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CHAPTER 5

AGENT ORANGE AND ITS DIOXIN CONTAMINATION

Alvin L. Young

Much of the concern over the widespread military use of tactical herbicides in South Vietnam, especially the use of Agent Orange, stemmed from the dioxin (2,3,7,8-tetrachlorodibenzo-*p*-dioxin, TCDD) contaminant in the 2,4,5-T herbicide. Our awareness of its toxicity, persistence in biological tissue, and environmental fate now spans at least 35 years. In that span of time, thousands of articles have been published on TCDD making it not only a chemical of intense regulatory interest but also one of the most researched molecules worldwide. Various agencies of the United States government have spent hundreds of millions of dollars on this tiny molecule, including expenditures by the Environmental Protection Agency (EPA), the National Institute of Environmental Health Sciences (NIEHS), the Centers for Disease Control and Prevention (CDC), the National Institute of Occupational Safety and Health (NIOSH) in conducting and monitoring superfund sites, in the support of regulation of pesticides (as contaminants), in studying and monitoring emissions from incineration systems, and on the conduct of numerous epidemiological studies of industrial populations. The Department of Defense (DOD) has expended hundreds of million dollars in the conduct of the Air Force Health Study, on the disposal of Agent Orange (Operation PACER HO), and on the numerous remediation and environmental monitoring programs conducted at the former sites where Agent Orange was stored in Mississippi and Johnston Island (see **Chapters 4,6,7**).

These expenditures by the above Agencies will however be dwarfed by the costs that will eventually be incurred by the United States Department of Veterans Affairs (DVA) on the Congressionally-mandated Agent Orange Act of 1991. This act established procedures that the DVA must follow in deciding whether to create new presumptions of service connection for disabilities suffered by Vietnam veterans that may be associated with exposure to Agent Orange and other herbicides in Vietnam [IOM 1994]. For the DVA, the determination of whether a disease (currently eleven) should be service connected is not based on determination of causation or proof of exposure, nor is it based on studies of veterans who served in Vietnam. Rather, it is based on whether the evidence, as judged following periodic reviews of the scientific literature by the National Academy of Sciences' Institute of Medicine, is sufficient to conclude there is a positive association [IOM 1994]. In making the final decision on whether an association exists, the Secretary of DVA must apply the standard, as mandated by Congress and the courts that any resolution of doubt favor the Vietnam veteran [Young 2002].

Vietnam, Agent Orange, and its associated dioxin are intense societal, emotional, legal, and public policy issues as much as medical and scientific issues – perhaps more so. There are strong societal concerns and public policies favoring our veterans, and rightly so. But our scientific principles ought not favor or disfavor anyone. However, as scientists, we cannot ignore the societal, emotional, or legal issues influencing public policies, because in today's environment those policies shape the research agenda (and hence funding), and if we are not careful, may affect even the research results.

Thus, it is appropriate to ask the question “How did we get ourselves into this situation?” This Chapter explores the history of Agent Orange and its dioxin contaminant and the conflict that exists between science and social concern.

5.1 Formation of the TCDD Contaminant

Polychlorodibenzo-*p*-dioxins may be contaminants of any of the chemical products that use chlorophenols in the manufacturing process [Young 1980]. The presence of 2,3,7,8-TCDD (and its concentration) was dependent upon the industrial process used in the manufacture of the basic chlorophenol, in this case, the production of sodium 2,4,5-trichlorophenate. The most common industrial process that was used for the production of 2,4,5-T herbicide is shown in **Figure 5.1**.

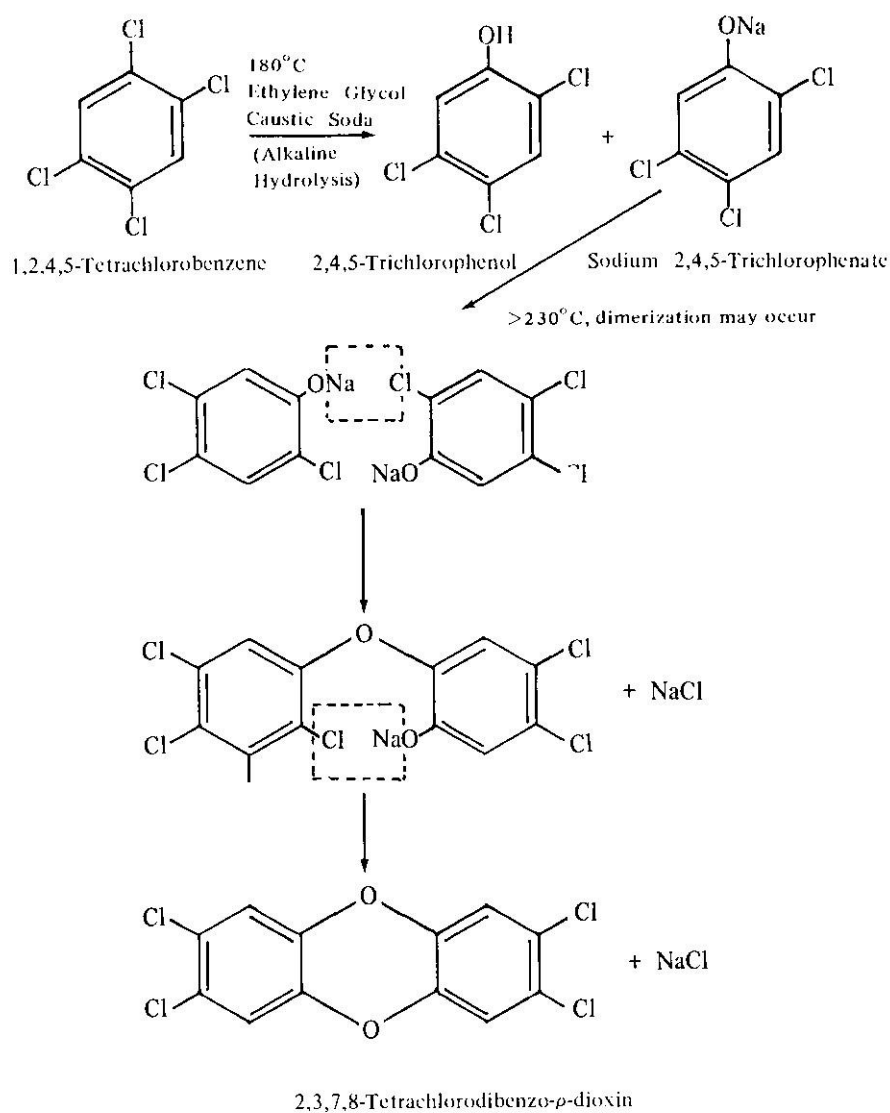


Figure 5.1 Formation of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin by the alkaline hydrolysis and subsequent dimerization of sodium 2,4,5-trichlorophenate [Young 1980]

The process of making 2,4,5-trichlorophenate by the hydrolysis of hexachlorobenzene was a process developed by Dow Chemical Company and was known as the 'Dow Process' [Plimmer 1973]. The reaction temperatures during the Dow Process had to be carefully maintained. If the temperature of the hydrolysate rose above the normal 180° C, an exothermic reaction occurred after any residual solvent, e.g., glycol, was removed by distillation. This reaction, attributed to the decomposition of sodium-2-hydroxethoxide, started at a temperature of 230° C and continued to 410° C. The heat generated by this reaction assisted in the formation of TCDD through the dimerization of two molecules of sodium trichlorophenate [Plimmer 1973; Young 1980]. The rapid temperature increase in the reaction vessel, results in a pressure increase; failure to release this pressure resulted in the Seveso accident of 1976 [Reggiani 1988]. In this case, the dimerization resulted in a 1% yield of 2,3,7,8-TCDD [Reggiani 1988].

The capability for accurately assessing the levels of TCDD in herbicide formulations did not exist in the years during the use of Agent Orange in Vietnam. Furthermore, no mention was made in the early scientific literature that dioxins might occur as contaminants in the commercial chlorinated phenols until 1959 [Julia and Baillarge 1959]. In 1962, the first description of the acnegenic potency of TCDD was published in the article: *A technique for testing acnegenic potency in rabbits, applied to the potent acnegen 2,3,7,8-tetrachlorodibenzo-p-dioxin* [Jones and Krizek 1962]. It is interesting to note that much controversy surrounded the preparation, properties, and identification of high purity samples of 2,3,7,8-TCDD that could be used as standards for analytical studies. The National Cancer Institute (NCI) took the lead in preparing purified standards, conducting studies of the chemistry, and searching for possible sources for human exposure. The NCI 4-volume publication "*Evaluation of the Carcinogenic, Tetragenic, and Mutagenic Activities of Selected Pesticides and Industrial Chemicals*" was released in 1967 [NCI 1967].

