 Clean Air Act Amendments (CAAA), the U.S. Environmental Protection Agency (EPA), state, local, and tribal agencies have spent considerable time and resources establishing regulations primarily through maximum achievable control technology (MACT) and mobile source standards, to reduce emissions for hazardous air pollutants (HAPs). Identification of the most important individual emission sources and source categories significantly contributing to potential health risks is challenging for many air quality managers. Large reductions in HAP emissions may not necessarily translate into significant reductions in health risk because toxicity varies by pollutant. For example, acetaldehyde mass emissions are more than double acrolein emissions on a national basis, according to EPA's 2005 National Emissions Inventory (NEI). However, according to the Integrated Risk Information System (IRIS), acrolein is 450 times more toxic in terms of respiratory noncancer risk than acetaldehyde. Thus, it is important to account for the toxicity as well as the mass of the targeted emissions when designing reduction strategies to maximize health benefits. One important tool for air quality managers is EPA's National-Scale Air Toxics Assessment (NATA), which uses HAP emissions from the NEI, meteorological data, background concentrations, population densities, and pollutants specific health data to calculate census-tract level toxicity risk by HAP and source category.

This paper analyzes the model performance of the 2005 NATA through a model-to-monitor comparison, in which receptor-level concentrations calculated from the NATA model are compared to 2005 annual average concentrations of individual HAPs for several hundred air toxics monitoring sites across the country. The preliminary results are summarized by HAP, and are important in understanding the strengths and limitations of air toxics modeling.

INTRODUCTION

Acute and chronic exposure to specific hazardous air pollutants (HAPs) can lead to cancer and/or noncancer effects. Since the passage of the 1990 Clean Air Act Amendments (CAAA),[1] EPA has spent considerable time and resources establishing federal regulations, primarily through maximum achievable control technology (MACT) standards and Risk and Technology Review (RTR) activities, to reduce emissions for HAPs. Atmospheric models, such as those executed for the National-Scale Air Toxics Assessment (NATA), are often used to characterize the nation's air toxics problem both in absolute as well as relative senses by geographic area and pollutant.

The most robust method for assessing what people may be breathing is through ambient air monitoring of HAPs. Ambient monitoring data can help identify pollutants and specific emission sources impacting an area's air quality, and track changes or identify trends in ambient concentrations.

Since 1990, the number of nationwide HAP monitors across the U.S. has increased dramatically (>50%). As a consequence, an increase representativeness of people's inhalation exposure to HAPs has increased. However, the majority of HAP monitors are generally clustered in urban areas. For example, 2005 ambient air monitoring for benzene, a national priority pollutant, occurred mainly in urban areas (Figure 1), accounting for less than 7% of all the counties in the country. This clustering of HAP monitors highlights the geographic disparity in truly assessing nationwide exposure.

Due to the resources required for ambient monitoring of HAPs, it is not feasible to place monitors all over the country, let alone in each county. However, EPA develops point source and county-level source emission inventories for all geographic areas in the country. Thus, emissions modeling can be performed to generate model ambient concentrations. NATA modeling is the "bridge" to assess national-level exposure trends at all geographic locations across the country.

This paper demonstrates the approach for the model-to-monitor comparison, as well as preliminarily evaluating the strengths/limitations of modeling specific HAPs. The following three questions were used to guide the study:

1. Which pollutants are in good agreement between the ambient concentrations and the NATA model?
2. Which pollutants are under-predicted between the ambient concentrations and the NATA model?
3. Which pollutants are over-predicted between the ambient concentrations and the NATA model?

DATA SOURCES

National-Scale Air Toxics Assessment (NATA)

NATA is EPA's ongoing comprehensive evaluation of air toxics in the U.S. EPA developed the NATA as a state-of-the-science screening tool for State/Local/Tribal Agencies to prioritize pollutants, emission sources and locations of interest for further study in order to gain a better understanding of risks.[2] NATA assessments do not incorporate refined information about emission sources, but rather, use general information about sources to develop estimates of risks which are more likely to overestimate impacts than underestimate them. NATA provides estimates of the risk of cancer and other serious health effects from breathing (inhaling) air toxics in order to inform both national and more localized efforts to identify and prioritize air toxics, emission source types and locations which are of greatest potential concern in terms of contributing to population risk. This in turn helps air pollution experts focus limited analytical resources on areas and/or populations where the potential for health risks are highest. Assessments include estimates of cancer and non-cancer health effects based on chronic exposure from outdoor sources, including assessments of non-cancer health effects for Diesel Particulate Matter (PM). Assessments provide a snapshot of the outdoor air quality and the risks to human health that would result if air toxic emissions levels remained unchanged.

