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(54) Titre : MICROEMULSIONS NON AQUEUSES A PHASE HUILEUSE CONTINUE PULVERISABLES ET LEURS  
 PROCEDES DE PRODUCTION  
 (54) Title: SPRAYABLE NON-AQUEOUS, OIL-CONTINUOUS MICROEMULSIONS AND METHODS OF MAKING SAME

(57) **Abrégé/Abstract:**

Non-aqueous, oil-continuous microemulsions and methods of making such microemulsions are provided. The non-aqueous, oil-continuous microemulsions comprise at least one oil component, at least one non-aqueous polar solvent component, at least one amphiphilic material component, and at least one solubilized polar compound component. The solubilized polar compound can be a polar-acidic agrochemical complex such as glyphosate. The microemulsions can be applied by either conventional mechanical, non-electrostatic or by electrostatic LV or ULV techniques, such as, for example, electrohydrodynamic spraying, and exhibit enhanced pesticidal activity.

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(54) Title: SPRAYABLE NON-AQUEOUS, OIL-CONTINUOUS MICROEMULSIONS AND METHODS OF MAKING SAME

(57) Abstract: Non-aqueous, oil-continuous microemulsions and methods of making such microemulsions are provided. The non-aqueous, oil-continuous microemulsions comprise at least one oil component, at least one non-aqueous polar solvent component, at least one amphiphilic material component, and at least one solubilized polar compound component. The solubilized polar compound can be a polar-acidic agrochemical complex such as glyphosate. The microemulsions can be applied by either conventional mechanical, non-electrostatic or by electrostatic LV or ULV techniques, such as, for example, electrohydrodynamic spraying, and exhibit enhanced pesticidal activity.

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5     **SPRAYABLE NON-AQUEOUS, OIL-CONTINUOUS MICROEMULSIONS AND**  
          **METHODS OF MAKING SAME**

          The present invention relates to oil-continuous microemulsions and, more particularly, to non-aqueous, oil-continuous microemulsions and methods of making same that are sprayable via electrostatic and non-electrostatic means.

10           Many chemical formulations used for crop protection and pest control, such as fungicides, herbicides, insecticides, acaricides, miticides, molluscicides, nematocides, rodenticides, plant-desiccants, plant-growth regulators, etc. (pesticides in general), are sprayable aqueous liquids that cover a target area (e.g., the surface of a plant) to effect their intended purpose. More specifically in  
15           some applications, an agricultural pesticide (either single or in combination with other compounds) needs to be in the liquid phase in order for it to penetrate the leaf cuticle of a target weed or plant or the cuticle of a target insect or similar pest. In order for the sprayed formulation to remain on the target, it needs to wet the surface. However, plant leaves often have waxy surfaces and both plants and  
20           insects can have tiny hairs that make it difficult for aqueous sprays to stick. Consequently, surface-active agents (e.g., wetting agents, surfactants, and other amphiphiles) are often utilized. These materials lower the interfacial tension between the sprayed aqueous liquid and the surface of the target, and permit the aqueous spray to stay on the target. Additionally, the formulation(s) may contain  
25           other ingredient(s) like spreaders, stickers, humectants, emollients, etc.

          In sprayed formulations that contain a contact herbicide, the active agent does not need to translocate to be effective. Alternatively, in systemic herbicidal formulations, the agent must penetrate the leaf cuticle and translocate (i.e., be transported) within the plant to the site where it may exert its intended biological  
30           function. PMG acid (N-phosphonomethylglycine or "glyphosate") and other similar acidic herbicides (e.g., glufosinate and 2,4-D) are known systemic herbicides. These materials, however, are insoluble solids. Several compounds of PMG have been prepared that are water-soluble and that lend themselves to application by

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5 mechanical spraying on the target vegetation. With the aid of special surface-  
active materials (e.g., alkylamine ethoxylates and propoxylates, alkyl  
polyglucosides, etc.), formulations of PMG are able to penetrate the leaf cuticle  
and become effective herbicides. Several commercial formulations of PMG  
10 compounds, which are intended as systemic herbicides, are available (i.e.,  
Roundup® herbicide available from Monsanto Company, St. Louis, MO and  
Touchdown® herbicide available from Syngenta Crop Protection, Greensboro,  
N.C.). The following U.S. patents and international application disclose PMG and  
other water-soluble herbicidal formulations: U.S. 3,977,860 and 3,948,975 to  
15 Franz; U.S. 5,668,085 to Forbes et al.; U.S. 5,300,680 and 5,410,074 to Jones et  
al.; U.S. 5,658,853 to Kassebaum et al.; U.S. 5,565,409 to Sato et al.; and WO  
01/89302 A2 by Lennon et al.

Water, which is used as a solvent/diluent or carrier in the above-referenced  
systems, has its volatility as a limiting factor. As such, the full potency of the  
herbicide may not be utilized. This is due to the fact that in an aqueous spray, the  
20 water may evaporate and a significant portion of the sprayed herbicide could be  
left (as solid) on the leaf surface. Furthermore, mechanical spraying of liquid  
formulations generates droplets of various sizes ranging from over 200 microns  
down to less than 10 microns. Larger droplets may hit the leaf surface and  
fragment and/or bounce off. The smaller droplets (less than 50 microns) can drift  
25 away or may dry or volatilize before contacting the leaf surface. The result is a  
lowering in the efficacy of the herbicide employed.

Non-volatile oils have been used for general or non-electrostatic, low-  
volume ("LV") and ultra-low volume ("ULV") application of herbicides. However,  
PMG and other similar herbicidal compounds are insoluble in such oils. In an  
30 attempt to solve such insolubility issues, water-in-oil emulsions containing water-  
soluble herbicides have been used for general mechanical as well as electrostatic  
LV and ULV application. (See, i.e., GB 2 022 418 A to Lawrence, Coffee and  
Robert). These formulations, however, still suffer from the volatility of the water

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5 droplets dispersed in the oil, as a large portion of the applied or sprayed active agent either remains on the leaf surface as a dry residue or drifts/diffuses away as a fine mist. Moreover, such water-in-oil emulsions are impractical for commercial spray application given that they are prone to separation upon standing and are highly viscous.

10 Accordingly, there is a recognized need for improvements in sprayable formulation design.

The present invention meets the above-mentioned need by providing sprayable, non-aqueous, oil-continuous microemulsions and methods of making such microemulsions, which contain a formulation with a desired biological  
15 function.

Although the present invention is not limited to specific advantages or functionality, it is noted that non-aqueous, oil-continuous microemulsions provide essentially non-volatile solvents for polar-acidic agrochemical complexes such as, for example, a PMG compound. By "essentially non-volatile" we mean that the  
20 vapor pressure of the solvents employed in the present invention is less than that of solvents containing water. Additionally, non-aqueous polar liquids are better solvents for PMG compounds than water, and the resulting solutions can contain up to 60% or more by weight acid equivalent. These completely non-aqueous solutions circumvent the volatility problems of known aqueous formulations of  
25 PMG. The latter solutions are used to prepare the non-aqueous, oil-continuous microemulsions of the present invention. The resulting microemulsions may be applied by either conventional mechanical, non-electrostatic or by electrostatic LV or ULV techniques. By "electrostatic techniques", we mean both conventional electrostatic spraying; and electric field spraying also known as electric field effect  
30 technology (EFET). EFET includes: electrohydrodynamic aerosolization (EHD) (also referred to herein as electrohydrodynamic spraying); and electrospinning; and any other conceivable technique that is effective in producing an electric field

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5 induced cone-jet or Taylor cone for application of particles or material to a target. These microemulsion formulations exhibit enhanced herbicidal activity.

As illustrated in further detail herein, post-emergence herbicidal spray compositions prepared in accordance with the present invention as non-aqueous, oil-continuous microemulsions show superior biological efficacy (i.e., long term  
10 weed control) with a reduction in the active ingredient dose applied when compared to conventional water-based formulations of PMG (i.e., Roundup® ULTRA MAX) in both glass-house and randomized, multiple-plot field trials. In contrast to such water-based formulations of PMG, the herbicidal performance exhibited by the non-aqueous formulations of the present invention is impervious  
15 of weed growth stage. Moreover, the onset of visual symptoms of herbicidal activity, including necrosis, is much more rapid and obvious when employing the non-aqueous, oil-continuous microemulsions of the present invention, as compared to conventional aqueous formulations of PMG. Without wishing to limit the invention to any particular theory or mechanism of operation, it is believed that  
20 the addition of particular oils, such as, for example, alkyl esters of fatty acids (i.e., methyl oleate), can have the effect of increasing uptake and translocation of certain solubilized polar herbicidal compounds within the plant, and thus improve long term weed control.

In one embodiment of the present invention, a non-aqueous, oil-continuous  
25 microemulsion is provided comprising at least one oil component, at least one non-aqueous polar solvent component, at least one amphiphilic material component, and at least one solubilized polar compound component. The solubilized polar compound can be a polar agrochemical complex and, more particularly, a polar-acidic agrochemical complex, such as, for example, a  
30 glyphosate complex, a 2,4-D complex, a glufosinate complex, and combinations thereof. As mentioned above, the oil component can be an alkyl ester of a fatty acid, such as, for example, methyl oleate.

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5           In another embodiment of the present invention, a method for solubilizing a polar compound into a non-aqueous, oil-continuous microemulsion is provided. The method comprises providing a polar portion, mixing an amphiphilic material (i.e., a cationic surfactant) with the polar portion to produce an intermediate mixture, and adding an oil component to the intermediate mixture, together with  
10 vigorous mixing. The polar portion can be prepared by: (a) providing a non-aqueous, polar solvent; (b) providing an insoluble, solid polar compound and combining the polar compound with the non-aqueous, polar solvent to form a reaction mixture; and (c) providing a compound having an amine, sulfonium, sulfoxonium, amide or ester functional group and adding it to the reaction mixture  
15 to form a salt, amide or ester of the polar compound, which forms a stock solution of a soluble salt, amide or ester of the polar compound by mixing and/or heating the reaction mixture. Optionally, a second amphiphilic material, such as a non-ionic surfactant, can be added together with vigorous mixing to form the non-aqueous, oil-continuous microemulsion of the present invention.

20           In yet another embodiment of the present invention, a post-emergence herbicidal spray composition comprising a non-aqueous, oil-continuous microemulsion is provided. The microemulsion comprises at least one oil component, at least one non-aqueous polar solvent component, at least one amphiphilic material component, and at least one solubilized polar compound  
25 component. The solubilized polar compound component can comprise a polar-acidic agrochemical complex selected from a glyphosate complex, a 2,4-D complex, a glufosinate complex, and combinations thereof.

          In still another embodiment of the present invention, a method of controlling weeds is provided which comprises applying a post-emergence herbicidal spray  
30 composition to a target plant. The composition comprises a non-aqueous, oil-continuous microemulsion comprising at least one oil component, at least one non-aqueous polar solvent component, at least one amphiphilic material component, and at least one solubilized polar compound component that

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5 comprises a polar-acidic agrochemical complex selected from a glyphosate complex, a 2,4-D complex, a glufosinate complex, and combinations thereof. The step of applying the post-emergence herbicidal spray composition to the target plant can be performed by either conventional mechanical, non-electrostatic or by electrostatic low-volume or ultra-low volume techniques.

10 In yet still another embodiment of the present invention, a method of improving the uptake and translocation of a systemic herbicidal compound in a target plant is provided which comprises applying a post-emergence herbicidal spray composition to the target plant. The composition comprises a non-aqueous, oil-continuous microemulsion comprising at least one oil component, at least one  
15 non-aqueous polar solvent component, at least one amphiphilic material component, and at least one solubilized polar compound component that comprises a polar-acidic agrochemical complex selected from a glyphosate complex, a 2,4-D complex, a glufosinate complex, and combinations thereof. The oil component, which can comprise methyl oleate, affects the improved uptake  
20 and translocation of the solubilized polar compound in the target plant.

In yet still another embodiment of the present invention, a post-emergence herbicidal spray composition comprising a non-aqueous, oil-continuous microemulsion is provided. The microemulsion is characterized by a pH of less than 7.00 at about 22°C.

25 In yet still another embodiment of the present invention, a method of controlling weeds is provided which comprises applying a post-emergence herbicidal spray composition to a target plant. The composition comprises a non-aqueous, oil-continuous microemulsion that is characterized by a pH of less than 7.00 at about 22°C. The step of applying the post-emergence herbicidal spray  
30 composition to the target plant is performed by either conventional mechanical, non-electrostatic or by electrostatic low-volume or ultra-low volume techniques.

In yet still another embodiment of the present invention, a post-emergence herbicidal spray composition comprising a non-aqueous, oil-continuous



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5 microemulsion is provided. The microemulsion is characterized by an octonal/water partitioning coefficient of between about 2 and about 4  $k_{ow}$ .

In yet still another embodiment of the present invention, a method of controlling weeds is provided which comprises applying a post-emergence herbicidal spray composition to a target plant. The composition comprises a non-  
10 aqueous, oil-continuous microemulsion that is characterized by an octonal/water partitioning coefficient of between about 2 and about 4  $k_{ow}$ . The step of applying the post-emergence herbicidal spray composition to the target plant is performed by either conventional mechanical, non-electrostatic or by electrostatic low-volume or ultra-low volume techniques.

15 In yet still another embodiment of the present invention, a method of controlling weeds is provided which comprises applying a post-emergence herbicidal spray composition to a target plant. The composition comprises a non-aqueous, oil-continuous microemulsion. The step of applying the post-emergence herbicidal spray composition to the target plant is performed by either  
20 conventional mechanical, non-electrostatic or by electrostatic low-volume or ultra-low volume techniques, which deliver a droplet size of between about 5 and about 30 microns to the target plant.

In yet still another embodiment of the present invention, a method of improving the uptake and translocation of a systemic insecticidal or fungicidal  
25 compound in a target plant is provided which comprises applying an insecticidal or fungicidal spray composition to the target plant. The composition comprises a non-aqueous, oil-continuous microemulsion that comprises at least one oil component, at least one non-aqueous polar solvent component, at least one amphiphilic material component, and at least one solubilized polar compound  
30 component that comprises a polar-acidic agrochemical complex. The oil component, which can comprise methyl oleate, affects the improved uptake and translocation of the solubilized polar compound in the target plant.

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5           In yet still another embodiment of the present invention, method of  
controlling insects or fungi is provided which comprises applying an insecticidal or  
fungicidal spray composition to a target plant. The composition comprises a non-  
aqueous, oil-continuous microemulsion, and the step of applying an insecticidal or  
fungicidal spray composition to a target plant is performed by either conventional  
10   mechanical, non-electrostatic or by electrostatic low-volume or ultra-low volume  
techniques, which deliver a droplet size of between about 5 and about 30 microns  
to the target plant.