Dr. Warren Crummett (a Dow Analytical Chemist) reported that the analytical limit of detection of 2,3,7,8-TCDD in herbicide formulations was 1 ppm in 1965, but the procedure for doing this analysis required rigorous cleanup and purification of the analyte, often using the rabbit ear test to validate TCDD [Crummett 2002]. It was not until late 1969 that 2,3,7,8-TCDD could be positively identified and quantified in herbicide formulations by the use of gas chromatography-mass spectroscopy [Crummett 2002]. Subsequently in 1970, they were able to confirm levels of 0.050 ppm with some consistency. However, it was not until 1974 that Dow Analytical Services was able to detect 0.001 ppm in ester, amine, and acid formulations of 2,4,5-T herbicide [Crummett 2002]. Buser of Switzerland reported the same level of detection that same year, indicating the global search that was occurring for methods of detection of the dioxins and furans [Buser 1974]. By 1975, the ability to detect TCDD in biological tissues and in other environmental samples had reached the limit of 10 parts-per-trillion, ppt (picograms/gram), but the cost of doing one analysis exceeded \$1000/sample [Young 1980]. Today the capability to detect 0.01 ppt of various dioxins and furans is common. However, as Crummett noted:

"Chemists seeking to measure small numbers of molecules will continue to develop more sensitive and specific instrumentation for doing so. Eventually, they will reach the ultimate limit – single molecule detection. And what will that mean in a practical sense? Nothing, of course! But as a point of interest, it may mean that at least one molecule of every substance that has ever existed in nature will be present in a glass of drinking water" [Crummett 2002].

In 1972, the United States Agricultural Research Service published data on the analysis of additional samples of 2,4,5-T herbicide [Woolson et al 1972]. Of 42 samples of 2,4,5-T, 22 samples

contained less than 0.5 ppm TCDD. Of the 20 samples containing more than 0.5 ppm of TCDD, 15 were obtained for the yearly survey of one manufacturer. The samples were from 1966 – 1970, with four samples usually collected each year. There was a 10-fold drop in TCDD content by this manufacturer between 1968 and 1969. However, their technical grade 2,4,5-T still contained 2 – 3 ppm of TCDD in 1970. The 1970 technical samples from another manufacturer contained less than 0.5 ppm [Woolson et al 1972]. Technical grade 2,4,5-T manufactured as a formulation for use in agricultural products, typically contained 90 – 92% 2,4,5-T (as the acid) and 8 – 10 % impurities; suggesting that the sample noted above, when made into a commercial form of 2,4,5-T herbicide, probably contained between 1 and 1.5 ppm TCDD [Bovey 1980]. Edmunds, Lee, and Nickels [1973] subsequently reported on 55 samples of butyl and octyl esters of 2,4,5-T from lots manufactured in the late 1960s and early 1970s. The mean concentration of TCDD in the 55 samples was 0.31 ppm.

In 1971, in a report on 2,4,5-T prepared by the President’s Science Advisory Committee [MacLoed 1971] obtained data on TCDD levels in technical grade 2,4,5-T from one manufacturer for samples analyzed from 1958 to 1969. These data were provided in **Table 5.1**.

Table 5.1 The history of TCDD concentrations, ppm, in technical grade 2,4,5-T acid manufactured by one company [MacLoed 1971]

Parts-per-Million, ppm, 2,3,7,8-TCDD in Technical Grade 2,4,5-T Manufactured by One Company	
1958	11
1959	11
1960	8
1961	5
1962	10
1963	11
1964	12
1965	5—32
1966	3—18
1967	1—25
1968	1—25
1969	<1

The data in **Table 5.1** represented only one of the seven major companies that produced 2,4,5-T for use in formulating Agent Orange. All seven companies simultaneously provided 2,4,5-T commercial formulations for US and international agriculture [WSSA]. Both the demand for 2,4,5-T for military and commercial uses increased during the period 1965-1970, at the same time that improvements were occurring in the industrial process and in the analytical methodology for the detection of the TCDD in the herbicide formulations.

5.2 Establishing Agent Orange and Its Contaminant as a Major Public Health Issue

The relationship between TCDD and Agent Orange first became a matter of public concern in the

fall of 1969 when the results of a study commissioned by the National Institutes of Health to the Bionetics Research Laboratories of Bethesda, Maryland, became known [Bionetics 1968; Reggiani 1988]. A portion of that report described the teratogenicity of 2,4,5-T in laboratory mice was subsequently published in Science Magazine in 1970 [Courtney et al 1970]. However, in June 1969 reports that Herbicide Orange had produced birth defects in humans had already appeared in Vietnamese newspapers [MacLeod 1971]. A subsequent analysis of the 2,4,5-T used by Bionetics revealed that the cause of the toxicity was the TCDD contaminant and that 2,4,5-T in itself was not teratogenic [Reggiani 1988; Crummett 2002]. The members of the scientific community that had been asked to examine the Bionetics data and the reports coming out of Vietnam concluded that the use of 2,4,5-T represented a potential risk to human health that outweighed the benefits of its use domestically, or by the Department of Defense in Vietnam [Nelson 1969; DuBridgde 1970].

In 1970, the United States Congress directed the Secretary of Defense to request that the National Academy of Sciences (NAS) conduct studies assessing the ecological and biological impact of the military use of herbicides in Vietnam [NRC, 1974]. A committee of the National Research Council (NRC) published their report “*The Effects of Herbicides in South Vietnam*” in February 1974 [NRC 1974]. Due to insufficient data it had not been possible to assess the potential impact of TCDD in Vietnam. For the next decade, studies funded by the US Federal Government and other governments on the toxicity, sources, environmental fate, and human risks of TCDD were debated and published in hundreds of forums [Young and Reggiani 1988; IOM 1994; Young 2002]. The public was bombarded by stories of the horror of dioxin (see **Figure 1**).



Figure 5.1 A cartoon criticizing the US Forest Service for its continued use of 2,4,5-T herbicide. Published by *The Daily Utah Chronicle*; Thursday, May 20, 1976, page 3

A large volume of toxicological data on 2,4,5-T and 2,4-D were available during the final years of US involvement in Vietnam, but woefully inadequate toxicological and environmental data on TCDD. Although scientists in 1969 had recognized that TCDD as acutely toxic and teratogenic (birth deforming) in laboratory animals, no studies were available on the effects of chronic, long-term, low-level exposures in lower mammalian species. Furthermore, numerous occupational exposures to TCDD were reported during the industrial production, but epidemiological studies were not available despite documented exposures as early as 1949 [Young et al 1978].

Reggiani [Reggiani 1988] described how the issue of TCDD and Agent Orange was refocused for the public:

“In 1978, with the help of a reporter from the Columbia Broadcasting system, Bill Kurtis, the issue of Agent Orange and its potential effects on human health was presented to the nation in a television documentary entitled “Agent Orange: Vietnam’s Deadly Fog”. In this way the public became aware of the magnitude of the veteran’s concern, and Agent Orange reached the dimensions of a public health problem. Thus, the public turned its attention to the scientific and policy decisions the government had taken or intended to take regarding this matter”.

In response to the documentary and numerous inquiries from Vietnam veterans, the United States Air Force’s Occupational and Environmental Health Laboratory (OEHL), Brooks Air Force Base, Texas published as a technical report a comprehensive review of **“The Toxicology, Environmental Fate, and Human Risk of Herbicide Orange and Its Associated Dioxin”** [Young et al 1978]. The significance of the document was that both historical and scientific analyses were available in a single publication on the tactical herbicides used in Vietnam and the dioxin contaminant. Of particular value was the assessment of how much herbicide had been procured and disseminated in Vietnam, and how much of the TCDD contaminant had likely been disseminated with the 2,4,5-T herbicide.