EPA made several methodological changes to its most recent assessment, the 2005 NATA. Although EPA is continually refining and updating the assessment methods, it is important to remember that NATA is a screening-level assessment. The intent is to identify HAPs resulting in high exposures or census tracts where population exposures may be of concern. These areas would then require more refined assessments, e.g., monitoring or site-specific risk assessments, to develop a more thorough understanding of these "hot-spot" exposures.

NATA 2005 Improvements

The following improvements have been made in the 2005 NATA:[3]

1. Point Sources
    a. The point source NATA inventory was based on 2005 emissions.
    b. Risk and Technology Review emissions inventory updates were included.
    c. Certain nonpoint categories are now modeled as point sources (i.e., forest and wildfires, chromium electroplating).
    d. Data for 19,000 airports were included.
2. Nonpoint sources
    a. The 2005 NEI was generally unchanged from 2002 (few minor edits).
    b. Emissions from forest fires, wildfires, and chromium electroplating were removed.
    c. Formaldehyde and benzene from pesticides were removed from the 2005 NEI inventory.
    d. Chromium Electroplating sources were moved to the point source inventory.
    e. Several minor adjustments to improve accuracy were made at the state/county level.
3. Mobile Sources
    a. The onroad and nonroad inventories were updated for 2005.
    b. The new MOVES (Motor Vehicle Emission Simulator) emissions model was used for some HAPs.
4. Modeling
    a. The secondary formation of formaldehyde, acetaldehyde, and acrolein were predicted using Community Multi-Scale Air Quality (CMAQ) model.
    b. The transformation of 1,3 butadiene to acrolein was accounted for using CMAQ.
    c. The mobile source modeling approach using AERMOD was improved.
    d. Emissions buoyancy for certain sources at coke oven facilities was accounted for.
5. Risk Characterization
    a. Dose-response values were updated with latest science (IRIS, CalEPA, ATSDR).
    b. The formaldehyde unit risk estimate was revised.

In addition to the census-tract level ambient concentrations predicted by the NATA 2005, EPA used the model to develop specific receptor-level HAP concentrations for over 1,000 locations which coincided with air toxics monitoring sites. These concentrations were the basis for the model-to-monitor comparison.

National Emissions Inventory (NEI)

EPA compiles the NEI, consisting of stationary (point and nonpoint area), mobile (onroad and nonroad), and biogenic source emissions for the entire United States.[4] These emission inventories are typically compiled for three years. Primary data sources for the point source NEI include:

1. State, local, and tribal agency emission inventories;
2. EPA's SPPD and Risk and Technology Review (RTR) Programs;
3. Department of Energy's (DOE) Energy Information Agency (EIA) and EPA's Clean Air Markets Division (CAMD) Emission Tracking System/Continuous Emissions Monitoring (ETS/CEM) data for electric generating utilities (EGUs);
4. EPA's Toxic Release Inventory (TRI); and
5. Data from other studies (e.g., trade associations, Bureau of Ocean Energy Management, Regulation, and Enforcement (BOEMRE) oil and natural gas platform data).

Pollutants in the NEI consist of HAPs and criteria air pollutants (CAPs) and their precursors (CO, NH_3, NO_x, PM, SO_2, and VOCs). Base year inventories are typically compiled every three years; the first NATA was for the 1996 NEI, with subsequent assessments in 1999 and 2002. EPA recently finished preparing the 2005 HAP emissions for the 2005 NATA.[5]

Ambient Monitoring Data

Air toxics ambient monitoring data were extracted from EPA's Phase VI Air Toxics Archive.[6] This archive contains over 26 million HAP concentration records, spanning from 1973 to 2007. For 2005, there were over 2.9 million HAP ambient records at varying measurement levels (1-hour, 3-hour, 4-hour, and 24-hour measurements) at over 800 monitoring sites. Nearly 92% of the HAP records for 2005 sampling dates were originally retrieved from EPA's Air Quality Subsystem (AQS) and less than 7% were extracted from the Interagency Monitoring of Protected Visual Environments (IMPROVE). The remaining data records (less than 2%) were taken from EPA's Phase V historical archive.[7]

METHODOLOGY

Calculating Annual Averages

To properly compare ambient monitoring data to the modeled receptor concentrations, annual average concentrations which represent the year 2005 must be calculated. Because the emissions that were modeled were annual estimates, the model-to-monitor comparison must also reflect concentrations for an entire year. Thus, from a temporal standpoint, it is not suitable to compare modeled concentrations (which are modeled from annual emissions) to ambient data that do not represent an entire year.

