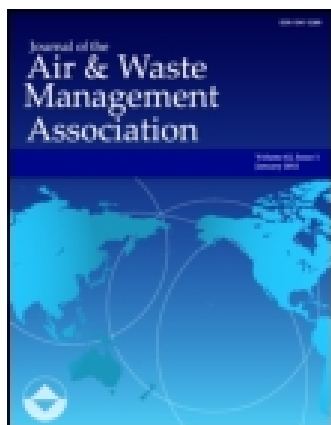


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2,4-D Herbicides in Central Washington

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Usage of 2,4-D herbicides has been extensive in central Washington wheat-growing areas for many years. The rapid development of a grape industry, both Concord and Vinifera, since the early 1960's has had periodic, widespread damage in vineyards due to drifting 2,4-D. Beginning in the Spring of 1973, programs of 2,4-D monitoring have been conducted in the lower Yakima Valley of central Washington. In 1976 a program was begun to test the drift potential of 2,4-D aircraft spray systems. Correlations between atmospheric monitoring data, observed vineyard damage, and weather patterns indicate that long distance transport—10 to 50 miles—is an important factor in the 2,4-D concentration pattern. Some simple models have been developed but many questions must be resolved.

The use of agricultural chemicals on crops and rangeland is an expanding activity that has frequently been a cause of controversy because of off target drift. In Washington State, 2,4-D herbicides are used widely on grain crops; and over the years, since at least 1955, there have been recurring problems from off target drift. The most seriously affected crop from 2,4-D drift damage in recent years has been grapes grown in the lower Yakima Valley. In the 1950's the 2,4-D grape damage problem prompted the research studies by Clore, *et al.*¹ on the sensitivity of grapes to 2,4-D. It was determined that grapes rank with cotton in being among the most sensitive plants to 2,4-D.

The use of 2,4-D continued to expand in the eastern Washington area and further research was done in the 1960's. This research resulted in the development of ambient air monitoring techniques by Bamesberger and Adams² at Washington State University (WSU) and also by Vernetti and Freed at Oregon State University.³ In the late 1960's and until about 1972 2,4-D drift problems seem to have attracted little attention although it might be incorrect to conclude that no damage occurred.

After the 1972 growing season, however, and continuing up to the present time there have been a number of instances of wide-spread 2,4-D damage to grapes, and this has brought about a considerable amount of field and laboratory research effort at WSU in an attempt to develop measures to combat the drift damage. These recent research studies at WSU are summarized in the following discussion.

The Nature of the 2,4-D drift

Drift of 2,4-D causes 2 different drift damage patterns in Washington, a localized type and a widespread type. The local pattern is characterized by severe damage symptoms typically in one part of a vineyard or field with a very clear gradient of decreasing symptom severity away from the severe zone. This pattern results from the application of 2,4-D close to the affected crop and then wind drift across the damaged area. The more severe damage zone identifies the upwind area closest to the application area and the rapid dilution of pollutant with distance from the source accounts for the gradient of damage effects across the susceptible crop. Although there may be considerable argument about the degree of damage or its economic value, local drift problems can generally be well defined in terms of the likely source and the extent of the affected receptor area.

In the grape vineyards of Washington there are a number of instances of 2,4-D damage from local drift every year; however this is not the most serious 2,4-D damage situation. In the Washington vineyard area, shown in Figure 1, the 2,4-D damage patterns that caused the major concern are cases where more or less uniform damage symptoms occur at the same time across the vineyards throughout much or all of the grape area. This situation does not have clearly definable gradients and appears to be attributable to the passage of a large, contaminated cloud with the dimensions of miles, at least. Thus this has been labeled as "air mass" damage. Most of the WSU 2,4-D research program has been directed toward finding explanations for this "air mass" damage pattern. This

