Targeted National Sewage Sludge Survey (TNSSS): Summary Statistics and Estimates of 95th Percentiles for 84 Additional Analytes

U.S. Environmental Protection Agency
Office of Water (4301T)
Office of Science and Technology
Health and Ecological Criteria Division
1200 Pennsylvania Avenue, NW
Washington, DC 20460

EPA 822-R-21-003

April 2021

Prepared by:

Battelle
505 King Avenue
Columbus, Ohio 43201

EPA Contract No. EP-W-09-024, Work Assignment 4-04

Table of Contents

List of Tables

List of Figures

Page

1. Introduction

The Targeted National Sewage Sludge Survey (TNSSS), collected and analyzed a total of 145 analytes in treated biosolids taken from a statistically representative subset of the nation's Publicly Owned Treatment Works (POTWs). The TNSSS statistical report (USEPA, 2009a) presented results of in-depth statistical analyses performed on the measurements of 34 prioritized analytes. This report presents the results of data analyses performed on measurements for 84 additional analytes, which were not prioritized in 2009. For each of the 84 analytes, this report assesses the distribution of measurements from the TNSSS and utilizes an appropriate statistical approach to estimate the 95th percentile of the distribution based on these data. EPA's ProUCL software serves as the tool for generating these estimates, while accounting for non-detected outcomes present among the measurements. Detections of 27 analytes were too limited to conduct statistical analyses; 16 analytes had zero detections and 11 analytes had one detection.

Following a brief overview of the TNSSS and the list of analytes measured in the sampled biosolids, Section 2 of this report discusses the statistical approaches considered for estimating the 95th percentiles. Section 3 presents the estimates that were calculated under these approaches. Section 4 presents key findings and conclusions.

1.1 TNSSS Design Overview

The target population for the TNSSS consisted of 3,337 POTWs that met the following criteria:

- Were in full operation in 2002 and/or 2004,
- Had flow rates greater than 1 million gallons per day (MGD),
- Employed a minimum of secondary treatment[1],
- Were located in the contiguous United States, and
- Were neither privately-owned, non-publicly owned, nor Tribal facilities.

EPA used statistical survey sampling techniques to select POTWs from which to collect biosolids samples in the TNSSS. Sample collection occurred from August 2006 to March 2007. To ensure that the sampled facilities covered the entire range of flow rates, the sampling design divided the sample frame into three strata defined by flow rate. Table 1 shows the three strata and the sample sizes resulting from each one. USEPA (2009a) contains more detail on the TNSSS design.

Table 1. Numbers of POTWs within the TNSSS, by Average Flow Rate.

Average Flow Rate	Number of POTWs Sampled in the TNSSS
>100 MGD	8
10 to 100 MGD	12
1 to 10 MGD	54
TOTAL	**74**

[1] At a POTW, all wastewater first must go through the primary treatment process, which involves screening and settling out large particles. The wastewater then moves on to the secondary treatment process, during which organic matter is removed by allowing bacteria to break down the pollutants.

Within each sampled POTW, EPA collected one grab sample of biosolids for analysis, except in the following situations where two grab samples were selected:

- At six facilities, duplicate grab samples were collected
- For four facilities that each utilized two treatment systems, EPA collected one grab sample from each system.

Therefore, EPA collected a total of 84 grab samples of treated biosolids in the TNSSS from the 74 sampled POTWs.

1.2 Compounds Analyzed in the TNSSS

The TNSSS statistical analysis report (USEPA, 2009a) presents nationally representative estimates of means and upper percentiles of the concentrations of 34 analytes measured in the biosolids samples. Table 2 lists these analytes, according to the class of chemicals in which they reside.

Table 2. Listing of 34 Analytes Measured in the TNSSS, Whose Measurements Were Subject to In-Depth Statistical Analysis in USEPA (2009a).

Metals	Barium Beryllium Manganese	Molybdenum Silver
Organics	4-Chloroaniline Fluoranthene	Pyrene
Classicals (Anions)	Nitrate/Nitrite	
PBDEs	BDE-47 (2,2',4,4'- tetrabromodiphenyl) BDE-99 (2,2',4,4',5-pentabromodiphenyl)	BDE-153 (2,2',4,4',5,5'-hexabromodiphenyl) BDE-209 (decabromodiphenyl)
Pharmaceuticals	4-Epitetracycline (ETC) Azithromycin Carbamazepine Cimetidine Ciprofloxacin Diphenhydramine Doxycycline	Erythromycin-Total Fluoxetine Miconazole Ofloxacin Tetracycline (TC) Triclocarban Triclosan
Steroids and Hormones	Beta Stigmastanol Campesterol Cholestanol Cholesterol	Coprostanol Epicoprostanol Stigmasterol

Along with the 34 analytes in Table 2, EPA measured the concentrations of 111 additional analytes in the biosolids samples. These 111 analytes were not subject to the in-depth data analysis featured in that report. Table 3 lists 27 of these non-prioritized analytes which had no more than one detected concentration reported among the 84 collected samples. The lack of a sufficient number of detected, quantifiable analysis outcomes for characterizing uncertainty in the measurements made it inappropriate to apply a rigorous statistical analysis to data for these 27 analytes (which were exclusively pharmaceuticals, steroids, and hormones).

Table 3. Listing of 27 Analytes Measured in the TNSSS, With No More than One Detected Outcome from Among the 84 Collected Samples of Treated Biosolids.

Pharmaceuticals	4-Epianhydrochlortetracycline (EACTC) 4-Epichlortetracycline (ECTC) Albuterol Anhydrochlortetracycline (ACTC) Carbadox Cefotaxime Chlortetracycline (CTC) Clinafloxacin Cloxacillin Digoxigenin Digoxin	Flumequine Isochlortetracycline (ICTC) Norgestimate Ormetoprim Oxacillin Oxolinic Acid Penicillin G Penicillin V Sulfamera-zine Sulfamethi-zole Sulfathiazole Tylosin Warfarin
Steroids and Hormones	17 Alpha-Dihydroequilin 17 Alpha-Ethinyl-Estradiol	Equilenin

Table 4 lists the remaining 84 analytes and the percentage of collected samples of biosolids in the TNSSS for which the analytical method yielded a detected outcome for that analyte. This report uses statistical techniques to estimate the 95th percentile of the concentration of these analytes in treated biosolids, based on data collected in the TNSSS.

Table 4. Listing of 84 Analytes Measured in the TNSSS, and the Percentage of Detected Outcomes from Among the 84 Collected Samples of Treated Biosolids.

Metals	Aluminum	100.0%	Magnesium	100.0%
	Antimony	86.5%	Mercury	100.0%
	Arsenic	100.0%	Nickel	100.0%
	Boron	97.3%	Phosphorus	100.0%
	Cadmium	100.0%	Selenium	100.0%
	Calcium	100.0%	Sodium	100.0%
	Chromium	100.0%	Thallium	94.6%
	Cobalt	100.0%	Tin	94.6%
	Copper	100.0%	Titanium	98.6%
	Iron	100.0%	Vanadium	100.0%
	Lead	100.0%	Yttrium	100.0%
			Zinc	100.0%
Organics	2-Methylnaphthalene	44.6%	Benzo(a)pyrene	77.0%
			Bis(2-ethylhexyl) phthalate	100.0%
Classicals (Anions)	Fluoride	100.0%	Water-Extractable Phosphorus	100.0%
PBDEs	BDE-028	100.0%	BDE-100	100.0%
	BDE-066	100.0%	BDE-138	67.9%
	BDE-085	100.0%	BDE-154	100.0%
			BDE-183	100.0%
Pharmaceuticals	1,7-Dimethylxanthine	5.1%	Metformin	7.8%
	4-EOTC	10.3%	Minocycline	43.3%
	4-Epianhydrotetracycline (EATC)	34.6%	Naproxen	51.3%
			Norfloxacin	33.3%
	Acetaminophen	2.6%	Oxytetracycline (OTC)	35.9%
	Anhydrotetracycline (ATC)	60.3%	Ranitidine	57.1%
	Caffeine	46.2%	Roxithromycin	2.6%
	Clarithromycin	53.8%	Sarafloxacin	2.6%
	Codeine	24.4%	Sulfachloropyridazine	2.6%
	Cotinine	44.9%	Sulfadiazine	3.9%
	Dehydronifedipine	21.8%	Sulfadimethoxine	6.5%
	Demeclocycline	3.8%	Sulfamethazine	2.6%
	Diltiazem	82.1%	Sulfamethoxazole	37.7%
	Enrofloxacin	15.4%	Sulfanilamide	10.4%
	Gemfibrozil	89.7%	Thiabendazole	69.2%
	Ibuprofen	62.8%	Trimethoprim	29.5%
	Lincomycin	3.8%	Virginiamycin	17.9%
	Lomefloxacin	2.6%		
Steroids and Hormones	17 Alpha-estradiol	6.8%	Equilin	17.8%
	17 Beta-estradiol	11.5%	Ergosterol	61.5%
	Androstenedione	41.1%	Estriol	21.6%
	Androsterone	65.8%	Estrone	76.7%
	Beta-Estradiol 3-Benzoate	23.0%	Norethindrone	6.6%
	Beta-Sitosterol	85.9%	Norgestrel	5.4%
	Desmosterol	66.7%	Progesterone	22.1%
			Testosterone	23.3%

2. Approach

This section describes the statistical methodology for estimating the 95th percentile of the concentration of the 84 additional analytes (listed in Table 4) in treated biosolids across the POTWs sampled in the TNSSS.

As noted in Section 2.4.3 of USEPA (2009a), the TNSSS aimed to collect a single sample of final treated biosolids from a facility; the measurements taken from this single sample represented the facility's average concentration for the pollutant at a single point in time. Therefore, in the ten instances when a facility had two biosolids samples collected, either for quality control purposes or because the facility generated two types of biosolids products, EPA investigated whether the two data values for a given analyte could be aggregated (averaged) into a single value prior to performing the data review and analysis. For the statistical analysis of the 34 prioritized analytes (USEPA, 2009a), EPA aggregated data values within a facility in the following instances:

- For all analytes, when the second sample was a field duplicate sample (6 facilities).
- For analytes within the classicals, metals, and organics classifications, when the two samples represented different treatment systems (4 facilities). Aggregation did not occur for other classifications (i.e., PBDEs, pharmaceuticals, steroids, and hormones) within these facilities because measurements often differed considerably between samples collected from different systems, especially between solid and liquid samples.

When data aggregation occurred, the criteria for classifying a facility's aggregated (average) value as a detect or non-detect result matched that used in USEPA (2009a) and is documented in Table 5.

Table 5. Criteria for Classifying Within-Facility Aggregated Measurements as a Detect or Non-Detect in the TNSSS.

If the two sample data values are …	The aggregated value is calculated as the …	The result is classified as a …
Both detected	Arithmetic average of the measured values	Detect
Both non-detected	Arithmetic average of the sample-specific detection limits	Non-detect
A mixture of detected and non-detected samples	Arithmetic average of the measured value (for the detected sample) and sample-specific detection limit (for the non-detected sample)	Detect

Thus, following this within-facility data aggregation, the 95th percentile was estimated using a set of 74 data values for each of the metals, organics, and classicals, and a maximum set of 78 data values for PDBEs, pharmaceuticals, steroids, and hormones. (For selected pharmaceuticals, steroids, and hormones, fewer than 78 data values were available for the calculation, as the laboratory did not report a value for certain samples/facilities.)

When the laboratory reported a non-detect outcome, it reported the sample-specific detection limit rather than a measured value for that sample. For a given analyte, different samples could have different detection limits whose values can overlap the distribution of detected outcomes. Table 6 lists the 84 analytes and some summary statistics on the observed detected measurements and on the reported detection limits (for non-detects).

Table 6. Summary Statistics for Detected Outcomes for 84 Analytes Measured in the TNSSS.