Annual averages are calculated using the following procedure:

1. Extract 2005 ambient HAP data from the Phase VI archive.
2. For sub-daily measurements, calculate valid daily concentrations.
3. Identify daily concentrations by site and pollutant which represent an entire year.
4. For non-detects, a zero was used as a surrogate.
5. Calculate annual average by HAP by site from the daily averages (including zeroes for non-detects).

Calculating Daily Averages

Over 2.9 million concentration records were extracted for the 2005 year from the Phase VI archive. Because these records were at differing temporal measurements, all records were converted to daily records. Sub-daily measurements (over 55% of the 2005 records) must have at least 75% temporal coverage within a day. Thus, to be considered a valid daily average:

1. At least eighteen of twenty-four 1-hour measurements must have a detected concentration.
2. At least six of eight 3-hour measurements must have a detected concentration.
3. At least five of six 4-hour measurements must have a detected concentration.

Calculating Annual Averages

Valid daily averages were then reviewed by site and HAP to assess whether there was adequate temporal coverage. To assess temporal coverage, we used a two-step procedure.

1. Ensured that within each calendar quarter (January 1-March 31, April 1-June 30, July 1-September 30, and October 1-December 31), six of eight prescribed sub-quarter zones by site and HAP must have a valid daily concentration.
    a. This approach allowed monitoring data which sampled once every twelve days (which, in theory, yields a minimum of seven sampling days in a calendar quarter).
    b. Sites which sampled more frequently than once every twelve days, such as 1-in-6 days or 1-in-3 days, will have more opportunity to meet this "sub-quarter zone" temporal coverage.
2. Ensured that a valid annual average by site and HAP consisted of three valid calendar quarters.

An annual average is simply the average of detected concentrations and non-detects which satisfied the above procedure. For non-detects, a zero value was used as a surrogate prior to calculating valid annual averages. This is another deviation from the 2002 NATA model-to-monitor comparison which used one-half the pollutant method detection limit as a surrogate for non-detects in calculating the annual average.[8]

PRELIMINARY RESULTS

Table 1 presents the number of monitoring sites used in the 2005 comparison, the 25th, 50th, and 75th ratios of the model-to-monitor average concentrations, and the model-to-monitor average concentration by HAP. The number of sites is the number of monitors with valid annual averages for 2005. A median ratio close to 1 implies the model overestimates the ambient concentrations about as often as it underestimates them. Not surprisingly, HAPs with a large number of monitors tended to have median model-to-monitor ratios closer to 1.

Only HAPs with a minimum of 25 monitors were included in this table. This is another deviation from the 2002 model-to-monitor assessment which required a minimum of 50 monitors.[8] In the 2002 assessment, model-to-monitor ratios for the metals were presented for the $PM_{2.5}$ and TSP size fractions. For the 2005 assessment, it was decided that modeled metal concentrations would be better compared to the PM_{10} size fraction. This is primarily because the vast majority of the $PM_{2.5}$ ratios used in the 2002 assessment were from monitors in the IMPROVE network, which were typically situated in Federal Class 1 and Class 2 areas (e.g., national parks, protected areas) where typical emission sources are not found, and where people do not live. In 2005, PM_{10} metals measurements were only being taken at around 70 monitoring sites, as opposed to over 400 $PM_{2.5}$ metals monitoring sites; thus, the minimum number of monitoring sites required needed to be reduced in order to characterize modeled metal concentrations. Finally, the number of monitoring sites measuring for PM_{10} metals has been increasing since 2005, especially in urban areas, and will continue to serve as a better comparison for modeled metal concentrations in future assessments.

Carbon tetrachloride, methyl chloride, and arsenic (PM_{10}) all had median ratios between 0.9 to 1.1. The percent of sites estimated "within a factor of 2" is the percent of sites for which the model estimate is somewhere between half and double the monitor average. HAPs in which 80% of their monitors were within a factor of 2 were benzene (82%), carbon tetrachloride (95%), methyl chloride (92%), and acetaldehyde (87%).

The "percent of sites estimated within 30%" is the percent of sites for which the model-to-monitor media ratio is between 0.7 and 1.3. Finally, the "percent of sites underestimated" is the percent of sites for which the model-to-monitor ratio is below 1.