Once the public was alerted to the controversy surrounding Agent Orange, it was only a matter of time before the US Congress and the Executive Office of the President expressed interest in taking action. Indeed, the importance to the Federal Government in resolving veteran health issues and addressing the potential risks of dioxin were demonstrated in December 11, 1979, when the Executive Office of the President (President Jimmy Carter) directed the establishment of an **“Interagency Work Group to Study the Possible Long-Term Health Effects of Phenoxy Herbicides and Contaminants”** [Eizenstat 1980]. Members of Interagency Work Group (IWG) included representatives from the Departments of Agriculture, Defense, Health and Human Services, Housing and Urban Development, and Labor, and representatives from the Environmental Protection Agency, Veterans Affairs, Office of Management and Budget, Council of Economic Advisors, and Office of Science and Technology Policy. In August 1981, the IWG was expanded and elevated to become the **Agent Orange Working Group (AOWG)** at the Cabinet Council level by President Ronald Reagan. The task assigned to the AOWG was...*“to guide and monitor all Federal research into the possible adverse health effects of Agent Orange and similar chemicals on humans, with a particular focus on the health of Vietnam veterans”* [Bowen 1988]. Secretary of Health and Human Services was appointed Chair of the AOWG, and the Director of the Centers for Disease Control was appointed Chair of the AOWG Science Panel. The Congressional Office of Technology Assessment and the General Accounting Office were invited to become observers and advisors to the Group.

The AOWG undertook a massive research effort encouraging, supporting, and monitoring studies conducted by VA, DOD (the Air Force Health Study of RANCH HAND personnel), the Centers for Disease Control and Prevention (CDC), other Federal Agencies, and the international community (e.g., Australia and New Zealand) [Davis 1983]. Subcommittees were formed to examine the use of TCDD as a bio-indicator of exposure to Agent Orange [Rall 1981], and the Science Panel of the AOWG undertook a comprehensive assessment of the feasibility of

conducting the major study of ground troops [Beach 1984].

The major issue facing all of the government-supported research was the confirmation of exposure to the phenoxy herbicides and the associated TCDD. The 1988 study released by the CDC compared levels of serum TCDD in 646 US Army veterans who served as ground troops in the most heavily sprayed regions of Vietnam with those of 97 Vietnam-era veterans who had not served in Vietnam [CDC 1988]. The distribution of TCDD levels were ‘nearly identical’ in the two groups, both having means and medians of about 4 ppt, which was well within the range of background at that time [CDC 1988]. The CDC concluded that neither military and spraying records nor self-reported history of exposure could reliably identify high or low exposure groups, and “*most US Army ground troops who served in Vietnam were not heavily exposed to TCDD, except perhaps men whose jobs involved handling herbicides*” [CDC 1988]. These results were consistent with other studies and so clear cut that a planned epidemiological study of ground troops and Agent Orange was discontinued as infeasible [Young 2004].

Subsequent publications by Buckingham [1982], Cecil [1986], and Young and Reggiani [1988] provided more insight into the details of Operation RANCH HAND. Publications by Westing [Westing, 1976, Westing, 1984] provided appraisals of the ecological impact of the use of herbicides in Southeast Asia. All of these publications became the primary sources of information on Agent Orange and RANCH HAND for the National Academy of Sciences’ Institute of Medicine’s publication in the 1994 on “*Veterans and Agent Orange: Health Effects of Herbicides Used in Vietnam*” [IOM 1994].

Disregarding the eight years of research conducted by the Federal Agencies in the United States, and the conclusion based upon that science, the Congress moved to find a “political solution” to Agent Orange [Hanson 1987]. The Congressionally-mandated (and signed by the President) Agent Orange Act of 1991, Public Law 102-4, established procedures that the Department of Veterans Affairs (DVA) must follow in deciding presumptive compensation; that is, whether to create new presumptions of service connection for disabilities suffered by Vietnam veterans that may be associated with exposure to Agent Orange and other herbicides in Vietnam. The procedures required that the DVA contract with the National Academy of Sciences’ Institute of Medicine to conduct reviews every two years of the scientific literature on the health effects of herbicides and TCDD [IOM 1994]. In response to the DVA, the IOM noted in its first report:

“Controversy has surrounded the study of Agent Orange since the first questions of herbicide-related health effects in Vietnam veterans were raised more than 20 years ago. In the course of its work, the Committee heard allegations of scientific misconduct and claims of government conspiracy to suppress information on health effects, as well as serious disagreements among scientists about the interpretation of laboratory and clinical data. The Committee was not charged with investigating or resolving these controversies, and it did not attempt to do so... Although the conclusions and recommendations presented here will not end the controversy surrounding this issue, it is the Committee’s hope that this report will crystallize the current scientific information on this important topic and prompt further research to answer the remaining questions being asked by veterans and their families, the Department of Veterans Affairs, and Congress” [IOM 1994].

The Academy's Institute of Medicine has now issued seven comprehensive reports [IOM 1994; IOM 1996; IOM 1998; IOM 2000; IOM 2002; IOM 2004; IOM 2006]. In accordance with their findings, DVA has prepared a list of conditions that are presumed to be service connected based on herbicide and/or TCDD exposure [DVA 2007]. The issue of whether a veteran was actually exposed to Agent Orange, and presumably dioxin, is irrelevant for establishing presumption. For any veteran who served in Vietnam between 9 January 1962 and 7 May 1975, and has one of the eleven diseases on that list, DVA must presume that they were exposed to herbicides (and associated TCDD) and their disease is service connected [DVA 2007].

5.3 Composition of Agent Orange and Associated Contaminants

In order to determine the quantity of TCDD that may have been present in the 2,4,5-T containing tactical herbicides, data on the TCDD contamination of the 2,4,5-T stocks used in formulating Agent Orange must first be gathered. Orange Herbicide was procured from numerous chemical companies. The USAF procured Orange under Purchase Description AFPID 6840-1, dated 23 February 1968, and Amendment 1, dated 11 April 1968. The Orange Purchase Description containing the changes and additions of Amendment I was published in the Final Environmental Statement on the "Disposition of Orange Herbicide by Incineration [Department of Air Force, 1974]. Since the most recent purchase description for "Herbicide Orange" was dated 11 April 1968, no reference was made of the TCDD contaminant. **Table 5.2** was the procurement specification for Herbicide Orange.

Table 5.2 The Military Procurement Specification for Herbicide Orange [Department of the Air Force 1974]

-
1. SCOPE: This purchase description prescribes requirements for a herbicide identified as Orange. The material is used as a systemic growth regulator to kill and defoliate vegetation.
 2. APPLICABLE DOCUMENTS:
 - PPP-D-729, Drums: Metal 55-gallon, for shipment of non-corrosive material.
 - MIL-H-51148, Herbicide N-Butyl 2,4,5-Trichlorophenoxyacetate.
 - MIL-H-51147, Herbicide N-Butyl 2,4-Dichlorophenoxyacetate.
 - MIL-STD-105, Sampling Procedure and Tables for Inspection of Attributes
 - MIL-I-45208, Inspection Systems Requirements
 3. REQUIREMENTS
 - 3.1 Materials. The herbicide shall be composed of the following two ingredient materials.
 - a. N-Butyl 2,4,5-Trichlorophenoxyacetate
 - b. N-Butyl 2,4-Dichlorophenoxyacetate
 - 3.1.1 The ingredient materials shall meet the following specifications:
 - a. Specification MIL-H-51148, N-Butyl 2,4,5-Trichlorophenoxy-acetate, except free acid will be 0.5% by weight.
 - b. Specification MIL-H-51147, N-Butyl 2,4-Dichlorophenoxyacetate except composition (purity) shall be 98% minimum by weight, acid equivalent shall not be less than 79.0% nor more than 80.0% and free acid shall be 0.5% maximum by weight.
 - 3.2 Finished Mixture (Orange)
 - 3.2.1 Composition
 - a. 50% by volume N-Butyl 2,4,5-Trichlorophenoxyacetate
 - b. 50% by volume N-Butyl 2,4-Dichlorophenoxyacetate

- 3.2.2 Tolerance. Tolerance range for amount of each composition ingredient contained in the final mixture will be $\pm 1.5\%$ including the precision allowance for the analytical method used.
- a. Range for N-Butyl 2,4,5-Trichlorophenoxyacetate is 48.5 to 51.5% by volume.
 - b. Range for N-Butyl 2,4-Dichlorophenoxyacetate is 48.5 to 51.5% by volume.
- 3.2.3 Free Acid. A maximum of 0.5% by weight.
- 3.2.4 Total Acid Equivalent (as 2,4-D Acid)
- 90.0% minimum by weight.
 - 94.0% maximum by weight.
- 3.2.5 Specific Gravity.
- 1.275 to 1.295 at 20° / 20° C
- 3.2.5 Color. A clear reddish brown color.
- 3.2.6 Weight per Gallon.
- 10.70 \pm 0.08 lbs at 20° C (55 gallons will weigh 584.10 to 592.90 pounds on a 20° C basis).
-

As part of the sampling protocol for TCDD analyses, the Air Force wanted to know how closely the military specifications had been met, and what other chemical compounds were present. **Table 5.3** provided a summary of the results from analyzing 12 randomly selected drums from one lot of drums that was part of the Agent Orange Inventory at the Naval Construction Battalion Center (NCBC), Gulfport, Mississippi. These samples were taken from Agent Orange produced by Dow Chemical Company (Dow Lot 10, or assigned number, ASN, 10). The data from these samples were presumably representative of the approximately 6,950 drums of Dow product in storage at NCBC in 1973. The average concentration of 2,3,7,8-TCDD in this lot was subsequently found to contain ≤ 0.05 ppm.