          In yet still another embodiment of the present invention, a method of  
controlling weeds is provided which comprises applying a post-emergence  
15   herbicidal spray composition to a target plant. The composition comprises a non-  
aqueous, oil-continuous microemulsion. The step of applying the post-emergence  
herbicidal spray composition to the target plant is performed by either  
conventional mechanical, non-electrostatic or by electrostatic techniques, which  
deliver a volume of formulated product to a target plant, at a potential volume rate  
20   ranging from ultra-low volumes (less than about 5 lts/ha) through to high volumes  
(greater than about 500 lts/ha).

          These and other features and advantages of the invention will be more fully  
understood from the following detailed description of the invention. It is noted that  
the scope of the claims is defined by the recitations therein and not by the specific  
25   discussion of features and advantages set forth in the present description.

          The following detailed description of the embodiments of the present  
invention can be best understood when read in conjunction with the following  
drawings, where like structure is indicated with like reference numerals and in  
which:

30           Figure 1 is a plot diagram showing soybean phytotoxicity after a second  
application of two experimental PMG-based formulations;

          Figure 2 is a plot diagram showing *Setaria* control after a second  
application of two experimental PMG-based formulations;

5 Figure 3 is a plot diagram showing broad-leaved weed control after a second application of two experimental PMG-based formulations;

Figure 4 is a plot diagram showing *Setaria* control after a first application of two novel post-emergence herbicidal spray compositions prepared in accordance with the present invention, which are compared to another experimental PMG-  
10 based formulation;

Figure 5 is a plot diagram showing *Setaria* control after a second application of two novel post-emergence herbicidal spray compositions prepared in accordance with the present invention, which are compared to another experimental PMG-based formulation;

15 Figure 6 is a plot diagram showing broad-leaved weed control after a first application of two novel post-emergence herbicidal spray compositions prepared in accordance with the present invention, which are compared to another experimental PMG-based formulation;

Figure 7 is a plot diagram showing broad-leaved weed control after a  
20 second application of two novel post-emergence herbicidal spray compositions prepared in accordance with the present invention, which are compared to another experimental PMG-based formulation;

Figure 8 is a plot diagram showing soybean phytotoxicity after a first application of two novel post-emergence herbicidal spray compositions prepared  
25 in accordance with the present invention, which are compared to another experimental PMG-based formulation;

Figure 9 is a plot diagram showing soybean phytotoxicity after a second application of two novel post-emergence herbicidal spray compositions prepared in accordance with the present invention, which are compared to another  
30 experimental PMG-based formulation;

Figure 10 is a plot diagram showing *Setaria* control after a first application of two novel post-emergence herbicidal spray compositions prepared in

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5 accordance with the present invention, which are compared to Roundup® ULTRA  
MAX;

Figure 11 is a plot diagram showing *Setaria* control after a second  
application of two novel post-emergence herbicidal spray compositions prepared  
in accordance with the present invention, which are compared to Roundup®  
10 ULTRA MAX;

Figure 12 is a plot diagram showing broad-leaved weed control after a first  
application of two novel post-emergence herbicidal spray compositions prepared  
in accordance with the present invention, which are compared to Roundup®  
ULTRA MAX;

15 Figure 13 is a plot diagram showing broad-leaved weed control after a  
second application of two novel post-emergence herbicidal spray compositions  
prepared in accordance with the present invention, which are compared to  
Roundup® ULTRA MAX;

Figure 14 is a plot diagram showing soybean phytotoxicity after a first  
20 application of two novel post-emergence herbicidal spray compositions prepared  
in accordance with the present invention, which are compared to Roundup®  
ULTRA MAX;

Figure 15 is a plot diagram showing soybean phytotoxicity after a second  
application of two novel post-emergence herbicidal spray compositions prepared  
25 in accordance with the present invention, which are compared to Roundup®  
ULTRA MAX;

Figure 16 is a plot diagram showing the rate response of broad-leaved  
weed control after a second application of one novel post-emergence herbicidal  
spray composition prepared in accordance with the present invention, which is  
30 compared to Roundup® ULTRA MAX;

Figure 17 is a plot diagram showing the speed-of-visual effect of *Setaria*  
control after a first application of one novel post-emergence herbicidal spray

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5 composition prepared in accordance with the present invention, which is compared to Roundup® ULTRA MAX;

Figure 18 is a plot diagram showing the speed-of-visual effect of total weed control after a second application of one novel post-emergence herbicidal spray composition prepared in accordance with the present invention, which is  
10 compared to Roundup® ULTRA MAX;

Figure 19 is a plot diagram showing the speed-of-visual effect of *Setaria* control in a glasshouse after application of two novel post-emergence herbicidal spray compositions prepared in accordance with the present invention, which are compared to Roundup® ULTRA MAX;

15 Figure 20 is a plot diagram showing the speed-of-visual effect of *Abutilon* control in a glasshouse after application of two novel post-emergence herbicidal spray compositions prepared in accordance with the present invention, which are compared to Roundup® ULTRA MAX;

Figure 21 is a photographic representation of soybean phytotoxicity and  
20 general weed control four days after a first application of one novel post-emergence herbicidal spray composition prepared in accordance with the present invention, which is compared to another experimental PMG-based formulation;

Figure 22 is a photographic representation of soybean phytotoxicity and  
25 general weed control four days after a first application of one novel post-emergence herbicidal spray composition prepared in accordance with the present invention, which is compared to an untreated control;

Figure 23 is a photographic representation of soybean phytotoxicity and  
general weed control four days after a first application of another novel post-emergence herbicidal spray composition prepared in accordance with the present  
30 invention, which is compared to Roundup® ULTRA MAX;

Figure 24 is a photographic representation of soybean phytotoxicity and  
general weed control three days after a second application of one novel post-

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5 emergence herbicidal spray composition prepared in accordance with the present invention, which is compared to Roundup® ULTRA MAX;

Figure 25 is a photographic representation of general weed control 31 days after a first application of another novel post-emergence herbicidal spray composition prepared in accordance with the present invention; and

10 Figure 26 is a photographic representation of early symptom expression in a glasshouse within three hours of applying one novel post-emergence herbicidal spray composition prepared in accordance with the present invention, which is compared to an untreated control.

Emulsions are dispersions of tiny distinct and discrete droplets of one liquid  
15 in another. More specifically, oil-in-water emulsions contain a fatty or oily material that is dispersed in water or in a very polar liquid (e.g., in polyol or glycol). The material inside of emulsion droplets does not move in and out of the droplet. Emulsions form when mechanical energy is applied to affect the dispersion of one liquid in another. A surfactant or emulsifier in an emulsion serves to slow down  
20 the rate of emulsion separation. Although some emulsions appear to be quite stable, in time all emulsions will break and release the energy (mechanical) that was exerted to prepare them.

Microemulsions, and more particularly water-continuous microemulsions, can be described as a special case of micellar solutions, wherein solvent and  
25 solute molecules are in constant mobility (i.e., thermal motion). In accordance with the present invention, by "micelle" we mean a *spherical* micelle. (In the chemical literature the word micelle has also been used to describe cylindrical as well as flat micelles. Cylindrical and flat micelles are better known, respectively, as hexagonal (or inverse-hexagonal) (nematic) and lamellar (smectic) lyotropic  
30 (i.e., solvent-formed) liquid crystals.) When a certain amount of amphiphilic material (e.g., soap or surfactant) is added to water or other polar liquids (e.g., simple alcohols, glycols and other polyols) the solvating forces of polar solvent molecules act very strongly only on the polar portion or heads of the dissolved

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5 amphiphilic molecules. Above a certain concentration (called the critical micelle concentration or "CMC"), the surfactant or soap molecules self-assemble to form spherical aggregates called micelles. The polar heads of the amphiphilic molecules align to the outside, towards the polar liquid. The hydrophobic portions or tails of the amphiphilic molecules are repelled by the polar molecules of the  
10 solvent and are thus forced to align close to each other and away from the water or polar liquid, towards the core of the spherical aggregates. In a water-continuous microemulsion, oil molecules are dissolved among the hydrocarbon tails of the amphiphiles. The micelles in water-continuous microemulsions are swollen to a much larger size compared to those in simple micellar solutions.

15 Non-polar liquids (e.g., liquid hydrocarbons), as compared to water or other similar polar liquids, are very poor solvents for polar materials. Although non-polar liquids may be able to dissolve very small, impractical amounts of the less polar amphiphiles (i.e., amphiphiles of "low hydrophilic-lipophilic balance"), the solubility of an amphiphile in a non-polar liquid is very greatly aided by the addition  
20 of a very small amount of water or very polar liquid such as a polyol or glycol. (A quantitative measure of the nature of amphiphiles is provided. As originally defined, it is the ratio of the molecular weight of the *polyoxyethylene* portion of the amphiphile molecule divided by the molecular weight of the entire molecule and then multiplied by 20. Thus, the least polar amphiphiles have an HLB value of  
25 less than about 10. The amphiphiles of intermediate polarity have HLB values between about 10 and about 15. The most polar amphiphiles have HLB values above about 15. Some very polar amphiphiles have HLB values as high as about 19.0. The latter have 95% of their composition made up of polyoxyethylene.) The molecules of water or polar solvent, by virtue of their thermal motion and physical  
30 surroundings, aggregate spontaneously into small molecular groupings throughout the system. These polar groupings attract the polar heads of amphiphile molecules, thus forcing them to align away from the bulk of the non-polar liquid or oil. The weak interaction of oil with the hydrocarbon tails of the amphiphiles is

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5 sufficient to solvate them. These resulting aggregates in non-polar liquids are called inverse-micelles. Oil-continuous microemulsions can be described as a special case of inverse-micellar solution. In oil-continuous microemulsions, the water or polar-solvent core of the inverse-micelles may be larger than those of simple inverse-micellar solutions and they may contain a solute.

10 Microemulsions, like micellar and inverse-micellar solutions, are transparent single-phase liquids, which are at thermodynamic equilibrium and therefore indefinitely stable over time. However, microemulsions may not remain stable if their temperature varies over a wide range. Among oil-continuous microemulsions, those that contain non-aqueous polar solvents possess better  
15 temperature stability than those containing water. Contrary to the apparent terminology, microemulsions are not simply emulsions with "micro" droplets inside of them. In microemulsions, the molecules forming the aggregates are continuously moving in and out of the aggregates because of their thermal motion, and microemulsions may contain additional amphiphilic materials. The additional  
20 amphiphiles may be other surfactants or co-surfactants such as pentanol or hexanol.

In accordance with one embodiment of the present invention, a non-aqueous, oil-continuous microemulsion is provided comprising at least one oil component, at least one non-aqueous, polar solvent component, at least one  
25 amphiphilic material component, and at least one solubilized polar compound component. The polar compound, which is solubilized in the non-aqueous, polar solvent, can be a polar agrochemical complex having herbicidal, insecticidal, fungicidal and/or other like properties. The microemulsion can be applied to a substrate, such as the leaves of a plant, by way of conventional mechanical,  
30 aerosol, and electrostatic spray techniques for LV and ULV application of liquid microemulsions containing active compounds such as the polar compound of the present invention.



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5           The oil component of the non-aqueous, oil-continuous microemulsion of the present invention can be a non-volatile oil and typically has a freezing point lower than about 0°C and a boiling point above about 300°C. The oil component can be selected from alkyl esters of fatty acids, fatty alcohols or esters of dicarboxylic acids (e.g., abietic acid, azelaic acid, fumaric acid, phthalic acid, adipic acid, 10 malonic acid, oxalic acid, succinic acid, and carbonic acid), guerbet alcohols, alcohol acetates, petroleum fractions, aliphatic paraffinic light distillates, hydrocarbon oils, vegetable oils, synthetic triglycerides, triethyl phosphate, and combinations thereof. The alkyl esters of fatty acids can be selected from methyl oleate, ethyl oleate, methyl soyate, ethyl soyate, soybean oil, castor oil, and 15 combinations thereof. The hydrocarbon oil is typically an aliphatic hydrocarbon, an aromatic hydrocarbon, or a combination thereof, and can be a branched-chain saturated or unsaturated hydrocarbon having between about 12 and about 20 carbon atoms. More particularly, the branched-chain hydrocarbon can have about 17 carbon atoms. Examples include diesel oil, Isopar® V (a synthetic oligomeric 20 high-purity isoparaffinic solvent), and Exxate® (an alkyl acetate ester), *inter alia*, which are available from ExxonMobil Lubricants & Petroleum Specialties Company, Fairfax, VA.

          The non-aqueous, polar solvent can be selected from an alcohol, an amine, an alkoxyated amine, an amide, a low-molecular weight ester (e.g.,  $\gamma$ - 25 butyrolactone), a nitrile (e.g., benzonitrile), a sulfoxide, sorbitol, urea (in a mixture with the aforementioned), and combinations thereof. The alcohol has more than one hydroxyl group and can be selected from dihydric alcohols, trihydric alcohols, polyhydric alcohols or polyols, and combinations thereof. The dihydric alcohol can be a glycol selected from ethylene glycol, propylene glycol, 1,3-butanediol, a 30 glycol derivative, and combinations thereof. The trihydric alcohol can be a glycerol or a glycerol derivative. The polyhydric alcohol or polyol can comprise the formula  $\text{CH}_2\text{OH}(\text{CHOH})_n\text{CH}_2\text{OH}$ , wherein  $n$  is between 2 and 5. In addition, components of the non-aqueous polar solvent can be, also, a monosaccharide

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5 like glucose or fructose. A disaccharide like sucrose or lactose, also, may be a  
component of the non-aqueous polar liquid. The amine can be selected from  
ethylene diamine, ethanolamine, diethanolamine, triethanolamine, and  
combinations thereof. More specifically, the amine can be tetra(2-  
hydroxypropyl)ethylenediamine or Quadrol®, which is available from BASF  
10 Corporation, Mount Olive, N.J. The amide can be selected from  
dimethylformamide, dimethylacetamide, N-methylpyrrolidone, and combinations  
thereof. The sulfoxide can be dimethylsulfoxide. Preferably, the non-aqueous,  
polar solvent is one that is approved as inert (for example, N-methylpyrrolidone)  
by the United States Environmental Protection Agency.