Figures 1 through 3 present the distribution statistics (25th, 50th, 75th, and average model-to-monitor ratios) for each HAP. Figure 1 presents gaseous HAPs with greater than 100 monitors, while Figure 2 presents gaseous HAPs with less than 100 monitors. Finally, Figure 3 presents metal HAPs. For example, if there are 295 monitors with valid annual average concentrations of benzene, there are 295 model-to-monitor ratios to compute. EPA then computed the median of these 295 ratios as well as the percentiles to create the plot. The bottom of the statistic is the 25th percentile, the top of the statistic is the 75th percentile, the horizontal line in the middle is the median (i.e., 50th percentile), and the "x" is the average model-to-monitor ratio. The yellow area represents the "factor of two" range. If the model consistently agrees with the monitored data for the pollutant, the 25th and 50th percentile lines will be narrow and centered at 1. Pollutants are organized alphabetically; this side-by-side display of pollutants facilitates comparison to indicate which pollutants are being overestimated and underestimated, and which are estimated consistently. As in the 1996, 1999, and 2002 comparisons, the box plots do not show extreme percentiles (e.g., 10th and 90th) of the ratios because the extreme percentiles were far from the center of the distribution.

Approximately 9% of all model-to-monitor ratios were within 10 % (i.e., ratios between 0.9 and 1.1), 17% were within 20% (ratios between 0.8 and 1.2), and 25% were within 30% (ratios between 0.7 and 1.3). "Factor of 2" ratios (ratios between 0.5 and 2.0) accounted for 44% of the model-to-monitor ratios.

These results show that the interquartile range of model-to-monitor comparisons was within a factor of two for acetaldehyde, arsenic (PM_{10}), benzene, carbon tetrachloride, formaldehyde, methyl chloride, and toluene. The remaining pollutants show various degrees of agreement. These results are similar to those found in the 2002 national-scale assessment comparisons. However, the model is still underestimating several pollutants (i.e., 75th percentile ratio is below 0.5), most noticeably, ethylene dichloride, *n*-hexane, 1,1,2-trichloroethane, acrylonitrile, carbon disulfide, chloroprene, 1,3-dichloropropene, ethylene dibromide, ethylidene dichloride, methyl isobutyl ketone, propionaldehyde, propylene dichloride, and vinylidene chloride.

As a whole, the PM_{10} metals appear to have good agreement with the NATA model, with the exception of beryllium (PM_{10}). Beryllium (PM_{10}) was the only HAP to have its 25th percentile ratio above 2.

Data Considerations - Uncertainties

Earlier in this analysis, we identified several data and model improvements for the 2005 NATA. These improvements are reflected in the increase of more pollutants approaching the median model-to-monitor ratio closer to 1. However, there are areas in which the confidence of the model performance can be improved. This confidence, or degree of agreement between model-to-monitor data, can be attributed to the following five uncertainties (which are the same identified in the 1996, 1999, and 2002 model-to-monitor comparison):

1. Emission characterization uncertainties (e.g., specification of source location, emission rates, and release characterization);
2. Meteorological characterization uncertainties (e.g., representativeness);
3. Model formulation and methodology uncertainties (e.g., characterization of dispersion, plume rise, deposition,);
4. Monitoring uncertainties; and
5. Uncertainties in background concentrations.

Data Considerations – Under-estimation

Approximately 10% of all model-to-monitor ratios were between 0.9 and 1.1, while the number of pollutants showed an increasing trend towards a median model-to-monitor ratio of 1, there were several pollutants identified that were under-estimated. The 1996, 1999, and 2002 model-to-monitor comparisons identified four possible reasons for pollutants to be under-estimated, which may be applicable for the 2005 model-to-monitor assessment. They are:

1. The NEI may be missing specific emissions sources (emissions parameters are missing for many of the sources in the NEI);
2. The emission rates may be underestimated. EPA believes the model itself contributed only in a minor way to the underestimation. In many tests evaluating the model performance, the modeled results compared favorably to monitoring data in cases where the emissions and meteorology were accurately characterized and the monitors made more frequent readings;
3. There is uncertainty in the accuracy of the monitor averages, which, in turn, have their own sources of uncertainty. Sampling and analytical uncertainty, measurement bias, and temporal variation can all cause the ambient concentrations to be inaccurate or imprecise representations of the true atmospheric averages; and
4. Background concentrations (pollutants transported large distances and/or formed by photochemical processes in the atmosphere) are poorly characterized. Most of the pollutants for which the model underestimated ambient concentrations were those for which background concentrations were not estimated. If background concentrations are a large fraction of ambient concentrations, the result would be large underestimations in model predictions.

CONCLUSIONS

EPA recently completed its fourth national-scale assessment for air toxics across the United States. In this paper, we evaluated model performance for several pollutants by comparing modeled concentrations to monitored concentrations. Approximately 9% of all model-to-monitor ratios were within 10 % (i.e., ratios between 0.9 and 1.1), 17% were within 20% (ratios between 0.8 and 1.2), and 25% were within 30% (ratios between 0.7 and 1.3).