Table 5.3 A 1973 Characterization of Agent Orange Produced by Dow Chemical Company and in the Inventory at NCBC, Gulfport, MS [Fee et al 1975]

Average Concentration (Relative % by Weight) ¹	Compound
0.30	Butanol
0.10	Toluene
0.03	Xylenes, Ethylbenzene
0.05	Butyl Chloride
0.12	Dichlorophenol
0.57	Peak D
0.23	Trichlorophenol
0.16	Butoxydichlorobenzene
0.16	Butoxytrichlorobenzene
46.87	Butyl dichlorophenoxyacetate
44.62	Butyl trichlorophenoxyacetate
1.38	Butyl monochlorophenoxyacetate
2.68	Butyl methoxydichlorophenoxyacetate
0.42	Butyl (bis-dichlorophenoxy) acetate
0.29	Octyl dichlorophenoxyacetate ²
0.42	Octyl trichlorophenoxyacetate ²
0.33	1,1-dibutoxy-2-trichlorophenoxyethane
1.27	Unidentified compounds
100.00	Sum

¹ The data represent the average concentration found in 12 samples

² Tentative identification based solely on gas chromatographic retention time

As noted for **Table 5.2** and **Table 5.3**, at the time that Orange Herbicide was procured for use in South Vietnam, TCDD was **NOT** recognized as either a contaminant or as an issue of quality control. Moreover, it was not until recently that other dioxins or furans became of importance to the biomonitoring of human populations [Schecter et al 2003; Sexton, Needham, and Pirkle 2004]. In an analytical study of 83 samples of 2,4,5-T herbicide produced from 1968–1971, the only readily quantifiable dioxin in 2,4,5-T was the 2,3,7,8-TCDD (see **Table 5.4**). Some of the samples contained trace quantities of 1,2,3,7,8-PnCDD; 1,2,3,4,7,8-HxCDD; 1,2,3,6,7,8-HxCDD; and, 1,2,3,6,7,8,9-HpCDD [Young and Andrews 2006].

Table 5.4 The Dioxin Congeners in 83 Samples of 2,4,5-T Acid Produced by One Manufacturer from 1968-1971 [Young and Andrews 2006]

Dioxin Congeners (parts-per-million, ppm) in 2,4,5-T Acid			
Congener	Analyzed	No. Non-detected	Range of Positive Samples
2,3,7,8-TCDD	47	26	0.5- 0.8 µg/g
PnCDD	3	2	0.17
HxCDD	11	3	0.16 ± 0.10
HpCDD	11	10	0.20

OCDD	11	9	0.40 ± 0.11
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To confirm that 2,3,7,8-TCDD was the principal congener of interest in blood serum, a study was conducted on 9 individuals who routinely sprayed 2,4,5-T herbicide [Smith et al 1998]. **Table 5.5** provided the data on lipid-adjusted serum levels of dioxins and furans from the nine applicators and their matched controls.

Table 5.5 Levels of PCDD and PCDF Congeners in Lipid-Adjusted Serum of Nine 2,4,5-T Applicators and Nine Matched Controls [Smith et al 1992]

Average level, ppt ± SE, § Not Detected		
PCDD/PCDF Congener	2,4,5-T Applicators	Matched Controls
Dibenzodioxins		
TCDD	53.3 ± 16.1	5.6 ± 1.1
1,2,3,7,8-PnCDD	12.4 ± 1.1	8.8 ± 0.7
1,2,3,4,7,8-HxCDD	6.8 ± 0.5	5.7 ± 0.4
1,2,3,6,7,8-HxCDD	28.6 ± 5.1	23.3 ± 4.9
1,2,3,7,8,9-HxCDD	9.9 ± 0.9	8.2 ± 0.6
1,2,3,4,6,7,8-HpCDD	121.9 ± 28.5	119.4 ± 18.4
OCDD	788.6 ± 82.3	758.7 ± 92.8
Dibenzofurans		
2,3,7,8-TCDF	1.6 ± 0.3	1.7 ± 0.3
1,2,3,7,8-PnCDF	<2.1§ ± 0.2	<2.0§ ± 0.2
2,3,4,7,8-PnCDF	8.0 ± 0.9	7.4 ± 0.8
1,2,3,4,7,8-HxCDF	5.4 ± 0.3	5.1 ± 0.5
1,2,3,6,7,8-HxCDF	5.5 ± 0.4	5.6 ± 0.6
1,2,3,7,8,9-HxCDF	<0.8§ ± 0.1	<0.8§ ± 0.1
2,3,4,6,7,8-HxCDF	<1.1§ ± 0.4	<1.7§ ± 0.2
1,2,3,4,6,7,8-HpCDF	14.2 ± 0.7	16.0 ± 2.3
1,2,3,4,7,8,9-HpCDF	<1.6§ ± 0.1	<1.9§ ± 0.3

5.4 Estimates of Quantities of Tactical Herbicides Procured by the Defense Supply Agency

For years there have many estimates published on the quantities of tactical herbicides purchased by the Defense Supply Agency from the various chemical companies for use in South Vietnam from January 1962 through October 1971. Differences in quantities of tactical herbicides disseminated and areas treated in South Vietnam varied among individual sources [Collins 1967; Irish et al 1969; NRC 1974; Westing 1976; Young et al 1978; Stellman et al 2003]. The differences were attributable to varying assumptions about the quantity expended on each mission, the number of missions, the loss of herbicide during the de-drumming and re-drumming of the residues, and the amount of herbicide spilled on the tarmac and in storage sites in Vietnam. In addition, the estimates varied because of reliance upon data that may have been uncertain, incomplete, or based on differing underlying assumptions, e.g., various revised HERBS Tapes. Only recently has it become possible to search and obtain actual procurement data from various record repositories (National Archives, Washington DC; Air Force Logistic Command at Kelly AFB, Texas and

Wright-Patterson AFB, Ohio; Air Force Historical Research Center, Montgomery, Alabama; and from the 1982 Pre-Trial Statements in Re: “Agent Orange” Product Liability Litigation). The best estimates of procurement data for Agent Orange taken from the above sources and for the nine chemical companies contracted with the Defense Supply Agency to manufacture the tactical herbicide were assembled in **Table 5.6**.¹ To obtain the estimated number of gallons, multiply the number of drums times 55; to obtain the estimated number of liters, multiply the number of drums time 208

As noted in **Table 5.6**, the first production of Agent Orange was in Fiscal Years (FY) 1963. Fiscal Year 1963 was from 1 October 1962 through 31 September 1963. The Army Chemical Corps first used Orange in the tests and evaluations conducted in Texas and Puerto Rico beginning in March 1963 [Young 2006]. The Air Force first used Orange in the tests and evaluation of the MC-1 and A/A 45Y-1 Spray Systems at Eglin AFB, Florida in Fiscal Year 1964 (1 October 1963 – 31 September 1964) [Young 2006]. The first use of Orange Herbicide in South Vietnam was in March 1965 [Cecil 1986]. Monsanto Company provided the majority of Agent Orange for use in South Vietnam in FY 1965 (1 October 1964 – 31 September 1965), but after 1965 five companies provided Orange to the Air Force Logistics Command (AFLC) for shipment and use in South Vietnam. The “history” of use of the tactical herbicides was provided in **Table 5.7**. Certainly not all of the tactical herbicide produced by the various manufacturers went to Vietnam.