Analyte	CAS Number	Total N	Detected Concentrations				
			N	Minimum	Median	Maximum	Mean
Metals (mg/kg)							
Aluminum	7429905	74	74	1,400.00	11,200.00	57,300.00	13,494.66
Antimony	7440360	74	64	0.45	1.71	20.50	2.53
Arsenic	7440382	74	74	1.18	4.96	49.20	6.94
Boron	7440428	74	72	5.70	33.00	131.00	41.48
Cadmium	7440439	74	74	0.21	1.76	11.80	2.64
Calcium	7440702	74	74	9,480.00	27,000.00	243,000.00	41,025.41
Chromium	7440473	74	74	6.74	32.68	1,160.00	80.16
Cobalt	7440484	74	74	0.87	4.59	290.00	10.73
Copper	7440508	74	74	115.00	456.00	1,720.00	553.13
Iron	7439896	74	74	1,580.00	15,650.00	131,000.00	26,252.50
Lead	7439921	74	74	5.81	46.15	350.00	76.19
Magnesium	7439954	74	74	713.50	4,460.00	18,050.00	4,956.61
Mercury	7439976	74	74	0.19	0.83	7.50	1.23
Nickel	7440020	74	74	7.61	23.45	526.00	48.32
Phosphorus	7723140	74	74	5,715.00	19,300.00	69,400.00	21,806.49
Selenium	7782492	74	74	1.10	6.20	24.20	7.00
Sodium	7440235	74	74	154.00	1,017.75	26,600.00	2,699.97
Thallium	7440280	74	70	0.02	0.13	1.68	0.18
Tin	7440315	74	70	7.50	36.15	522.00	49.08
Titanium	7440326	74	73	18.50	86.90	4,805.00	281.73
Vanadium	7440622	74	74	2.04	12.65	617.00	36.19
Yttrium	7440655	74	74	0.70	3.89	26.30	4.82
Zinc	7440666	74	74	216.00	784.00	8,550.00	970.01
Organics (µg/kg)							
2-Methylnaphthalene	91576	74	33	10.00	250.00	4,600.00	498.12
Benzo(a)pyrene	50328	74	57	63.00	360.00	4,000.00	810.69
Bis(2-ethylhexyl) phthalate	117817	74	74	657.35	24,000.00	310,000.00	52,862.48
Anions (mg/kg)							
Fluoride	16984488	74	74	14.70	54.10	234.00	59.42
Water-Extractable Phosphorus	C055	74	74	11.00	420.75	9,550.00	988.08
BDEs (ng/kg)							
BDE 028	41318756	78	78	2,200.00	8,900.00	160,000.00	15,348.72
BDE 066	189084615	78	78	1,800.00	12,000.00	110,000.00	17,396.79
BDE 085	182346210	78	78	3,200.00	23,000.00	150,000.00	27,943.59
BDE 100	189084648	78	78	13,000.00	120,000.00	1100000.00	150,365.38
BDE 138	182677301	78	53	1,900.00	7,900.00	40,000.00	10,247.17
BDE 154	207122154	78	78	7,700.00	46,500.00	440,000.00	59,900.00
BDE 183	207122165	78	78	2,100.00	10,000.00	120,000.00	16,664.74
Pharmaceuticals (µg/kg)							
1,7-Dimethylxanthine	611596	78	4	1,130.00	2,245.00	9,580.00	3,800.00
4-Epianhydrotetracycline (EATC)	4465650	78	27	126.00	299.00	2,160.00	434.29
4-Epioxytetracycline (EOTC)	14206587	78	8	35.70	45.80	54.90	45.60
Acetaminophen	103902	78	2	1,120.00	1,210.00	1,300.00	1,210.00
Anhydrotetracycline (ATC)	4496859	78	47	94.30	205.00	1,960.00	330.06
Caffeine	58082	78	36	72.90	262.50	1,110.00	369.16
Clarithromycin	81103119	78	42	8.68	34.50	617.00	65.53
Codeine	76573	78	19	10.70	35.80	328.00	61.28
Cotinine	486566	78	35	11.40	21.00	690.00	99.36
Dehydronifedipine	67035227	78	17	3.48	5.96	21.65	7.97
Demeclocycline	127333	78	3	96.00	164.00	200.00	153.33
Diltiazem	42399417	78	64	1.81	18.25	225.00	44.45
Enrofloxacin	93106606	78	12	12.55	32.20	66.00	34.42
Gemfibrozil	25812300	78	70	12.10	115.00	2,650.00	234.12
Ibuprofen	15687271	78	49	99.50	255.00	11,900.00	920.67
Lincomycin	154212	78	3	12.85	29.10	33.40	25.12
Lomefloxacin	98079517	78	2	33.30	36.55	39.80	36.55
Metformin	657249	77	6	550.00	756.00	1,160.00	781.50
Minocycline	10118908	67	29	351.00	475.00	8,650.00	883.40
Naproxen	22204531	78	40	20.90	75.75	1,020.00	137.37
Norfloxacin	70458967	78	26	99.30	203.00	995.50	297.30

Analyte	CAS Number	Total N	Detected Concentrations				
			N	Minimum	Median	Maximum	Mean
Oxytetracycline (OTC)	79572	78	28	21.05	62.50	467.00	83.07
Ranitidine	66357355	77	44	3.85	18.15	2,250.00	81.98
Roxithromycin	80214831	78	2	14.30	18.33	22.35	18.33
Sarafloxacin	98105998	78	2	179.00	1,079.50	1,980.00	1,079.50
Sulfachloropyridazine	80320	77	2	26.00	42.35	58.70	42.35
Sulfadiazine	68359	77	3	22.90	77.30	140.00	80.07
Sulfadimethoxine	122112	77	5	3.58	7.35	62.20	18.30
Sulfamethazine	57681	77	2	21.50	22.35	23.20	22.35
Sulfamethoxazole	723466	77	29	3.91	12.30	651.00	43.26
Sulfanilamide	63741	77	8	191.00	1,593.50	15,600.00	3,651.50
Thiabendazole	148798	78	54	8.42	22.05	238.00	46.32
Trimethoprim	738705	78	23	12.65	38.90	204.00	51.37
Virginiamycin	11006761	78	14	43.50	125.25	469.00	162.56
Steroids/Hormones (µg/kg)							
17 Alpha-Estradiol	57910	73	5	14.45	21.90	48.80	26.13
17 Beta-Estradiol	50282	78	9	22.00	33.20	222.25	60.89
Androstenedione	63058	73	30	108.00	387.50	1,520.00	495.15
Androsterone	53418	73	48	17.65	107.50	1,030.00	157.26
Beta-Estradiol 3-Benzoate	50500	74	17	30.20	145.00	1,850.00	449.16
Beta-Sitosterol	83465	78	67	24,400.00	260,000.00	1640000.00	333,643.28
Desmosterol	313042	78	52	2,730.00	14,700.00	94,400.00	19,037.69
Equilin	474862	73	13	22.30	36.75	100.30	48.31
Ergosterol	57874	78	48	4,530.00	21,700.00	91,900.00	27,988.33
Estriol	50271	74	16	7.56	77.85	232.00	79.24
Estrone	53167	73	56	26.70	74.90	965.00	133.78
Norethindrone	68224	76	5	21.00	41.10	1,360.00	305.82
Norgestrel	6533002	74	4	43.80	113.75	1,300.00	392.83
Progesterone	57830	77	17	143.00	757.00	1,290.00	753.50
Testosterone	58220	73	17	30.80	97.90	2,040.00	291.79

Section 2.1 introduces the ProUCL software used to calculate the 95th percentile estimates for the 84 analytes in Table 6. Sections 2.2 and 2.3 present goodness-of-fit distributional tests and statistical outlier tests, respectively, which were used in preparing the datasets for analysis and determining an appropriate statistical approach for estimating the 95th percentile. Section 2.4 presents brief overviews of these statistical approaches; the results of applying these approaches to data for the 84 analytes follow in Section 3.

2.1 ProUCL Software

The analysis in this report utilized Version 4.1.00 of EPA's ProUCL software, an open-source statistical estimation software tool available for download at https://www.epa.gov/land-research/proucl-software. ProUCL offers a variety of parametric and nonparametric statistical approaches for calculating estimates of the upper percentiles of a statistical distribution. These approaches differ in the assumed underlying distribution of the data and in how non-detects are treated. Most approaches regard non-detects as left-censored at the reported sample-specific detection limit (i.e., the reported result is known only to fall below the limit), and some can handle multiple values for detection limits among the non-detects. Because the 95th percentile was of interest to estimate here, because ProUCL offers approaches that are more rigorous than simple substitution methods for handling non-detects and which have demonstrated good performance in peer reviewed publications, and because the reported data in the TNSSS contain non-detects at multiple detection limits (when non-detects were present) for a given analyte, EPA considered ProUCL to be an appropriate tool for estimating 95th percentiles for the 84 analytes in this report.

ProUCL was designed to analyze environmental concentration data associated with a localized site characterization. Thus, it was not designed to analyze data from complex sampling designs, such as stratification or the use of sampling weights. The in-depth statistical analysis performed in USEPA (2009a) did account for the sampling weights, and thus, generated nationally representative estimates.

2.2 Methods for Testing Distributional Goodness-of-Fit

ProUCL considers three different data distributions as a basic assumption in its parametric estimation methods: normal, lognormal, and gamma distributions. Thus, ProUCL offers goodness-of-fit tests for each of these three distribution models. ProUCL recommends that the results of these tests be reviewed with histograms or quantile-quantile (Q-Q) plots of the data in order to get a more complete assessment of distributional goodness-of-fit. These data plots also provide useful information about the presence of potential outliers and influential data values. This, histograms of detected measurements for the individual analytes can be found at the end of Section 3.

Because of the unknown quantitative value of non-detects, the goodness-of-fit tests were applied only to the set of detected measurements for each analyte. That is, any non-detects were excluded from the test.

ProUCL uses the Shapiro-Wilk test for normality (Gilbert 1987), as well as Lilliefors test (Dudewicz and Misra, 1988; Conover, 1999) when the sample size exceeds 50. (ProUCL indicates that Lilliefors test performs well for samples of this size, while recognizing that the Shapiro-Wilk test can be applied to samples with larger sample sizes.) Lognormality tests are equivalent to normality tests performed on log-transformed data.

To test for goodness-of-fit to a gamma distribution, ProUCL uses two empirical distribution function (EDF)-based methods: the Kolmogorov-Smirnov (K-S) test and the Anderson-Darling (A-D) test (D'Agostino and Stephens, 1986; Stephens, 1970). The critical values for these two test statistics originate from Monte Carlo simulation experiments.

Conclusions derived solely from goodness-of-fit tests need to be made with caution. Because the null hypothesis of these tests is that the given distributional model holds (e.g., normality) and the alternative is that it does not hold, then the outcome of these tests is either the distributional model can or cannot be rejected based on the data. Thus, if one fails to reject the given distribution, this does not mean that the distribution is the best fit to the data, only that it cannot be outright rejected. Furthermore, the outcome of the test is highly influenced by the sample size -- fewer data points make it more difficult to reject the distributional model, thereby making it more likely to conclude that the distribution is reasonable when in fact it is not, while many data points can result in rejecting the distribution with high likelihood, even when the distribution is appropriate. The test outcomes can also be influenced by extreme data values. Thus, these goodness-of-fit tests provide only a general indication of the relevance of a given distributional assumption.

2.3 Methods for Identifying Statistical Outliers

The presence of outliers among the collected concentration data could distort the estimates of distributional parameters such as upper percentiles. To identify and assess potential outliers in the measurements for the 84 analytes, the following outlier detection tests were accessed in ProUCL:

- Dixon's Extreme Value test (Dixon, 1953), when the sample size is less than 25.
- Rosner's test (Gilbert, 1987), which can detect up to 10 outliers for sample sizes of 25 or more.

The outcomes of both outlier tests are sensitive to the assumption that the data are normally distributed in the absence of outliers. Therefore, the extent to which normality holds in the data was checked along with the results of the outlier tests (or equivalently, lognormality if the tests are performed on log-transformed data). Furthermore, using outlier tests to identify a single (i.e., most extreme) outlier often suffer from masking effects when multiple outliers are present, as these outliers inflate the standard deviation, which makes it more difficult to identify the most extreme data point as an outlier. For both tests, non-detects can be either excluded from the dataset or represented by one-half of the detection limit. (In this analysis, non-detects were excluded; that is, outlier tests were performed only on detected outcomes, and thus, the sample size refers to the number of detected outcomes.)

As always, the decision regarding the proper disposition of outliers (e.g., to include or not to include outliers in statistical analyses) should consider the extent to which conditions associated with sampling and analysis, as well as facility conditions on the day of collection, are not typical, and thus, warrant exclusion. Because no data exclusions could be warranted for such reasons, no outliers were excluded from the calculation of 95th percentiles based on applying these outlier tests.

2.4 Methods for Estimating the 95th Percentile

ProUCL provides four basic statistical approaches to calculating the 95th percentile. They typically calculate a 95th percentile as:

$$\hat{x}_p = \hat{\mu} + c\sqrt{\hat{\sigma}^2} \quad (1)$$

where $\hat{\mu}$ and $\hat{\sigma}^2$ represent the estimate of the mean and variance, respectively, of the underlying distribution, and *c* is a multiplier that is linked to the percentile of interest (i.e., 95th). The four approaches differ in the treatment of non-detects, as follows:

- A traditional substitution approach (Section 2.4.1) that assumed either normality or lognormality, where non-detects are substituted by one-half of the detection limit and treated as detected outcomes in the estimation.
- A maximum likelihood estimation (MLE) approach (Section 2.4.2)
  - For normal or lognormal distributions, the MLE approach was used only when at least one non-detect was; non-detects were treated as left-censored values at the detection limit.
  - For a gamma distribution, the MLE approach was used only when 100% detected outcomes occurred.
- An approach that assumes lognormality or a gamma distribution, where non-detects are substituted by values obtained from extrapolated regression on order statistics (ROS) techniques (Section 2.4.3).
- A nonparametric approach that utilizes Kaplan-Meier (KM) estimates for the mean and standard deviation (Section 2.4.4).

The following sections describe each of the four approaches in more detail.

2.4.1. Detection Limit Substitution Methods

ProUCL makes available substitution methods that replace non-detects with either the detection limit or one-half of the detection limit and then treat the result as detected when calculating the mean and variance of the data. Over the years, scientists have frequently used this approach with datasets containing non-detects due to its simple and straightforward application. However, the arbitrary nature of the substitution makes it less appealing than more statistically rigorous (and computer intensive) approaches. Furthermore, the performance of substitution methods is considerably degraded when multiple detection limits are present, as they are with the TNSSS data. Thus, estimates of the 95th percentiles using substitution methods were included in this analysis as a means of comparison only.

Using substitution methods, ProUCL can calculate 95th percentiles in all instances, as long as the sample size is sufficient to calculate a sample variance. These calculations are as follows:

- Under normality, Equation (1) is applied to the data after substituting non-detects with one-half of the detection limit, $\hat{\mu}$ and $\hat{\sigma}^2$ are the sample mean and variance of these data, and *c*=1.645.
- Under lognormality, Equation (1) is applied to the log-transformed data after substituting non-detects with one-half of the detection limit, $\hat{\mu}$ and $\hat{\sigma}^2$ are the sample mean and variance of these log-transformed data, and *c*=1.645. The result is then exponentiated.