The following three questions were used to guide the study:

- ***Which pollutants are in good agreement between the ambient concentrations and the NATA 2005 model?*** Good agreement (i.e., interquartile values within a factor of two) were seen for the following pollutants: acetaldehyde, arsenic (PM_{10}), benzene, carbon tetrachloride, formaldehyde, methyl chloride, and toluene.
- ***Which pollutants are under-predicted between the ambient concentrations and the NATA 2005 model?*** Under-prediction (upper bound of the interquartile range less than a factor of two) was seen for the following pollutants: ethylene dichloride, *n*-hexane, 1,1,2-trichloroethane, acrylonitrile, carbon disulfide, chloroprene, 1,3-dichloropropene, ethylene dibromide, ethylene dichloride, methyl isobutyl ketone, propionaldehyde, propylene dichloride, and vinylidene chloride.
- ***Which pollutants are over-predicted between the ambient concentrations and the NATA 2005 model?*** Over-prediction (lower bound of the interquartile greater than a factor of two) was seen for beryllium (PM_{10}).

HAPs in which 80% of their monitors were within a factor of 2 were benzene (82%), carbon tetrachloride (95%), methyl chloride (92%), and acetaldehyde (87%). “Factor of 2” ratios (ratios between 0.5 and 2.0) accounted for 44% of the model-to-monitor ratios.

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KEYWORDS

Hazardous Air Pollutants (HAPs)
National Emissions Inventory (NEI)
Annual Averaging
Model Evaluation
Model-to-Monitor
National-Scale Air Toxics Assessment (NATA)

Figure 1. Benzene Emission Sources and Monitoring Sites in 2005.