15 The amphiphilic material can function as a surfactant to reduce the surface  
and interfacial tension between two immiscible liquids, or as an agricultural  
adjuvant to enhance the activity of the active ingredient. The amphiphilic material  
can be selected from cationic surfactants, non-ionic surfactants, quaternary  
surfactants, amphoteric surfactants, zwitterionic surfactants, and combinations  
20 thereof. The cationic surfactant can be an alkylamine having a carbon chain  
length of between 8 and 18 (including unsaturated carbon chains such as an oleyl  
group), an alkoxyated amine having between 8 and 18 carbon atoms, such as  
Ethomeen® S/12 (bis(2-hydroxyethyl)soyaalkylamine), Ethomeen® S/15  
(polyoxyethylene (5) soyaalkylamine), and Ethomeen® S/25 (polyoxyethylene  
25 (15) soyaalkylamine), which are available from Akzo Nobel Surface Chemistry  
LLC, Chicago, IL, and combinations thereof. The non-ionic surfactant can be a  
polyoxyethylene alcohol such as Brij™ 93 (polyoxyethylene (2) oleyl ether) or  
Brij™ 97 (polyoxyethylene (10) oleyl ether), a polyoxyethylene sorbitan fatty acid  
ester such as Tween™ 80 (polyoxyethylene (20) sorbitan monooleate), and  
30 combinations thereof, which are available from Uniqema, New Castle, DE,  
acetylenic and ethoxylated acetylenic diol surfactants such as Surfynol™ 104,  
Surfynol™ 420, Surfynol™ 440, etc., which are available from Air Products and  
Chemicals, Inc. of Allentown, PA, and combinations thereof. In addition, the

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5 amphiphilic material can be selected from alkylamines, alkylamine ethoxylates, alkylamine propoxylates, alkylamine propoxylates-ethoxylates, fatty alcohol ethoxylates, fatty alcohol propoxylates, fatty alcohol propoxylates-ethoxylates, fatty acid ethoxylates, fatty acid propoxylates, fatty acid propoxylates-ethoxylates, synthetic long-chain alcohol ethoxylates, synthetic long-chain alcohol  
10 propoxylates, synthetic long-chain alcohol propoxylates-ethoxylates, synthetic long-chain acid ethoxylates, synthetic long-chain acid propoxylates, synthetic long-chain acid propoxylates-ethoxylates, alkylphenol ethoxylates, alkylphenol propoxylates, alkylphenol propoxylates-ethoxylates, alkylpolyglucosides, sorbitol esters, sorbitan esters, sorbitol ester ethoxylates, sorbitan ester ethoxylates,  
15 polyoxypropylene-polyoxyethylene block copolymers [e.g., BASF's "PLURONICS™"], ethylenediamine-polyoxypropylene-polyoxyethylene block copolymers [e.g., BASF's "TETRONICS™"], and combinations thereof.

The solubilized polar compound can be a polar agrochemical complex and, more particularly, a polar-acidic agrochemical complex, which can be herbicidal in  
20 nature. The equivalent acid content of the solubilized polar compound present in the microemulsion is between less than about 5 and about 20% by weight. Examples of such herbicidal, polar-acidic agrochemical complex include amino acids (e.g., glyphosate and glufosinate), phenoxies (e.g., 2,4-D), and combinations thereof. The glyphosate complex can be selected from a  
25 glyphosate ester, a glyphosate amide, a glyphosate alkylamide, a glyphosate salt, and combinations thereof. The glyphosate salt can be a glyphosate-amine salt, a glyphosate-alkylamine ethoxylate salt, a glyphosate-alkylaminepropoxylate salt, a glyphosate-alkylaminepropoxylate-ethoxylate salt, a glyphosate sulfonium salt, or a glyphosate sulfoxinum salt, *inter alia*. The glyphosate-amine salt can be  
30 selected from glyphosate-diamine salts, glyphosate-polyamine salts, glyphosate-primary, secondary and tertiary amine salts, glyphosate-quaternary ammonium salts, glyphosate-hydroxylamine salts, glyphosate-ammonium salts, glyphosate-long-chain alkylamine salts, and combinations thereof. The glyphosate-primary

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5 amine salt can be selected from a glyphosate-low molecular weight primary amine salt, a glyphosate-high molecular weight primary amine salt, and combinations thereof. Low molecular weight primary amine salts of glyphosate can include those containing up to and including 7 carbon atoms. High molecular weight primary amine salts of glyphosate can include those containing between 8 and 18  
10 carbon atoms, which can be saturated, mono-unsaturated, or poly-unsaturated chains. The glyphosate-low molecular weight primary amine salt can be selected from glyphosate-monomethylamine salts, glyphosate-ethylamine salts, glyphosate-propylamine salts, glyphosate-isopropylamine salts, glyphosate-butylamine salts, glyphosate-amylamine salts, glyphosate-hexylamine salts,  
15 glyphosate-heptylamine salts, glyphosate-ethanolamine salts, and combinations thereof. The glyphosate-high molecular weight primary amine salt can be a glyphosate-fatty amine salt, such as glyphosate-oleylamine salt. The glyphosate-secondary amine salt can be selected from glyphosate-low molecular weight secondary amine salts, glyphosate-high molecular weight secondary amine salts,  
20 and combinations thereof. The glyphosate-low molecular weight secondary amine salt can be a glyphosate-dimethylamine salt, and the glyphosate-high molecular weight secondary amine salt can be a glyphosate-fatty amine salt. The glyphosate-tertiary amine salt can be selected from glyphosate-low molecular weight tertiary amine salts, glyphosate-high molecular weight tertiary amine salts,  
25 and combinations thereof. The glyphosate-low molecular weight tertiary amine salt can be a glyphosate-trimethylamine salt.

In accordance with the present invention, the glyphosate complex can also comprise a glyphosate ester and an alcohol selected from primary alcohols, secondary alcohols, tertiary alcohols, and combinations thereof. In addition, the  
30 glyphosate complex can comprise a glyphosate alkylamide and an amine. The amine can be selected from primary amines, secondary amines, and combinations thereof. The secondary amines can include N-methyl-alkylamines.

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5 In accordance with another embodiment of the present invention, a method for preparing a non-aqueous, oil-continuous microemulsion is presented. The polar portion of the microemulsion, which comprises a polar compound such as a polar-acidic agrochemical complex as described above, in soluble form, plus a non-aqueous, polar solvent, which is also described above, is prepared as  
10 described below.

A typical method of preparing the soluble form of the polar compound (i.e., polar-acidic agrochemical complex such as glyphosate, glufosinate, or 2,4-D) is the *in-situ* method. Here, a predetermined amount of the non-aqueous solvent is weighed into a reaction vessel. The (insoluble) solid polar compound is also  
15 weighed and added into a reaction vessel. If an amine salt of the polar compound is sought, the desired amine is weighed and added to the reaction container. The amine can be selected from simple amines including diamines and polyamines, primary-, secondary-, and tertiary-amines, ammonium and quaternary-ammonium compounds, hydroxylamines, long-chain alkylamines, and combinations thereof.  
20 Sulfonium salts, sulfoxonium salts, amides and esters of the polar compound can also be prepared. An overage (i.e., up to 5% extra) of amine may be used to ensure that variations in amine purity do not become a limiting factor for the neutralization reaction. The reaction may start at room temperature and the mixture may heat up due to the reaction. Typically, the reaction mixture needs  
25 warming and stirring to bring the reaction to completion. When the reaction is complete, the entire amount of solid polar compound will go into solution. This description is typical of the preparation of soluble salts of a polar-acidic agrochemical complex, such as PMG. The most convenient procedure is to make a stock solution of the salt in the non-aqueous, polar solvent.

30 The polar solution may be mixed with an amphiphile. The amphiphile can be any of the amphiphilic materials described above, and typically comprises an alkylamine surfactant (e.g., octylamine, oleylamine), or an alkoxyated amine surfactant such as alkylamine ethoxylates and alkyl-1,3-propanediamine

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5 ethoxylates. A predetermined amount of the amine surfactant is weighed and added into the polar solution, which contains the solubilized polar compound, to form an intermediate mixture. This intermediate mixture may appear slightly turbid. Consequently, it typically requires warming and mixing.

10 The intermediate mixture, described above, and the oil component are mixed together. In one embodiment, a predetermined amount of oil is weighed and a desired amount of the intermediate mixture is added. The combination is then subjected to vigorous mixing to form the oil-continuous microemulsion. When more than one amphiphilic material is desired, a second amphiphilic material can be added to the microemulsion. Typically, this second amphiphilic material is a non-ionic surfactant. In one embodiment, a predetermined amount of such second amphiphilic material is weighed and added to the reaction mixture, which is then subjected to vigorous mixing, to form the non-aqueous, oil-continuous microemulsion, as described herein.

20 The resultant microemulsion could be further diluted with the oils to give formulations containing lower concentrations of the polar compound. However, since these formulations are typically intended for LV and ULV application, further dilution of the oil-continuous microemulsions is not required. In addition, it is contemplated that the manufacturer of the non-aqueous, oil-continuous microemulsions of the present invention will produce formulations that are ready for application by electrostatic spray techniques. However, it is further contemplated that the intermediate mixture described above could be packaged for later dilution by the end user. Methods for making such a concentrate are described in the modular approach below.

30 In accordance with yet another embodiment of the present invention, a post-emergence herbicidal spray composition is provided comprising the non-aqueous, oil-continuous microemulsion described herein. As such, the microemulsion can comprise at least one oil component, at least one non-aqueous polar solvent component, at least one amphiphilic material component,

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5 and at least one solubilized polar compound component. Each of these components are defined herein, however, more specifically regarding the present post-emergence herbicidal spray composition, the solubilized polar compound component can comprise a polar-acidic agrochemical complex selected from a glyphosate complex, a 2,4-D complex, a glufosinate complex, and combinations  
10 thereof, and the oil component can comprise methyl oleate. As illustrated in the examples below, this post-emergence herbicidal spray composition shows superior biological efficacy while reducing the active ingredient dose applied as compared to conventional water-based systemic herbicides, such as Roundup® ULTRA MAX.

15 In accordance with still another embodiment of the present invention, a method of controlling weeds is provided comprising applying the post-emergence herbicidal spray composition that is described directly above to a target plant. Application of the post-emergence herbicidal spray composition can be performed by either conventional mechanical, non-electrostatic or by electrostatic low-volume  
20 or ultra-low volume techniques, which are defined herein.

In accordance with still yet another embodiment of the present invention, a method of improving the uptake and translocation of a systemic herbicidal compound in a target plant is provided which comprises applying a post-emergence herbicidal spray composition to the target plant, that spray  
25 composition comprising the non-aqueous, oil-continuous microemulsion described herein. As such, the microemulsion can comprise at least one oil component, at least one non-aqueous polar solvent component, at least one amphiphilic material component, and at least one solubilized polar compound component that comprises a polar-acidic agrochemical complex selected from a glyphosate  
30 complex, a 2,4-D complex, a glufosinate complex, and combinations thereof.

The oil component, which lyses plant cell membranes, is a known phytotoxic agent when employed as a contact herbicide. However, without wishing to limit the invention to any particular theory or mechanism of operation, it

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5 is believed that the oil component, which can be, for example, an alkyl ester of  
fatty acid (i.e., methyl oleate) also affects the improved uptake and translocation  
of the solubilized polar compound (i.e., the systemic herbicidal compound) in the  
target plant. It is contemplated that the oil component more effectively penetrates  
the hydrophobic cuticle of the plant as compared to a conventional water-based  
10 herbicidal formulation, thereby facilitating the uptake of more systemic herbicide  
into the plant. This is a result of the particle size (e.g., between about 5 and about  
30 microns), pH (slightly acidic), and octonal/water partitioning coefficient (e.g.,  
between about 2 and about 4  $k_{ow}$ ) of the polar-acidic herbicidal compound, which  
enables its transport into the plant phloem (slightly alkaline) more freely, as is  
15 described in further detail below.

In accordance with yet still another embodiment of the present invention,  
the non-aqueous, oil-continuous microemulsion of the present invention can be  
characterized by a pH that is slightly acidic (such as, for example, less than 7.00  
at about 22°C), which enables transport of the solubilized polar compound  
20 component into the phloem of a target plant, which phloem is slightly alkaline.  
There are two proposed mechanisms by which solutes, both natural and  
xenobiotic (e.g., pesticides such as glyphosate) might enter the phloem for  
transport throughout the plant. These mechanisms appear to be species specific.  
In the first mechanism, or symplastic, the solute may enter any cell (e.g.,  
25 mesophyll, parenchymal, sieve element/companion cell) in the transport pathway  
by crossing the cell's membrane. The material is then transported along the  
pathway [Mesophyll → Vein Epidermal Cells → Vascular Parenchymal Cells →  
Sieve Element/Companion Cell Complex (SE-CC)] from cell to cell through  
plasmodesmatal cytoplasmic connections. The driving mechanism for this  
30 transport is both passive concentration (osmotic) and actively (membrane carriers)  
driven. In the second, or apoplastic mechanism, the material is transported from  
cell to cell up to the SE-CC where it is released into the intracellular space (free



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5 space or apoplasm) and then taken up by the SE-CC by active membrane transport mechanisms.

One of the main driving forces for membrane transport here is a proton-co-transport carrier where H<sup>+</sup> ions are driven across the membrane by a pH gradient and carry the solute with them. The gradient may be as much as 2 pH units or  
10 more where the average pH of the xylem and apoplast may be between about 5.5 and about 5.8 or lower, while that of the phloem (SE-CC) is between about 7.8 and about 8.0. This is maintained by active extrusion of K<sup>+</sup> and H<sup>+</sup> ions. A slightly acidic material (+ charged) will therefore have a higher likelihood of transport into the phloem than a neutral or basic moiety. Further, the pKa's of, for  
15 example, glyphosate (2.74, 5.63, and 10.18) would also indicate that at a pH of about 5.5 or lower the molecule would have an overall positive charge.

In accordance with yet still another embodiment of the present invention, the non-aqueous, oil-continuous microemulsion of the present invention can also be characterized by an octanol/water partitioning coefficient of between about 2  
20 and about 4  $K_{ow}$ , which enables transport of the solubilized polar compound component into the phloem of a target plant because it more easily passes through the surrounding membrane. The octanol/water partitioning coefficient, which is usually expressed as a log function, is a measure of the polarity or lipophobicity of a molecule (i.e., the higher the value the more the molecule will  
25 partition into the non-polar octanol). Cell membranes are composed paired lipid sheets with embedded protein and carbohydrate molecules. The non-polar portions of the lipid molecules are turned inward creating a non-polar zone into which only other non-polar molecules would be able to pass. For example, glyphosate by itself has a  $K_{ow}$  of 0.00000381 while the  $K_{ow}$  of methyl oleate, the  
30 adjuvant or oil component added to the present formulation, is about 7.45. Based on  $K_{ow}$  alone, the methyl oleate easily penetrates the membrane or diffuses into a plant leaf's cuticle, while the passage of glyphosate is much slower. Methyl oleate has been shown to increase penetration of herbicides through plant cuticles (see

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5 Santier, S. and Chamel, A. 1996. Weed Research 36(2):167-174). The non-  
aqueous, oil-continuous microemulsion of the present invention therefore aids in  
the penetration of the solubilized polar compound (e.g., glyphosate) into the cells  
of the leaf or the vascular bundles of the target plant, thus shortening the path to  
the phloem and the time for export from the target leaf to the rest of the plant.  
10 Further, the use of the amine salt of glyphosate may add to the amount that  
penetrates into the cell by allowing the compound to utilize the same membrane  
carriers present for amino compounds (e.g., amino acids).

