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Discussion Points on the Significance of Environmental Fate on Exposure to the Four Component Herbicides

Compensation Service

Department of Veterans Affairs
810 Vermont Ave., NW
Washington, DC 20420

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Agent Orange Briefs:

Special Topics

Brief No. 5

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3 February 2014

Mr. Michael D. Pharr
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Department of Veterans Affairs
810 Vermont Ave., NW
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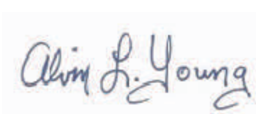
Dear Mr. Pharr,

Please find attached to this letter Agent Orange Brief No. 5 titled **"Discussion Points on the Significance of Environmental Fate on Exposure to the Four Component Herbicides."** This brief is the fifth of many briefs that will be prepared upon request by Compensation Services to address special topics that are germane to issues supporting the ***Agent Orange Investigative Report Series.*** These briefs are prepared in fulfillment of Contract VA-101-12-C-0006, *Development of an Archival Directory of Agent Orange Documents.*

The preponderance of scientific studies and publications on the environmental fate of 2,4-D, 2,4,5-T, picloram, and cacodylic acid has provided ample evidence that within days after these herbicides were sprayed, the lack of bioavailability, even at the application rates used in Vietnam, would have minimized human exposure.

Sincerely,



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DISCLAIMER

The conclusions reached in this report are based upon a comprehensive review of the historical records maintained in the publicly available files of the National Archives and Records Administration, and other archival repositories. However, the conclusions reached do not necessarily represent those of the Department of Veterans Affairs or any other Department or Agency of the United States Government.

This "Discussion Point Brief" is part of the ***Agent Orange Investigative Report Series***, and should be considered as an amendable or living document. If additional authenticated documents or records are found that address the topic of this report, a re-evaluation of the conclusions may be necessary.

DISCUSSION POINTS ON THE SIGNIFICANCE OF ENVIRONMENTAL FATE ON EXPOSURE TO THE FOUR COMPONENT HERBICIDES

INTRODUCTION

In 1991, Congress passed Public Law 102-4, **The Agent Orange Act of 1991**, to address the uncertainty about the long-term health effects on Vietnam veterans, who during their service in Vietnam, were exposed to any of the four herbicides that constituted the various tactical herbicides used in military operations in Vietnam, 9 January 1962 – 7 May 1975, namely, 2,4-D, 2,4,5-T, picloram and cacodylic acid. Previous reports for Compensation Service have been prepared on all four of these component herbicides [1, 2, 3, 4]. This Report focuses on the studies of the environmental fates of these four herbicides and the significance of such studies to exposure to Vietnam-era veterans *outside of Vietnam*, especially at locations where the components of the tactical herbicides were tested and evaluated, and veterans have alleged exposure to such herbicides.

US ARMY CHEMICAL CORPS TESTING OF THE TACTICAL HERBICIDES

From 1959 through 1967, the US Army Chemical Corps maintained an extensive program to develop, test and evaluate the above four herbicides as proposed components for the tactical herbicides to use or be used in Vietnam. Six major comprehensive reports have been published on the Army Chemical Corps efforts. In 1963, 1964, and 1965, the Army Chemical Corps Biological Laboratories sponsored "Defoliation Conferences" at Fort Deterick Maryland in which the published proceedings described the results of tests [5, 6, 7]. In 1968, the Agricultural Research Service published a "***Research Report…Response of Tropical and Subtropical Woody Plants to Chemical Treatments***." This research via USDA was sponsored by the Department of Defense's Advanced Research Projects Agency [8]. In 2006, the Department of Defense published the report "***The History of the US Department of Defense Programs for the Testing, Evaluation, and Storage of Tactical Herbicides***" [9]. In May 2013, as part of Compensation Service's Agent Orange Investigative Report Series, a report was prepared on "***Investigations into Sites Where Agent Orange Exposure to Vietnam-Era Veterans Has Been Alleged***" [10].