2.4.2. MLE Methods

ProUCL utilized MLE methods in two situations:

- Under the assumption of normality and at least one non-detect outcome,
- Under the assumption of a gamma distribution and 100% detected outcomes.

In the first situation (i.e., normality and at least one non-detect outcome), ProUCL estimates a 95th percentile using Cohen's MLE method (Cohen, 1950, 1959) for those analytes having data that can accommodate the method's numerical analysis. Among the 84 analytes, 21 had sufficient data to calculate MLE estimates for the 95th percentile under the normality assumption. Here, $\hat{\mu}$ and $\hat{\sigma}^2$ are the MLEs of the mean and variance, with non-detects assumed to be left-censored at their respective detection limits. The value of c=1.645, and $\hat{x}_p$ from Equation (1) is the estimated 95th percentile.

In the second situation (i.e., gamma distribution and no non-detects), the 95th percentile estimate corresponds to the 95th percentile of the gamma distribution with shape and scale parameters estimated by their MLEs.

2.4.3. ROS Substitution Method

ProUCL applies the ROS method (Gilliom and Helsel, 1986; Helsel, 1990) only when non-detects are present. The method is applied under a specific distributional assumption (either normality, lognormality, or gamma). ProUCL fits an ordinary least squares regression line to the normal (or lognormal, or gamma) scores of the order statistics for the detected outcomes, and then uses values extrapolated from the fitted line to replace each of the non-detects. As a result, at least three detected outcomes are needed to apply ROS methods, to allow the regression line to be fitted. The extrapolated values for non-detects are then treated as detected outcomes when estimating the mean and variance, and the 95th percentile is then calculated using the standard formulas for the given distribution (e.g., Equation 1 for normal and lognormal distributions).

The ROS method can handle situations where multiple detection limits are present, and when some of the detection limits (for non-detects) exceed the observed detected values. This makes it appealing for use with the TNSSS data.

2.4.4. Kaplan-Meier Nonparametric Method

The nonparametric Kaplan Meier (K-M) approach (Kaplan and Meier, 1958) is also applicable when non-detects are present at multiple detection limits. It was initially developed for survival analysis applications. Because these applications often involve censored data at multiple time points (typically right censored data, such as time until disease occurs or the end of the study, whichever occurs first), the K-M approach accounts for such outcomes. The K-M approach estimates the cumulative distribution function of the underlying parameter of interest, from which percentiles and other distributional-related parameters can be estimated.

The flexibility and distribution-free nature of the K-M approach have led analysts to recognize its potential for analyzing concentration data that include non-detects at multiple detection limits. However, because non-detected outcomes are left-censored in nature, concentration data need to be "flipped" to resemble right-censored data when applying the K-M approach (i.e., subtracted from a large positive value). When the smallest value of a concentration dataset is a non-detect, the K-M approach can yield mean estimates that are biased high, although this does not cause estimates of upper percentiles to be biased (Helsel, 2005).

As modified to apply to left-censored data with possibly multiple detection limits, the K-M approach estimates the cumulative distribution function in the following manner. Let $x_1 \le x_2 \le \ldots \le x_n$ represent the

(observed) measured concentrations (the detection limits for non-detects) for n samples for which the concentrations originate from a common underlying distribution, and assume $y_1 < y_2 < \ldots < y_p$ represent the p distinct values among the detected concentrations (where $1 \le p \le n$. Thus, assume that at least one detected value exists among the n samples). For $j = 1, \ldots, p$, let m_j represent the number of samples whose measured concentrations are classified as detected and are equal to y_j, and let n_j represent the number of samples with reported detected measurements (if detected), or reported detection limits (if non-detected), that are less than or equal to y_j. Then the cumulative distribution function $F(x)$, as estimated by the K-M approach, equals the following:

$F(x) = 1$ if $x \ge y_p$ (i.e., x exceeds the maximum observed detected value)

$$= \prod_{j \text{ such that } y_j > x} \frac{n_j - m_j}{n_j}$$ if $y_1 \le x < y_p$ (i.e., x falls between the smallest and largest observed detected values)

$= F(y_1)$ if $x_1 \le x < y_1$ (i.e., x falls between the smallest detection limit and the smallest detected value)

$= 0$ if $0 \le x < x_1 = y_1$ (i.e., x falls below all observed detected measurements, and the smallest value is detected)

= undefined if $0 \le x < x_1 < y_1$ (i.e., x falls below all observed detected measurements, and the smallest observed value is a non-detect).

Therefore, the estimate $F(x)$ is a step function that is calculated from the highest observed measurement down to the smallest, as follows:

- if $y_{p-1} \le x < y_p$, then $F(x) = (n_p - m_p)/n_p$
- if $y_{p-2} \le x < y_{p-1}$, then $F(x) = [(n_{p-1} - m_{p-1})/n_{p-1}] * [(n_p - m_p)/n_p]$
- etc.

Note that when x is below the smallest of the n reported measurements (x_1), $F(x)$ is undefined if x_1 is a non-detect, and is zero if x_1 is a detected value.

Using the estimate $F(x)$ and the set of p detected measurements, the mean of the distribution is estimated as follows:

$$\hat{\mu} = \sum_{i=1}^{p} y_i [F(y_i) - F(y_{i-1})] \qquad \text{(where } F(y_0)=0\text{)} \qquad (2)$$

The variance is estimated as follows:

$$\hat{\sigma}^2 = \sum_{i=1}^{p} y_i^2 [F(y_i) - F(y_{i-1})] - \hat{\mu}^2 \quad \text{(where } F(y_0)=0\text{)} \qquad (3)$$

One estimate of the 95th percentile based on the K-M approach is the value of x for which $F(x) = 0.95$. (If multiple values of x satisfy this criterion, then any of these values could be chosen, such as the midpoint or minimum value.)

Alternatively, the 95th percentile could be estimated from K-M estimates and standard normal z-scores by calculating the K-M mean and variance from Equations (2) and (3) and inserting those values into Equation 1, letting c=1.645. This alternative approach assumes an underlying normal distribution in the data.

2.4.5. Summary of 95th Percentile Calculation Methods

Table 7 contains a summary of the statistical methods used in ProUCL to calculate 95th percentiles, as discussed in Sections 2.4.1 through 2.4.4. Section 3 applies these methods to the measurement data for the 84 analytes.

Table 7. Summary of 95th Percentiles Calculation Methods in ProUCL.

Normal 95th Percentile (DL/2 Sub.)	95th percentile calculated from the sample mean and standard deviation (sd) with non-detects replaced by one-half of the detection limit: mean + 1.645*sd. Here, 1.645 is the 95th percentile of the standard normal distribution. *ProUCL calculates this estimate in all situations, but does not recommend this substitution method and includes this calculation only for historic reasons.*
Normal 95th Percentile (MLE)	95th percentile calculated from Cohen's MLE estimates of the mean and standard deviation (sd): mean + 1.645*sd. *ProUCL calculates this value only when at least one non-detected sample result exists and when a sufficient number of detected sample results exist to perform the MLE estimation technique.*
Lognormal 95th Percentile (DL/2 Sub.)	95th percentile calculated from the sample mean and standard deviation (sd) of log-transformed concentrations with non-detects replaced by one-half of the log-transformed detection limit: (exp[mean + 1.645*sd]). *ProUCL calculates this estimate in all situations, but does not recommend this substitution method and includes this calculation only for historic reasons.*
Lognormal-ROS 95th Percentile	Regression on order statistics (ROS) approach assuming that non-detected outcomes follow a lognormal distribution. 95th percentile calculated as (exp[mean + 1.645*sd]), where the mean and standard deviation (sd) are calculated as the sample mean and standard deviation with non-detects replaced by estimates obtained from a linear regression fitted to detected measurements paired with lognormal quantiles. *ProUCL calculates this value only when at least one non-detect and three detected sample results exist.*
Gamma 95th Percentile (MLE)	95th percentile calculated by y*theta/2, where y is the 95th percentile of a chi-square distribution with 2*k degrees of freedom (where k is the MLE of the shape parameter of the Gamma distribution), and theta is the MLE of the scale parameter of the Gamma distribution. *ProUCL calculates this value only when all sample results are detected.*
Gamma-ROS 95th Percentile	Regression on order statistics (ROS) approach assuming that non-detected outcomes follow a gamma distribution with shape and scale parameters (y and theta, respectively) represented by their MLEs calculated from detected data. 95th percentile calculated as y*theta/2, with non-detects replaced by estimates obtained from a linear regression fitted to detected measurements paired with quantiles from the same gamma distribution. *ProUCL calculates this value only when at least one non-detect and four detected sample results exist.*
Nonparametric 95th Percentile (Order stats.)	95th percentile calculated by $(0.95*n)^{th}$ order statistic. If (0.95*n) is not an integer, then if l is the next lowest integer and e=(0.95*n)-l, and if x(k) denotes the k^{th} order statistic, then the 95th percentile is x(l)+e*(x(l+1)-x(l)). *ProUCL calculates this value only when all sample results are detected.*
K-M 95th Percentile	95th percentile calculated from the Kaplan-Meier estimate of the cumulative distribution function (Section 2.4.4). *ProUCL calculates this value only when at least one non-detected sample result exists.*

3. Results

Using ProUCL, the methods of Section 2 were applied to the set of concentration data summarized in Table 6, for each of the 84 non-prioritized analytes having at least two detected outcomes. Table 8 summarizes the results of the tests of goodness-of-fit discussed in Section 2.2 and presents estimates of the 95th percentile of the underlying distribution of data under the various approaches presented in Section 2.4 and summarized in Table 7.

For a given analyte, the estimates in Table 8 can vary considerably among the different approaches. In fact, some of these approaches may not be suitable for estimating an analyte's 95th percentile due to the data failing to satisfy important underlying assumptions related to the distribution of the data. This section investigates the distributional properties of the analyte data in order to make a proper decision on an approach for a final estimate of the 95% percentile for each analyte. To assist the decision-making, the detected measurements for each analyte are plotted in histograms within Figures 1 through 5 at the end of this section. (Each bar within these figures represents the number of samples/facilities whose data values fall within a specified range, with the median of the range specified to the left of the bar.)

Goodness of fit test outcomes. For pollutant measurements in environmental media, lognormal or gamma distributions are often good models for the underlying concentration distribution, as they cover only positive values and are skewed toward low values, with long right-hand tails to represent possible large measurements. Table 8 includes the results of goodness-of-fit tests (described in Section 2.2) for the normal, lognormal, and gamma distributions when applied to the detected measurements for each of the 84 analytes. For a given analyte, an "X" is specified in a given column of the table if the distribution specified in the column heading cannot be rejected at a 0.05 significance level. Thus, if no X is specified for a given distribution, then the approaches that require this distribution to hold should not be used to estimate a 95th percentile.

Table 8 shows that when considering the detected observations only, the lognormal and gamma distributions are most frequently deemed satisfactory for the 84 analytes (i.e., could not be rejected by the goodness-of-fit tests). However, nearly one-third of the analytes (27) had neither the lognormal nor gamma distributions as sufficient representations of the observed data based on the outcomes of the goodness-of-fit tests. Nevertheless, the histograms (Figures 1 through 5) demonstrate a skewed distribution for most analytes that resembles a lognormal or gamma distribution.

Of the 84 analytes, the majority of the 35 metals, anions, organics, and PBDEs had 100% detected outcomes, and only one of these analytes was below 50% detected. The lognormal distribution could not be rejected for 23 of these analytes, the gamma distribution fitted satisfactorily to two additional analytes, and all three distributions were rejected for the remaining 10 analytes. In USEPA (2009a), a lognormal assumption was made for the metals, organics, and PBDEs. One could, therefore, recommend using a lognormal-based approach to calculate 95th percentiles among the metals, classicals, organics, and PBDEs, given their high detection percentages and to be consistent with the approach taken in USEPA (2009a). However, for the 12 analytes for which the goodness-of-fit tests for lognormality were rejected, it would be worthwhile to compare the lognormal-based estimates with those under the nonparametric approach and note any differences.

Of the 84 analytes, for the 49 pharmaceuticals, steroids, and hormones, the detection percentages were considerably lower than for the other analytes. Thus, it was more difficult to characterize these distributions. When identifying a common approach to calculating the 95th percentile across these analytes, the overall conclusion from the distributional goodness-of-fit tests is that nonparametric techniques (e.g., Kaplan-Meier) are more appropriate for the pharmaceuticals and steroids/hormones that have a relatively high proportion of non-detects. This conclusion is consistent with ProUCL's recommendations for calculating 95% upper confidence limits on the means when detection percentages were low. It differs, however, from USEPA (2009a), where a lognormal approach was used for the prioritized pharmaceuticals, steroids, and hormones (for which the detection percentages were higher).

Table 8. Outcome of Goodness-of-Fit Tests, and Estimates of 95th Percentiles Using Various Statistical Methods and Assumptions, for 84 Analytes Measured in the TNSSS.