Applying the non-aqueous, oil-continuous microemulsion of the present  
invention in the form of small droplets (e.g., between about 5 and about 30  
15 microns) enables direct penetration of herbicidal compounds into stomata of a  
target plant, and thus faster penetration into the plant. From the stomata, the  
surrounding cells within the sub-stomatal cavity have thinner cuticles and cell  
walls, thus facilitating penetration and enabling faster transport into the mesophyll  
or vascular tissue of the target plant. The size of the stomatal pore, or aperture,  
20 can vary widely between species and can also depend on the physiological state  
of the leaf. Cross sectional diameters of stomates can vary from 2 to 12 microns  
in size with the pore areas in excess of 60 square microns. These sizes may  
allow for direct penetration of droplets smaller than 10 microns into the  
substomatal cavity inside the leaf. The cells which line this area, usually  
25 mesophyll, are responsible for gas (CO<sub>2</sub>/H<sub>2</sub>O) and water exchange for the plant  
and will generally have moistened, thinner cell walls with greatly reduced cuticles  
compared to those on the surface of the leaf. These surfaces will therefore  
present less of a barrier for penetration of the formulation and will be closer to the  
sites of translocation from the leaf (SE-CC of minor veins).

30 It is further contemplated that the oil component of the microemulsion of the  
present invention insulates the systemic herbicidal compound, which has a slight  
negative charge, thus further assisting its penetration of the stoma and  
translocation of the herbicidal compound within the plant. All cell walls are

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5 charged because of their composition which will include proteins, amino sugars,  
and adsorbed ions such as  $\text{Ca}^{++}$  or  $\text{K}^+$ . These will tend to give plant cells a net  
positive charge. This includes the cell walls of the stomatal guard cells which line  
the aperture. This positive charge is often increased through the exudation of  $\text{K}^+$   
important to the osmotically driven opening and closing of the aperture. The cells  
10 may therefore be capable of repelling like charged molecules and small droplets  
from the aperture. Lipid emulsions are essentially neutral in charge and could  
pass these cells with no or little electrostatic interference.

Although the oil component has a contact herbicidal effect, it is also  
contemplated that the relatively small volume of oil component applied to the plant  
15 allows systemic herbicidal action to begin before the uptake system of the plant is  
damaged. By themselves, both systemic herbicides (e.g., glyphosate) and the oil  
component (e.g., methyl oleate) are phytotoxic. However, where oleic acid will  
cause direct damage to cell membranes, it will only damage those cell it comes in  
contact with. In contrast, a systemic herbicide such as, for example, glyphosate  
20 works through disruption of the aromatic amino acid (shikimic acid) pathway of the  
target plant and is capable of intercellular and intracellular transport. Therefore,  
while it may take days to demonstrate symptomology, it will affect a much greater  
area within the plant. The assistance in penetration provided by the small amount  
of oil, (e.g., methyl oleate) in the present formulation could permit a higher "dose"  
25 of the systemic herbicidal compound to enter into the symplasm of the plant  
before the onset of symptomology of the transport system, thus causing greater  
damage to the entire plant and a higher probability of lethality. As a result, as  
illustrated in further detail in the examples to follow, this herbicidal spray  
composition provides superior long term weed control at a lower concentration of  
30 active ingredient than conventional aqueous formulations (i.e., Roundup® ULTRA  
MAX). Moreover, the superior herbicidal performance exhibited by the present  
embodiment is unaffected by weed growth stage and provides visual symptoms of  
herbicide activity more rapidly than such conventional aqueous formulations

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5 containing a systemic herbicide.

In order that the invention may be more readily understood, reference is made to the following examples, which are intended to illustrate the invention, but not limit the scope thereof.

By way of example, Table 1, below, gives the compositions of two solutions  
10 of PMG salts in a non-aqueous solvent.

**Table 1. PMG Salts in Non-Aqueous Solvent**

<b>Ingredient</b>	<b>Weight %</b>		<b>Ingredient</b>	<b>Weight %</b>
Methylamine ( <i>anh.</i> )	10.70		Monoethanolamine	21.67
PMG-Acid	54.98		PMG-Acid	56.33
Propylene glycol	34.32		Propylene glycol	22.00
Total	100.00		Total	100.00

15 For the sake of comparison, the solubility of the isopropylamine salt of glyphosate in water is given as 1050 grams of salt in a liter of water at 25°C and pH 4.3. This is equivalent to 778 grams of equivalent PMG-acid per liter (i.e., per 1000 g water). The weight percent "acid" in this saturated solution would be 37.95%.

20 Several examples of prepared polyol-in-oil microemulsions are presented below. These formulations possess a range of electric and viscosity properties that make them readily sprayable by electrostatic methods. (See "Pesticide Application Methods", G. A. Matthews (Author) – Blackwell Publishers, Second Edition (1992): Electrostatically Charged Sprays – Chapter 9, pp. 202-93.) The  
25 PMG-based formulations were subjected to comprehensive testing for biological efficacy, including glass-house testing and testing in the field.

A PMG-based formulation can be considered superior if additional value is created by such a formulation. To match current standards a 100% "ultimate death" of a target plant (weed) is the minimum requirement, i.e., complete  
30 biological efficacy for a novel PMG-based formulation. Additional value is created

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5 by a novel PMG-based formulation if the speed of kill, both perceived as well as  
 actual time to kill, is accelerated through enhanced biological performance.  
 Additional value is also created if novel formulations reduce the amount of user  
 and environmental exposure through a reduction in the required quantities of  
 active ingredient and/or formulated product required, while simultaneously  
 10 maintaining biological efficacy at least equivalent to current  
 commercial/conventional water-based PMG formulations.

The following tables present the compositions of an assortment of oil-  
 continuous microemulsions. In all cases, the polar portions of these  
 microemulsions are completely non-aqueous and essentially non-volatile liquids.  
 15 Several of these liquids (or a mixture of such liquids), which include propylene  
 glycol, may be utilized as the polar portion of these non-aqueous microemulsions.  
 The oil portion of some of the following microemulsions is methyl oleate or  
 Isopar® V. The latter is a branched-chain saturated hydrocarbon having about 17  
 carbon atoms. Several other oils may be used. These are the same oils that  
 20 have been traditionally used for low-volume and ultra-low volume applications.  
 (See Barlow, F. and Hadaway, A.B. (1974) Some aspects of the use of solvents in  
 ULV formulations. *British Crop Protection Council Monograph 11*, 84-93.)

25 **Table 2. Compositions of Various Oil-Continuous Microemulsions with Non-  
 Aqueous Polar Parts**

<b>A</b>		<b>B</b>		<b>C</b>	
<b>Ingredients</b>	<b>Weight %</b>	<b>Ingredients</b>	<b>Weight %</b>	<b>Ingredients</b>	<b>Weight %</b>
PMG Acid	11.89	PMG Acid	10.20	PMG Acid	10.25
Trimethylamine ( <i>Anh.</i> )	4.26	Methylamine ( <i>Anh.</i> )	1.87	Monoethanolamine	3.92
Propylene Glycol	21.35	Propylene Glycol	7.93	Propylene Glycol	10.83
Ethomeen® S/12	12.50	Ethomeen® S/12	12.50	Ethoduomeen® T/13	22.50
Brij™ 93	16.67	Brij™ 93	16.67	Brij™ 93	7.50
Methyl Oleate	33.33	Methyl Oleate	50.83	Methyl Oleate	45.00
<b>Total</b>	<b>100.00</b>	<b>Total</b>	<b>100.00</b>	<b>Total</b>	<b>100.00</b>

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5 **Table 3. Compositions of Various Oil-Continuous Microemulsions with Non-Aqueous Polar Parts**

A		B		C	
Ingredients	Weight %	Ingredients	Weight %	Ingredients	Weight %
PMG Acid	10.25	PMG Acid	10.25	PMG Acid	10.25
Monoethanolamine	3.92	Monoethanolamine	3.92	Monoethanolamine	3.92
Propylene Glycol	10.83	Propylene Glycol	10.83	Propylene Glycol	10.83
Ethoduomeen® T/13	15.00	Ethoduomeen® T/13	10.00	Ethoduomeen® T/13	25.00
Brij™ 93	20.00	Brij™ 93	40.00	Brij™ 93	25.00
Methyl Oleate	40.00	Methyl Oleate	25.00	Isopar® V	25.00
Total	100.00	Total	100.00	Total	100.00

10 **Table 4. Compositions of Various Oil-Continuous Microemulsions with Non-Aqueous Polar Parts**

A		B		C	
Ingredients	Weight %	Ingredients	Weight %	Ingredients	Weight %
PMG Acid	10.25	PMG Acid	12.24	PMG Acid	7.82
Monoethanolamine	3.92	Methylamine ( <i>Anh.</i> )	2.35	Trimethylamine ( <i>Anh.</i> )	2.80
Propylene Glycol	10.83	Propylene Glycol	9.41	Propylene Glycol	14.04
Ethoduomeen® T/13	15.00	Ethoduomeen® T/13	10.00	Ethomeen® S/15	26.02
Brij™ 93	30.00	Brij™ 93	40.00	Tween™ 80	24.66
Isopar® V	30.00	Methyl Oleate	26.00	Methyl Oleate	24.66
Total	100.00	Total	100.00	Total	100.00

15 **Table 5 Compositions of Various Oil-Continuous Microemulsions with Non-Aqueous Polar Parts**

A		B		C	
Ingredients	Weight %	Ingredients	Weight %	Ingredients	Weight %
PMG Acid	12.23	PMG Acid	10.20	PMG Acid	10.20
Methylamine ( <i>Anh.</i> )	2.36	Methylamine ( <i>Anh.</i> )	1.96	Methylamine ( <i>Anh.</i> )	1.96
Propylene Glycol	9.41	Propylene Glycol	7.84	Propylene Glycol	7.84
Ethoduomeen® T/13	10.00	Ethoduomeen® T/13	10.00	Ethomeen® S/12	12.50
Brij™ 93	40.00	Brij™ 93	40.00	Brij™ 93	16.50
Methyl Oleate	26.00	Methyl Oleate	30.00	Methyl Oleate	51.00
Total	100.00	Total	100.00	Total	100.00

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5 **Table 6. Compositions of Various Oil-Continuous Microemulsions with Non-aqueous Polar Parts**

A		B		C	
Ingredients	Weight %	Ingredients	Weight %	Ingredients	Weight %
PMG Acid	10.20	PMG Acid	10.25	PMG Acid	10.25
Methylamine ( <i>Anh.</i> )	1.96	Monoethanolamine	3.92	Monoethanolamine	3.92
Propylene Glycol	7.84	Propylene Glycol	10.83	Propylene Glycol	10.83
Ethoduomeen® T/13	10.00	Ethoduomeen® T/13	10.00	Ethomeen® S/12	12.50
Brij™ 93	40.00	Brij™ 93	40.00	Brij™ 93	16.50
Methyl Oleate	30.00	Methyl Oleate	25.00	Methyl Oleate	46.00
Total	100.00	Total	100.00	Total	100.00

10 **Table 7. Identification of Ingredients and Components Used in Formulations**

Name	Nature / Structure	Utility	Source
Brij™ 93	Oleyl alcohol/2EO	Surfactant	Uniqema
Brij™ 97	Oleyl alcohol/10EO	Surfactant	Uniqema
Emerest™ 2301	Methyl Oleate	Oil Solvent	Cognis
Ethoduomeen® T/13	Tallow-1,3-propanediamine/3EO	Adjuvant/Surfactant	Akzo-Nobel USA
Ethomeen® S/12	Soya-amine/2EO	Adjuvant/Surfactant	Akzo-Nobel USA
Ethomeen® S/15	Soya-amine/5EO	Adjuvant/Surfactant	Akzo-Nobel USA
Ethomeen® S/25	Soya-amine/15EO	Adjuvant/Surfactant	Akzo-Nobel USA
Isopar® V	Synthetic Isoparaffinic Hydrocarbon	Oil Solvent	Exxon
Isopropylamine salt of PMG: 40% Aq.		Herbicide	Sigma-Aldrich
Methylamine [40% Aq.]		Base	Sigma-Aldrich
Monoethanolamine		Base & Polar Solvent	Sigma-Aldrich
PMG Acid	N-Phosphonomethylglycine	Herbicide	Chem Pacific & "in-house"
Propylene Glycol		Polar Solvent	J. T. Baker
Trimethylamine [45% Aq.]		Base	Fluka
Tween™ 80	Sorbitan Monooleate/20EO	Surfactant	Uniqema

15 A glasshouse trial was performed (under controlled conditions) to determine the biological efficacy of the novel PMG-based formulations of the present invention across a broader spectrum (three species) of commonly available weeds. The following weeds were selected as target species: giant foxtail, *Setaria faberi*, *Herm.*; common ragweed, *Ambrosia elatior*, and velvetleaf, *Abutilon theophrasti*, *Medik.*

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5 Each glasshouse treatment represents a single application of formulated PMG product to four weeds of each species (a total of 12 weeds treated in each treatment). The weeds measured between 10-12 inches high at the time of treatment. At four weeks after treatment all weeds were removed from their containers, the soil was shaken and then washed off the roots and the remaining  
10 biomass was air dried. Once dry, the four weeds from each species were weighed together and the cumulative weight (in grams) was noted for each species in each treatment. The novel PMG formulation results (weights) were compared to conventional glyphosate (Roundup® ULTRA MAX) and to the control (untreated) weeds.

15 Tables 8 and 9 illustrate the effectiveness of the novel PMG formulations compared to commercial water-based formulations and the controls. The compositions of the novel PMG formulations referenced in Tables 8 and 9 are provided in Tables 8A and 9A below.