In order to validate a location where Agent Orange or other tactical herbicides were developed, field evaluated, sprayed in equipment testing operations, or buried or destroyed, it was necessary to conduct a comprehensive search for the historical records. The search identified 13 US military installations in the Continental United States (CONUS) where Agent Orange or other tactical herbicides were developed, tested, sprayed, stored or buried. Appropriate and authenticated documents were available for the following military installations:

Aberdeen Proving Grounds, MD: Field tests of tactical herbicides 1965 – 1966
Dugway Proving Ground, UT: Loading and testing of aerial spray equipment, 1964
Fort Chaffee, AR: Field Tests of tactical herbicides, 1967
Fort Detrick, MD: Tests in greenhouse & small plots; disposal of Orange and White in 1972
Fort Drum, NY: Field tests of Purple, 1959
Fort Gordon, GA: Field tests of tactical herbicides, 1967
Fort Meade, MD: Field tests of tactical herbicides, 1963 – 1964
Fort Ritchie, MD: Field tests of tactical herbicides, 1963 – 1964
Fort Smith, AR: Field tests of tactical herbicides, 1967
Naval Construction Battalion Center, MS: Storage of surplus Agent Orange, 1968 – 1977
Eglin AFB, FL: Loading and testing of aerial spray equipment, 1962 -1971
Hill AFB, UT: Soil biodegradation tests at the AFLC Test Range Complex, 1972
Kelly AFB, TX: Storage of Pink and Blue, 1969 – 1974

All of the above military installations were locations where Vietnam-era veterans were stationed, where tactical herbicides were developed, tested, evaluated, or stored, and where technical reports and other supporting documentation were found in the archives and other repository sources. The circumstances involved for these various tests or military operations are critical to understanding the potential for Vietnam-era veterans to have been exposed to the tactical herbicide.

FACTORS INFLUENCING ENVIRONMENTAL FATE

The phenoxy herbicides 2,4-D and 2,4,5-T, as well as the herbicides picloram and cacodylic acid, were used for decades in agricultural and forestry programs worldwide. In the case of 2,4-D and picloram, their use continue today [11]. As a consequence, a large of amount data were available on their environmental fate and how those data have provided a better understanding of human exposure to these herbicides.

The factors important in determining the fate of herbicides in the environment especially as they relate to the potential of a chemical for causing harm to humans were the following: adsorption, leaching, runoff, plant uptake, chemical decomposition, microbial decomposition, volatilization, and photodecomposition [12].

- **Adsorption**: Refers to how tightly bound a chemical is to soil components. If a compound is tightly bound, only a small fraction of the applied material is available for the other modes of dissipation;
- **Leaching**: Refers to ability of a chemical compound to move throughout the soil profile. The water solubility of a compound influences its downward mobility;
- **Runoff**: Low adsorption and high water solubility of a compound within the first week of application will result in significant runoff of the compound;
- **Plant Uptake**: Generally less than 5% of herbicide in soil is taken up by plants. Plants are able to degrade many of the herbicides;
- **Chemical Decomposition**: Several chemical reactions occur in higher plants to dissipate the presence of herbicides;
- **Microbial Decomposition**: Microbial decomposition is one of the major routes for the destruction of most herbicides;
- **Volatilization**: Compounds with high vapor pressures are more volatile than those with low values. However, before a herbicide is free to leave the soil through volatilization, it must escape the binding capacity of the adsorptive soil complex; and,
- **Photodecomposition**: Photodeomposition degrades many herbicides. On soil and leaf surfaces it represents a major avenue of loss [12].

THE ENVIRONMENTAL FATE OF 2,4-D and 2,4,5-T

As noted, the above factors influence the persistence of 2,4-D and 2,4,5-T in the environment. As the 2,4-D and 2,4,5-T degraded or were removed from the environment, the chances decreased that humans would contact it as dislodgeable residue from leaf, soil surfaces, or in food. There were many excellent reviews available on the environmental fate of 2,4-D and 2,4,5-T, including those by Young et al in the 1978 USAF report "**The Toxicology, Environmental Fate, and Human Risk of Herbicide Orange and Its Associated Dioxin**" [13], by the Veterans Administration in the various "**Reviews of Literature on Herbicides**…" [14], and by Lavy in the 1987 VA monograph on "**Human Exposure to Phenoxy Herbicides"** [12]. In summarizing the conclusions from these reports, the following factors influenced the persistence of 2,4-D and 2,4,5-T in the environment:

- Soil Adsorption – The acid formulation of 2,4-D and 2,4,5-T possessed moderate aqueous solubility (~250 mg/l), but weakly adsorbed to soil, while the ester formulations had low water solubility and were more strongly absorbed;
- Leaching – The n-butyl ester of 2,4-D and 2,4,5-T were resistant to leaching in all soils, although some movement occurred in sand. However, even there it seldom leached below the top 5 cm (~ 2 inches) of the soil surface;
- Runoff - Total loss of 2,4-D or 2,4,5-T due to runoff over substantial periods of time did not generally account for more than 5 percent of the applied herbicide;
- Plant Uptake – Both 2,4-D and 2,4,5-T were most effective when applied as the n-butyl ester because of rapid absorption into the leaf surface. Once inside the leaf, the butyl ester of was readily degraded (within hours) through transesterification and β oxidation;
- Chemical decomposition – The n-butyl ester of 2,4-D and 2,4,5-T completely hydrolyzed within a few days in moist soil or lake water;
- Microbial decomposition – The half-lives of 2,4-D and 2,4,5-T in previously untreated soil were from 3 to 16 days (2,4-D) to about 30 days (2,4,5-T), but when the same soil was re-treated the half-life was 1.5 to 15 days. In aquatic or moist environments microorganisms readily degraded 2,4-D and 2,4,5-T. Rates of breakdown increased with increased nutrients, sediment load and dissolved organic carbon, conditions typical of those found in moist agricultural and forestry ecosystems;
- Volatilization – approximately 30% of the n-butyl ester of both herbicides were collected as vapor drift 250 feet from the ground application point; and,

- Photodecomposition – The photo-oxidaton of 2,4-D and 2,4,5-T resulted in rapid (hours) deactivation and low persistence, a process that minimized the use of Agent Orange in some environments in Vietnam, thus, requiring retreatment of some targets.

Although the issue of volatility is important, what is really crucial to understanding exposure to individuals on the ground is the dispersion of the droplets or particles of herbicide from aerial applications. Veterans have repeatedly claimed that they were exposed miles from where the actual applications occurred, e.g., Eglin AFB, but the data on dispersal of the herbicide does not support such claims [15]. Aerial Tests at Eglin AFB, Florida with the UC-123B RANCH HAND aircraft showed that 87% of the herbicide (droplet size > 300 µm) would have impacted the vegetation (ground) within one minute and within or near to the swath (~260 feet) [16]. The remaining 13% of the herbicide would have taken longer to settle due to vortices at the wing tips, drift, or evaporation [16]. Calculations made using Stokes Law showed that even the <100 µm size droplets, would have had a settling velocity of over 30 cm/sec indicating that droplets would have impacted the vegetation or ground less than 3 minutes after spraying. Since spray missions were always undertaken in calm or near-calm conditions, there was insufficient time for significant lateral movement or ‘spray drift’. Any significant lateral movement of spray would have required the herbicide to remain in the air for extended periods of time and they would therefore have been subject to rapid degradation by ultraviolet light [16].

Summary: The extensive available data indicated that both 2,4-D and 2,4,5-T had short persistence time in the environment, and when combined with the toxicological data (>350 mg/kg) did not pose a threat to human health. The uptake of a measureable dose was possible only when handling the actual liquid.

THE ENVIRONMENTAL FATE OF PICLORAM

A tremendous amount of research was conducted on picloram between 1969 and the early 1980s. As data began to emerge on how particles of herbicides behaved when aerially disseminated, interest also increased in the ecological impact of woody plant control with picloram [8, 17]. Several ecologists expressed concern that the use of herbicides as defoliants in Vietnam might result in soil erosion and soil laterizations. The assumptions were that the selective nature of these herbicides would result in the death of all higher plants, and that the residues, especially of picloram, would prevent re-vegetation [17]. The research in Puerto Rico by USDA clearly did not validate these assumptions in areas where the tactical herbicides, including Agent White, were sprayed on jungle vegetation at concentrations similar to those used in Vietnam [8, 17].

The tests conducted by the USDA's Agricultural Research Service in Texas (1964-66) and in Puerto Rico (1964-66) [8] concluded:

- The broad spectrum of woody species susceptible to picloram made it an extremely important herbicide for woody plant control;
- The use of picloram was particularly appropriate for the defoliation of forest types characterized by high species diversity;
- Picloram provided greater suppression of lateral and basal dormant buds than did other herbicides;
- The combined advantages of effectiveness on many species and greater bud suppression made picloram the best single herbicide available for woody plant control;
- Experience with picloram indicated that gallon for gallon it equaled or exceeded the control obtained with any other herbicide;
- In greenhouse and field studies, picloram was translocated throughout woody plant species more rapidly than 2,4-D or 2,4,5-T;
- The most effective herbicide applied at relatively low rates to soil in dry areas was picloram, and in wet sites, picloram was unquestionably most effective;
- Research indicated that picloram was mobile in the soil, but did not degrade rapidly;
- In sandy soils, leaching soon carried picloram to soil depths that roots did not usually penetrate;
- Ultraviolet light decomposed picloram rapidly (hours); and
- The degree of revegetation following application of picloram to either the foliage or to the soil depends primarily on the amount of rainfall that occurred in the treated area [8].