Table 5.6 Orange Productions and Shipment of Estimated Number of Drums from the Nine Chemical Companies with Contracts to the Defense Supply Agency, Fiscal Years 1963-1969

Company	1963	1964	1965	1966	1967	1968	1969	Number Drums
Dow			5,465	3,720	32,115	36,935		78,235
Monsanto	15	1,085	15,490	9,960	18,520	20,875	1,120	67,065
Hercules			6,005	12,885	14,505	16,550		49,945
Thompson-Hayward					5,875	15,180		21,055
Diamond Alkali				1,000	4,920	6,595	40	12,555
UniRoyal				1,635	8,180	1,820		11,635
Thompson					2,985	3,365	835	7,185
Aggrasit							1,875	1,875
Hoffman-Taff							410	410
Gallons	825	59,675	1,482,800	1,606,000	4,790,500	5,572,600	235,400	
Drums	15	1,085	26,960	29,200	87,100	101,320	4,280	249,960¹
Liters	3,120	225,680	5,607,680	6,073,600	18,116,800	21,074,560	890,240	

Table 5.7 History of the Use and Disposition of the Tactical Herbicides Procured by the Defense Supply Agency for the US Army Chemical Corps and the US Air Force Logistics Command

Estimated Number of Drums of Tactical Herbicides							
	Orange	White	Blue	Purple	Pink	Green	Blue¹
Total Procured	249,960	105,700	30,130	12,780²	2,025³	365	95
Test Programs							
Eglin	345	75	80	285			
Others ⁴	60	15	20	15	2		
Thailand				5	3		
Korea, 1968 ⁵	380		625				
Hurricane ⁶ Camille, 1968	75		170				
Disposal Options	180	810 ⁷					
NCBC	15,370				705 ³		
Johnston Island	25,220						
Vietnam	208,330	104,800	29,235	12,475	1,315	365	95

¹ The first “Blue” produced in 1961 and shipped to Vietnam was a powdered formulation. In subsequent tables, the 95 drums of Ansul 138® were added to the total of liquid Blue, Phytar 560G, to bring the total of Blue used in Vietnam to 29,330 drums

² Questions remain as to the total production of Purple, especially as to the source of Purple used at Eglin AFB, Florida. One source indicated that the Purple used at Eglin was manufactured in 1953-54 [Young and Newton 2004]

³ The tactical herbicide that was identified as “Pink” by AFLC [Craig 1975], was actually 2,4,5-T formulations remaining after AFLC terminated contracts in FY 1969 for the procurement of Orange II. The 705 drums were shipped to Kelly AFB, Texas awaiting final disposition

⁴ “Others” refer to the various test and evaluation programs conducted by the Army Chemical Corps throughout the United States, Puerto Rico and Canada [Young 2006]

⁵ In 1968, the Department of State requested that AFLC provide tactical herbicides for the control of vegetation adjacent to the Demilitarized Zone in Korea [Young 2006]

⁶ In 1968, Hurricane Camille destroyed 75 drums of Orange and 170 drums of Blue [Young 2006]

⁷ Approximately 810 drums from the final procurement of White Herbicide from Dow Chemical Company in 1971 were not shipped to South Vietnam, but directed by the Armed Forces Pest Management Board to be sold as “Tordon 101” for use by Base Civil Engineers [Craig 1975; Young 2006]

5.5 The Initial Analysis of Dioxin Contamination in the Agent Orange Inventory

5.5.1 Sampling the NCBC and Johnston Island Inventories of Agent Orange

The most controversial issue associated with Agent Orange has been the concentration of the “unacceptable levels of impurities.” The procurement specifications provided no information on potential impurities, including 2,3,7,8-TCDD. The Orange herbicide returned from South Vietnam to Johnston Island in 1972 (Operation PACER IVY) was stored until a decision was made by AFLC for its final disposition. However, before a decision could be made about the method of disposing of the Agent Orange, data on the level of the dioxin contamination was required. Because of the extraordinary toxicity of 2,3,7,8-TCDD and its association with 2,4,5-T herbicide, it was one of the earliest dioxin isomers available in sufficient quantity to be used as an analytical standard [Tiernan 1983]. Mass spectrometry was selected as the method for the detection and quantification of TCDD in Agent Orange [Hughes et al 1975].

In 1973, the USAF assigned responsibility for characterizing the dioxin concentrations in the Agent Orange Inventories to the Environmental Health Laboratory at Kelly AFB, Texas, and the Aerospace Research Laboratory at Wright-Patterson AFB, Ohio [Fee et al 1975; Department of Air Force 1974]. The Environmental Health Laboratory at Kelly AFB later became the Air Force Occupational and Environmental Health Laboratory (OEHL) and was located at Brooks AFB, Texas. OEHL was subsequently given the responsibility for conducting Operation PACER HO [Thomas et al 1978]. The analytical team at the Aerospace Research Laboratory that characterized Agent Orange and its associated dioxin subsequently became the Brehm Laboratory, part of the Department of Chemistry at Wright State University in Dayton, Ohio [OEHL 1977]. As an academic institution, it supported many of the analytical requirements for Operation PACER HO [Tiernan 1983].

Two different types of sampling procedures were used to obtain Agent Orange samples for characterization and analyses. Because of re-drumming operations in Operation PACER IVY, and the continual maintenance requirements, the Agent Orange Inventory at Johnston Island could not be separated into identifiable processing lots. Therefore, two hundred separate samples were collected to represent the entire population (of drums). It was assumed that these 200 samples were a random representative sample of the Johnston Island Agent Orange inventory [Department of Air Force 1974].

Figure 5.2 is a photograph of a Bioenvironmental Engineer from the Environmental Health Laboratory sampling an Agent Orange drum at Johnston Island in October 1973. The samples at Johnston Island were sent to the Analytical Services Laboratory of Dow Chemical Company in Midland, Michigan, for analyses of 2,3,7,8-TCDD [Department of Air Force 1974]. Dow Analytical Services was used because only a few analytical laboratories were capable of handling large numbers of samples of liquid herbicide for TCDD analyses in 1973 [Young 1980].

Unlike Johnston Island, the samples of Agent Orange taken at the Naval Construction Battalion Center could be grouped to represent concentrations of TCDD in stocks supplied by certain manufacturers [Department of the Air Force 1974]. Initially, six to twelve samples were taken to represent each manufacturer’s stocks (later more than 80 samples were taken to characterize the stocks).



Figure 5.2 Sampling drums of Agent Orange for Dioxin content, 1973, Johnston Island. A random sample of 200 drums was assumed to represent a population of 25,220 drums (Photograph courtesy of USAF OEHL, Brooks AFB, Texas)

Figure 5.3 is a photograph of a team of Bioenvironmental Engineers from the Environmental Health Laboratory inspecting the NCBC inventory. Note that the drums on the left have a number painted in white on the lids (in this case number 10, representing the stock manufactured by Dow Chemical Company). There were seven major stocks identified by both their TCN (Transportation Control Number) and the DSA (Defense Supply Agency) Contract Number at NCBC when the samples were collected in June 1973. The Air Force Aerospace Research Laboratory, Wright-Patterson AFB, and Wright State University's Department of Chemistry performed the analysis for dioxin (2,3,7,8-TCDD), and for characterizing the herbicide at NCBC [Fee et al 1975; Hughes et al 1975; OEHL 1977]. **Table 5.8** provided a list of the manufacturers, the TCN Number, the DSA Contract Number, and the approximate quantities of 208-liter drums in the Agent Orange Inventory at the Naval Construction Battalion Center in 1973.



Figure 5.3 Inspection of the Agent Orange Inventory at NCBC in 1975
(Photograph courtesy of USAF OEHL, Brooks AFB, Texas)

Table 5.8 Manufacturers of Agent Orange Identified by TCN and DSA Numbers, and the Number of 208-liter Drums for each Stock or Lot at the Naval Construction Battalion Center, 1973 [Department of the Air Force 1974; OEHL 1977]

Manufacturer	TCN ¹	DSA Contract No.	Number of Drums
Dow	8155-X052CXX	400-68-C-6163	6,949
Diamond Shamrock	8156-0001AA	400-68-C-5898	507
Hercules	8192-0001	400-68-C-6093	2,734
Monsanto	8183-X002	400-68-C-6607	2,138
Thompson Chemical	8155-X012	400-68-C-6250	468
Thompson-Hayward	8155-X032XX	400-68-C-6166	1,560
Monsanto	7163-X001XX	400-67-C-9087	724
Unknown	718-X011XX	Unknown	138
Unknown	8066-X031XX	Unknown	69
<i>Total</i>			<i>15,287²</i>

¹Each of the TCN had prefix FY-9463, except Diamond Shamrock Co. (FY-9461), and Hercules, Inc (FY-9464)

²The total number of drums at NCBC fluctuated over time due to drums being received from Eglin AFB, Florida and Kelly AFB, Texas, and drums removed for disposal option studies

5.5.2 Air Force Results of Johnston Island Analyses for Dioxin

When the 200 samples were collected in 1973, the Agent Orange Inventory on Johnston Island was estimated to be 26,689 208-liter drums. This number was incorrect because in Project PACER IVY the actual number of drums was determined to be 25,220; in PACER HO 24,795 drums were emptied, and it was estimated that the remaining 425 drums had leaked or were spilled in the coral of the storage area. The arithmetic mean value for TCDD concentration was 1.909 mg/kg (ppm). Based on the estimated inventory of 26,689 drums, the total TCDD in the Orange stocks at

Johnston Island was estimated to be 13.63 kg [Department of Air Force 1974].