20 **Table 8: Novel PMG formulations compared to the conventional glyphosate formulation (and untreated weeds) at ~25% of the recommended field rate\***

Formulation Ref.	AI Dose (kg/Ha)	Volume applied per treatment (ml)	Ragweed	Velvetleaf	Giant Foxtail	Total of all 3 weed species
			Total Wt (grams)	Total Wt (grams)	Total Wt (grams)	Total Wt (grams)
D2	0.42	0.33	9.5	3.5	12.7	25.7
B3	0.38	0.15	14.8	4.6	16.4	35.8
K2	0.39	0.18	28.4	19.3	13.1	60.8
Roundup®	0.41	1.90	22.0	25.2	14.6	61.8
Control 1	Nil	Nil	35.8	67.3	186.6	289.7
Control 2	Nil	Nil	42.4	40.8	197.6	280.8

\*\*A sub-optimal rate was used to provide a more accurate indicator of biological effect

25 **Table 8A: Compositions of Various Novel PMG Formulations**

Ingredients	Weight %	Ingredients	Weight %	Ingredients	Weight %
PMG Acid	4.23	PMG Acid	4.09	PMG Acid	4.17
Methylamine (Anh.)	0.81	Isopropylamine (Net)	1.43	Monoethanolamine	1.60



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Propylene Glycol	14.96	Water	8.28	Propylene Glycol	7.56
Ethomeen® S/12	10.00	Ethomeen® S/15	21.56	Ethomeen® S/15	20.83
Brij™ 93	17.00	(This sample has <i><b>NO</b> Brij™</i> )		(This sample has <i><b>NO</b> Brij™</i> )	
Methyl Oleate	53.00	Methyl Oleate	64.64	Methyl Oleate	65.84
Total	100.00	Total	100.00	Total	100.00
<b>D2</b>		<b>B3</b>		<b>K2</b>	

5

10 **Table 9: The top three glasshouse test formulations (of all formulations tested), which showed the greatest degree of biological control (as determined by interim visual observations and the ultimate desiccation of the weeds). Treatments were at ~100% of recommended field rate.**

Formulation Ref	AI Dose (kg/Ha)	Volume applied per treatment (ml)	Ragweed	Velvetleaf	Giant Foxtail	Total of all 3 weed species
			Total Wt (grams)	Total Wt (grams)	Total Wt (grams)	Total Wt (grams)
B2	1.80	0.54	1.7	1.6	3.6	6.9
L3	1.41	0.37	3.5	2.0	5.0	10.5
E1	1.46	0.69	9.5	3.9	1.1	14.5
Control 1	Nil	Nil	35.8	67.3	186.6	289.7
Control 2	Nil	Nil	42.4	40.8	197.6	280.8

15 **Table 9A: Compositions of Various Novel PMG Formulations**

Ingredients	Weight %	Ingredients	Weight %	Ingredients	Weight %
PMG Acid	4.09	PMG Acid	4.08	PMG Acid	4.17
Isopropylamine (Net)	1.43	Methylamine (Anh.)	0.79	Monoethanolamine	1.60
Water	8.28	Propylene Glycol	3.13	Propylene Glycol	7.56
Ethomeen® S/15	21.56	Ethomeen® S/12	6.25	Ethomeen® S/12	10.00
(This sample has <i><b>NO</b> Brij™</i> )		Brij™ 93	15.00	Brij™ 93	19.17
Methyl Oleate	64.64	Methyl Oleate	70.75	Methyl Oleate	57.50
Total	100.00	Total	100.00	Total	100.00
<b>B2</b>		<b>L3</b>		<b>E1</b>	

20 Randomized, multiple-plot field trials were performed on test plots of the annual grass weed giant foxtail, *Setaria faberi*, Heour. Various novel PMG formulations were compared to a commercial standard, Roundup® ULTRA MAX, applied at the normal recommended field rate (100% of recommended label dose

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5 rate), as well as at a sub-optimal rate (50% of recommended label dose rate). A single application (treatment) was made when the weeds were 6-12 inches tall. Observations were made at 4 (4-DAT) and at 14 days after treatment (14-DAT).

10 Tables 10 and 11 illustrate superior field performance of the novel PMG formulations compared to commercial water-based formulations. The compositions of the novel PMG formulations referenced in Tables 10 and 11 are provided in Tables 10A, 10B and 11A below.

**Table 10: Novel PMG formulations showing superior biological performance with a reduction in the active ingredient dose applied (kg/Ha PMG-acid equivalent)**

15

Formulation Reference	AI Dose (kg/Ha)	Visual Observation % Effective Kill	
		(4-DAT)	(14-DAT)
G2	0,50	60%	100%
E2	0,61	60%	100%
E1	1,06	66%	100%
G1	1,09	66%	100%
Roundup® (recommended rate)	2,08	55%	100%
Roundup® (sub-optimal rate)	1,04	33%	100%
Control Plots (untreated)	Nil	0%	0%

**Table 10A: Compositions of Various Novel PMG Formulations**

Ingredients	Weight %	Ingredients	Weight %
PMG Acid	4.17	PMG Acid	4.09
Monoethanolamine	1.60	Isopropylamine (Net)	1.43
Propylene Glycol	7.56	Water	8.28
Ethomeen® S/12	10.00	Ethomeen® S/15	21.60
Brij™ 93	19.17	(This sample has <b><u>NO</u></b> Brij™)	
Methyl Oleate	57.50	Methyl Oleate	64.60
Total	100.00	Total	100.00
Sample E1 (Applied at 100% field dose)		Sample G1 (Applied at 100% field dose)	

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5 **Table 10B: Compositions of Various Novel PMG Formulations**

Ingredients	Weight %	Ingredients	Weight %
PMG Acid	2.08	PMG Acid	4.09
Monoethanolamine	0.80	Isopropylamine (Net)	1.43
Propylene Glycol	3.78	Water	8.28
Ethomeen® S/12	5.00	Ethomeen® S/15	21.60
Brij™ 93	9.58	(This sample has <b><u>NO</u></b> <i>Brij™</i> )	
Methyl Oleate	78.76	Methyl Oleate	64.60
Total	100.00	Total	100.00
Sample E2		Sample G2 (Applied at half flow-rate as G1)	

10 **Table 11: Novel PMG formulations showing superior biological performance with a reduction in the volume of formulated product applied (ml of formulated product applied per square meter)**

Formulation Reference	Volume (ml/sqm)	Visual Observation % Effective Kill	
		(4-DAT)	(14-DAT)
O3	1.25	55%	100%
E1	2.65	66%	100%
G2	1.30	60%	100%
Roundup® (recommended rate)	19.20	55%	100%
Control Plots (untreated)	Nil	0%	0%

**Table 11A: Compositions of Various Novel PMG Formulations**

Ingredients	Weight %	Ingredients	Weight %	Ingredients	Weight %
PMG Acid	10.14	PMG Acid	4.17	PMG Acid	4.09
Methylamine ( <i>Anh.</i> )	1.97	Monoethanolamine	1.60	Isopropylamine (Net)	1.43
Propylene Glycol	7.88	Propylene Glycol	7.56	Water	8.28
Ethomeen® S/12	12.50	Ethomeen® S/12	10.00	Ethomeen® S/15	21.60
Brij™ 93	16.67	Brij™ 93	19.17	(This sample has <b><u>NO</u></b> <i>Brij™</i> )	
Methyl Oleate	50.84	Methyl Oleate	57.50	Methyl Oleate	64.60
Total	100.00	Total	100.00	Total	100.00
<b>O3</b> (Applied at 100% field dose)		<b>E1</b> (Applied at 100% field dose)		<b>G2</b> (Applied at half flow-rate as G1)	

15

In addition, the results of a field trial to assess the performance of electric-field-effect-technology (EFET) applications of the post-emergence herbicide glyphosate are presented. The field trial compared several experimental EFET

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5 glyphosate formulations, each applied by an EFET device, with the commercial Monsanto formulation Roundup® ULTRA MAX, applied conventionally using a knapsack sprayer, at two times of application.

The EFET formulation G, which was provided in accordance with the present invention, was the most efficacious formulation tested. Overall, control of  
10 the predominant weed species on the trial site (*Setaria*, *Abutilon*, and *Ambrosia*) was achieved at 0.5 kg ai/ha at both times of application, at least 2-4 times more efficacious than that provided by Roundup® ULTRA MAX, at volumes of application 20 times lower (10 liters/hectare). The herbicidal performance of EFET formulation G was unaffected by weed growth stage, in contrast to  
15 Roundup® ULTRA MAX where higher rates of application were required to control broad-leaved weeds, especially *Abutilon*, at more advanced growth stages. At these more advanced weed growth stages, EFET formulation G controlled all weeds at 0.3 kg ai/ha, the lowest rate tested, 3-6 times more efficacious than Roundup® ULTRA MAX.

20 The onset of visible symptoms was also much more rapid and obvious, including necrosis, than that for Roundup® ULTRA MAX. It is contemplated that improvements to the experimental EFET device, and optimization of the novel PMG-based formulation G, are likely to result in further improvements in herbicidal performance.

25 Two experimental EFET-compatible formulations (M1 and N1) were less aggressive in producing early herbicidal symptoms and less efficacious for long-term weed control than G. The two formulations M1 and N1 were broadly similar in performance at the first time of spray application (T1) and only M1 was selected for further evaluation at the second time of application (T2). At T2, the 10% M1  
30 formulation was diluted with an appropriate solvent to produce a 7.5% formulation, M2, which, surprisingly, significantly improved weed control. M2, however, was significantly less efficacious than the formulation G.

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5 The field trial site was sown to the Roundup Ready® soybean variety 03422310RR; all of the EFET formulations described below were phytotoxic to the crop and would be unsuitable for use on Roundup Ready® crops.

## **MATERIALS AND METHODS**

### **10 The Trial Site**

The site selected for the field trial, on the farm of Mr. Ralph Yeager, Somersville, Ohio 43340, USA, was a conventionally-tilled site, previously cropped to wheat, planted with Roundup Ready® soybeans (Var. 03422310RR) at 7" row spacings.

15 The scientific rationale for selecting such a site in preference to a non-crop site was to try to separate the herbicidal effect of the formulation adjuvants in the novel formulations (to which Roundup Ready® crops have no bio-engineered resistance) from those of the active ingredient glyphosate (to which Roundup Ready® crops have strong bio-engineered resistance).

20 The trial site was also selected because the predominant weeds:

- *Setaria faberi* (Giant Foxtail),
- *Ambrosia elatior* (Common Ragweed), and
- *Abutilon theophrasti* (Velvetleaf)

were the weed species used in the preceding glasshouse screening experiments  
25 because of their commercial importance.

### **The Experimental Treatments**

The experimental design was a randomized complete block with three replicates. There were 60 plots in each block. The plot size was 15 ft long by 10  
30 ft wide. The width actually sprayed varied with EFET formulation and was measured for each treatment to determine accurate application rates; it always provided a wide guard area from neighboring plots.

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5 In total, 36 experimental EFET formulations were included in the field trial, comparing 10 different formulation families.

To simplify presentation, the less efficacious or less developed formulations have been excluded; only 12 EFET formulations are reported representing seven EFET formulation families. This abbreviated treatment list is described in Table  
10 12 below.

Each candidate EFET formulation was applied using an experimental EFET device at various rates of application and, usually, at two times of application to evaluate the effect of weed growth stage on herbicidal performance.

The EFET formulations were compared to a modern, conventional  
15 glyphosate formulation, Roundup® ULTRA MAX commercialized by Monsanto, applied at the recommended field rate to control the predominant annual weeds, and at one half and at one quarter this rate.

20 **Table 12. The Treatment List**

Formulation Family	Formulation Code	% ai+	Method of Application	ai Rate (kg/ha)	Total Spray Volume (l/ha)	Time of Application
1	G	4.0	EFET	1.087	27.2	1
1	G	4.0	EFET	0.503	12.6	1
1	G	4.0	EFET	1.066	26.7	2
1	G	4.0	EFET	0.473	11.8	2
1	G	4.0	EFET	0.348	8.7	2
2	E	4.0	EFET	1.060	26.5	1
2	E	2.0	EFET	0.610	30.5	1
2	E	4.0	EFET	1.496	37.4	2
2	E	4.0	EFET	0.626	15.7	2
2	E	4.0	EFET	0.449	11.2	2
3	F	1.5	EFET	0.977	65.1	1
3	F	1.5	EFET	0.550	36.7	1
3	F	3.5	EFET	1.253	35.8	2
3	F	3.5	EFET	0.638	18.2	2
3	F	3.5	EFET	0.316	9.0	2
6	N2	7.5	EFET	1.216	16.2	1
7	M1	10.0	EFET	1.043	10.4	1
7	M1	10.0	EFET	0.994	9.9	2
7	M1	10.0	EFET	0.615	6.2	2
7	M1	10.0	EFET	0.325	3.3	2
8	M2	7.5	EFET	1.205	16.1	1

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8	M2	7.5	EFET	0.964	12.9	2
8	M2	7.5	EFET	0.668	8.9	2
8	M2	7.5	EFET	0.565	7.5	2
9	N1	10.0	EFET	1.148	11.5	1
17	ULTRA MAX*	1.1	Conventional	2.082	192.3	1
17	ULTRA MAX*	0.6	Conventional	1.043	192.3	1
17	ULTRA MAX*	0.3	Conventional	0.521	192.3	1
17	ULTRA MAX*	1.1	Conventional	2.082	192.3	2
17	ULTRA MAX*	0.6	Conventional	1.043	192.3	2
17	ULTRA MAX*	0.3	Conventional	0.521	192.3	2
-	Untreated	-	Untreated	-	-	-
-	Untreated	-	Untreated	-	-	-
-	Untreated	-	Untreated	-	-	-

5 + Refers to concentration of ai in applied spray.

\* Commercial glyphosate formulation Roundup® ULTRA MAX (50.2% glyphosate, N (phosphonomethyl) glycine as isopropylamine salt, 49.8% other ingredients).

10 At the first time of application (T1), for practical reasons, rates of application (kg ai/ha) for the EFET formulations were varied either by dilution with appropriate formulation components or by varying walking speed. (Table 12, above, describes the spray concentrations used.)

At the second time of application (T2), rates of application of all EFET formulations were varied only by walking speed.

15 At both times of application, the actual rate of application was very accurately determined by measuring the plot area actually sprayed and the actual volume of spray used.

### The Prevailing Conditions

20 The first spray application was made when the soybean crop was in the V-6 to V-7 stage and the weeds were, on average, 12-14 inches tall. The second application was made two weeks later when the soybean crop was at the R-1 stage and the weeds were, on average, 14-21 inches tall.

25 The conditions at the time of commencement of spraying on both application dates are described below and in Table 13 below.

**Table 13. The Prevailing Conditions**

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	Fist Application	Second Application
Commence Spraying	11:00 a.m.	9:30 a.m.
Air Temperature (Start)	78.3°F	81.5°F
Air Temperature (Finish)	Low 90s °F	Low 90s °F
% Relative Humidity (Start)	64	43
Wind Velocity	4-15 mph	6-15 mph
Soil Type	Clay loam	Clay loam
Soil Temperature	71.2°F	74.6°F
Soil Moisture	Dry	Dry
% Cloud Cover	25%	10%

5

After both applications, the weather continued to be dry with the temperature in the low 90s °F for prolonged periods.

The gusty winds, up to 15 mph at both applications, were considered marginal-severe with respect to crop spraying guidelines to minimize spray drift.

10

No fertilizer or maintenance pesticides were applied to the trial area throughout the trial to avoid any possible chemical interactions.