Analyte	n	% Detected	Goodness-of-Fit Test Outcomes (on Detected Results Only)			Normal-Based 95th Percentile Estimates		Lognormal-Based 95th Percentile Estimates		Gamma-Based 95th Percentile Estimates		Nonparametric 95th Percentile Estimates		Minimum 95th Percentile	Obs. Max. Detected Conc.
			Normal Test	Log-normal test	Gamma test	DL/2 Sub.	MLE	DL/2 Sub.	ROS Extrapolation	MLE	ROS Extrapolation	Order stats.	K-M		
Metals (mg/kg)															
Aluminum	74	100.0%		X	X	29,773		34,255		30,870		29,960		29,773	57,300
Antimony	74	86.5%		X		6.93	7.16	14.9	6.49		9.85		6.89	6.49	20.5
Arsenic	74	100.0%		X		17.9		15.7		16.1		14.0		14.0	49.2
Boron	74	97.3%		X	X	94.0	135	115	112		119		93.5	93.5	131
Cadmium	74	100.0%				6.65		6.68		6.54		8.31		6.54	11.8
Calcium	74	100.0%		X		111,421		100,753		103,717		109,700		100,753	243,000
Chromium	74	100.0%				323		227		253		265		227	1,160
Cobalt	74	100.0%				68.1		22.0		36.0		20.4		20.4	290
Copper	74	100.0%		X	X	1,146		1,298		1,202		1,248		1,146	1,720
Iron	74	100.0%		X		70,814		78,323		71,689		91,795		70,814	131,000
Lead	74	100.0%		X		195		220		201		241		195	350
Magnesium	74	100.0%		X	X	10,402		12,096		11,050		11,945		10,402	18,050
Mercury	74	100.0%		X		3.28		3.09		3.09		3.56		3.09	7.50
Nickel	74	100.0%				197		115		148		189		115	526
Phosphorus	74	100.0%		X	X	40,871		44,114		42,278		40,780		40,780	69,400
Selenium	74	100.0%		X	X	13.8		15.6		14.4		14.5		13.8	24.2
Sodium	74	100.0%				10,899		7,653		8,934		10,128		7,653	26,600
Thallium	74	94.6%		X	X	0.517	0.592	0.439	0.439		0.446		0.515	0.439	1.68
Tin	74	94.6%				155	175	109	108		175		155	108	522
Titanium	74	98.6%				1,555	1,550	675	674		1,063		1,547	674	4,805
Vanadium	74	100.0%				162		101		118		111		101	617
Yttrium	74	100.0%		X	X	11.8		12.8		11.9		14.2		11.8	26.3
Zinc	74	100.0%		X		2,622		2,087		2,178		1,839		1,839	8,550
Organics (ug/kg)															
2-Methyl-naphthalene	74	44.6%		X	X	1,349		1,124	728		1,334		1,229	728	4,600
Benzo(a)pyrene	74	77.0%		X		2,220	2,838	2,397	2,194		3,252		2,207	2,194	4,000
Bis(2-ethylhexyl) phthalate	74	100.0%		X	X	161,178		266,644		180,771		184,000		161,178	310,000
Anions (mg/kg)															
Fluoride	74	100.0%		X	X	124		135		128		131		124	234
Water-Extractable Phosphorus	74	100.0%			X	3,792		4,910		3,628		3,733		3,628	9,550
PBDEs (ng/kg)															
BDE-028	78	100.0%				54,936		38,006		43,681		55,600		38,006	160,000
BDE-066	78	100.0%		X		47,906		45,781		44,914		57,300		44,914	110,000
BDE-085	78	100.0%		X		64,134		69,656		64,202		61,150		61,150	150,000

Table 8. (cont.)

Analyte	*n*	*% Detected*	Goodness-of-Fit Test Outcomes (on Detected Results Only)			Normal-Based 95th Percentile Estimates		Lognormal-Based 95th Percentile Estimates		Gamma-Based 95th Percentile Estimates		Nonparametric 95th Percentile Estimates		Minimum 95th Percentile	Obs. Max. Detected Conc.
			Normal Test	Log-normal test	Gamma test	DL/2 Sub.	MLE	DL/2 Sub.	ROS Extrapolation	MLE	ROS Extrapolation	Order stats.	K-M		
BDE-100	78	100.0%			X	386,860		387,979		363,164		314,500		314,500	1,100,000
BDE-138	78	67.9%		X		22,310		23,144	19,114		39,832		18,787	18,787	40,000
BDE-154	78	100.0%		X	X	155,163		149,085		143,689		130,000		130,000	440,000
BDE-183	78	100.0%				50,338		41,314		44,090		57,300		41,314	120,000
Pharmaceuticals (µg/kg)															
1,7-Dimethylxanthine	78	5.1%	X	X	X	2,439		1,125	107		1,071		2,868	107	9,580
4-EOTC	78	10.3%	X	X	X	40.2		40.0	38.8		31.3		43.5	31.3	54.9
4-Epianhydrotetracycline (EATC)	78	34.6%		X	X	694	839	531	517		872		702	517	2,160
Acetaminophen	78	2.6%				528		406					1,156	406	1,300
Anhydrotetracycline (ATC)	78	60.3%				694	848	638	640		1,144		693	638	1,960
Caffeine	78	46.2%		X	X	604	723	587	585		993		602	585	1,110
Clarithromycin	78	53.8%			X	168	192	117	123		204		168	117	617
Codeine	78	24.4%		X		90.0		48.9	49.5		87.0		89.8	48.9	328
Cotinine	78	44.9%				242	679	125	134		260		241	125	690
Dehydronifedipine	78	21.8%				9.03	8.71	6.95	6.55		10.1		9.42	6.55	21.7
Demeclocycline	78	3.8%	X	X		94.1		83.2	50.5				121	50.5	200
Diltiazem	78	82.1%		X		127	147	182	173		187		126	126	225
Enrofloxacin	78	15.4%	X	X	X	44.1		32.4	26.9		56.0		33.1	26.9	66.0
Gemfibrozil	78	89.7%		X		904	920	885	791		993		900	791	2,650
Ibuprofen	78	62.8%				3,300	3,641	1,515	1,799		3,338		3,291	1,515	11,900
Lincomycin	78	3.8%	X	X		36.7		29.5	18.3				18.7	18.3	33.4
Lomefloxacin	78	2.6%				25.4		18.9					34.6	18.9	39.8
Metformin	77	7.8%	X	X	X	716		742	445		341		709	341	1,160
Minocycline	67	43.3%				2,224	2,167	1,038	1,075		2,226		2,261	1,038	8,650
Naproxen	78	51.3%		X		305	361	248	255		409		305	248	1,020
Norfloxacin	78	33.3%			X	763		426	345		575		448	345	995
Oxytetracycline (OTC)	78	35.9%				136	161	96.6	97.9		176		137	96.6	467
Ranitidine	77	57.1%				469	501	91.3	98.8		271		467	91.3	2,250
Roxithromycin	78	2.6%				12.2		11.0					15.9	11.0	22.4
Sarafloxacin	78	2.6%				775		295					538	295	1,980
Sulfachloropyridazine	77	2.6%				18.6		10.8					32.6	10.8	58.7
Sulfadiazine	77	3.9%	X	X		36.8		13.8	1.11				49.1	1.11	140
Sulfadimethoxine	77	6.5%		X	X	14.0		3.90	0.683		6.86		15.6	0.683	62.2
Sulfamethazine	77	2.6%				14.5		7.68					21.8	7.68	23.2
Sulfamethoxazole	77	37.7%				142	156	31.4	36.1		94.9		142	31.4	651
Sulfanilamide	77	10.4%		X	X	3,650	2,620	451	82.3		2,096		3,715	82.3	15,600

Table 8. (cont.)

Analyte	*n*	*% Detec-ted*	Goodness-of-Fit Test Outcomes (on Detected Results Only)			Normal-Based 95th Percentile Estimates		Lognormal-Based 95th Percentile Estimates		Gamma-Based 95th Percentile Estimates		Nonparametric 95th Percentile Estimates		Mini-mum 95th Percent-ile	Obs. Max. Detected Conc.
			Normal Test	Log-normal test	Gamma test	DL/2 Sub.	MLE	DL/2 Sub.	ROS Extra-polation	MLE	ROS Extra-polation	Order stats.	K-M		
Thiabendazole	78	69.2%				113	124	107	110		177		112	107	238
Trimethoprim	78	29.5%		X	X	76		65.1	50.7		88.1		74.2	50.7	204
Virginiamycin	78	17.9%		X	X	284		234	93.6		167		183	93.6	469
Steroids/Hormones (µg/kg)															
17 Alpha-estradiol	73	6.8%	X	X	X	21.5		18.3	21.1		40.3		23.4	18.3	48.8
17 Beta-estradiol	78	11.5%				69.2		41.0	18.8		40.3		67.0	18.8	222
Androstenedione	73	41.1%		X	X	785	1,049	795	736		1,184		774	736	1,520
Androsterone	73	65.8%		X	X	365	442	390	366		579		363	363	1,030
Beta-Estradiol 3-Benzoate	74	23.0%		X	X	652	826	245	192		591		650	192	1,850
Beta-Sitosterol	78	85.9%		X	X	756,638	786,498	3,422,184	1,069,746		1,492,776		751,640	751,640	1,640,000
Desmosterol	78	66.7%		X	X	40,327	46,638	47,737	42,371		73,162		40,145	40,145	94,400
Equilin	73	17.8%	X	X	X	52.6		49.4	34.1		49.7		51.7	34.1	100
Ergosterol	78	61.5%		X	X	51,969	58,455	77,101	60,380		100,132		51,566	51,566	91,900
Estriol	74	21.6%	X	X	X	93.9	121	70.3	51.2		99.0		91.7	51.2	232
Estrone	73	76.7%				376	460	339	328		544		375	328	965
Norethindrone	76	6.6%		X		397		87.4	114		293		293	87.4	1,360
Norgestrel	74	5.4%		X	X	289		65.8	2.29		120		302	2.29	1,300
Progesterone	77	22.1%	X		X	810		731	600		949		797	600	1,290
Testosterone	73	23.3%		X		544	356	273	161		390		526	161	2,040

X: The hypothesis that the given distribution holds cannot be rejected at the 0.05 level.

In those few instances where normality could not be rejected at a 0.05 level (i.e., 11 pharmaceuticals and steroids/hormones), only a small number of detected outcomes (less than 25%) were available for the goodness-of-fit test. As a result, for these analytes, there is typically not sufficient power to declare that a given distributional form is not appropriate, as these tests require the data to demonstrate that the distribution model does not hold. Thus, normality was not considered to be a viable distributional assumption for the analytes in Table 8.

Identifying possible statistical outliers. Section 2.3 noted the two outlier tests that ProUCL uses to identify statistical outliers among a set of detected outcomes: Dixon's test (which identifies a maximum of one outlier and is applied when the number of detected outcomes is less than 25), and Rosner's test (which can identify up to 10 outliers and is applied when at least 25 detected outcomes are available). These tests were applied to the set of log-transformed detected measurements for each of the 84 analytes (as Figures 1 through 5 indicate that the log-measurements are more likely to resemble a normal distribution compared to the untransformed measurements, and these outlier tests assume normality in the data being analyzed). When Rosner's test was applied in this analysis, a maximum of five outliers was specified given the sample sizes.

Outlier testing resulted in identifying one or more statistical outliers at the 0.05 significance level for 13 of the 84 analytes. Table 9 lists these analytes and those measurements identified as statistical outliers (with the ID number for the surveyed facility that was linked to the outcome in parentheses following each measurement). Because the number of detected outcomes for each of these 13 analytes exceeded 25, Rosner's test was used to identify the outliers (listed in the last column of Table 9). As a means of comparison, Table 9 also includes the largest detected measurement for the analyte which was not labeled as an outlier – each outlier listed in the last column of Table 9 ranged from 50% higher (BDE 028) to over 14 times higher (Rantidine) than the analyte's highest non-outlier measurement. These outliers are clearly visible in the histograms within Figures 1 through 5. Finally, Table 9 indicates that the outliers are associated with a variety of facilities, and no one facility tends to be the source of many outliers (which would have suggested a possible issue with that facility which would make its measurements incompatible with the distribution of measurements from the other facilities).

Table 9. Detected Facility Measurements Labeled as Statistical Outliers by Outlier Tests for 84 Analytes Measured in the TNSSS.

Analyte	Number of Detected Measure-ments	Highest detected measurement not classified as an outlier	Detected measurements labeled as outliers at a 5% Significance Level (survey ID of facility is in parentheses)		
Metals (mg/kg)					
Antimony	64	9.89			**20.5 (20)**
Arsenic	74	29.8			**49.2 (55)**
Cobalt	74	23.9		**97.2 (57)**	**290 (37)**
Nickel	74	255		**508 (2)**	**526 (71)**
Thallium	70	0.50			**1.68 (55)**
Tin	70	226			**522 (7)**
Titanium	73	732	**1,930 (4)**	**4,510 (27)**	**4,805 (18)**
Vanadium	74	190			**617 (4)**
Zinc	74	2,479			**8,550 (57)**
PBDEs (ng/kg)					
BDE 028	78	77,000		**120,000 (70)**	**160,000 (48)**
Pharmaceuticals (µg/kg)					
Minocycline	29	1,590			**8,650 (9)**
Oxytetracycline (OTC)	28	139			**467 (62)**
Ranitidine	44	154			**2,250 (47)**

While the presence of large outliers has the potential for impacting the 95th percentile estimates considerably, no evidence was apparent to exclude any of the measurements listed in Table 9 from the calculation of 95th percentile estimates due to quality concerns. However, if other concerns remain for these outliers, nonparametric approaches tend to be less impacted by the presence of outliers compared to the approaches that are specific to a distributional model form.