The TCDD concentrations in the 200 samples were not normally distributed. Of the 200 samples, 153 or 76.5% contained TCDD concentrations of 1.0 mg/kg (1.0 ppm) or less. Of the 200 samples, 195 or 97.5% had TCDD concentrations of 10.0 mg/kg (10.0 ppm) or less. Five samples (2.5%) had TCDD concentrations larger than 10.0 mg/kg (10.0 ppm). Those five samples had values of 13, 17, 22, 33, and 47 mg/kg. The “outliers” were included in computing the arithmetic mean of 1.909 mg/kg (1.91 ppm) [Department of the Air Force 1974].

5.5.3 Results of the Naval Construction Battalion Center Analyses

Table 5.9 was a compilation of the results of the TCDD analyses of the seven major manufacturer’s Herbicide Orange stock at the NCBC Gulfport. The number of drums (15,326) was obtained from the inventory at the time of the sampling in 1973.

The arithmetic mean concentration of TCDD in the NCBC inventory was calculated by summing the cumulative concentration of TCDD, and dividing by the sum of the number of kg of Agent Orange (7,265,980 mg divided by 4,100,226 Kg). By this method, the average concentration of TCDD in the Agent Orange Inventory at NCBC was 1.772 mg/kg or 1.77 ppm [Department of Air Force 1974]. When the samples were collected in 1973, the total Air Force inventory of Agent Orange at NCBC and Johnston Island was estimated at 42,015 208-liter drums or approximately 8.5 million liters. The weighted average concentration was 1.859mg/kg or 1.86 ppm. The total amount of TCDD in the entire USAF inventory at NCBC and Johnston Island was estimated to be 20.1 kg [Department of Air Force 1974].

Table 5.9 TCDD Analyses of Stock at the Naval Construction Battalion Center [Fee et al 1975]

Number of Drums	Kg Agent Orange	PPM TCDD	Mg of TCDD	Cumulative Mg TCDD
2,652	709,500	0.05	35,475	35,475
6,981	1,867,655	0.12	224,119	259,594
934	249,877	0.17	42,479	302,073
1,560	417,353	0.32	133,557	435,630
2,185	584,562	7.62	4,454,360	4,889,990
984	263,253	8.62	2,269,244	7,159,234
30	8,026	13.30	106,746	7,265,980
Total 15,326¹	4,100,226		7,265,980	7,265,980

¹ Represented 98% of the total NCBC stock in 1973 at time of sampling

Based upon the above data, the Occupational and Environmental Health Laboratory (OEHL) estimated that 167 kg of TCDD may have been in the 2,4,5-T-containing tactical herbicides used in South Vietnam [Young et al 1978]. In 1974, the National Research Council had estimated the total amount of TCDD disseminated in Vietnam to be between 106 – 163 kg [NRC, 1974]. The Columbia University group estimated 366 kg [Stellman et al 2003]. In July 2003, Dwernychuk of Hatfield Consultants, Ltd., stated: “*The equivalent of about 600 kg of pure TCDD was sprayed and spilled in Vietnam during the war*” [Hileman 2003].

5.6 A Re-analysis of TCDD In Agent Orange Stocks

5.6.1 A Re-evaluation of the NCBC and Johnston Island Agent Orange Inventories

In 1977, in preparation for Operation PACER HO, there were questions raised on the analyses of some of the stocks in the NCBC Inventory. The Project Director for Monitoring Programs, Major James Tremblay of the USAF Environmental Health Laboratory, requested clarification of data in Volume II of Technical Report ARL TR-75-0110 [Hughes et al 1974]. Dr. Michael Taylor, Research Associate Professor at Wright State University, wrote the following response in a 9 March 1977 letter:

“Dear Maj. Tremblay:

In response to your inquiries made by telephone concerning the concentrations of TCDD in samples of Herbicide Orange reported in ARL-75-0110, we offer the following information in order to confirm and to supplement our telephone conversations of 8 March 1977.

Regarding the raw data included in the technical report as Appendix F, we must underscore the fact that this was raw data and therefore was interpreted by the analyst before a final value for each determination was reported. As we discussed in our telephone conversation, the raw data in Appendix F had not been corrected for such factors as carry-over from the analysis of a sample or standard to the analysis of the succeeding sample. In addition, variations in various operating parameters bring about shifts in retention time of the TCDD peak and changes instrument response and other subtleties in the raw data, all of which must be taken into consideration by the analyst at the time of data reduction. These considerations make interpretation of the raw data a task that can be properly addressed only by analysts with first-hand knowledge of the analytical instrumentation and circumstances prevailing during the actual analysis.

Concerning the TCDD concentration in the samples from ASN #s 5, 8, 10, and 14, we have tabulated the data on a barrel-by-barrel basis and the tabulated data are enclosed. It must be noted that the TCDD included in Volume II of ARL TR-75-0110 (page 5) are as is stated on Page 4 of the report “TCDD levels reported earlier by Dow Chemical”(Analytical Services Laboratory). Based on the eighty analyses that we have performed, we have determined the average concentration of TCDD in the Dow ASN 10 Herbicide Orange is 0.25 µg TCDD/g Herbicide Orange or 0.25 ppm. The average TCDD concentration in the Thompson ASN 5 Herbicide Orange, based on our analyses, is 0.13 µg/g or 0.13 ppm”.

The attachment provided new data on the analyses of manufacturers of Agent Orange that differed from those in **Table 5.9**. The new data are presented in **Table 5.10**.

Table 5.10 Revised Estimates of TCDD Concentration by Manufacturer

ANS Number	Manufacturer	TCN Number	Number of Samples Analyzed	Estimated TCDD Concentration (ppm)
5	Thompson	FY-9463-8155-X012	60	0.13 ppm
8	Hercules	FY-9464-8156-0001	57	≤ 0.02 ppm
10	Dow	FY-9463-8155-X052	80	0.25 ppm
14	Hercules	FY-9464-8192-001	52	≤ 0.02 ppm

Source: Information provided to Major James Tremblay in a 9 March 1977 letter from Dr. Michael Taylor clarifying data in Volume II of Technical Report ARL TR-75-0110

5.6.2 Statistical Methodology for Air Force Data

Historical records indicated that the Air Force had collected 525 samples from the Agent Orange inventories stored on Johnston Island in the Central Pacific Ocean and the Naval Construction Battalion Center (NCBC) in Gulfport, Mississippi, prior to incineration at sea. TCDD concentrations were determined for each sample. It was recognized that the samples were likely to have been most representative of the herbicide in use after 1967 because stocks shipped to Vietnam earlier were probably disseminated to support military operations.

Because the distributions were positively skewed (see **Figure 5.4**), bootstrapping techniques were used to obtain a reliable estimate of the upper 95th percentile mean value of TCDD in the stockpiles, corresponding to an estimate of the upper 95 percent confidence interval of the mean concentration of TCDD in the stockpiles. Bootstrapping was a way of creating pseudo-replicate datasets by randomly re-sampling the original data for statistical analysis. A total of 5,000 pseudo-replicate datasets with 525 observations each were randomly generated from the 525 observations of the Johnson Island and NCBC combined dataset by re-sampling these data with replacement. Sample concentration values that were below the detection limit of the quantification technology were replaced with a value equal to ½ of the detection limit. This resulted in a total of 2,625,000 randomly selected total observations for the 5,000 pseudo-replicate datasets. The mean TCDD concentration for each of the 5,000, 525 observation datasets was determined. The 95th percentile mean TCDD concentration of the 5,000 datasets was calculated and used to estimate the quantity of TCDD dispensed in Vietnam.