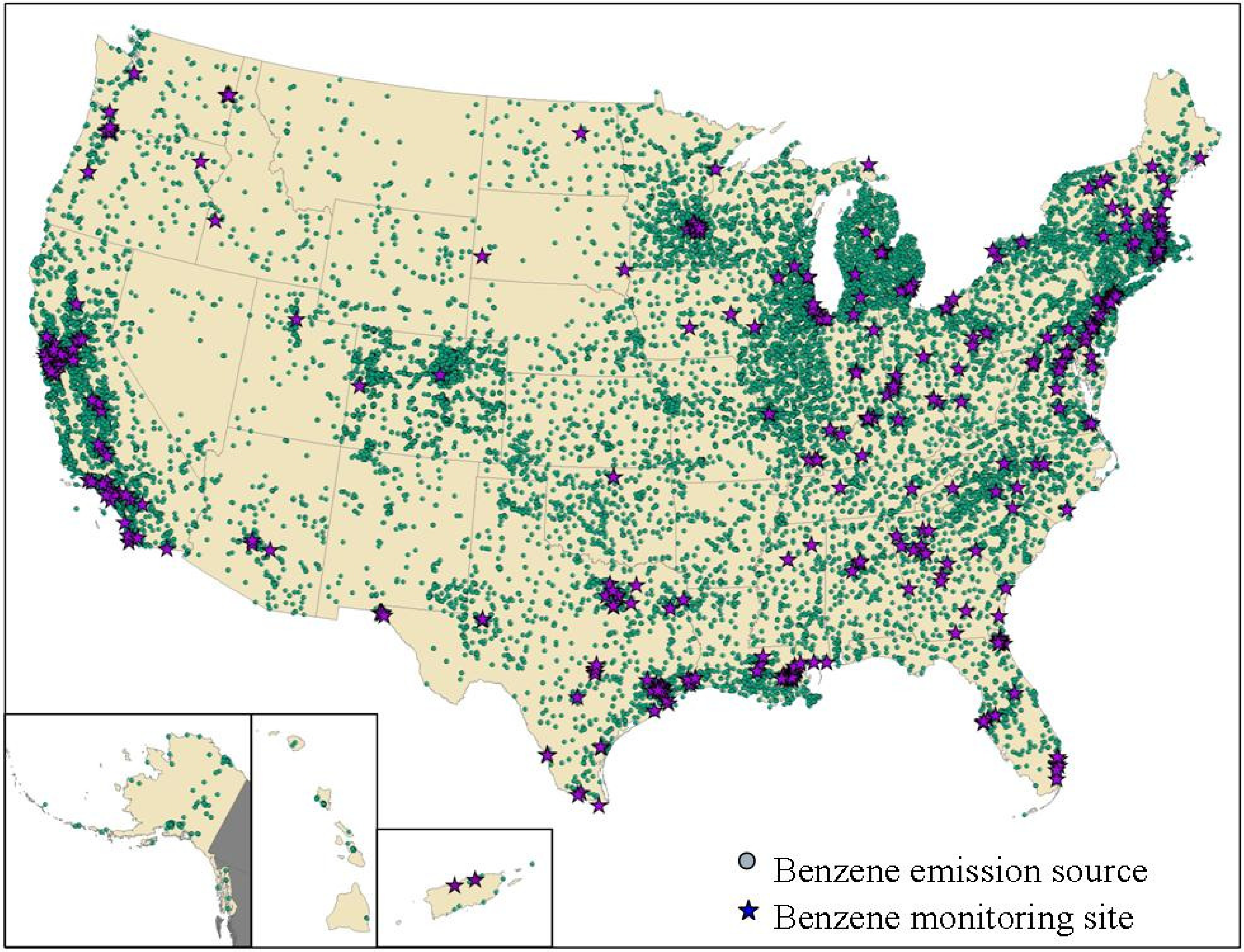

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Table 1. 2005 NATA Model-to-Monitor Comparison.

	Model-to-Monitor Ratios								
Pollutant	# Monitors	25th Percentile	50th Percentile	75th Percentile	Average	Average % Difference	% Monitors Within 30%	% Monitors Within Factor of 2	% Under-estimated
Benzene	295	0.634	0.826	1.141	0.984	-2	49	82	65
Toluene	295	0.579	0.812	1.244	1.071	7	39	76	66
Xylenes	265	0.748	1.284	2.041	3.465	247	29	59	33
Ethylbenzene	242	0.289	0.471	0.771	1.275	27	20	41	85
Carbon Tetrachloride	221	0.939	1.018	1.173	1.135	14	87	95	48
Methyl Chloride	206	0.914	1.030	1.181	1.083	8	85	98	42
Styrene	195	0.178	0.397	0.762	1.402	40	15	32	83
Methylene Chloride	190	0.395	0.524	0.693	0.726	-27	11	48	85
1,3-Butadiene	176	0.425	0.697	0.955	0.962	-4	31	56	76
Tetrachloroethylene	174	0.289	0.449	0.687	0.560	-44	20	42	93
Chloroform	169	0.383	0.554	0.807	0.746	-25	18	55	81
1,1,1-Trichloroethane	163	1.166	1.622	3.993	2.646	165	13	63	18
Formaldehyde	162	0.610	0.783	1.001	0.919	-8	48	77	75
n-Hexane	162	0.160	0.262	0.439	0.398	-60	6	18	93
Acetaldehyde	160	0.878	1.159	1.439	1.235	23	49	87	36
Trichloroethylene	145	0.185	0.411	0.901	0.562	-44	22	41	79
Bromomethane	143	0.316	0.817	1.244	0.979	-2	37	66	62
2,2,4-Trimethylpentane	111	0.215	0.490	1.112	0.942	-6	17	35	73
Ethylene Dichloride	111	0.066	0.092	0.222	0.237	-76	6	15	98
Chlorobenzene	102	0.003	0.005	0.021	0.017	-98	0	0	100
p-Dichlorobenzene	102	0.113	0.226	0.546	0.435	-56	9	25	91
Propionaldehyde	98	0.029	0.060	0.130	0.094	-91	0	2	100
Methyl *tert*-butyl ether	88	0.088	0.567	0.930	0.800	-20	25	43	78
Vinyl chloride	87	0.009	0.026	0.089	0.143	-86	3	7	98
Cumene	78	0.003	0.009	0.195	0.211	-79	3	10	94
Cis-1,3-Dichloropropylene	77	0.003	0.149	1.066	0.850	-15	4	17	74
Trans-1,3-Dichloropropylene	77	0.004	0.156	1.066	0.849	-15	4	17	74
1,1,2,2-Tetrachloroethane	74	0.027	0.080	0.089	0.067	-93	0	0	100
Ethylene Dibromide	73	0.005	0.017	0.028	0.029	-97	0	1	100
1,1,2-Trichloroethane	69	0.0003	0.003	0.004	0.011	-99	0	1	100
Propylene Dichloride	67	0.013	0.025	0.038	0.031	-97	0	0	100
Ethylidene Dichloride	65	0.004	0.005	0.006	0.008	-99	0	0	100

Table 1. 2005 NATA Model-to-Monitor Comparison (Cont.).