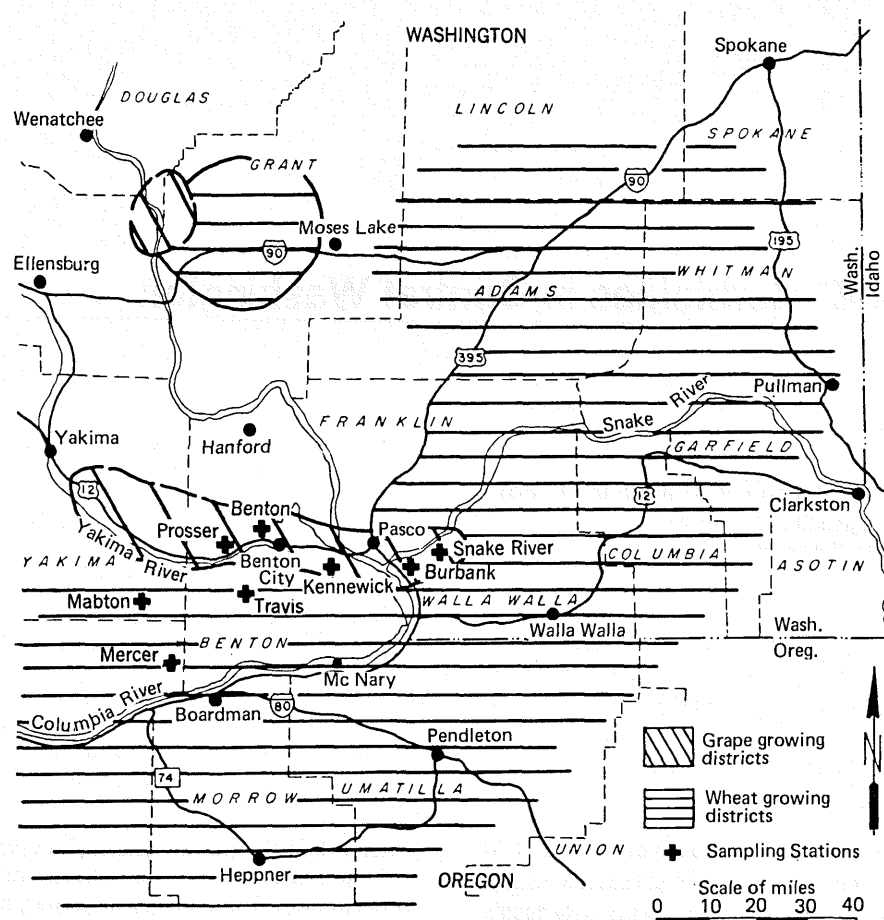


Figure 1. Map of Central Washington showing 1974 2,4-D sampling network, vineyard areas, and grain growing areas.

research has included ambient air monitoring in the grape and grain areas as illustrated by the monitoring station network shown in Figure 1. The research has also included meteorological studies to identify transport systems. Recently because of continuing local drift problems, a program of aerial application studies has been begun to determine the relative drift potential of various application systems currently being used in Washington.

2,4-D Monitoring Program

The 2,4-D monitoring program was based on the operation of continuous collectors at each station in the network. The number of stations has varied from 8 to 12 in different years. Each sampling site was visited on a regular daily schedule from April through June and the sample collector at each station changed. The daily visit schedule means that the 2,4-D concentration data represent 24 hr average concentrations and consequently, no indications of intermediate concentration values during the day are available. Since the average "sample-changing" time was approximately 12 P.M., a sample dated May 22 would have been collected between noon on May 22 and noon on May 23.

The field samples were shipped to the Air Pollution Section of Washington State University on a regular weekly schedule for subsequent chemical extraction and analysis.

Air Sampler for 2,4-D

The 2,4-D sampler used in the initial phases of the program, 1973 and 1974, was of the differential impactor-impinger type developed by Bamesberger and Adams.² However, beginning in 1975, a collection device using solid adsorbent material was

employed.⁴ Figure 2 shows a schematic drawing of this sampling apparatus. It consists of a glass tube (15 cm in length and 19 mm in diameter) filled with 3–4 grams of a dry adsorbent resin. Glass wool plugs and a pair of small stainless steel screens serve to hold the resin in the glass collection tubes. The solid adsorbent, known as Amberlite XAD-2, is a porous polystyrene-divinylbenzene copolymer from Rohm and Haas Chemical Co. as 20- to 60-mesh beads. Before the solid adsorbent was packed into the glass tubes, it was cleaned by a water and an acetone rinse and then extracted for 6 to 24 hr with methanol. After drying the clean XAD-2 was immediately packed and sealed in the glass tubes.