Summary: The extensive available data on the environmental fate of picloram provided ample evidence that it was rapidly taken up by the vegetation and although more persistent in the soil, it's availability for human uptake was minimal. When combined with the toxicological data (i.e., >8,000 mg/kg) picloram did not pose a threat to human health.

THE ENVIRONMENTAL FATE OF CACODYLIC ACID

The fate of organoarsenicals in plants and soils has been extensively investigated [18]. Much of the research on the fate of cacodylic acid was conducted in the 1970s. Cacodylic acid was a highly effective tool for thinning over-stocked young stands of conifers (e.g., Douglas Fir), and thousands of acres were treated yearly in forests of Oregon and Washington State [19]. Two major reports were prepared and published by the Pacific Northwest Forest and Range Experiment Station (1977), and the School of Forestry, at Oregon State University, Corvallis, Oregon (1986) [19, 20].

Studies indicated that the cacodylic acid moved rapidly (days) into the treated trees and was detected in the needles and twigs (41 ppm arsenic), and subsequently in the forest floor litter and soil (5.0 ppm) within a few months. Results also indicated that the cacodylic acid would persist in the dead trees for years [19, 20]. The cacodylic acid was mobile in water throughout the forest litter, but was essentially immobile when bound within the surface of the mineral soil. Little or no leaching occurred in mineral soil. Contamination of groundwater by leaching was considered unlikely [20]. The levels of cacodylic acid declined rapidly with time and distance from the treated trees [20]. Additional studies have also shown that when the herbicide was intercepted by soil it was “deactivated” by absorption to soil colloids [20]. While the initial soil absorption of cacodylic acid or sodium cacodylate was rapid (hours), long-term changes (within weeks) resulted in redistribution of the water-soluble cacodylic acid into less soluble factions associated with aluminum and iron [19, 20].

Additional studies have noted that when cacodylic acid was added to various soils at concentrations as high as 100 ppm, its persistence after 32 weeks varied from 62% to 23% depending upon soil type [18, 21]. The loss of cacodylic acid was related to its conversion in soils to a volatile alkyl arsine. It was thought that this degradation (transformation) was due primarily to microbiological action [18, 19]. The significance is that the arsenic in Agent Blue (as cacodylic acid or sodium cacodylate) would have been bound within the soil matrix making it immobile, but its persistence over time would have decreased because of its slow conversion to volatile arsenical compounds which would have been released into the atmosphere [21].

Summary: The extensive available data on the environmental fate of cacodylic provided ample evidence that it was rapidly taken up and bound by the vegetation. When in contact with soil, the organic form of arsenic rapidly became bound and immobile due to the mineral content of the soil. Its slow disappearance was due to conversion to the volatile alkyl arsine. When combined with the toxicological data (i.e., >2,000 mg/kg) cacodylic acid did not pose a threat to human health.

A LONG-TERM ECOLOGICAL STUDY OF THE COMPONENTS

The most important long-term ecological study of the fate of 2,4-D, 2,4,5-T, picloram and cacodylic acid, components herbicides of the tactical herbicides sprayed in Vietnam was sponsored by the Air Force Armament Laboratory, Eglin AFB Florida beginning in 1969 and concluding in 1994 [22]. From 1961 – 1971, the Air Development Test Center, Eglin AFB Florida developed, tested, and calibrated the aerial spray systems used in support of Operation RANCH HAND and the US Army Chemical Corps in Vietnam. The tests, conducted under climatic and environmental conditions similar to those in Vietnam, included the use of the military herbicides Agents Orange, Purple, White and Blue.

Approximately 165,000 pounds of 2,4,5-T and 168,000 pounds of 2,4-D, 2,200 pounds of picloram, and 8,400 pounds of cacodylic acid were aerially disseminated on a test area (Test Area C-52) of less than one (1) square mile [22]. As a comparison, a “typical” mission of spraying Agent Orange in Operation RANCH HAND would have resulted in 12.6 pounds of 2,4-D per acre, most of which would have been intercepted by forest canopy, versus the 1,900 pounds 2,4-D per acre received on the Eglin test grids [22].