### The Spray Devices

The experimental EFET sprayer contained many novel features that are described in a series of PCT International Patent Applications, which are identified by the following Attorney Docket Nos.: BAT 0078 PB / 40078.255 entitled "FLUID CONTAINER FOR ELECTROHYDRODYNAMIC SPRAY DEVICE AND METHOD OF USING SAME" and BAT 0079 PB / 40078.256 entitled "SPRAY HEAD FOR ELECTROHYDRODYNAMIC SPRAY DEVICE AND ELECTROHYDRODYNAMIC SPRAYER SYSTEM".

20

The knapsack sprayer used to apply the conventional Roundup® ULTRA MAX formulation was a conventional field trial device using CO<sub>2</sub> as the propellant.

Details of the application characteristics for both sprayers are described in Table 14 below.

25



5

**Table 14. Sprayer Specification**

	Conventional	EFET
Operating Pressure	30 p.s.i.	Not relevant
Nozzle type	Flat fan 8002VS	Experimental, multi-site
Nozzles/Boom	3	3
Band Width	5 feet	3 - 6 feet
Boom Height	18 inches	6 – 12 inches
Ground Speed	3 mph	Varied
Spray Volume	20 gal/ac	Varied
Spray Water pH	7.0	Not relevant

**Methods of Assessment of Herbicidal Effects**

10

Crop injury and weed control assessments were made by the same assessor for all treatments on all the assessment dates.

The relationship between the dates of assessment and the spray application dates is described in Table 15 below.

15

**Table 15. Relationship Between Spray Dates and Herbicidal Performance Assessment Dates**

Spray Application Dates	Performance Assessment Dates	Days After Treatment (DAT)
1st Application	1st Assessment	4 DAT (1st application)
2nd Application	2nd Assessment	17 DAT (1st application) 3 DAT (2nd application)
	3rd Assessment	31 DAT (1st application) 17 DAT (2nd application)

20

At all times of assessment, herbicidal damage was recorded on a linear 0-6 rating scale where 0 = no observable effect and 6 = complete control. Ratings were made separately on each occasion for:

25

- grass weed control (exclusively *Setaria faberi*);
- broad-leaved weed control (predominantly *Abutilon theophrasti* and *Ambrosia elatior*); and
- phytotoxicity to soybean.

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5 In addition, observations were made to record effects on other weed species and other less quantifiable effects.

Other ratings for crop injury and weed control were made by the spray contractor at 7, 14, and 21 DAT.

10

### **RESULTS AND DISCUSSION**

Detailed results are presented in Tables 16 – 21.

15

**Table 16. Results of First Spray Application (T1): Control of Giant Foxtail (*Setaria faberi*)**

(0-6 score, where 0 = no observable effect, 6 = complete control)

Experimental Formulation Family	ai Rate (kg/ha)	Application Method	Grass Weed Control Score		
			3 DAT	17 DAT	31 DAT
G	1.09	EFET	4.0	6.0	6.0
	0.89	EFET	3.0	6.0	6.0
	0.50	EFET	3.7	6.0	6.0
E	1.06	EFET	4.0	6.0	6.0
	0.61	EFET	3.7	6.0	6.0
F	0.98	EFET	4.7	6.0	6.0
	0.55	EFET	3.7	6.0	6.0*
N2	1.22	EFET	2.7	5.7	6.0
M1	1.04	EFET	3.0	5.3	6.0
M2	1.20	EFET	3.0	5.7	6.0
N1	1.15	EFET	2.7	6.0	6.0
Monsanto Roundup® ULTRA MAX	2.08	Conventional	3.3	6.0	6.0
	1.04	Conventional	2.0	6.0	6.0
	0.52	Conventional	1.7	6.0*	1.7

\* Includes one missing plot.

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5 **Table 17. Results of First Spray Application (T1): Control of Broad-Leaved Weeds (*Abutilon theophrasti* and *Ambrosia elatior*)**  
(0-6 score, where 0 = no observable effect, 6 = complete control)

Experimental Formulation Family	ai Rate (kg/ha)	Application Method	Weed Control Score			
			3 DAT*	17 DAT	31 DAT	Average
G	1.09	EFET	—	5.0	6.0	5.5
	0.89	EFET	—	4.7	6.0	5.3
	0.50	EFET	—	5.0	6.0	5.5
E	1.06	EFET	—	4.3	6.0	5.1
	0.61	EFET	—	4.7	4.7	4.7
F	0.98	EFET	—	4.3	4.7	4.5
	0.55	EFET	—	5.0	5.0	5.0
N2	1.22	EFET	—	4.7	5.3	5.0
M1	1.04	EFET	—	4.3	5.3	4.8
M2	1.20	EFET	—	4.0	5.0	4.5
N1	1.15	EFET	—	4.3	5.7	5.0
Monsanto Roundup® ULTRA MAX	2.08	Conventional	—	5.0	6.0	5.5
	1.04	Conventional	—	5.7	6.0	5.8
	0.52	Conventional	—	3.3	5.3	4.3

\* Broad-leaved weeds not assessable; relatively uncommon and irregularly distributed.

Note: Although Velvetleaf (*Abutilon theophrasti*) and Common Ragweed (*Ambrosia elatior*) were abundant across the trial site, they were not, in contrast to Giant Foxtail (*Setaria faberi*), uniformly distributed and this made accurate weed control ratings more difficult. No assessment was attempted at 3 DAT due to apparent sparsity.

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5

**Table 18. Results of First Spray Application (T1): Phytotoxicity on Soybean (Var. 03422310RR)**

(0-6 score, where 0 = no observable effect, 6 = complete control)

Experimental Formulation Family	ai Rate (kg/ha)	Application Method	Soybean Phytotoxicity Score			
			3 DAT	17 DAT	31 DAT	Average
G	1.09	EFET	2.0	2.0	2.0	2.0
	0.89	EFET	2.0	1.0	2.0	1.6
	0.50	EFET	1.0	1.3	1.0	1.1
E	1.06	EFET	2.0	2.3	2.0	2.1
	0.61	EFET	1.0	2.7	1.0	1.6
F	0.98	EFET	4.3	5.3	4.3	4.6
	0.55	EFET	3.3	5.3	3.3	4.0
N2	1.22	EFET	1.3	1.7	1.3	1.4
M1	1.04	EFET	1.3	2.0	1.3	1.5
M2	1.20	EFET	2.3	2.7	2.3	2.4
N1	1.15	EFET	1.7	2.3	1.7	1.9
Monsanto Roundup® ULTRA MAX	2.08	Conventional	0	1.0*	0	0.3
	1.04	Conventional	0	1.3*	0	0.4
	0.52	Conventional	0	1.0*	0	0.3

\* No specific symptoms were noted. Positive scores were recorded on one replicate only and were probably an environmental, not a treatment-related effect.

10

**Table 19. Results of Second Spray Application (T2): Control of Giant Foxtail (*Setaria faberi*)**

(0-6 score, where 0 = no observable effect, 6 = complete control)

Experimental Formulation Family	ai Rate (kg/ha)	Application Method	Grass Weed Control Score	
			3 DAT	17 DAT
G	1.07	EFET	4.7	6.0
	0.47	EFET	2.7	6.0

15

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	0.35	EFET	2.0	6.0
E	1.50	EFET	2.0	4.7
	0.63	EFET	2.0	6.0
	0.45	EFET	1.3	3.3
F	1.25	EFET	4.3	6.0
	0.64	EFET	3.0	6.0
	0.32	EFET	2.7	6.0
M1	0.99	EFET	1.7	6.0
	0.62	EFET	1.7	5.7
	0.32	EFET	1.0	4.7
M2	0.97	EFET	3.0	6.0
	0.67	EFET	2.7	6.0
	0.56	EFET	2.3	5.7
Monsanto Roundup® ULTRA MAX	2.08	Conventional	1.7	6.0
	1.04	Conventional	2.0	6.0
	0.52	Conventional	2.3	6.0

5

**Table 20. Results of Second Spray Application (T2): Control of Broad-Leaved Weeds**

(0-6 score, where 0 = no observable effect, 6 = complete control)

Experimental Formulation Family	ai Rate (kg/ha)	Application Method	Weed Control Scores	
			3 DAT	17 DAT
G	1.07	EFET	4.0	6.0
	0.47	EFET	2.7	6.0
	0.35	EFET	2.3	6.0
E	1.50	EFET	2.7	6.0
	0.63	EFET	3.0	6.0
	0.45	EFET	1.0	4.0

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F	1.25	EFET	4.3	6.0
	0.64	EFET	3.7	6.0
	0.32	EFET	2.7	6.0
M1	0.99	EFET	2.0	4.3
	0.62	EFET	1.7	5.3
	0.32	EFET	1.3	4.6
M2	0.97	EFET	2.3	5.7
	0.67	EFET	3.0	6.0
	0.56	EFET	2.0	5.0
Monsanto Roundup® ULTRA MAX	2.08	Conventional	2.0	6.0
	1.04	Conventional	2.0	5.7
	0.52	Conventional	2.3	5.3

5

**Table 21. Results of Second Spray Application (T2): Phytotoxicity on Soybean (Var. 03422310RR)**

(0-6 score, where 0 = no observable effect, 6 = complete control)

Experimental Formulation Family	ai Rate (kg/ha)	Application Method	Soybean Phytotoxicity Score	
			3 DAT	17 DAT
G	1.07	EFET	4.0	2.0
	0.47	EFET	2.3	1.7
	0.35	EFET	1.7	0.7
E	1.50	EFET	1.7	1.0
	0.63	EFET	2.0	0.7
	0.45	EFET	1.0	0
F	1.25	EFET	3.3	2.3
	0.64	EFET	2.7	1.0
	0.32	EFET	2.0	1.0

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M1	0.99	EFET	1.3	0
	0.62	EFET	1.0	0
	0.32	EFET	0.7	0
M2	0.97	EFET	2.7	2.0
	0.67	EFET	1.7	0.7
	0.56	EFET	1.3	0
Monsanto Roundup® ULTRA MAX	2.08	Conventional	0	0
	1.04	Conventional	0	0.3
	0.52	Conventional	0	0

5

The field trial was designed to achieve four objectives:

- 10           1.     To compare the relative field efficacy of several families of  
experimental EFET glyphosate formulations created in accordance  
with the present invention, all sprayed using an experimental hand-  
held EFET boom sprayer.
- 15           2.     To compare the relative field efficacy of novel EFET glyphosate  
formulations applied using an experimental hand-held EFET device,  
with the commercial glyphosate formulation, Roundup® ULTRA-  
MAX, applied using a conventional, CO<sub>2</sub>-propelled, knapsack  
sprayer.
- 20           3.     To compare the relative field performance of early applications  
(weeds 12-14 inches average height) with late applications (weeds  
14-21 inches average height) to evaluate the effect of weed growth-  
stage on the herbicidal performance of EFET applications.

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4. To compare the relative speed of herbicidal action of the most efficacious EFET formulations, applied using an experimental EFET device, with that of the commercial glyphosate formulation Roundup® ULTRA MAX, applied using a conventional, CO<sub>2</sub>-propelled, knapsack sprayer.

10

These four objectives are addressed separately in the following sections.

#### **A Comparison of Experimental EFET Formulations**

15

The novel PMG-based formulations of the present invention were compared directly at both times of application. At the first time of application (T1), the formulations were compared only at 1.0 kg ai/ha, in contrast to the second time of application (T2) where the formulations were nominally compared at three rates of application (1.0, 0.5, and 0.25 kg ai/ha).

20

The results were generally consistent at the two times of application. M1 and N1 were compared only at T1 and were broadly similar in activity. M1, a 10% formulation, was selected for further evaluation at T2, alongside M2, a 7.5% derivative of M1, achieved by solvent-dilution.

25

M2 was the most "aggressive" formulation in terms both of its speed-of-effect on all weeds, grass and broad-leaved, and its more phytotoxic effects on soybeans. This is manifest photographically in Figure 21.

30

Comparisons of the performance of M2 and M1 are shown in Figure 1. Soybean Phytotoxicity T2: A Comparison of M1 and M2; Figure 2. *Setaria* Control T2: A Comparison of M1 and M2; and Figure 3. Broad-Leaved Weed Control T2: A Comparison of M1 and M2. The difference in performance was most marked for soybean phytotoxicity and is illustrated in Figure 1, where the values for M2 at 0.6 kg ai/ha are the average values for the 0.67 and 0.56 kg ai/ha rates actually achieved in the field.



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5           The field trial studied weed control in a Roundup Ready® soybean crop (a genetically modified soybean variety (Var. 032422310RR) with a high level of bio-engineered tolerance to the herbicidal action of glyphosate). The soybean variety was not bio-engineered to be tolerant to the EFET formulation components, however, hence the damage to soybeans provides an indicator to the damaging effects of formulation components, in the absence of glyphosate effects. The only difference between M1 and M2 was the addition of a single diluent (i.e., methyl oleate). The increased damage on soybean from M2 over M1 was severe and persistent especially at the higher 1.0 kg ai/ha rate. At this higher rate, the symptoms were also conspicuously different; the soybean leaves showed obvious leaf-edge necrosis with M2 but not M1. It is extremely unlikely that this symptom is a result of run-off on the leaf surface to the leaf edge because of the exceedingly low applied volume (16 liters/hectare). A more probable explanation is that increased uptake and translocation of the formulation to the leaf edge resulted in internal diluent concentrations which were damaging. It is contemplated herein, in accordance with one embodiment of the present invention, that this increased uptake and translocation of the diluent was associated with increased uptake and translocation of glyphosate. Accordingly, this would explain the surprisingly improved long-term weed control of M2 over M1.

25           M2 was, however, slower to express herbicidal symptoms, its long-term weed control was inferior and soybean damage was conspicuously less than that from the most efficacious novel EFET formulations F and G.

          These comparisons are presented in Figures 4-9.

30           The clear difference in soybean phytotoxicity at T1 is shown photographically in Figure 21.

          The difference in weed control performance is illustrated in Figure 4 for *Setaria* control at T1, where the actual field-applied dose of M2 was 1.2 kg ai/ha compared to 1.0 and 1.1 kg ai/ha for F and G respectively.

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5 It is probable that formulation optimization of M2 (and also N2) will lead to improved herbicidal performance.

**A Comparison of the Most Efficacious EFET Formulations with Roundup® ULTRA MAX, Applied Conventionally**

10 The results were generally consistent at both application times. The novel EFET formulations F and G were distinctly more “aggressive” in terms of speed-of-effect on weeds (both grass and broad-leaved) and phytotoxic effects on soybean than Roundup® ULTRA MAX. This is vividly illustrated photographically in Figures 22-23 and 24 at T1 and T2, respectively.

15 EFET formulation E was generally less efficacious than either F or G and is not discussed further.

These effects are presented in Figures 10-15.

Long-term weed control was also improved substantially compared to Roundup® ULTRA MAX, particularly the control of the broad-leaved weed  
20 *Abutilon* at advanced growth stages at T2. This difference in weed control performance is illustrated on broad-leaved weeds at T2 in Figure 13.