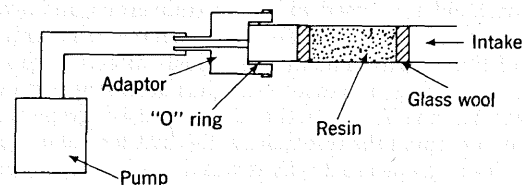


Figure 2. Dry collector sampler design.

At the sampling site, the sampling cartridge was unstoppered and attached to the sample holder. The sample holder was constructed from a Cajon Ultra-Torr 12-UT-A-16 adaptor that was modified to include a Swagelok hose connector. A critical orifice was incorporated into the hose connector in order to control the flow rate at 5.5 liters per minute. In operation, air is drawn through the sampling tube and the porous

adsorbent collects and retains the organic herbicides from the moving air stream. The overall collection efficiency (collection vs. extraction) of XAD-2 for 2,4-D compounds was studied in our laboratory, and the efficiency results ranged from 84% to 100%.⁴

This change in sampling technology from the liquid-filled system of Bamesberger and Adams² to the more convenient dry system was possible because a number of porous polymers such as the Chromosorb century series 101-108, the Porapak series, the Amberlite XAD series, and Tenax-GC have been shown to be effective adsorbents for the sampling of organic pollutants in air. The use of adsorbents in air samplers is practicable due to their ability to retain selectively the organic compounds while allowing water vapor and the common air components to pass on through the collection column. These polymeric adsorbents also offer such distinctive advantages as: (a) no interferences from water vapor because of the hydrophobic nature of the porous polymer beads, (b) their relatively large surface areas provide efficient sampling, (c) a lower surface adsorption strength than most common adsorbents permits desorption efficiencies greater than 90%, (d) ambient air samples collected in columns filled with these porous supports can often be sealed and stored for later analysis, (e) the collection of organics by dynamic enrichment on the polymer adsorbents allows the complete adsorption of organic compounds having a retention volume lower than the sample volume passing through the collection column, and (f) the solid-based adsorbent sampling tubes are of simple construction and permit easy sample handling in the field with a minimized chance of inadvertent contamination.

Adsorbent Elution and Gas Chromatographic Analysis

The recovery of the adsorbed 2,4-D compounds from the XAD-2 collection tubes was performed by a sequential solvent extraction. First, the 2,4-D esters were removed from the collection tubes by elution with 60 ml of nanograde hexane. This hexane solution was subsequently transferred to a 75°C water bath where the solvent was evaporated down to about 5 ml. Then, 1 ml of a 1 ppm solution of 2,4-D *n*-amyl ester was added for an internal chromatographic retention time standard, and the sample was brought to a final volume of 10 ml with additional hexane. Five microliters (μ l) of the sample solution were injected into the gas chromatograph for analysis.

After extraction of the 2,4-D esters, the adsorbent tubes were eluted with 60 ml of a 0.05N KOH in 20% aqueous methanol solution. This basic solvent elutes the non-volatile types of 2,4-D from the resin. The basic methanol solution was then evaporated to approximately 10 ml in a 75°C water bath and was extracted with three 5 ml aliquots of diethyl ether. 10 ml of a 14% BF₃/methanol solution was added to the combined ether extracts and the entire solution was evaporated to approximately 10 ml in a 35°C water bath. Then, 10 ml of distilled water and 5 ml of nanograde benzene were added to the concentrated ether solution. After shaking, the two solvent layers were separated and the benzene portion was brought to a final volume of 10 ml with benzene. 5 ml samples of the benzene solutions were injected into the gas chromatograph for analysis.

The preceding BF₃/methanol esterification procedure converts all of the nonvolatile 2,4-D to the methyl ester. Therefore, the ensuing gas chromatographic analysis of the 2,4-D methyl ester is directly proportional to the total non-volatile 2,4-D in the individual samples.