Pollutant	Model-to-Monitor Ratios								
	# Monitors	25th Percentile	50th Percentile	75th Percentile	Average	Average % Difference	% Monitors Within 30%	% Monitors Within Factor of 2	% Under-estimated
Vinylidene Chloride	65	0.001	0.004	0.004	0.011	-99	0	0	100
Methyl Isobutyl Ketone	63	0.019	0.105	0.486	5.386	439	2	11	86
Carbon disulfide	53	0.0002	0.002	0.007	0.010	-99	0	0	100
Acrolein	51	0.062	0.103	0.271	0.265	-74	4	10	96
Acrylonitrile	49	0.008	0.015	0.037	0.023	-98	0	0	100
Chloroprene	46	0.0003	0.004	0.005	0.003	-100	0	0	100
Manganese (PM_{10})	38	0.276	0.459	0.700	0.801	-20	13	37	84
Arsenic (PM_{10})	37	0.721	1.037	1.368	1.112	11	43	78	38
Lead (PM_{10})	37	0.448	0.623	0.870	0.704	-30	30	65	81
Nickel (PM_{10})	37	0.307	0.724	1.242	0.982	-2	32	46	57
Chromium (PM_{10})	36	0.265	0.455	0.879	0.984	-2	14	28	81
Cadmium (PM_{10})	32	0.683	1.179	1.859	1.320	32	31	69	41
Beryllium (PM_{10})	26	9.160	19.718	43.517	29.551	2855	8	8	8
Selenium (PM_{10})	26	0.196	0.381	0.597	0.518	-48	8	23	92
Cobalt (PM_{10})	25	0.346	0.490	1.612	1.117	12	16	32	64

Figure 2. Model-to-Monitor Comparisons of Gaseous HAPs (>100 Monitors)

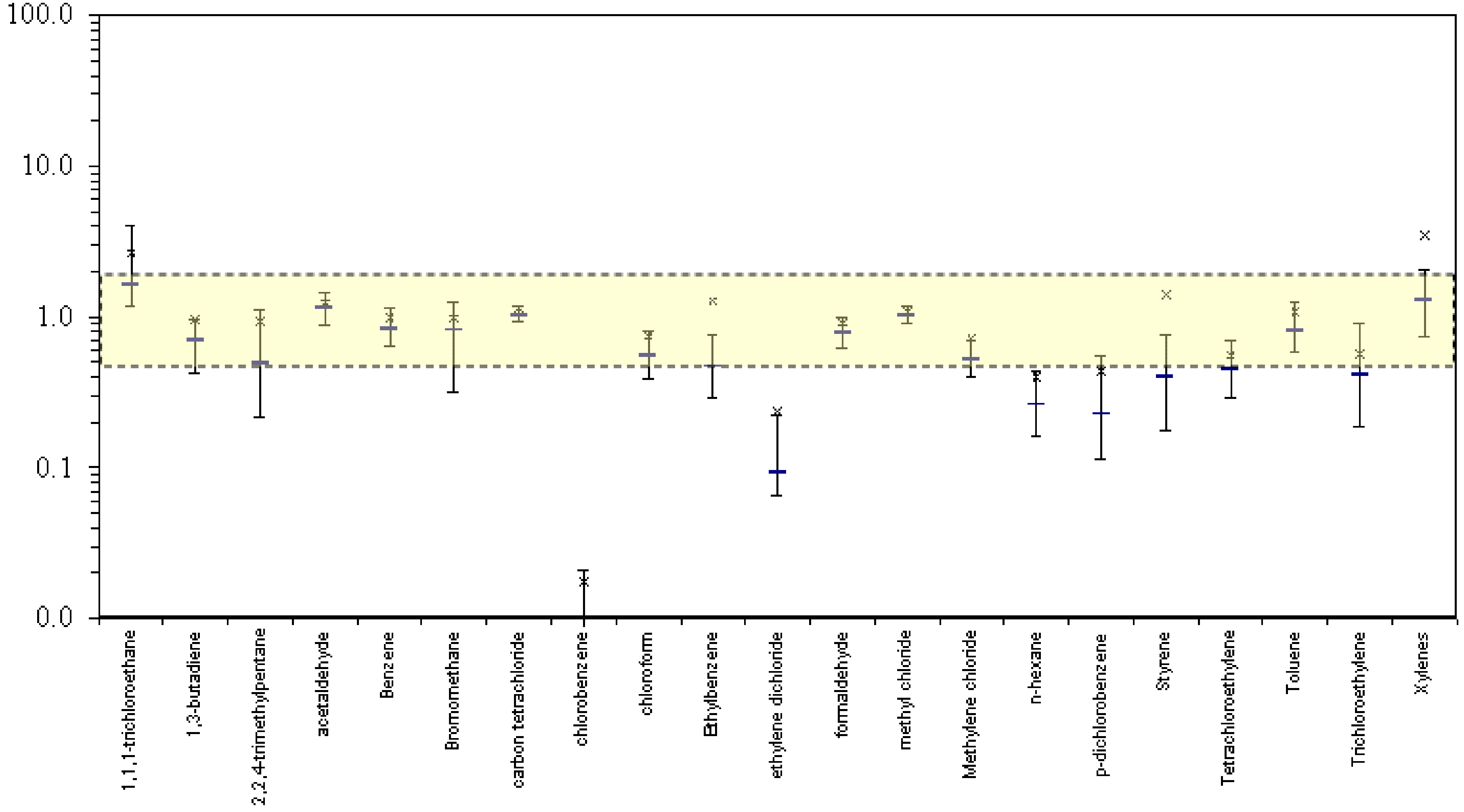

Figure 3. Model-to-Monitor Comparisons of Gaseous HAPs (<100 Monitors)