The difference is further illustrated in Figure 16, which compares the rate response of the most efficacious EFET formulation G with Roundup® ULTRA MAX at the earliest (3 DAT) and last (17 DAT) dates of assessment. EFET  
25 formulation G was clearly superior in weed control performance at all rates of application at both assessment times. Here, the weed species not fully controlled by Roundup® ULTRA MAX at 0.5 and 1.0 kg ai/ha was *Abutilon theophrasti*, the most tolerant weed to glyphosate of the abundant species on the trial site (see Table 22).

30 There were also clear differences in damage to soybeans at both times of application. This effect was unsurprising because the Roundup Ready® soybean (Var. 03422310RR) was engineered to be tolerant to glyphosate and to the

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5 Roundup® ULTRA MAX formulation but it demonstrates the herbicidal potency of the other formulation components in the EFET formulations.

The difference in soybean phytotoxicity was particularly striking, especially at T1, and is illustrated in Figure 14. Here, Roundup® ULTRA MAX did not produce any phytotoxicity at any rate of application up to 2.1 kg ai/ha at any time  
10 of assessment.

The clear conclusion from this single field trial is that the EFET applications were at least as effective or more effective than Roundup® ULTRA MAX at both weed growth stages. This is illustrated in Figure 16. Overall, EFET formulation G was the most efficacious formulation tested in terms of long-term weed control,  
15 though it was less “aggressive” than F in early symptom expression. This combination of properties, the rapid initiation of early visual symptoms without detriment to long-term weed control is unexpected and exceptionally difficult to achieve in glyphosate formulations; it merits further optimization. It is surprising that this early, severe necrosis did not impair the longer-term, systemic activity of  
20 glyphosate in EFET formulation G since it is well known that the addition of compounds which cause rapid damage and disruption to weed tissue does not usually result in additive weed control effects but to a reduction in the translocation of glyphosate within the weed (which is an active transport process) and a consequent reduction in long-term weed control. EFET formulation F  
25 illustrates this point; it was more aggressive in terms of early damage but less effective at long-term weed control.

At both times of application, EFET formulation G was at least 2-4 times more efficacious than Roundup® ULTRA MAX and at T2, where the lower rate of 0.3 kg ai/ha was included, at least 3-6 times more efficacious. Optimization of the  
30 formulation is likely to result in further improvements in efficacy.

The complete weed control achieved by G at T1 at 0.5 kg ai/ha is shown photographically in Figure 25.

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5 **The Effect of Weed Growth Stage on Herbicidal Performance**

The field trial was not designed to evaluate the effect of weed growth stage on herbicidal efficacy in detail. It was designed, however, to determine whether EFET applications were as efficacious on tall weeds (14-21 inches) at advanced growth stages as conventional applications of Roundup® ULTRA MAX applied according to label recommendations.

The Roundup® ULTRA MAX label describes in detail the rates of application required to control weeds at different growth stages. This is summarized below in Table 22 for the main weed species on the trial site.

15

**Table 22. Label Recommended Rates (kg ai/ha) for Roundup® ULTRA MAX to Control Annual Weed Species of Different Heights (inches)**

Weed Species	Maximum Weed Height (inches)			
	3"	6"	12"	18"
<i>Setaria</i>	–	0.45	0.7	0.9
<i>Abutilon</i>	0.7	0.9	1.4	>1.4
<i>Ambrosia</i>	–	0.7	0.9	1.4

20

The results from the final weed control assessments, comparing weed control performance at T1 (when weeds were, on average, 12-14 inches tall) and T2 (when weeds were, on average, 14-21 inches tall), are presented in Table 23 below.

25 **Table 23. Total Weed Control: A Comparison of the Effect of Weed Growth Stage on the Herbicidal Effectiveness of the Most Efficacious EFET Formulation (G) with Roundup® ULTRA MAX**

Treatment	Application Rate (kg ai/ha)			
	0.3	0.5	1.0	2.0

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	blw	<i>Setaria</i>	blw	<i>Setaria</i>	blw	<i>Setaria</i>	blw	<i>Setaria</i>
T1 G	–	–	6.0	6.0	6.0	6.0	–	–
T1 ULTRA MAX	–	–	5.3	1.7	6.0	6.0	6.0	6.0
T2 G	6.0	6.0	6.0	6.0	6.0	6.0	–	–
T2 ULTRA MAX	–	–	5.3	6.0	5.7	6.0	6.0	6.0

5 Note: blw = broad-leaved weeds

The results presented are the final weed control scores, 31 DAT for T1 and 17 DAT for T2.

10 The results show that the long-term weed control from EFET formulation G was unaffected by weed growth stage; 0.5 kg ai/ha controlled all weeds completely at both times of application. At the second time of application only, a lower rate of EFET formulation G, 0.3 kg ai/ha, was included and this rate also gave complete long-term weed control. In contrast, Roundup® ULTRA MAX was less effective on broad-leaved weeds (especially *Abutilon*) at more advanced  
15 growth stages where 1.0 – 2.0 kg ai/ha was required for complete weed control.

At the first spray application (T1), weeds were, on average 12-14" tall; they had grown to 14-21" tall at the time of the second application (T2). To consistently control *Abutilon theophrasti*, the most tolerant weed to glyphosate of the widespread species on the test site, when it is more than 12 inches tall, the  
20 recommended rate of Roundup® ULTRA MAX is ca 1.5 kg ai/ha. This is shown in the table below and compared to the most efficacious EFET formulation, G.

25 **Table 24. A Comparison of the Application Rate (kg ai/ha) of EFET Formulation G and Roundup® ULTRA MAX which is Required to Provide 100% Control of All Weeds**

Formulation	<i>Setaria</i>		Broad-Leaved Weeds	
	T1	T2	T1	T2
G	Less than 0.5	Less than 0.3	Less than 0.5	Less than 0.3

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Roundup® ULTRA MAX	0.5 – 1.0	Less than 0.5	0.5 – 1.0	1.0 – 2.0
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5

The results of the field trial are, therefore, consistent with the Roundup® ULTRA MAX label recommendation both for in-crop and non-crop applications. The superior performance of EFET formulation G is not related in any way to a less-than-expected performance of Roundup® ULTRA MAX.

10

**A Comparison of the Speed of Herbicidal Action of the Most Efficacious EFET Formulations, with Roundup® ULTRA MAX Applied Conventionally**

The onset of visually obvious herbicidal effects was significantly faster with the EFET formulation G than with Roundup® ULTRA MAX at both times of application on all weed species. This is illustrated in Figure 17 for the first time of application (T1) and in Figure 18 for the second application (T2). The comparative overall weed control at T2 is illustrated in Figure 18. The rate response scores for *Setaria* and broad-leaved weeds were very similar hence the data presented are averages for all weeds.

20

At both times of application, 0.5 kg ai/ha of the EFET formulation G produced herbicidal symptoms more rapidly than 2.0 kg ai/ha of Roundup® ULTRA MAX without any detriment to long-term herbicidal efficacy. At the second time of application, where the lower rate of 0.3 kg ai/ha of EFET formulation G was included, it produced early herbicidal effects equivalent in severity to 1.0 – 2.0 kg ai/ha of Roundup® ULTRA MAX. This combination of herbicidal properties, early symptom expression, and improved long-term weed control is unexpected and warrants further study and optimization.

25

The earliest assessment date following both spray applications was three-four days after treatment. Symptom expression was studied closely only at T1. At this time, the EFET formulation G produced conspicuous leaf-edge necrosis on soybean at the higher rate of application (achieved by applying 27 l/ha spray

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5 volume) compared to generalized leaf speckling and spotting at the lower rate (0.5 kg ai/ha, achieved by applying 13 l/ha spray volume). Clearly, application volume has a significant effect on the severity and type of symptom expressed. (The leaf-edge necrosis symptom on soybean was also noted with formulation M2 (applied at 16 l/ha) which was produced by solvent-dilution from M1 (applied at 10 l/ha)).

10 The symptoms on all the weed species were also clearly visible 3-4 DAT but they varied with species. On *Setaria*, there was obvious wilting and necrosis; on *Ambrosia*, significant necrosis and terminal chlorosis; on *Abutilon*, necrosis and leaf cupping. On the other weeds which were occasionally present, chlorotic terminals were observed on *Cirsium*, *Chenopodium*, *Taraxacum*, and *Euphorbia*.

15 Symptoms were much less obvious with Roundup® ULTRA MAX 3-4 DAT but terminal chlorosis was noted on *Ambrosia*, *Abutilon*, *Cirsium*, and *Taraxacum*.

This rapid expression of symptoms with EFET formulations had been noted earlier in the glasshouse screening trial (Born, G. L., 2002b) which preceded the field trial. The results of this trial are summarized for *Setaria* and *Abutilon* in Table  
20 24 and in Figures 19 and 20 respectively. The effects on *Setaria* and *Abutilon* are described in the table below.

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5 Table 25. Glasshouse Trial: Percent Control of *Setaria* and *Abutilon* by EFET Formulations F and G Compared to that from Roundup® ULTRA MAX

Formulation	kg ai*/ha	1 DAT		3 DAT		7 DAT		14 DAT		28 DAT	
		Setaria	Abutilon	Setaria	Abutilon	Setaria	Abutilon	Setaria	Abutilon	Setaria	Abutilon
F	1.6	48.8 b	58.8 c	72.5 c	70.0 a	80.0 ab	83.8 ab	95.0 a	90.0 abc	100.0 a	100.0 a
G	1.5	40.0 c	57.5 c	42.5 hi	58.8 b	65.0 cde	77.5 abc	82.5 d	87.5 abc	98.5 abc	100.0 a
Roundup® ULTRA MAX	1.3	0 i	0 i	0 q	7.5 ghi	33.8 i	31.3 jk	86.3 cd	66.3 fgh	82.5 f	61.3 f

\* Actual applied rate.

Note: Means followed by same letter do not differ significantly (P = .05, Duncan's New Multiple Range Test).  
Source: Extracted from Born, G. L., 2002b.



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5           In the glasshouse environment, symptoms were expressed very rapidly indeed. Within three hours, significant leaf blackening was noted on *Setaria* and leave cupping on *Abutilon* with both EFET formulations F and G at 1.5 kg ai/ha. These early symptoms are shown photographically in Figure 26. Roundup® ULTRA MAX produced no symptoms at this time.

10           It is clear that the speed of onset of symptom expression in the field with EFET formulations occurs very rapidly, perhaps within hours, in contrast to the relatively slow activity of Roundup® ULTRA MAX. The symptoms: leaf blackening, cupping, and necrosis are also more visually obvious than the terminal chlorosis, yellowing, and general debility produced by Roundup® ULTRA  
15   MAX.

          With the preferred EFET formulation (G), the early symptoms were achieved without any loss of long-term weed control, especially of *Abutilon*. This combination of herbicidal properties, whilst simultaneously improving rapid damage followed by long-term, systemic weed control is exceptionally difficult to  
20   achieve with glyphosate because the early damage normally antagonizes the systemic movement of glyphosate.

### 3.5   General Discussion

          The field trial results demonstrate that experimental EFET-formulations of  
25   glyphosate from different formulation families, applied by EFET devices, can significantly out-perform the glyphosate formulation Roundup® ULTRA MAX, applied conventionally, in terms both of speed-of-onset of symptoms and final weed control. The differences were large, apparent at both application times, and consistent for both grass- and broad-leaved weeds.

30           The field trial is the first reported trial on EFET applications of glyphosate. The results are unexpectedly superior to those in an earlier published report on EFET applications of other herbicides in which Parham, M. R. (1982) described only comparable weed control from EFET applications of the pre-emergence

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5 herbicides trifluralin and chloridazon and the post-emergence herbicides fluazifop-butyl, mecoprop, and ioxynil octonate compared to conventional applications.

The most efficacious experimental EFET formulations, F and G, are unoptimized representatives from different formulation families. Both achieved complete weed control at volumes of application around 10 liters/ha. The  
10 percentage active ingredient in the F and G formulations was 3.5% and 4% respectively, providing a significant opportunity for the development of higher concentration formulations in the formulation optimization process. In stark contrast, Roundup® ULTRA MAX was applied at 192 liters/ha (equivalent to 17 gallons/acre), a volume of application in the middle of the water carrier  
15 volumes label-recommended by Monsanto for annual weed control (10-40 gallons/acre).

The preferred EFET formulations, therefore, achieved superior (at least 2-4 times) weed control to Roundup® ULTRA MAX at volumes of application twenty times lower and with the real prospect of further spray volume reduction. This has  
20 obvious implications for speed and convenience of application.

The lower rates of application of glyphosate (kg ai/ha) required for complete weed control with EFET applications should also result in substantial environmental benefits including reduced contamination of surface waters.

## 25 CONCLUSIONS

The field trial results showed that EFET applications of glyphosate offer substantial improvements over Roundup® ULTRA MAX applied conventionally at equivalent rates of active-ingredient application.

In particular, EFET formulations:

- 30 • were at least 2-4 times more efficacious than Roundup® ULTRA MAX,
- produced herbicidal symptoms much more rapidly and obviously than Roundup® ULTRA MAX, and

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- 5           • were effective at volumes of application 20 times lower (10  
liters/hectare).

It is probable that optimization of the EFET formulations and the EFET application device will result in further improvements in relative performance.

10

Oil-Continuous, Non-Aqueous Microemulsions: Exemplary Method of Preparation

The first step in preparing the subject microemulsion formulation involves the active ingredient(s) or pesticide(s). If the pesticide or agrochemical complex as described herein is, itself, an oily liquid, then this initial step is not necessary. If  
15 the pesticide is an oil-soluble solid, then it is dissolved in a suitable oil solvent in accordance with the oil component of the present invention. If the pesticide is soluble in a polar solvent, then it is made into a solution with one of the non-aqueous polar solvents described above. If the pesticide, in its original form, is a solid that is not soluble in any solvent, then it may have to be chemically modified,  
20 such that a soluble derivative of it is obtained. A solubilizing agent such as a (organic) low-molecular weight amine may be needed to convert the pesticide into a soluble form.

One particular pesticide that, in its original solid form, is practically insoluble in polar as well as non-polar liquids is the herbicide N-phosphonomethylglycine or  
25 "PMG". PMG herbicide is a white solid [zwitterionic] amino-acid. It can be described as having a solubility of 11.6 grams per liter of water at 25°C and as insoluble in common organic solvents like acetone, alcohol and xylene. The first step in its formulation is to convert it into a soluble form. One available method is to form the salt of PMG with a simple (organic) amine, i.e., (mono-)methylamine,  
30 which itself is a gaseous material at room temperature. The gaseous (mono-)methylamine may be dissolved into an essentially non-volatile polar solvent like propylene glycol when the appropriate facilities and safety precautions are available for running such a reaction. A water-solution of the (mono-)

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5 methylamine may be equally useful, but the water solvent would have to be stripped off at the end of the reaction by heating the system under vacuum.