Analytical Instrumentation

A Perkin-Elmer Model 3920 gas chromatograph equipped with a ⁶³Ni electron capture detector was used for the routine analysis of the 1975 field samples. A 10 ft $\times$ 1/8 in. glass column packed with 1.5% SP-2250/1.95% SP-2401 and 100/120-mesh

Supelcon AW-DMCS was employed in the GC/ECD analysis. The GC/ECD was connected to a Hewlett-Packard Model 3380 A recording integrator for automatic peak integration and analog recording. As previously indicated, *n*-amyl 2,4-D was added to all of the analyzed field samples for use as an internal chromatographic retention time standard. The *n*-amyl 2,4-D internal standard was only utilized to improve the qualitative accuracy of the GC/ECD-recording integrator system and was not used for quantitation. Farwell, *et al.*⁵ gives more information on the analytical techniques used.

Although a gas chromatograph with a ⁶³Ni electron capture detector was used for routine analysis of the field samples, our gas chromatograph/quadrupole mass spectrometer was employed to substantiate the qualitative and quantitative data obtained with the GC/ECD.

Monitoring Results

There are a number of compounds of 2,4-D that are used as herbicides in south central Washington and surrounding areas.⁵ They are generally classed according to volatility as "high volatile" (HV), "low volatile" (LV), and "non-volatile" (NV) according to vapor pressure ranges. HV compounds include esters with 5 or less carbon atoms and have a vapor pressure of 10⁻⁵ mm Hg at 20°C. In our area *n*-butyl 2,4-D seems to be the most important HV detected. LV 2,4-D are ester compounds having a vapor pressure in the range of 10⁻⁵ to 10⁻⁶ mm Hg with the most common ones in Washington being isooctyl, 2-ethyl hexyl, butoxyethanol (BE), and propylene glycol butyl ether (PGBE). Salts and amine compounds of 2,4-D are the common NV materials and the NV class is based on the vapor pressure of 10⁻⁷ mm Hg or less.

One of the important results of the 2,4-D monitoring program was the fact that different 2,4-D volatility types as described above were determined and that high volatile esters of 2,4-D accounted for a majority of the total 2,4-D in many situations in 1973 and 1974. Table I, for example, shows the monthly average values for the seven operating stations for May, 1973. In this case 74% of the total 2,4-D was high volatile ester. This finding was considered quite significant because previous regulatory action had prohibited the use of high volatile 2,4-D esters in the areas adjacent to the grape vineyards. In fact the areas where these esters could be used legally was more than 20 miles away. In the case of southerly winds this would put the source in the State of Oregon.

Table I. Average 2,4-D concentrations (mg/m³) by volatility type, May, 1973.

Sampler location	High volatile	Low volatile	Non-volatile	Total 2,4-D
Prosser	0.23	0.07	0.01	0.31
Kennewick	0.23	0.06	0.02	0.31
Benton City	0.23	0.09	0.02	0.34
Snake River	0.32	0.05	0.01	0.38
Sunnyside	0.24	0.08	0.01	0.33
Plymouth	0.18	0.02	0.01	0.21
Mabton	0.20	0.07	0.01	0.28
7-Station average	0.23	0.06	0.01	0.31

There have been major differences in the general levels of atmospheric 2,4-D from year to year and also in the severity of 2,4-D damage symptoms in grape vineyards. The years 1973 and 1974 were years with high levels of 2,4-D and serious, widespread grape damage symptoms. In 1975 concentrations were generally very low, and there was little or no grape damage. A cool, stormy spring that changed the average dispersion pattern and also reduced the use of 2,4-D is credited with this result. In 1976 there were a number of instances of local-type 2,4-D damage but no air mass contamination.

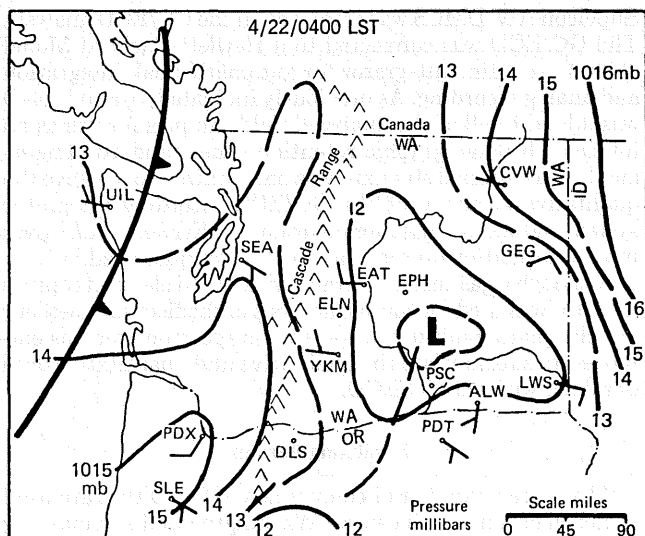


Figure 3. Typical synoptic weather pattern for relatively high concentrations of 2,4-D.