Tests of soil persistence of 2,4-D and 2,4,5-T were first conducted at the conclusion of the last spray equipment test using Agent Orange in December 1969. In April 1970 the combined residues of 2,4-D and 2,4,5-T were in the top 6 inches of soil at levels of approximately 2.8 ppm, while in December 1970, no residues of 2,4-D or 2,4,5-T were detected, but picloram was detected at 11 ppb and cacodylic acid was detected at 1.6 ppm [22, 23]. A subsequent study found that little or no movement of the arsenicals occurred into the adjacent aquatic watersheds or ecosystems [24].

Studies of the soils, fauna, flora, and aquatic ecosystems of the test areas and associated perimeters were initiated in 1969 and concluded in 1984. More than 340 species of organisms were observed and identified within the test area over the 15 years of observations. The ecological studies conducted and documented on Test Area C-52A found no long-term adverse ecological effects despite the massive quantities of herbicides (and TCDD) that were applied to the site. Forty years after the establishment of the test areas in 1962, the site had returned to the original climax forest [22].

DISLODGEABLE RESIDUES WHEN WALKING THROUGH TREATED VEGETATION

The fraction of a chemical residue that is available for cutaneous (skin) uptake from the surface of plant leaves is called the 'dislodgeable foliar residue'. For chemicals that are absorbed into plant tissues, this fraction decreases as the chemicals penetrate in the leaves. A number of human studies have been conducted on exposure to the phenoxy herbicides using dermal exposure pads attached to the clothing in the chest area, on the back near the shoulder blades, on each arm in the biceps area, and on each leg of the upper thigh [12, 25]. Only 8% of the residues were detected on individuals that entered a treated area 1 hour after application, and this dropped to 1% of the total 24 hours after application. Moreover, studies with human volunteers confirmed that after 2 hours of saturated contact with bare skin, only 0.15 – 0.46% of 2,4,5-T entered the body and was eliminated in the urine[12, 25].

SPRAY PENETATION AND DEPOSITION OF PARTICLES IN FOREST CANOPIES

Scientists from USDA studied the penetration and distribution of herbicide sprays through forest canopies in Puerto Rico and Texas [8]. Although the two areas were widely separated geographically, the forests were similar in terms of structure. The test site in Puerto Rico was typical of moist forest formation consisting of three canopies, with an upper canopy of >50 feet above ground level. In Texas, the forest had a dense and relatively unbroken canopy of >16 feet above ground level. Spray materials (including tactical herbicides Orange and White) were aerially applied at levels consistent with the use in Vietnam. On the average, about 21% of the spray volume penetrated the upper canopies in both locations, and about 6% penetrated to ground level. Thus, exposure to the four component herbicides in CONUS tests and evaluations by the US Army Chemical Corps would have been further minimized.

CONCLUSION

A large preponderance of scientific studies and publications on the environmental fate of 2,4-D, 2,4,5-T, picloram, and cacodylic acid provided ample evidence that within days after these herbicides were sprayed into the environment, the lack of bioavailability, even at application rates used in Vietnam, would have minimized human exposure.

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BRIEF BIOGRAPHY OF THE AUTHORS

For more than 40 years, Dr. Alvin L. Young has been involved in issues surrounding the use of Herbicide Orange and other tactical herbicides in Vietnam. He completed his PhD in Herbicide Physiology and Environmental Toxicology at Kansas State University in 1968. In his 21 years with the USAF (obtaining the rank of Colonel), he was involved with the testing and evaluation of the equipment used in Operation RANCH HAND, Vietnam, and with the environmental and human health studies with the USAF School of Aerospace Medicine and the Department of

Veterans Affairs. He served as a Science Advisor on environmental issues including Agent Orange with the President's Office of Science and Technology Policy. He was the Director of the Department of Energy's Center for Risk Excellence. He was a Visiting Professor at the University of Oklahoma, 2001-2007, and has served as the Senior Consultant on Herbicide Orange for the Office of the Deputy Under Secretary of Defense (Installations and Environment). He has more than 300 publications in the scientific literature, including five books on issues related to Herbicide Orange and/or dioxins and furans. From 2000 to 2012, He was the Editor of the international journal *Environmental Science and Pollution Research*.

For the past ten years, Kristian L. Young has been the Principal Researcher for A.L. Young Consulting. He received his Bachelor of Arts in Political Science from DePaul University, Chicago (Magna Cum Laude, Phi Kappa Phi, and Pi Sigma Alpha). He received the Master of Arts in International Relations in 2010 through Webster University's Global Program having studied in Europe and China. He has provided support to the company in areas of public policy, technical issues, archival research, and the coordination of national and international projects.