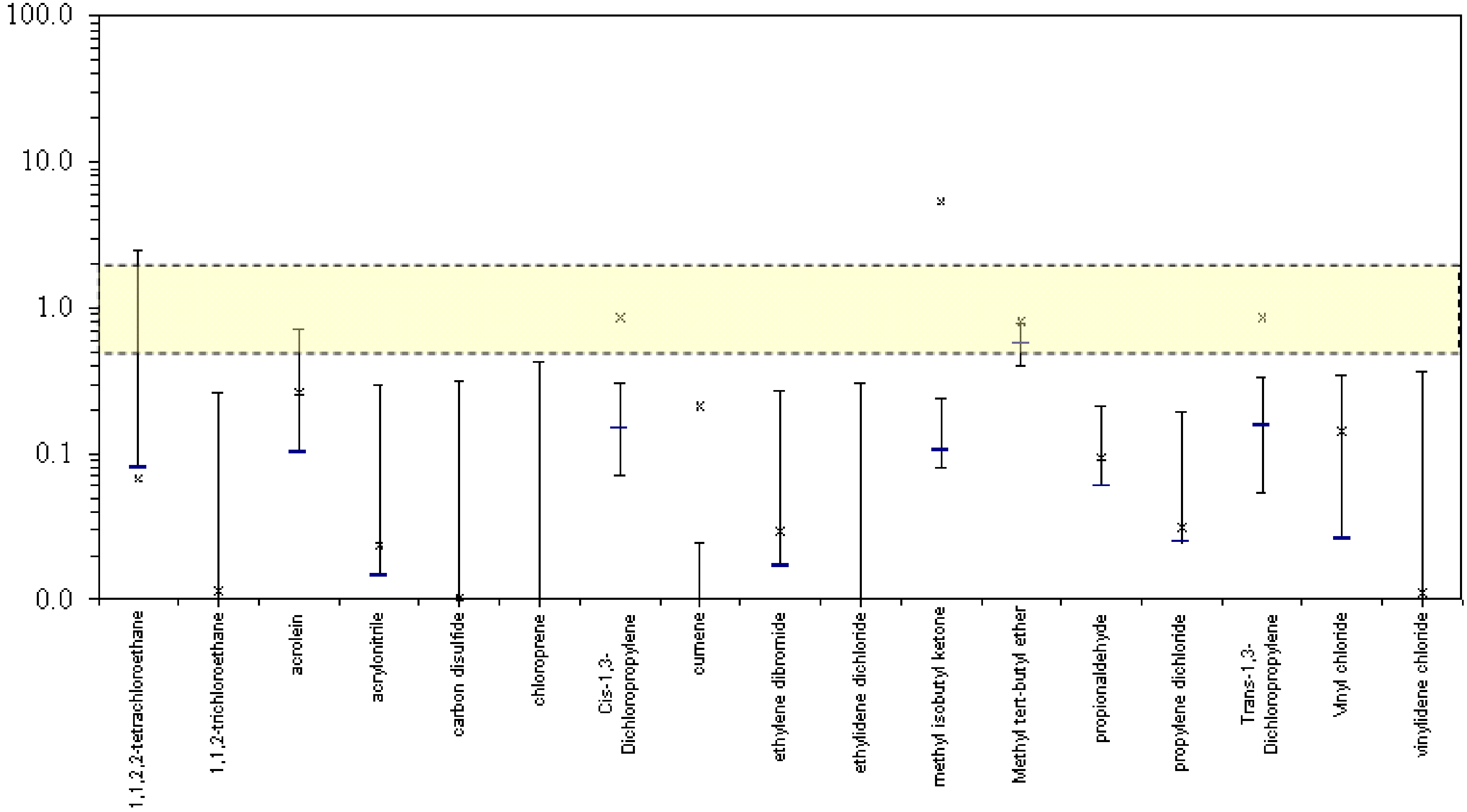

Figure 4. Model-to-Monitor Comparisons of Metal HAPs