Meteorological Analysis

The current meteorological research shows that there are three distinct weather patterns characterizing three levels of 2,4-D concentration severity in the area: "high," "moderate," and "low."⁶ The severity of observed 2,4-D concentrations over the sampling network was originally used to guide the classification of damage situations. Higher concentrations were generally accompanied by observable herbicide damage symptoms in grape vineyards.

Meteorological data used in the analysis of the research results were obtained from the various governmental and private weather stations in south central Washington. Daily weather maps published by the National Weather Service and synoptic data provided by Battelle-Northwest at Hanford were invaluable inputs to the assessment of both synoptic or large scale weather patterns and the smaller mesoscale weather patterns.

As originally recognized by Peckenpaugh,⁷ the highest 2,4-D concentration/damage periods have very similar weather patterns associated with them. One to two days prior to the "high" concentration day, the model (Figure 3) is characterized by a weak low pressure development or troughing in eastern Washington (leeside trough) and a weak frontal system approaching the Washington coast. The transport wind pattern associated with this situation is characterized by a gradual backing from a westerly direction to a more southerly, prefrontal direction. Both transport and surface winds are relatively light throughout most of the prefrontal period. Clouds increase such that by the day of high concentration surface heating has been significantly reduced and vertical mixing is governed mainly by mechanical mixing processes until the front passes on the day of high concentration or early on the following day. Relatively poor spraying conditions often follow these frontal situations—which, together with the usual shift to a more westerly wind, helps to explain why high 2,4-D levels seldom persist to a second day.

This basic model accompanied the two highest concentration days in 1973 and 1974.

The second weather pattern, Figure 4, associated with the "moderate"-concentration days, was characterized by a high pressure system off the Oregon or southwest Washington coast with a low pressure trough in eastern Washington.⁶ This combination produces a dynamic or pressure gradient effect which is often accompanied by strong winds throughout the area of north-central Oregon and southern Washington. The basic southwest-to-northeast pressure orientation produces

a rather constant air flow over the entire region. This second weather pattern accompanied moderate damage to the grape vineyards in the Lower Yakima Valley in 1974 and in 1975.

There are two separate weather patterns that were correlated with the occurrences of low levels of 2,4-D at the monitoring stations.

The first "low" concentration weather type is produced by a high pressure system, Figure 5, situated northwest through east of the Yakima Valley monitoring area. This produces north through east winds throughout the region. Skies are generally fair and wind speeds are relatively light. This is similar to air pollution "episode" weather.

The second "low" concentration weather pattern is associated with a weak low pressure trough in eastern Washington and Oregon and thus is somewhat similar to the "high" concentration weather pattern. The important difference is that there is no surface front in the area. The absence of a frontal system usually results in clear sky and no organized wind flow from the south or the southwest. The surface winds are relatively light and variable—thus, no significant transport wind movement. Sky conditions are similar to the high pressure situation in that mostly sunny skies prevail. Apparently good vertical mixing accompanies both of the low concentration situations.

In comparing the three major weather patterns identified as a result of the Washington State University research program, certain conclusions can be reached. The mechanical mixing produced by the low level winds appeared to be the only significant 2,4-D dispersion mechanism for both the "high" concentration and "moderate" concentration weather patterns. However, in both of the "low" concentration weather patterns, convection, which would be expected to produce a deep mixing layer, appears to be the main dispersion mechanism, since relatively sunny days with light winds accompany these situations.

It is widely recognized that the typical urban or regional high concentration air pollution episode is associated with a high pressure system accompanied by weak winds, a subsidence inversion and sunny skies, whereas the primary pattern for our particular (2,4-D) episodes is associated with low pressure, cloud cover, and a well defined wind flow pattern. The high pressure system in the Lower Yakima Valley produced a deep mixing layer giving way to good dilution of pollution. We concluded from this comparison of typical synoptic conditions that transport rather than local sources, was an important part of the overall 2,4-D impact system.