95th percentile estimates for the pharmaceuticals, steroids, and hormones (use of nonparametric estimation techniques). For the pharmaceuticals, steroids, and hormones, the relatively high non-detect percentages warranted that the 95th percentile estimates should be based upon nonparametric K-M techniques (e.g., MLE techniques tend to yield unstable estimates when the percentage of non-detects is high). Table 10 lists the recommended estimates of the 95th percentile for these analytes, along with their maximum observed values.

Table 10. Recommended (Nonparametric) Estimates of the 95th Percentile for the Pharmaceuticals, Steroids, and Hormones, Along with the Maximum Observed Concentration

Analyte	95th Percentile	Observed Maximum Conc.	Analyte	95th Percentile	Observed Maximum Conc.
Pharmaceuticals (µg/kg)					
1,7-Dimethylxanthine	2,868	9,580	Sulfachloro-pyridazine	32.6	58.7
4-EOTC	43.5	54.9	Sulfadiazine	49.1	140
4-Epianhydrotetra-cycline (EATC)	702	2,160	Sulfadimethoxine	15.6	62.2
Acetaminophen	1,156	1,300	Sulfamethazine	21.8	23.2
Anhydrotetracycline (ATC)	693	1,960	Sulfamethoxazole	142	651
Caffeine	602	1,110	Sulfanilamide	3,715	15,600
Clarithromycin	168	617	Thiabendazole	112	238
Codeine	89.8	328	Trimethoprim	74.2	204
Cotinine	241	690	Virginiamycin	183	469
Dehydronifedipine	9.42	21.7	**Steroids/Hormones (µg/kg)**		
Demeclocycline	121	200	17 Alpha-estradiol	23.4	48.8
Diltiazem	126	225	17 Beta-estradiol	67.0	222
Enrofloxacin	33.1	66.0	Androstenedione	774	1,520
Gemfibrozil	900	2,650	Androsterone	363	1,030
Ibuprofen	3,291	11,900	Beta-Estradiol 3-Benzoate	650	1,850
Lincomycin	18.7	33.4	Beta-Sitosterol	751,640	1,640,000
Lomefloxacin	34.6	39.8	Desmosterol	40,145	94,400
Metformin	709	1,160	Equilin	51.7	100
Minocycline	2,261	8,650	Ergosterol	51,566	91,900
Naproxen	305	1,020	Estriol	91.7	232
Norfloxacin	448	995	Estrone	375	965
Oxytetracycline (OTC)	137	467	Norethindrone	293	1,360
Ranitidine	467	2,250	Norgestrel	302	1,300
Roxithromycin	15.9	22.4	Progesterone	797	1,290
Sarafloxacin	538	1,980	Testosterone	526	2,040

Note that the 95th percentile estimates (second column of Table 10) are, on average, 43 percent of the size of the observed maximum concentration (last column). These estimates range from 21 percent (Ranitidine, which has a large outlier as noted in Table 9) to 94 percent (Sulfamethazine) of the observed maximum. These estimates tended to be in line with the estimates from other techniques, and more importantly, do not appear to be underestimates.

95th percentile estimates for the non-prioritized metals, anions, organics, and PBDEs (use of lognormal estimation techniques). The metals, anions, organics, and PBDEs had relatively high percentages of detected measurements which tended to be well-represented by a lognormal distribution. Table 11 lists the recommended lognormal-based estimates of the 95th percentiles for these analytes along with their maximum observed values. Like the pharmaceuticals, steroids, and hormones in Table 10, the 95th percentiles in Table 11 are 43% of the observed maximum concentrations, on average. They range from 8% (cobalt, which had two large outliers as noted in Table 9) to 86% (Bis(2-ethylhexyl) phthalate) of the observed maximum. They are similar in magnitude to the nonparametric estimates for these analytes.

Table 11. Recommended (Lognormal-Based) Estimates of the 95th Percentile for the 84 Metals, Organics, Anions, and PBDEs, Along with the Maximum Observed Concentration.

Analyte	95th Percentile	Observed Maximum Conc.
Metals (mg/kg)		
Aluminum	34,255	57,300
Antimony	6.49	20.5
Arsenic	15.7	49.2
Boron	112	131
Cadmium	6.68	11.8
Calcium	100,753	243,000
Chromium	227	1,160
Cobalt	22.0	290
Copper	1,298	1,720
Iron	78,323	131,000
Lead	220	350
Magnesium	12,096	18,050
Mercury	3.09	7.50
Nickel	115	526
Phosphorus	44,114	69,400
Selenium	15.6	24.2
Sodium	7,653	26,600
Thallium	0.439	1.68
Tin	108	522
Titanium	674	4,805
Vanadium	101	617
Yttrium	12.8	26.3
Zinc	2,087	8,550

Analyte	95th Percentile	Observed Maximum Conc.
Organics (µg/kg)		
2-Methyl-naphthalene	728	4,600
Benzo(a)pyrene	2,194	4,000
Bis(2-ethylhexyl) phthalate	266,644	310,000
Classicals (mg/kg)		
Fluoride	135	234
Water-Extractable Phosphorus	4,910	9,550
PBDEs (ng/kg)		
BDE-028	38,006	160,000
BDE-066	45,781	110,000
BDE-085	69,656	150,000
BDE-100	387,979	1,100,000
BDE-138	19,114	40,000
BDE-154	149,085	440,000
BDE-183	41,314	120,000

Note from Table 8 that only modest differences in the 95th percentile estimates occur between the lognormal-based and nonparametric approaches for the 12 analytes in Table 11 for which the goodness-of-fit test for lognormality was rejected. Thus, taking a lognormal approach to estimating 95th percentiles for each of the analytes in Table 11 is not highly impactful when lognormality is rejected.

Updated 95th percentile estimates for an analyte in the 2009 report having an outlier excluded. The 95th percentile estimates in Tables 10 and 11 utilized all available data without excluding any of the outliers listed in Table 9. In contrast, USEPA (2009a) presented the 95th percentile estimate for silver upon excluding one outlier (856 mg/kg) from the calculation. This outlier was suspected to be the result of an anomaly to normal operations at the POTW, although the value of the sample analysis was confirmed with the facility (USEPA, 2009b). The 95th percentile estimates for silver were as follows:

- 95th percentile estimate with outlier excluded: 57 mg/kg (as reported in USEPA, 2009a).
- 95th percentile estimate with outlier included: 74 mg/kg (a 30 percent increase).

Note that among the other analytes in the 2009 report, one sample measurement for cimetidine and two sample measurements for fluoxetine were also omitted from estimation in USEPA (2009a), but the exclusions were due to failing chemical quality assurance criteria rather than classification as a statistical outlier.

Comparing 95th percentile estimates with estimates that result from applying the analysis approach used on prioritized analytes in the 2009 report. Table B-7 of USEPA (2009a) included preliminary estimates of the 95th percentile for the 84 analytes in this report using the statistical techniques that were applied to the 34 prioritized analytes. Table 12 replicates the estimates from this table, as a means of comparing to the 95th percentile estimates given in Tables 10 and 11. The 2009 statistical analysis accounted for the survey weights assigned to the sampled POTWs and the survey's stratified sample design.

Table 12. Weighted Summary Statistics and 95th Percentile Estimates for the 84 Analytes, Using Statistical Techniques Applied in the Weighted (Preliminary) Analysis Performed in USEPA (2009a).

Analyte	# Sampled POTWs	Mean	Standard Deviation	Median	95th Percentile
Metals (mg/kg)					
Aluminum	74	13,477.80	10,020.66	11,200.00	34,525.52
Antimony	74	2.26	2.99	1.42	14.18
Arsenic	74	6.76	6.84	4.95	15.13
Boron	74	43.25	33.70	33.00	122.42
Cadmium	74	2.48	2.28	1.72	6.09
Calcium	74	39,539.11	39,847.24	25,950.00	96,371.30
Chromium	74	78.15	152.58	30.60	212.92
Cobalt	74	10.99	36.71	4.44	21.51
Copper	74	558.54	368.89	449.00	1,330.71
Iron	74	24,742.64	27,716.08	13,250.00	71,425.51
Lead	74	73.96	73.51	44.40	210.31
Magnesium	74	4,705.62	2,978.38	4,300.00	11,295.55
Mercury	74	1.27	1.29	0.83	3.20
Nickel	74	47.38	92.09	22.80	108.42
Phosphorus	74	21,668.72	11,761.54	18,300.00	43,262.02
Selenium	74	7.10	4.18	6.20	15.97
Sodium	74	2,873.59	5,102.50	1,110.00	8,344.24
Thallium	74	0.17	0.21	0.13	0.41
Tin	74	43.54	40.38	36.20	102.33
Titanium	74	221.31	601.17	80.90	627.73
Vanadium	74	33.94	79.63	11.60	86.76
Yttrium	74	4.55	3.63	3.54	12.07
Zinc	74	969.77	1,054.80	759.00	2,110.95
Organics (µg/kg)					
2-Methylnaphthalene	74	449.04	746.50	200.00	1,111.65
Benzo(A)Pyrene	74	661.00	849.06	320.00	2,259.31
Bis(2-Ethylhexyl) Phthalate	74	48,142.54	65,207.23	23,000.00	226,937.29
Anions (mg/kg)					
Fluoride	74	58.20	35.87	54.20	132.68
Water-Extractable Phosphorus	74	1,062.09	1,770.57	480.00	5,012.47
PBDEs (ng/kg)					
BDE 28	78	13,990.24	20,783.92	8,500.00	33,076.02
BDE 66	78	16,536.70	16,088.17	12,000.00	41,134.17
BDE 85	78	27,824.89	20,202.11	23,000.00	66,312.15
BDE 100	78	148,973.10	125,545.38	120,000.00	362,133.61
BDE 138	78	10,807.30	12,722.42	7,000.00	20,822.02
BDE 154	78	58,730.15	50,756.61	49,000.00	143,826.47
BDE 183	78	15,079.78	17,215.83	10,000.00	36,522.57
Pharmaceuticals (ug/kg)					
1,7-Dimethylxanthine	78	1,180.46	1,088.76	986.50	1,440.00
4-Epioxytetracycline (EOTC)	78	45.30	11.66	41.50	68.60
4-Epianhydrotetracycline (EATC)	78	251.31	301.11	140.00	797.00
Acetaminophen	78	461.80	200.38	395.50	973.00
Anhydrotetracycline (ATC)	78	262.91	283.89	153.00	680.00
Caffeine	78	231.59	239.33	103.00	881.00
Clarithromycin	78	41.58	81.76	13.40	141.00
Codeine	78	30.63	40.75	19.90	70.40
Cotinine	78	57.97	120.79	13.20	332.00
Dehydronifedipine	78	5.03	3.12	4.04	10.70
Demeclocycline	78	105.97	24.36	99.20	147.00
Diltiazem	78	40.20	56.35	14.80	199.00
Enrofloxacin	78	27.87	30.69	19.80	66.00

Table 12. (cont.)

Analyte	# Sampled POTWs	Mean	Standard Deviation	Median	95th Percentile
Gemfibrozil	78	213.56	437.13	101.00	665.00
Ibuprofen	78	652.80	1,703.48	143.00	2,980.00
Lincomycin	78	30.20	27.43	19.90	85.10
Lomefloxacin	78	22.93	15.84	19.80	33.30
Metformin	77	533.68	451.80	546.00	1,160.00
Minocycline	67	660.76	1,090.03	433.00	1,180.00
Naproxen	78	86.20	146.58	31.60	316.00
Norfloxacin	78	274.57	699.46	109.00	684.00
Oxytetracycline (OTC)	78	57.87	53.47	43.15	113.00
Ranitidine	77	57.66	276.53	12.50	89.80
Roxithromycin	78	8.10	9.17	4.72	22.35
Sarafloxacin	78	293.65	718.92	91.90	1,150.00
Sulfachloropyridazine	77	11.96	9.91	9.84	14.00
Sulfadiazine	77	13.61	18.22	9.84	22.90
Sulfadimethoxine	77	3.57	7.67	2.01	7.35
Sulfamethazine	77	7.38	12.57	4.02	21.50
Sulfamethoxazole	77	21.65	81.60	4.32	67.70
Sulfanilamide	77	536.88	2,110.35	99.20	2,390.00
Thiabendazole	78	36.59	49.33	16.50	137.00
Trimethoprim	78	30.37	37.72	10.80	114.00
Virginiamycin	78	137.50	233.05	73.30	469.00
Steroids and Hormones (ug/kg)					
17 Alpha-Estradiol	73	22.54	6.45	21.40	27.20
17 Beta-Estradiol	78	34.33	40.48	21.50	131.00
Androstenedione	73	326.82	325.94	158.00	1,100.00
Androsterone	73	120.29	130.72	84.90	332.00
Beta-Estradiol 3-Benzoate	74	146.80	345.64	23.20	695.00
Beta-Sitosterol	78	291,398.60	294,849.73	207,000.00	885,000.00
Desmosterol	78	15,654.68	16,484.25	10,800.00	38,500.00
Equilin	73	34.77	22.37	23.00	80.60
Ergosterol	78	19,829.93	18,535.97	12,600.00	56,100.00
Estriol	74	38.70	38.78	24.80	128.00
Estrone	73	105.97	160.61	51.20	326.00
Norethindrone	76	101.84	338.51	22.30	146.00
Norgestrel	74	66.94	155.02	42.00	111.00
Progesterone	77	322.37	355.78	139.00	1,260.00
Testosterone	73	162.85	270.69	95.20	511.00

Taken from Table B-7 of USEPA (2009a).