5.6.3 Results for NCBC and Johnston Island Agent Orange Inventories

The 95th percentile value for the arithmetic means of the Johnston Island inventory was 2.46 ppm TCDD, while the 95th percentile of the arithmetic means of the NCBC inventory was 1.75 ppm TCDD. The 95th percentile of the mean concentrations of the TCDD in the pooled datasets was calculated to be 1.88 ppm. This value can be used to infer the mean concentration of TCDD in the approximately 40,665 drums (8.48 million liters) of Agent Orange returned or not sent to Vietnam. The question remained, would the Agent Orange stocks purchased before FY 1967 have a similar mean level of TCDD? One solution to the question was to look at the data collected by the National Institute for Occupational Safety and Health (NIOSH) for their Dioxin Registry Study [Piacitelli et al 2000].

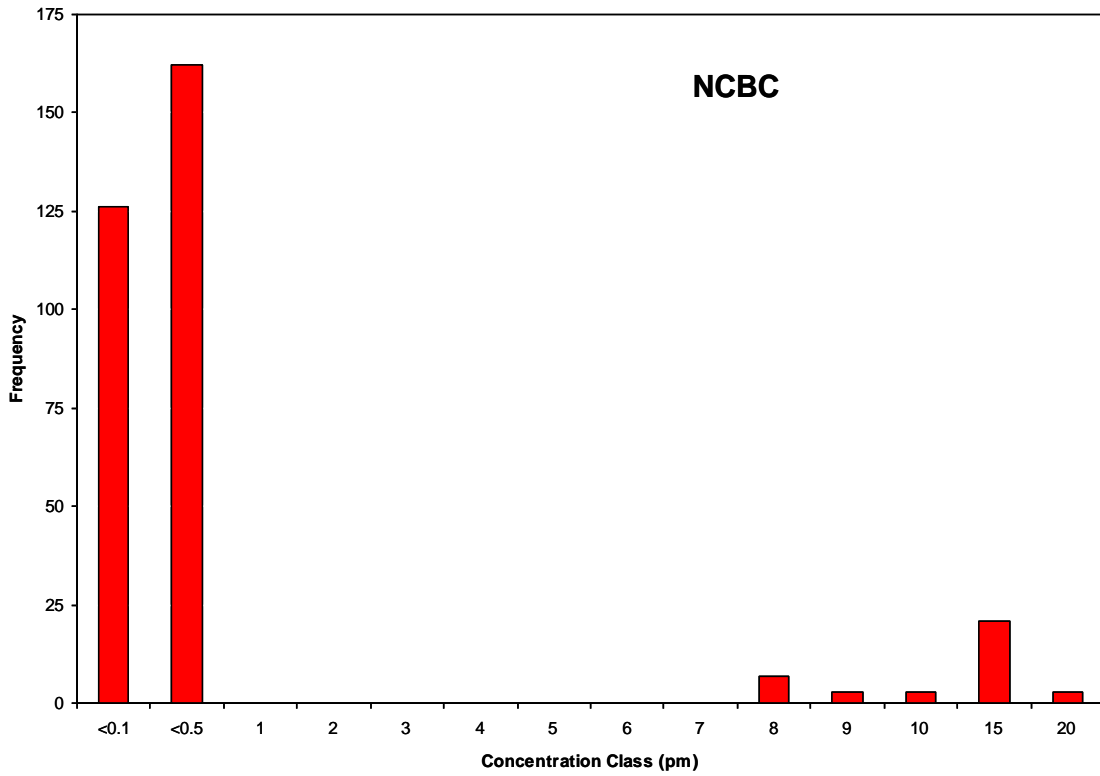
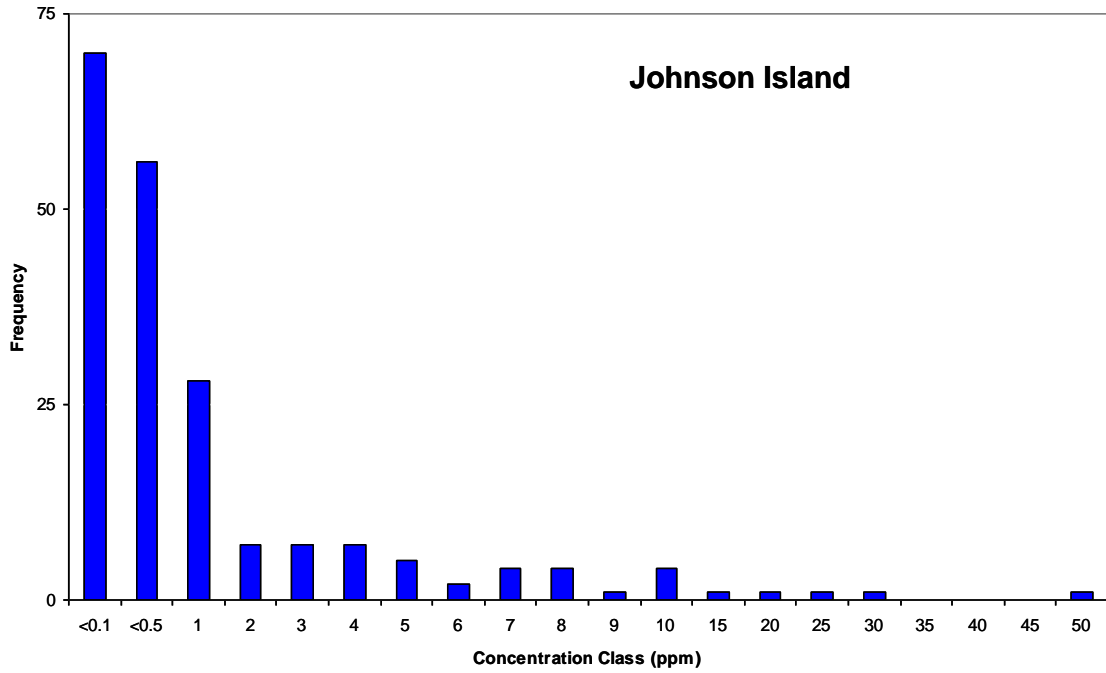


Figure 5.4 Frequency distribution of observed TCDD concentrations in the Agent Orange inventory at Johnson Island and NCBC, Gulfport, Mississippi [OEHL 1977]

5.7 TCDD Data from the NIOSH Studies of 2,4,5-T Production

In 1984, the Industrywide Study Branch of the National Institute for Occupational Safety and Health (NIOSH) in Cincinnati, Ohio began construction of a “Dioxin Registry”, a compilation of demographic and work histories information of all US production workers who have synthesized products known to be contaminated with 2,3,7,8-TCDD and/or hexachlorodibenzo-*p*-dioxins (HxCDD). Fourteen production sites and approximately 7,000 workers were included in the Registry [Piacitelli et al 2000]. Six of these production sites produced Agent Orange including: Diamond Alkali Company/Diamond Shamrock Corporation, Newark, New Jersey [Marlow and Fingerhut 1986]; Thompson-Hayward Company, Kansas City, Kansas [Marlow et al 1990]; Thompson Chemical Company, Saint Louis, Missouri [Marlow and Fingerhut 1991]; Hercules Incorporated, Jacksonville, Arkansas [Marlow et al 1991a]; Dow Chemical Company, Midland, Michigan [Marlow et al 1991b]; and, Monsanto Chemical Company, Sauget, Illinois [Marlow 1997]. These six companies produced 236,040 drums of Agent Orange out of a total of 249,960, or 94% of all Orange produced.

5.7.1 Statistical Analysis of Dioxin Levels in Production Samples of 2,4,5-T Formulations

The data considered from the NIOSH documents in this analysis consisted of dioxin (2,3,7,8-TCDD) values determined for 2,4,5-T acid, 2,4,5-T butyl ester, 2,4,5-T isooctyl ester, and in a few cases the sodium salt of 2,4,5-trichlorophenol, a precursor compound. When considering dioxin concentrations it was assumed that 2,4,5-T acid, esters and precursor were equivalent in the sense that the dioxin measurement in these would be the same as the dioxin concentration in the final 2,4,5-T used in the production of Agent Orange (or Orange II). Since Agent Orange was one-half 2,4,5-T by weight, and there was no dioxin in the other component of Agent Orange, 2,4-D, it was assumed that dioxin concentrations in Agent Orange were ½ the concentration measured in 2,4,5-T or its precursors. Finally for samples with dioxin levels less than the limit of detection (LOD) it was assumed that the dioxin concentration was ½ the LOD. This approach provided 557 dioxin values for the statistical analysis, **Table 5.11**.