In commenting on these weather models for 2,4-D incidents in central Washington, Decker and Hennessey⁸ have argued for more emphasis to be placed on local or close-by source

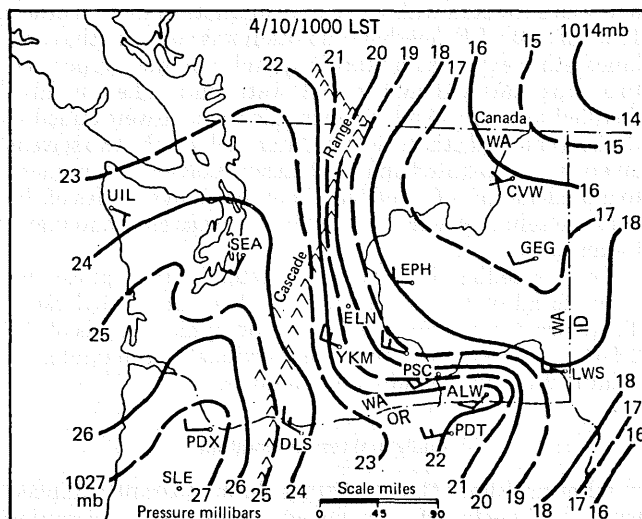


Figure 4. Typical synoptic weather pattern for moderate concentration levels of 2,4-D.

areas rather than long distance transport. It seems necessary to invoke the transport phase in order to correlate observed increased air concentrations with the windier, low pressure situations and lower concentrations with weak winds and high pressure situations.⁶

Aerial Application Experiments

Attempts to reduce or eliminate damage from 2,4-D drift characteristically take the form of legal regulations on chemical formulations and application methods. These actions have been based on research from a number of investigators.⁹⁻¹¹ To meet the apparent needs of the industry in Washington, it was decided to carry out a program of aerial application experiments. It is expected that the results of these experiments, which will be done over a period of several years, will serve as useful guides to the establishment of effective and efficient pesticide usage and legal regulations. A review of available data has shown in general that the Washington program was not a duplication of other research and that the results should be generally useful in the pesticide regulatory program.

The research effort involved two phases: 1) the design and construction of an experimental facility and 2) the aerial application research studies.

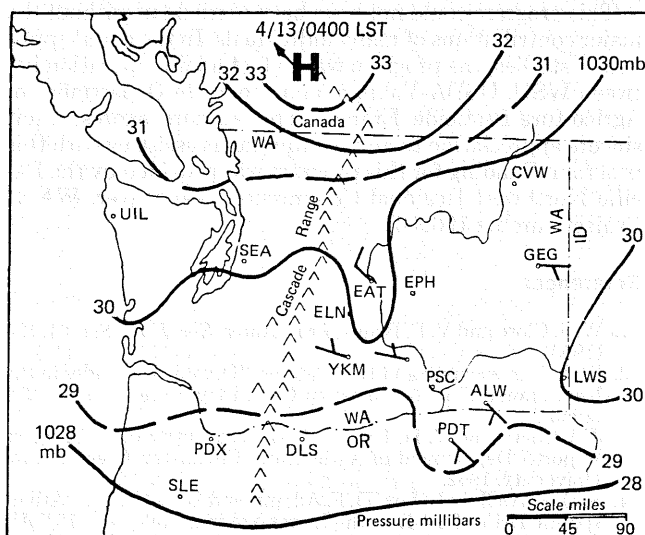


Figure 5. Typical anticyclonic weather pattern characterized by low concentration levels of 2,4-D.

The facility for aerial application research consisted of a pattern of air samplers and ground deposition plates. These were set up in a pattern to measure the on-target or swath deposition and the off-target deposition and airborne drift out to a distance of 650 m from the swath center line. Figure 6 shows the general design of the drift sampling grid.