Like the analyses presented in Tables 10 and 11, the weighted analysis estimates presented in Table 12 utilized a nonparametric approach for pharmaceuticals, steroids, and hormones, and a lognormal-based approach for all other analytes:

- The weighted lognormal approach is documented in Section C.1 of Appendix C of USEPA (2009a). This approach used Cohen's MLE techniques when non-detects were present.
- The nonparametric approach is documented in Section C.2 of Appendix C of USEPA (2009a). It utilized a weighted order statistics approach to identifying the 95th percentile, but substituted non-detects with the detection limit.

In general, the 95th percentile estimates in the last column of Table 12 compared favorably with the estimates given in Tables 10 and 11. The following specific findings were noted:

- For metals, organics, anions, and PBDEs, the estimates differed on average by about one percent. The weighted analysis tended to yield larger estimates than the above unweighted analyses. The largest observed difference was a 54% decrease, from 14.2 to 6.5 mg/kg, in the 95th percentile estimate for antimony from the weighted analysis estimate to the unweighted estimate in this report.
- For pharmaceuticals and steroids/hormones, the difference was about 16 percent, on average. Larger differences between the two methods were observed, in part due to the nonparametric approach and the smaller number of detected outcomes compared to the other analytes. The number of analytes with estimates from the weighted analysis that were lower than the estimates presented in Table 10 was about equal to the number that had higher estimates.

Table 13 lists the 34 prioritized analytes and estimates of the 95th percentile under the in-depth (weighted) analysis used in USEPA (2009a), as well as both the lognormal-based and nonparametric (unweighted) approaches used for the non-prioritized analytes in this report. The lognormal-based unweighted estimates averaged about 7% lower than the weighted estimates for these analytes. (The weighted analysis for all but one of these analytes was lognormal-based.) The nonparametric unweighted estimates averaged 17% lower than the weighted estimates. Thus, using techniques that utilize a lognormal distributional assumption, the 95th percentile estimates differ as a whole in only a minor way between the weighted and unweighted approaches.

Thus, as a result of this investigation, it is not apparent that accounting for the weighting and stratified sample design as was done by using the in-depth analysis approach (Tables 12) would lead to considerably different estimates for the 95th percentile compared to the results from the unweighted analysis that are presented in Tables 10 and 11.

Table 13. 95th Percentile Estimates for the Prioritized Analytes, as Reported in USEPA (2009a), and Unweighted Estimates Generated by ProUCL.

Analyte	95th Percentile Estimates		
	As reported in USEPA (2009a)[1]	Unweighted Estimates from ProUCL -- Lognormal	Unweighted Estimates from ProUCL -- Nonparametric
Metals (mg/kg)			
Barium	1,396	1,336	1,674
Beryllium	1.04	1.06	0.99
Manganese	4,156	4,020	3,430
Molybdenum	40.5	40.9	43.5
Silver	57	71.5	63.6
Organics (µg/kg)			
4-Chloroaniline	4,762	3,541	2,648
Fluoranthene	5,256	5,774	5,374
Pyrene	6,184	6,398	6,477
Classicals (mg/kg)			
Nitrate/Nitrite	960	473	712
PBDEs (ng/kg)			
BDE-47	1,688,881	1,776,508	1,575,000
BDE-99	1,713,370	1,812,193	1,530,000
BDE-153	166,454	170,769	150,000
BDE-209	7,360,103	8,029,037	7,606,248
Pharmaceuticals (ug/kg)			
4-Epitetracycline (ETC)	3,787	3,513	2,470
Azithromycin	3,172	2,689	2,484
Carbamazepine	497	468	1,317
Cimetidine*	4,789	3,631	3,429
Ciprofloxacin	36,095	34,531	21,690
Diphenhydramine	2,696	2,662	2,005
Doxycycline	3,082	2,348	1,988
Erythromycin-Total	123	103	82.8
Fluoxetine*	778	688	863
Miconazole	4,652	3,643	3,417
Ofloxacin	32,363	27,133	19,753
Tetracycline (TC)	4,458	4,185	2,823
Triclocarban	131,079	144,599	95,475
Triclosan	62,217	63,043	40,268
Steroids and Hormones (ug/kg)			
Beta Stigmastanol	632,009	631,228	504,913
Campesterol	360,119	360,990	257,550
Cholestanol	2,629,149	2,519,426	1,446,500
Cholesterol	4,369,111	3,355,221	1,976,463
Coprostanol	16,626,022	16,249,696	8,001,500
Epicoprostanol	5,143,938	5,948,141	2,716,385
Stigmasterol	1,157,099	365,893	281,498

[1] In-depth analysis was based on a lognormal approach for all but nitrate/nitrite, for which a nonparametric approach was used.

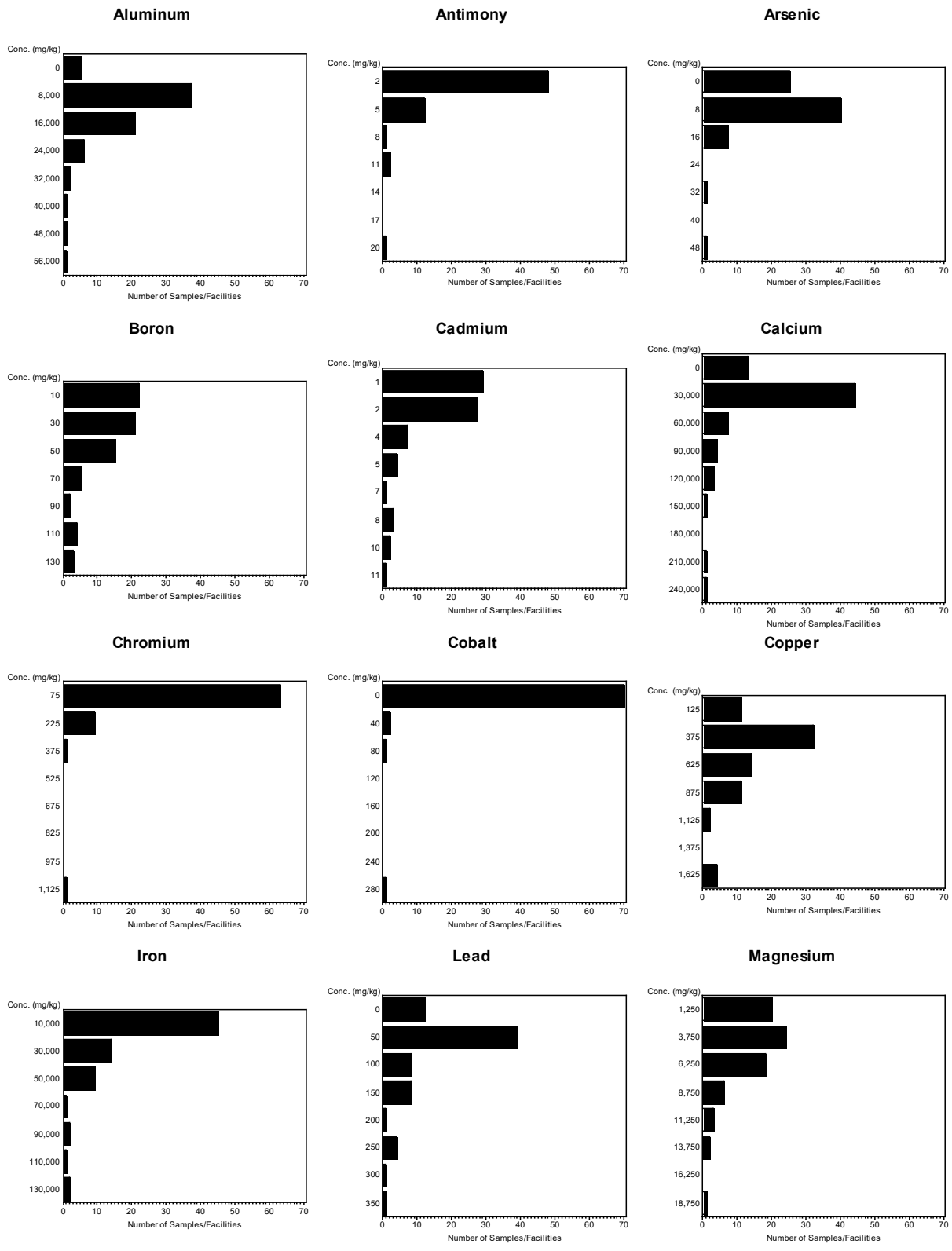


Figure 1. Histograms of Facility-Specific Concentrations for Non-Prioritized Metals in the TNSSS.

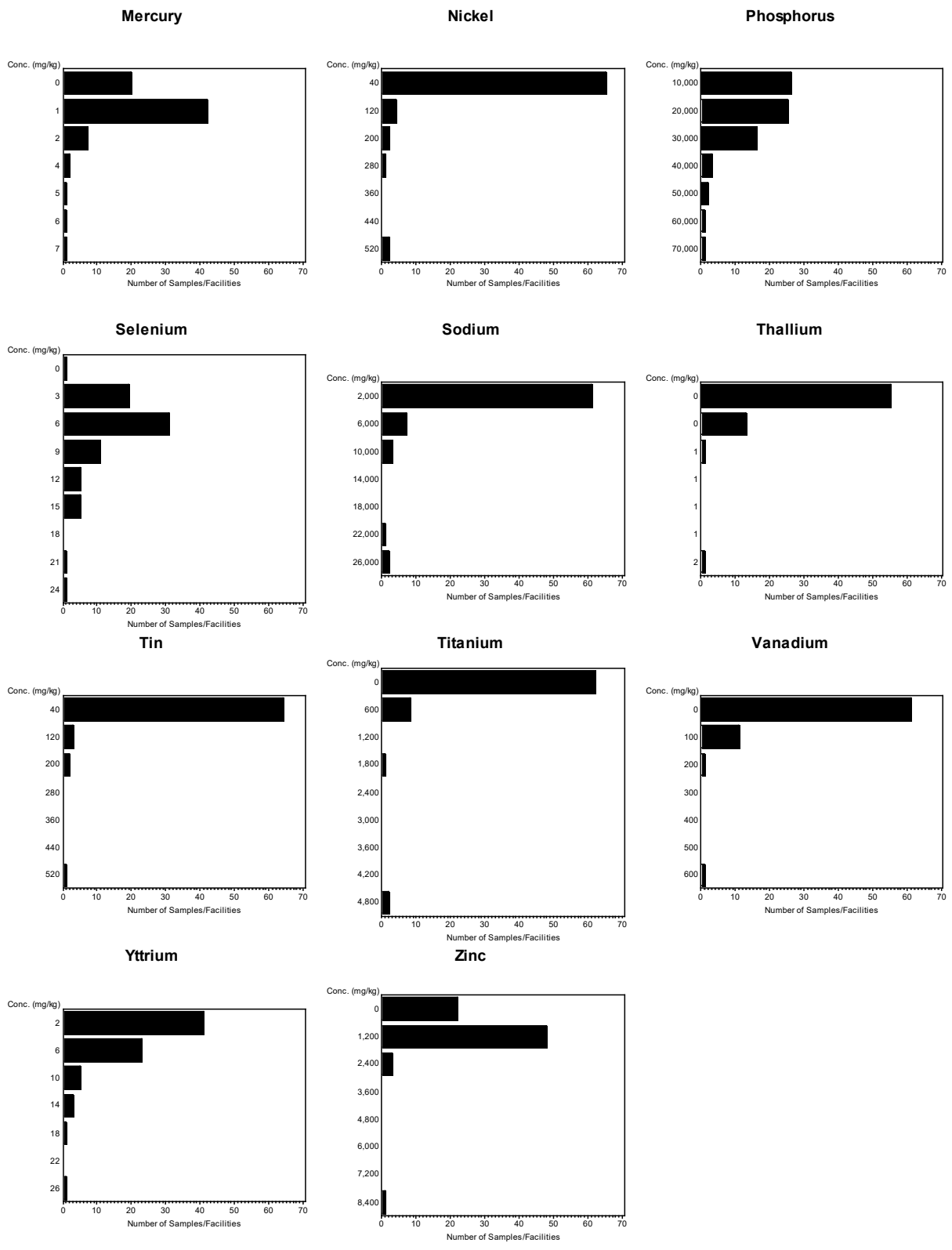


Figure 1. (cont.)