For purposes of analysis each manufacturer/year combination was considered as a separate data set. There were several reasons for this. First, dioxin levels changed from both manufacturer-to-manufacturer, and year-to-year. Also, dioxin detection limits changed over time. Most importantly the object of this analysis was to get a good estimate of the total amount of dioxin present in the 2,4,5-T procured for use in Agent Orange in Vietnam, and manufacturers production volumes changed dramatically over time (**Table 5.11**). Thus the arithmetic mean level was taken for a given manufacturer/year combination which was the best estimate of dioxin level for those data and weight it by the total production for that year. If the average dioxin level was defined for a given manufacturer (i) and year (j) combination as $M_{i,j}$, a weighted arithmetic mean, W , level can be obtained as:

$$W = \left(\sum_i \sum_j P_{ij} M_{i,j} \right) / \left(\sum_i \sum_j P_{ij} \right) \quad \text{[Equation 1]}$$

Where P_{ij} is the production for manufacturer i in year j. If one additional assumption was made, namely, that the probability of a given production unit of Agent Orange being actually used in Vietnam was the same for all production units, W would allow the estimate for the total dioxin (D) used in Vietnam as:

$$D = W U$$

Where U was the total amount of Agent Orange used in Vietnam, by weight.

Table 5.11 NIOSH Data Sets for TCDD from the Production of 2,4,5-T by Five Manufacturers

Company/Year	Sample Size	TCDD Detects	Mean TCDD (ppm)	Liters Produced
Diamond Alkali/Diamond Shamrock				
1966	11	11	8.27	208,198
1967	28	28	0.53	1,024,332
1968	11	11	2.03	1,373,272
1969	2	2	1.50	8,536
DOW Chemical Company				
1965	150	27	0.64	1,137,876
1966	4	0	0.18	774,628
1967	3	0	0.01	6,685,037
1968	14	0	0.20	7,688,171
Hercules Incorporated				
1965	12	0	0.25	1,249,716
1966	12	1	0.03	2,682,437
1967	12	0	0.03	3,020,531
1968	12	0	0.03	3,445,766
Monsanto Chemical Company				
1963	1	1	5.50	3,199
1964	1	1	6.00	225,629
1965	18	18	11.53	3,224,981
1966	27	27	5.27	2,073,175
1967	120	120	4.73	3,853,549
1968	32	32	2.61	7,688,171
1969	83	83	1.04	4,345,652
Thompson - Hayward				
1968	4	4	0.32	5,980,950
Total	557	366		56,693,806

The manufacturer/year combinations were quite variable in the amount of data available. **Table 5.11** shows both total numbers of samples and total numbers of detected values for all manufacturer/year combinations. Total samples ranged from 1 to 150 and total detected values range from zero to 120. A combined analysis for the overall mean could have been done by simply applying Equation 1 to the data. However, a combined analysis of such diverse data to get an upper bound on the mean required a hybrid approach. The following conventions were adopted:

- For complete year/company samples (all detects) with 10 or more observations 5000 bootstrap means were generated for each sample.
- For the Dow 1965 data where there were a total of 27 detects out of 150 samples, a

tail-augmented bootstrap was used to generate 5000 random means [Ginevan 2003]. Here, for each mean, with replacement, 150 random numbers between 1 and 150 were generated. If the number was 124 or greater, the data value associated with that rank was selected; if it was less than 124 a random number was assigned from a uniform zero-one distribution. Note that this assumed that concentrations were uniformly distributed between the detection limit, which was 1, and zero, which was the same assumption inherent in assigning non-detects the value of $\frac{1}{2}$ LOD.

- For all other samples, 5000 random means were generated per sample assuming that the distribution was log-normal truncated at the 99th percentile (e.g. no random variables could be greater than about 2.33 standard deviations above the mean) with mean equal to the natural log of the mean estimator (LM; if all values were ND, this was $\frac{1}{2}$ LOD) and logarithmic standard deviation equaled to 1. The later assumption was based on the large samples from Monsanto that suggested that logarithmic standard deviations were generally less than 1 for these sorts of data. To calculate a mean, N observations were generated, where N was the sample size for the company/year combination being considered, from a truncated log-normal distribution with mean LM and standard deviation 1. These logarithmic values (L) were then transformed to the original scale X, using the formula:

$$X = \exp(L)$$

A random mean was then generated as the mean of the N randomly generated X's.

At this point there were 5000 randomly generated mean values for each company year combination. Each set of the resulting manufacturer-year means together with manufacturer-year production values were then used to calculate a random production weighted mean. The random production weighted means were sorted and the 95% upper bound was the 4750th largest value. The result was a 95% upper bound on the overall mean that reflected the uncertainties in the data.

The conventions adopted here reflected the fact that applying methods based on purely log-normal assumptions when estimating upper bounds on the arithmetic mean may result in substantial positive bias in the upper bound. That is, the upper bound is often much larger than any credible value [Ginevan and Splitstone 2002]. Thus either a bootstrap procedure was employed or a log-normal distribution truncated at the 99th percentile (the logarithmic mean, which was taken here as $\ln(\frac{1}{2} \text{ LOD})$, plus about 2.33. because it was assumed a logarithmic standard deviation of 1). Truncated distributions were produced by generating standard normal variates (mean=0; standard deviation=1), and randomly replacing all values greater than 2.33 with another standard normal variate until all values were less than 2.33 [Gentle, 2003].

5.7.2 Results and Discussion of NIOSH Data Sets

The best estimate for the average dioxin concentration produced using Equation 1 was 1.88 ppm, while the upper bound on this mean, produced using the procedures described above, was 2.14 ppm. That is, the 95% upper bound was only about 14% higher than the central estimate.

Note that the bootstrap analysis of the combined Johnson Island/NCBC data gave a best estimate of 1.58 ppm and a 95% upper bound of 1.88 ppm. It has been suggested that dioxin levels in the samples from Johnson Island/NCBC were biased low because they represented dioxin levels in late production runs that were lower than dioxin levels in early runs. While it was true that the Johnson

Island/NCBC data did show slightly lower dioxin levels, the difference was not large. Thus it can be said that these two large data sets give quite comparable answers in terms of both average levels and 95% upper bounds on average levels, which in turn suggests that estimates of average dioxin levels in Agent Orange of much greater than 2.2 ppm would not be very credible.

5.8 Conclusions as to the Amount of TCDD Disseminated in South Vietnam

The frequency distribution of the TCDD data for the Agent Orange samples from Johnston Island and NCBC was skewed toward the high concentrations of dioxin, and thus the statistical method employed was a tail-augmented bootstrap method to estimate the mean. This approach resulted in a mean of 1.58 ppm and a 95% upper bound of 1.88 ppm for the combined inventory of 40,665 drums of Orange. As noted, the 557 individual samples of 2,4,5-T from the NIOSH reports spanned the years 1963 through 1969. The mean TCDD concentration was 1.88 ppm, with an 95% upper bound of 2.14 ppm **IF** these samples of 2,4,5-T had been used in the production of Agent Orange. Thus, these two large data sets gave comparable estimates, suggesting that an estimate of the total amount of the contaminant TCDD in Agent Orange used in South Vietnam would be between 105 kg and 119 kg.

As noted previously, in addition to Agent Orange, other herbicides were used on a limited basis that also contained the herbicide 2,4,5-T and the contaminant TCDD. Reliable historical sampling data quantifying the TCDD concentrations in Agents Purple, Pink, and Green were unavailable. Agents Pink and Green contained 100% 2,4,5-T while Purple contained 50% 2,4,5-T making the concentration of TCDD in Pink and Green double the concentration in Agent Purple. Samples of 2,4,5-T from early production runs were also available from some of the chemical companies that produced Herbicides Green, Pink, and Purple. Statistical analyses of these samples resulted in an estimated (95% confidence level) mean concentration in Pink and Green of 12.2 mg/kg, and 6.1 mg/kg for Purple [Young, Van Houten, and Andrews 2008]. The total estimated contribution from these early tactical herbicides was 25 kg. ***The total estimated amount of the contaminant 2,3,7,8-TCDD associated with the 2,4,5-T-containing tactical herbicides used in Vietnam therefore was between 130 kg and 144 kg.***

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