The grid network in 1976 consisted of 114 deposition plates, 100 spaced at 1 ft intervals across the aircraft flight line and 14 at locations where air samples were taken. The air sampling included 12 sampling stations where samples were taken at a height of 4 ft above the ground and three towers where vertical concentration profile data could be obtained. A total of 26 air samplers were used. The three towers included two 70 ft towers at 225 m and 500 m from the flight line and one 110 ft tower at 600 m. The 70 ft towers had samplers at heights of 4, 15, 40, and 70 ft while the 110 ft tower had samplers at these heights plus a fifth sampler at 110 ft. The sampling grid was set up on a west to east line so that the expected west winds at the site would provide suitable conditions for drift experiments.

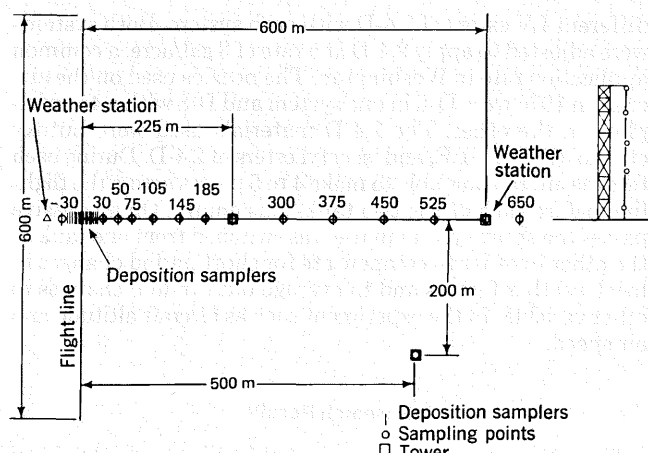


Figure 6. The schematic design of the 2,4-D applications grid.

Weather measurements were made at two locations, just up wind, i.e., west, of the flight line and 600 m downwind at the 110 ft tower. These measurements included surface wind and vertical temperature gradients. The station near the flight line was a micrometeorological recording station provided by the USDA-Yakima Research Center.

The three towers were designed so that they could be assembled on the ground and then tilted into place using a gin-pole and a portable winch. The tower samplers were hoisted up the towers using a simple rope and pulley system.

The air samples were obtained using the same XAD-resin adsorption and gas chromatographic analysis system as was used in the air monitoring program. Air flow through the samplers was obtained by attaching each sampler to one of two central manifold lines within which the necessary vacuum was maintained by two centrally located pumps. Portable electrical generators were used to power the pumps.

The application aircraft used in the experiment was provided by USDA-Yakima Research Center. It was a Pawnee equipped with dual tanks and spray boom systems.

The deposition plate samplers were provided by USDA-Yakima and consisted of numbered 3 by 6 in. stainless steel plates slipped into small stands so that the plate was held level and about 1 ft off the ground. The plates were carried in closed boxes to the laboratory, washed with solvent and the solvent analyzed for 2,4-D compounds by gas chromatography.

The experimental test plan called for the spray aircraft to be equipped with two different nozzle systems and to apply

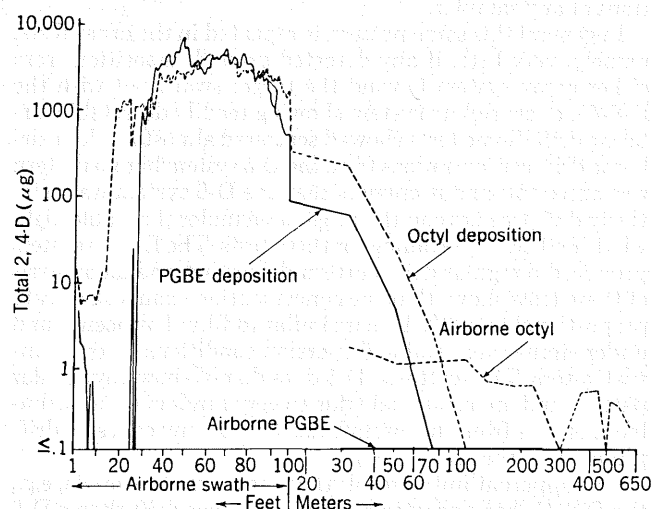


Figure 7. Deposition and airborne drift results from Flight No. 8, October 28, 1976.

different LV esters of 2,4-D with each system. Both systems were adjusted to apply 2,4-D at a rate of 3 gal/acre, a common application rate in Washington. The nozzles used on the aircraft in 1976 were D-5 in one system and D-8 with a 45 swirlplate in the other. The 2,4-D materials used were butoxy ethanol (BE), PGBE, and isooctyl esters of 2,4-D. During each test the aircraft was able to make 4 to 5 passes across the flight line before each of the two tanks was empty. On successive passes the spray system in use was switched from one tank to the other in order to compensate for short period changes in local weather factors and to average out random changes in other variables in the experiment such as aircraft altitude and air speed.