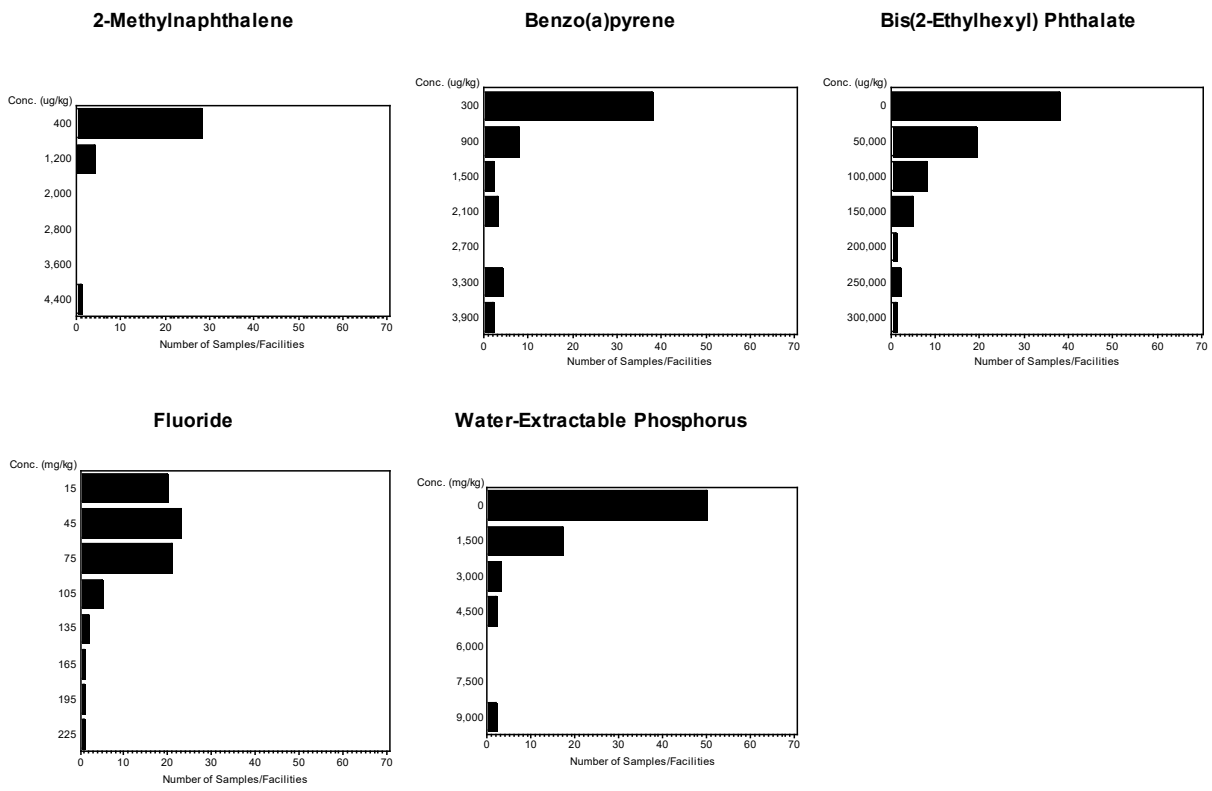


Figure 2. Histograms of Facility-Specific Concentrations for Non-Prioritized Organics and Classicals (Anions) in the TNSSS.

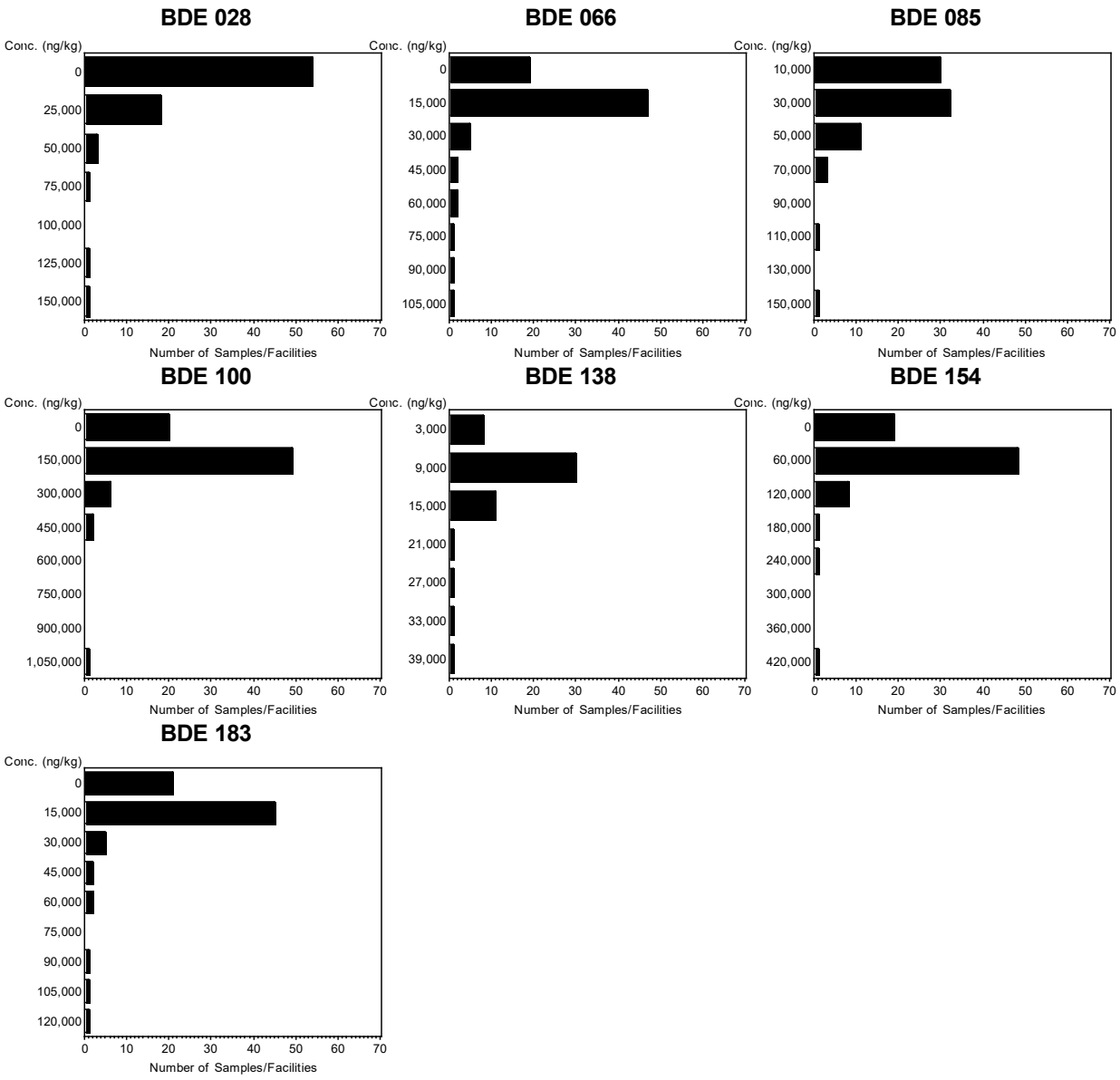


Figure 3. Histograms of Facility-Specific Concentrations for Non-Prioritized PBDEs in the TNSSS.

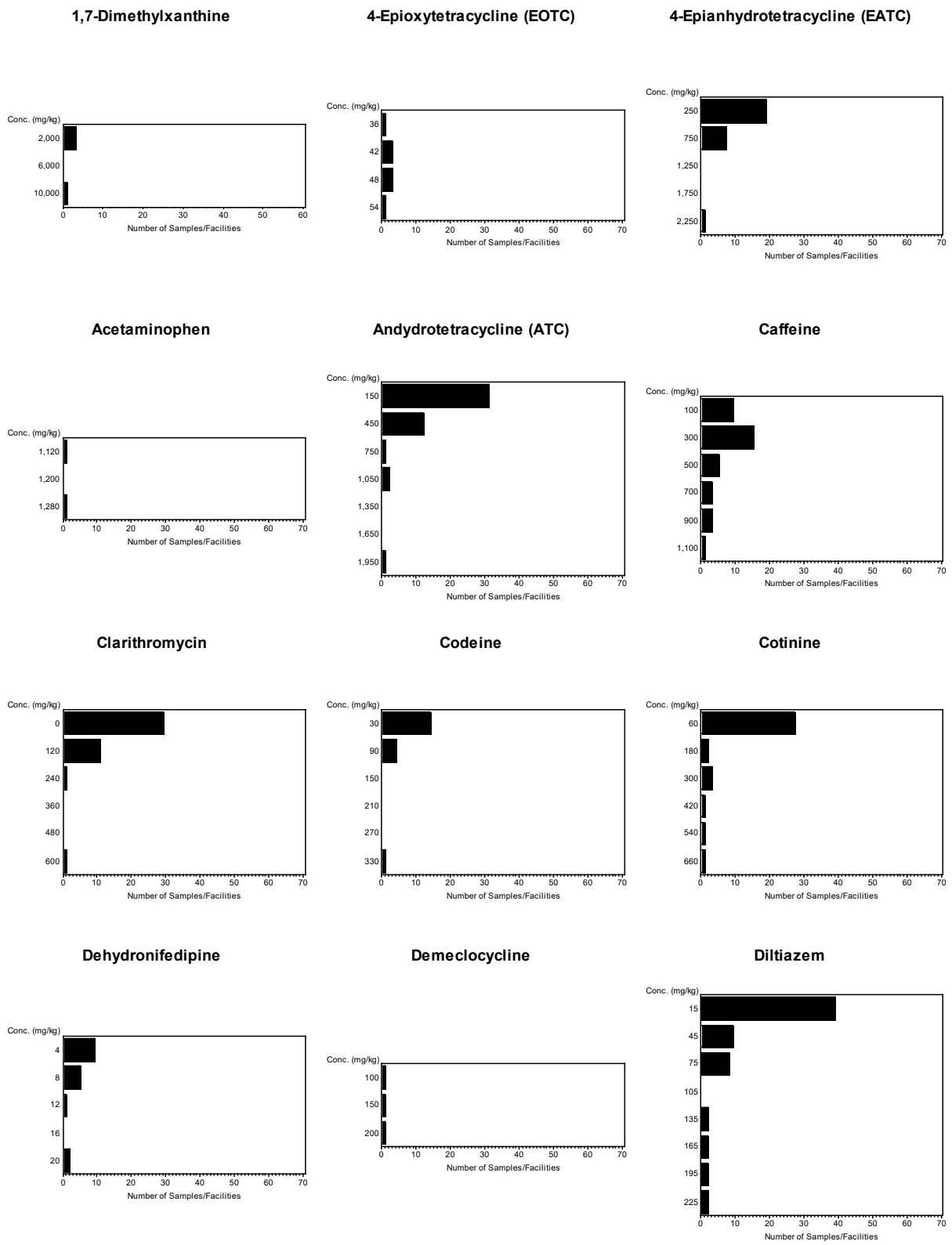


Figure 4. Histograms of Facility-Specific Concentrations for Non-Prioritized Pharmaceuticals in the TNSSS.

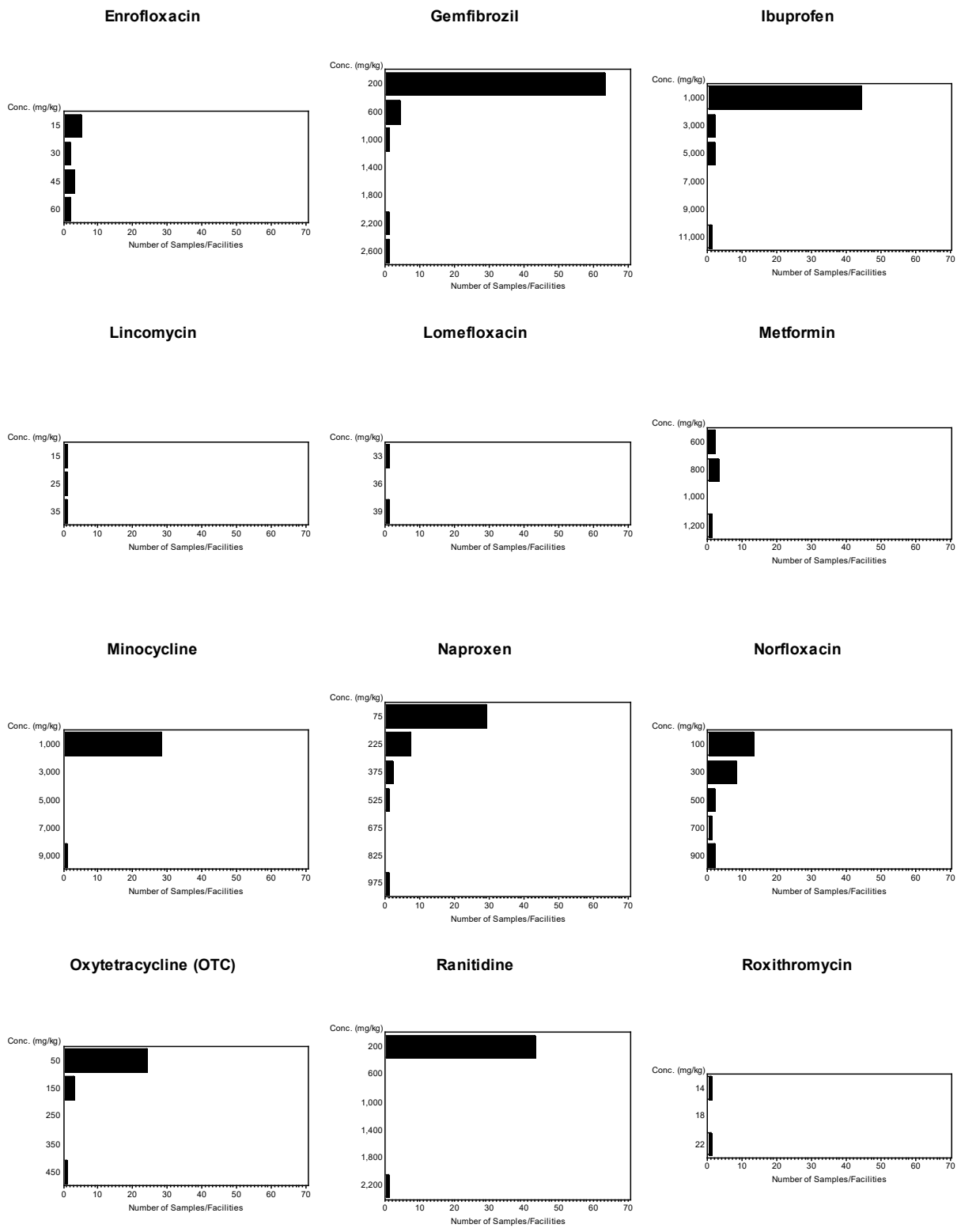
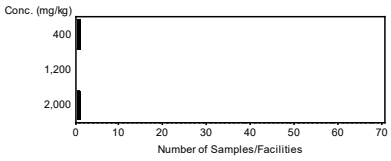
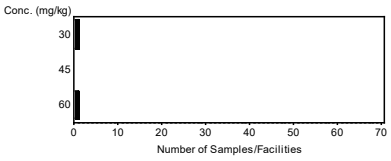


Figure 4. (cont.)