Research Results

Even though in the first operational season only seven potentially useful tests were flown, this research study has produced a considerable amount of data. We have concentrated our assessment on the airborne drift aspects.

Specific details for an example test flight are given below:

Flight 8: October 28, 1976

Application time: 0753-0804

Application altitude: 8-12 ft

Estimated weather: Wind, west/3-6 mph

Temperature, -1° to 0°C at 4 ft height stable turbulence regime.

Spray system: Tank No. 1, D8-45 nozzles with octyl ester 2,4-D

Tank No. 2, D5 nozzles with PGBE 2,4-D

As indicated above Flight 8 used two spray nozzle systems and switched from one to the other on alternate passes over the application swath. Four passes were flown with each system.

Figure 7 shows the data for Flight 8 and can serve as a basis for a discussion of the type of conclusions that will be coming from these tests. First, with regard to deposition on the flight line "target" area, both the D-5, or octyl, and the D-8-45, or PGBE system showed about the same degree of performance. In the downwind area, as well, the ground deposition was not detectable beyond 75 m for the D-8-45 system and 50 m for the D-5 system. However, as shown in Figure 7 a major and apparently significant difference between the systems occurred in the off-target airborne concentration measurements. Here the D-8-45 system produced readily measurable drift to 650 m, the far end of the sampling grid and the concentrations decrease with distance in a regular manner. In contrast, the D-5 system did not produce measurable airborne concentrations at any sampler.

In general this same pattern is repeated in the other tests, namely, very little if any detected ground deposition from either spray system beyond the target swath but with the D-8-45, or swirlplate system showing readily detectable airborne drift. Some tests showed scattered almost random airborne drift concentrations from the D-5 system but no pattern was apparent, and it appears that the D-5 system was relatively drift free beyond the target area under the stable, light wind conditions prevailing for these tests. The D-8-45 system produced a regular drift pattern downwind and an analysis of these tests shows that the concentration changes at a rate proportional to $x^{-0.6}$. This is similar to what has been found under similar very stable dispersion conditions by other investigators. The scattered D-5 data do not show any regular pattern and are most likely due to very random wake turbulence effects from the aircraft rather than any cohesive drift plume from the application.

The apparent initial result that the swirlplate nozzle, e.g., the D-8-45, has a greater potential to cause drift than a D-5 is not a new finding and in fact confirms results gathered by a number of other investigators. The lack of drift for the D-5

system may be due to the weak winds and the strong, early morning stability, or to the test procedures. Yates, *et al.*¹¹ found measurable drift with D-6 systems using 1 m² deposition targets during unstable mid-day conditions. The results in Washington, if confirmed by the more detailed assessment of the rest of the flight data, are important because they show that these two nozzle systems, the D-5 and the D-8-45, legal in Washington for 2,4-D application, have significantly different drift potential.

Conclusions

This research program has contributed useful information towards understanding 2,4-D problems in Washington. The monitoring data from the lower Yakima Valley show that high volatile 2,4-D is prevalent in the area. Grape damage and airborne concentrations tend, in recent years, to be more isolated situations than in prior seasons.^{5,6} This may indicate more close-in or local problems during times when weather does not favor long distance drift. The research on the drift potential of D-5 and D-8-45 spray systems at 3 gal/acre application rates showed that the D-8-45 system had significantly more off-target airborne drift. However, much more work must be done in this phase of the research.

Acknowledgments

This program could not have been carried out without the major contributions of many individuals. In the aerial application studies, the program was in fact a joint operation between WSU, USDA-Yakima, and the State Department of Agriculture-Pesticide Branch. The authors express their sincere appreciation to all the individuals assisting with this program. Funding for this program was provided by the Pacific Northwest Regional Commission, Vancouver, WA as Project number 1069044.

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