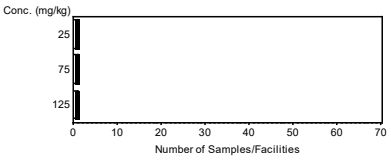
Sarafloxacin



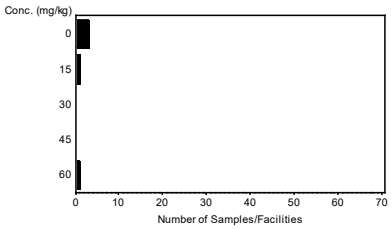
Sulfachloropyridazine



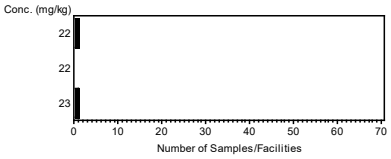
Sulfadiazine



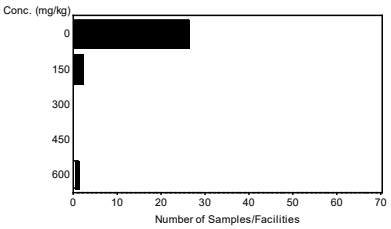
Sulfadimethoxine



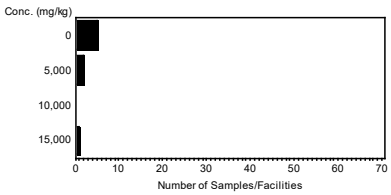
Sulfamethazine



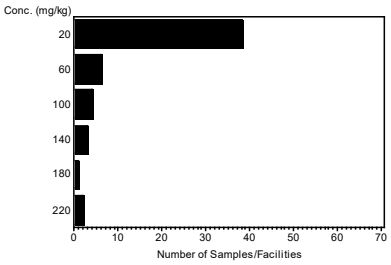
Sulfamethoxazole



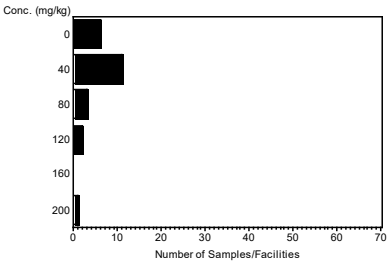
Sulfanilamide



Thiabendazole



Trimethoprim



Virginiamycin

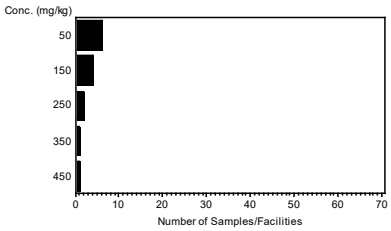


Figure 4. (cont.)

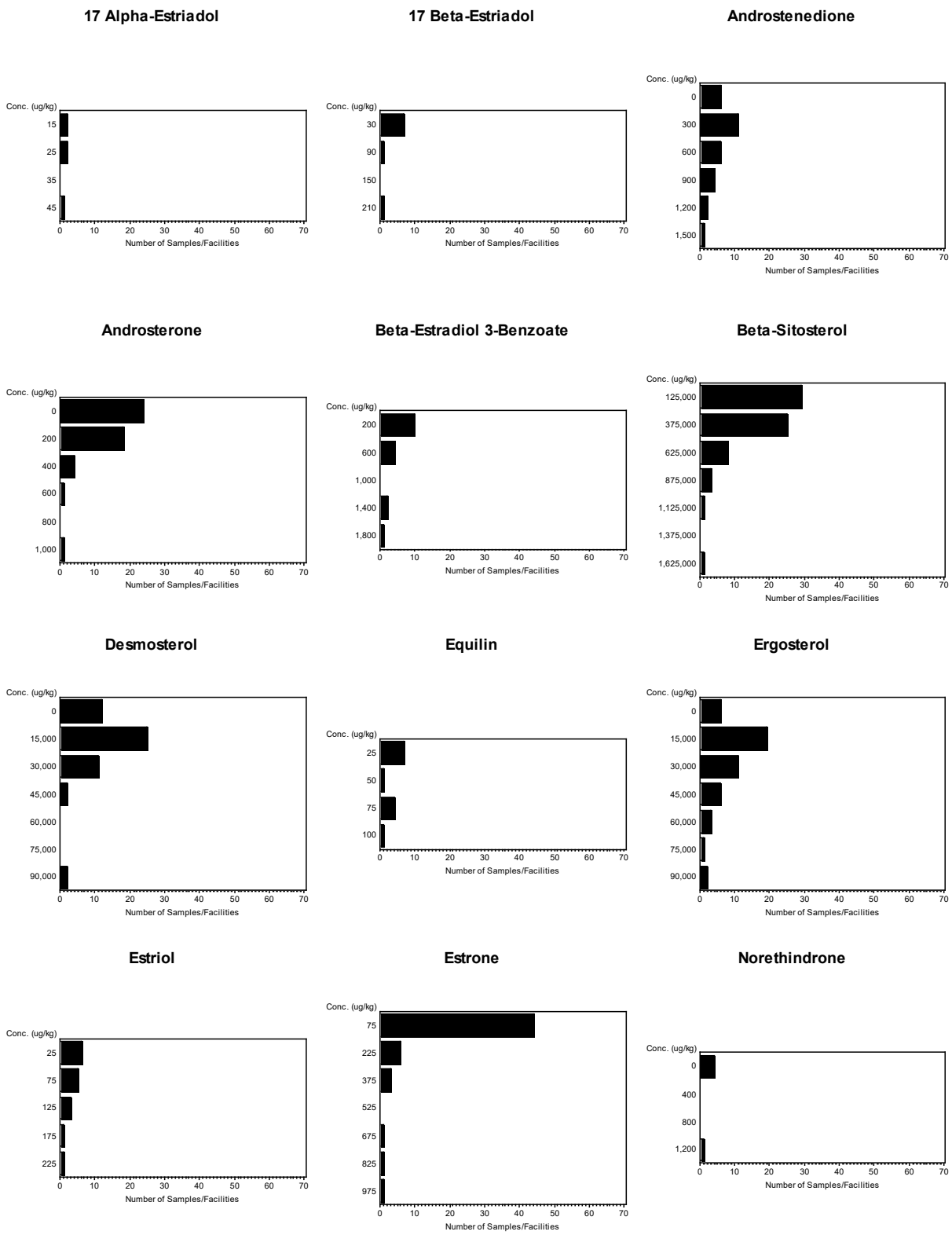


Figure 5. Histograms of Facility-Specific Concentrations for Non-Prioritized Steroids/Hormones in the TNSSS.

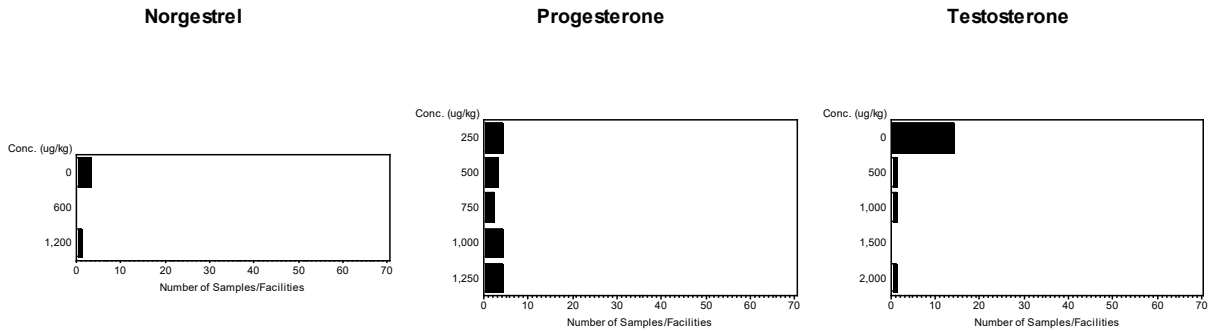


Figure 5. (cont.)

4. Key Findings and Conclusions

This report presented estimates of the 95th percentile for 84 additional analytes which were measured in the treated biosolids sampled within the TNSSS. These 84 analytes had at least two detected outcomes among the tested biosolids from the sampled facilities. The statistical techniques available within EPA's ProUCL open-source software tool were applied to yield the 95th percentile estimates. Because the measurements in the TNSSS were frequently below detection limits, and because multiple detection limit values were observed for a given analyte, the ProUCL software was especially relevant for use here. ProUCL offers rigorous statistical estimation techniques that handle non-detects more appropriately than simple substitution methods that treat non-detects as detected outcomes. These estimation techniques allow for non-detects at multiple detection limits and include the Kaplan-Meier nonparametric technique and regression on order statistics (ROS) methods that extrapolate values for non-detects based on information available from the detected outcomes.

Conclusions for the 84 analytes:

- For the metals, organics, anions, and PBDEs, which tended to have a high prevalence of detected outcomes, a lognormal-based approach was recommended for estimating the 95th percentile. When non-detects were present for a given analyte, ROS estimates were assigned to the non-detects. These ROS estimates were obtained by extrapolating from a fitted ordinary least squares regression line that was fitted to the observed log-transformed detected outcomes and corresponding normal scores.
- For the pharmaceuticals, steroids, and hormones, which often had detection percentages that fell below 50%, a nonparametric Kaplan-Meier approach was recommended for estimating the 95th percentile. The low detection percentages resulted in less stable and defensible percentile estimates from parametric-based approaches, and goodness-of-fit test outcomes were less certain due to limited detected data and non-consistent across the analytes. This is in accord with ProUCL recommendations, where nonparametric techniques are recommended when detection percentages are low.

While the sample data occasionally contained large measurement values for selected analytes, evidence was insufficient to warrant excluding these measurements from the analysis. In addition, outliers were not clustered among one or more facilities, nor were outliers flagged with data qualifiers in the survey database which would have suggested invalidity. However, it is appropriate to assess how the presence of large values may impact the estimates by performing the analysis both with and without outliers.

5. References

Cohen, A. C. Jr. 1950. *Estimating the Mean and Variance of Normal Populations from Singly Truncated and Double Truncated Samples*. Ann. Math. Statist., Vol. 21, pp. 557-569.

Cohen, A. C. Jr. 1959. *Simplified Estimators for the Normal Distribution When Samples Are Singly Censored or Truncated*. Technometrics, Vol. 1, No. 3, pp. 217-237.

Conover W.J. 1999. Practical Nonparametric Statistics, 3rd Edition, John Wiley & Sons, New York.

CSC, 2007. CSC's Sampling and Analysis Report for the 2006–2007 Targeted National Sewage Sludge Survey. Technical report prepared by CSC Environmental Solutions to Rick Stevens, Office of Water, U.S. Environmental Protection Agency. 30 September 2007.

D'Agostino, R.B. and Stephens, M.A. 1986. *Goodness-of-Fit Techniques*. Marcel Dekker, Inc.

Dixon, W.J. 1953. Processing data for outliers. *Biometrics*. Vol. 9, No. 1, pp. 74-89.

Dudewicz, E.D. and Misra, S.N. 1988. *Modern Mathematical Statistics*. John Wiley, New York.

Gilbert, R.O. 1987. *Statistical Methods for Environmental Pollution Monitoring*. Van Nostrand Reinhold, New York.

Gilliom, R. H. and Helsel, D.R. 1986. *Estimations of Distributional Parameters for Censored Trace Level Water Quality Data 1. Estimation Techniques*. Water Resources Research, 22, pp. 135-146.

Helsel, D.R. 2005. *Nondetects and Data Analysis*. Statistics for Censored Environmental Data. John Wiley and Sons, NY.

Helsel, D.R. 1990. *Less Than Obvious, Statistical Treatment of Data Below the Detection Limit*. ES&T Features Environmental Sci. Technol., Vol. 24, No. 12, pp. 1767-1774.

Kaplan, E.L. and Meier, O. 1958. *Nonparametric Estimation from Incomplete Observations*. Journal of the American Statistical Association, Vol. 53, pp. 457-481.

NRC, 2002. *Biosolids Applied to Land: Advancing Standards and Practices.* Report by the National Research Council. Washington, DC: National Academy Press. July 2002.

Stephens, M. A. 1970. *Use of Kolmogorov-Smirnov, Cramer-von Mises and Related Statistics Without Extensive Tables*. Journal of Royal Statistical Society, Vol. B 32, pp. 115-122.

USEPA. 2009a. Targeted National Sewage Sludge Survey: Statistical Analysis Report. Washington, DC: Office of Water, U.S. Environmental Protection Agency. EPA-822-R-08-018.

USEPA. 2009b. Targeted National Sewage Sludge Survey: Sampling and Analysis Technical Report. Washington, DC: Office of Water, U.S. Environmental Protection Agency. EPA-822-R-08-016.