A. The Pesticide Concentrate as an Intermediate – A Modular Approach

10 When a manufacturing process or a preparation involves several complex or difficult steps, running quality control or tracing the source of a possible error may be prohibitively difficult. A modular approach avoids these problems. Such an approach is a preferred embodiment of the present invention.

The first step or module may be aptly described as the “taming” of the insoluble pesticide. It involves an *in-situ* preparation of a soluble form that yields a concentrated solution of the pesticide in a polar solvent. Propylene glycol, in a predetermined amount, was weighed into an E.M. flask. Monomethylamine (40% aqueous solution) was weighed into the propylene glycol. A stoichiometric amount of amine plus a 2.5% overage was used to ensure that any deviation from the label concentration would not leave any un-reacted PMG. The solution was swirled to mix the ingredients. PMG-acid (the predetermined amount) was then weighed into the flask and the mixture was swirled to wet the powdered solid. The chemical reaction started before any heating was applied. This was evidenced by a rise in temperature of the ingredients upon mixing and by the majority of solid going into solution upon swirling the flask. A magnetic stirring bar was then introduced into the flask and it was set on a heater stirrer. The flask was heated with stirring for about 25 minutes. The reaction temperature was kept about 100°C and heating was continued until the solid had dissolved. The flask with contents was then cooled in air until its temperature reached about 50°C. The flask was then introduced into a vacuum evaporator. The temperature of the evaporator was kept about 50°C and the vacuum gage indicated a residual pressure of about 10mm Hg. After about one hour the flask attained a constant weight indicating that the water had been completely driven off.

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5           This was the procedure used to prepare the (mono-)methylamine salt  
example described in Table 1 above. The monoethanolamine salt of PMG was  
prepared in a similar procedure. Monoethanolamine (anhydrous) was used for the  
reaction. The vacuum evaporation step was not needed in the preparation of the  
latter compound. The monoethanolamine salt example in Table 1 was prepared  
10 according to this procedure.

The concentrates resulting from the latter procedure contain the active  
ingredient (i.e., the pesticide) in a liquid (solution). These concentrates are stable  
to storage and they lend themselves, easily, to quality control tests. Furthermore,  
they may be (warmed and then) pumped or they may be transported (as needed)  
15 to other locations where the final formulation can be made.

B.    The Polar Sub-Assembly: Concentrate Solution Plus Adjuvant(s)

The solution from part A, above, was used to prepare the polar part of the  
formulation. The two ingredients, the concentrate plus the adjuvant, were  
20 weighed in the predetermined amounts. The addition of the fatty amine (or the  
amine ethoxylate, amine propoxylate, quaternary amine, etc.) produced the polar  
part of the formulation. This was a simple mixing procedure. Warming the  
(viscous) concentrate aided in effecting complete mixing. Steps A and B may be  
combined into a single step. The resulting polar sub-assembly is a stable liquid  
25 that may be stored and used at a later stage for preparing the final formulation.

C.    The Non-Polar Sub-Assembly: Oil Plus Surfactant

When a second surfactant (amphiphile) was called for, it was mixed in the  
predetermined amount with the oil component. This step, also, involved a very  
30 simple mixing.

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5 D. The Final Mixing Step: Combining the Two Sub-Assemblies

This final mixing step was the combining of the two sub-assemblies to form the final formulation. Since the final mixture is an oil-continuous recipe, preferably the polar mixture is added into the non-polar mixture followed by warming and vigorous mixing, as needed.

10

While the invention has been described by reference to certain preferred embodiments, it should be understood that numerous changes could be made within the spirit and scope of the inventive concepts described. Accordingly, it is intended that the invention not be limited to the disclosed embodiments, but that it  
15 have the full scope permitted by the language of the following claims.

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CLAIMS

1. A non-aqueous, oil-continuous microemulsion comprising:  
at least one oil component;  
at least one non-aqueous polar solvent component;  
10 at least one amphiphilic material component; and  
at least one solubilized polar compound component.
2. The microemulsion of claim 1 wherein said oil component is selected from  
alkyl esters of fatty acids, fatty alcohols, esters of dicarboxylic acids, guerbet  
15 alcohols, alcohol acetates, petroleum fractions, aliphatic paraffinic light distillates,  
hydrocarbon oils, vegetable oils, synthetic triglycerides, triethyl phosphate, and  
combinations thereof.
3. The microemulsion of claim 2 wherein said esters of dicarboxylic acids are  
20 selected from abietic acid, azelaic acid, fumaric acid, phthalic acid, adipic acid,  
malonic acid, oxalic acid, succinic acid, carbonic acid, and combinations thereof.
4. The microemulsion of claim 2 wherein said alkyl esters of fatty acids are  
selected from methyl oleate, ethyl oleate, methyl soyate, ethyl soyate, soybean oil,  
25 castor oil, and combinations thereof.
5. The microemulsion of claim 2 wherein said hydrocarbon oils are selected  
from an aliphatic hydrocarbon, an aromatic hydrocarbon, and combinations  
thereof.
- 30 6. The microemulsion of claim 1 wherein said oil component comprises a  
branched-chain hydrocarbon having between about 12 and about 20 carbon  
atoms.

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7. The microemulsion of claim 1 wherein said non-aqueous, polar solvent is selected from an alcohol, a low-molecular-weight ester, an amine, an alkoxyated amine, an amide, a nitrile, a sulfoxide, sorbitol, urea, and combinations thereof.

10

8. The microemulsion of claim 1 wherein said non-aqueous, polar solvent comprises an alcohol having more than one hydroxyl group selected from a dihydric alcohol, a trihydric alcohol, a polyhydric alcohol (polyol), and combinations thereof.

15

9. The microemulsion of claim 8 wherein said dihydric alcohol is selected from ethylene glycol, propylene glycol, 1,3-butanediol, a glycol derivative, and combinations thereof.

20

10. The microemulsion of claim 8 wherein said trihydric alcohol is a glycerol or a glycerol derivative.

25

11. The microemulsion of claim 8 wherein said polyhydric alcohol (polyol) comprises the formula  $\text{CH}_2\text{OH}(\text{CHOH})_n\text{CH}_2\text{OH}$ , and wherein  $n$  is between 2 and 5.

30

12. The microemulsion of claim 7 wherein said amine is selected from ethylene diamine, ethanolamine, diethanolamine, triethanolamine, and combinations thereof.

13. The microemulsion of claim 7 wherein said amide is selected from dimethylformamide, dimethylacetamide, N-methylpyrrolidone, and combinations thereof.



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5 14. The microemulsion of claim 7 wherein said sulfoxide is dimethylsulfoxide.

15. The microemulsion of claim 7 wherein said low-molecular-weight ester is  $\gamma$ -butyrolactone.

10 16. The microemulsion of claim 7 wherein said nitrile is benzonitrile.

17. The microemulsion of claim 1 wherein said amphiphilic material is selected from cationic surfactants, non-ionic surfactants, quaternary surfactants, amphoteric surfactants, zwitterionic surfactants, and combinations thereof.

15

18. The microemulsion of claim 17 wherein said cationic surfactant is selected from an alkylamine having between 8 and 18 carbon atoms, an alkoxyated amine having between 8 and 18 carbon atoms, and combinations thereof.

20 19. The microemulsion of claim 17 wherein said non-ionic surfactant is selected from a polyoxyethylene alcohol, an alcohol polyoxypropylene-polyoxyethylene, a polyoxyethylene sorbitan fatty acid ester, an acetylenic diol, an ethoxylated acetylenic diol, and combinations thereof.

25 20. The microemulsion of claim 1 wherein said amphiphilic material is selected from alkylamines, alkylamine ethoxylates, alkylamine propoxylates, alkylamine propoxylate-ethoxylates, fatty alcohol ethoxylates, fatty alcohol propoxylates, fatty alcohol propoxylate-ethoxylates, fatty acid ethoxylates, fatty acid propoxylates, fatty acid propoxylate-ethoxylates, synthetic long-chain alcohol ethoxylates,  
30 synthetic long-chain alcohol propoxylates, synthetic long chain acid propoxylate-ethoxylates, synthetic long-chain acid ethoxylates, synthetic long-chain acid propoxylates, synthetic long chain acid propoxylate-ethoxylates, alkylphenol ethoxylates, alkylphenol propoxylates, alkylphenol propoxylate-ethoxylates,

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5 alkylpolyglucosides, sorbitol esters, sorbitan esters, sorbitol ester ethoxylates, sorbitan ester ethoxylates, polyoxypropylene-polyoxyethylene block copolymers, ethylenediamine-polyoxypropylene-polyoxyethylene block copolymers, and combinations thereof.

10 21. The microemulsion of claim 1 wherein said solubilized polar compound is a polar agrochemical complex.

15 22. The microemulsion of claim 21 wherein said polar agrochemical complex is a polar-acidic agrochemical complex is selected from an amino acid, a phenoxy, and combinations thereof.

20 23. The microemulsion of claim 22 wherein said polar-acidic agrochemical complex is selected from a glyphosate complex, a 2,4-D complex, a glufosinate complex, and combinations thereof.

24. The microemulsion of claim 23 wherein said glyphosate complex is selected from a glyphosate ester, a glyphosate amide, a glyphosate alkylamide, a glyphosate salt, and combinations thereof.

25 25. The microemulsion of claim 23 wherein said glyphosate complex comprises a glyphosate ester and an alcohol.

26. The microemulsion of claim 23 wherein said glyphosate complex comprises a glyphosate alkylamide and an amine.

30

27. The microemulsion of claim 1 wherein the equivalent acid content of solubilized polar compound present in said microemulsion is between less than about 5 and about 20% by weight.

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5

28. A method of making a non-aqueous, oil-continuous microemulsion containing a solubilized polar compound comprising:

providing a non-aqueous polar portion comprising said polar compound solubilized in a non-aqueous, polar solvent;

10 providing an amphiphilic material and mixing said amphiphilic material with said non-aqueous polar portion to produce an intermediate mixture; and

providing an oil component and mixing said oil component with said intermediate mixture to produce said non-aqueous, oil-continuous microemulsion.

15 29. The method of claim 28 further comprising heating said amphiphilic material with said non-aqueous polar portion.

30. The method of claim 28 wherein said polar compound comprises a polar-acidic agrochemical complex.

20

31. The method of claim 28 wherein said amphiphilic material comprises a cationic surfactant.

25 32. The method of claim 31 wherein said cationic surfactant comprises an alkylamine surfactant or an alkoxyated amine surfactant.

33. The method of claim 28 wherein said non-aqueous polar portion is prepared by:

providing said non-aqueous, polar solvent;

30 providing an insoluble, solid polar compound and combining said polar compound with said non-aqueous, polar solvent to form a reaction mixture; and

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5 providing a compound having an amine, sulfonium, sulfoxonium, amide or ester functional group and mixing said compound with said reaction mixture to form a soluble salt, amide, or ester of said polar compound.

10 34. The method of claim 33 further comprising heating said compound with said reaction mixture.

15 35. The method of claim 33 wherein said soluble salt of said polar compound is selected from diamine salts, polyamine salts, primary amine salts, secondary amine salts, tertiary amine salts, ammonium and quaternary ammonium salts, hydroxylamine salts, ammonium salts, long-chain alkylamine salts, sulfonium salts, sulfoxonium salts, and combinations thereof.

20 36. The method of claim 28 further comprising providing a second amphiphilic material and mixing said second amphiphilic material with said intermediate mixture to produce said non-aqueous, oil-continuous microemulsion.

37. The method of claim 36 wherein said second amphiphilic material comprises a non-ionic surfactant.

25 38. A post-emergence herbicidal spray composition comprising a non-aqueous, oil-continuous microemulsion, said microemulsion comprising:  
at least one oil component;  
at least one non-aqueous polar solvent component;  
at least one amphiphilic material component; and  
30 at least one solubilized polar compound component.

39. The post-emergence herbicidal spray composition of claim 38 wherein said oil component comprises methyl oleate.

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5

40. The post-emergence herbicidal spray composition of claim 38 wherein said solubilized polar compound component comprises a polar-acidic agrochemical complex selected from a glyphosate complex, a 2,4-D complex, a glufosinate complex, and combinations thereof.

10

41. The post-emergence herbicidal spray composition of claim 38 wherein said microemulsion is characterized by an octanol/water partitioning coefficient of between about 2 and about 4  $k_{ow}$ .

15

42. The post-emergence herbicidal spray composition of claim 38 wherein said microemulsion is characterized by a pH of less than 7.00 at about 22°C.

20

43. A method of controlling weeds which comprises applying a post-emergence herbicidal spray composition to a target plant, said composition comprising a non-aqueous, oil-continuous microemulsion comprising:

at least one oil component;

at least one non-aqueous polar solvent component;

at least one amphiphilic material component; and

at least one solubilized polar compound component that comprises a polar-

25

acidic agrochemical complex selected from a glyphosate complex, a 2,4-D complex, a glufosinate complex, and combinations thereof.

30

44. The method of claim 43 wherein said applying a post-emergence herbicidal spray composition to a target plant is performed by either conventional mechanical, non-electrostatic or by electrostatic low-volume or ultra-low volume techniques.

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- 5 45. A method of improving the uptake and translocation of a systemic herbicidal compound in a target plant which comprises applying a post-emergence herbicidal spray composition to said target plant, said composition comprising a non-aqueous, oil-continuous microemulsion comprising:
- 10 at least one oil component;  
at least one non-aqueous polar solvent component;  
at least one amphiphilic material component; and  
at least one solubilized polar compound component that comprises a polar-acidic agrochemical complex selected from a glyphosate complex, a 2,4-D complex, a glufosinate complex, and combinations thereof, wherein
- 15 said oil component affects said improved uptake and translocation of said solubilized polar compound in said target plant.
46. A method of improving the uptake and translocation of a systemic insecticidal or fungicidal compound in a target plant which comprises applying an
- 20 insecticidal or fungicidal spray composition to said target plant, said composition comprising a non-aqueous, oil-continuous microemulsion comprising:
- at least one oil component;  
at least one non-aqueous polar solvent component;  
at least one amphiphilic material component; and
- 25 at least one solubilized polar compound component that comprises a polar-acidic agrochemical complex, wherein
- said oil component affects said improved uptake and translocation of said solubilized polar compound in said target plant.
- 30 47. The method of claim 45 or 46 wherein said oil component comprises methyl oleate.

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5 48. A method of controlling weeds which comprises applying a post-emergence herbicidal spray composition to a target plant, wherein  
said composition comprises a non-aqueous, oil-continuous microemulsion,  
and

10 said applying a post-emergence herbicidal spray composition to a target plant is performed by either conventional mechanical, non-electrostatic or by electrostatic low-volume or ultra-low volume techniques, which deliver a droplet size of between about 5 and about 30 microns to said target plant.

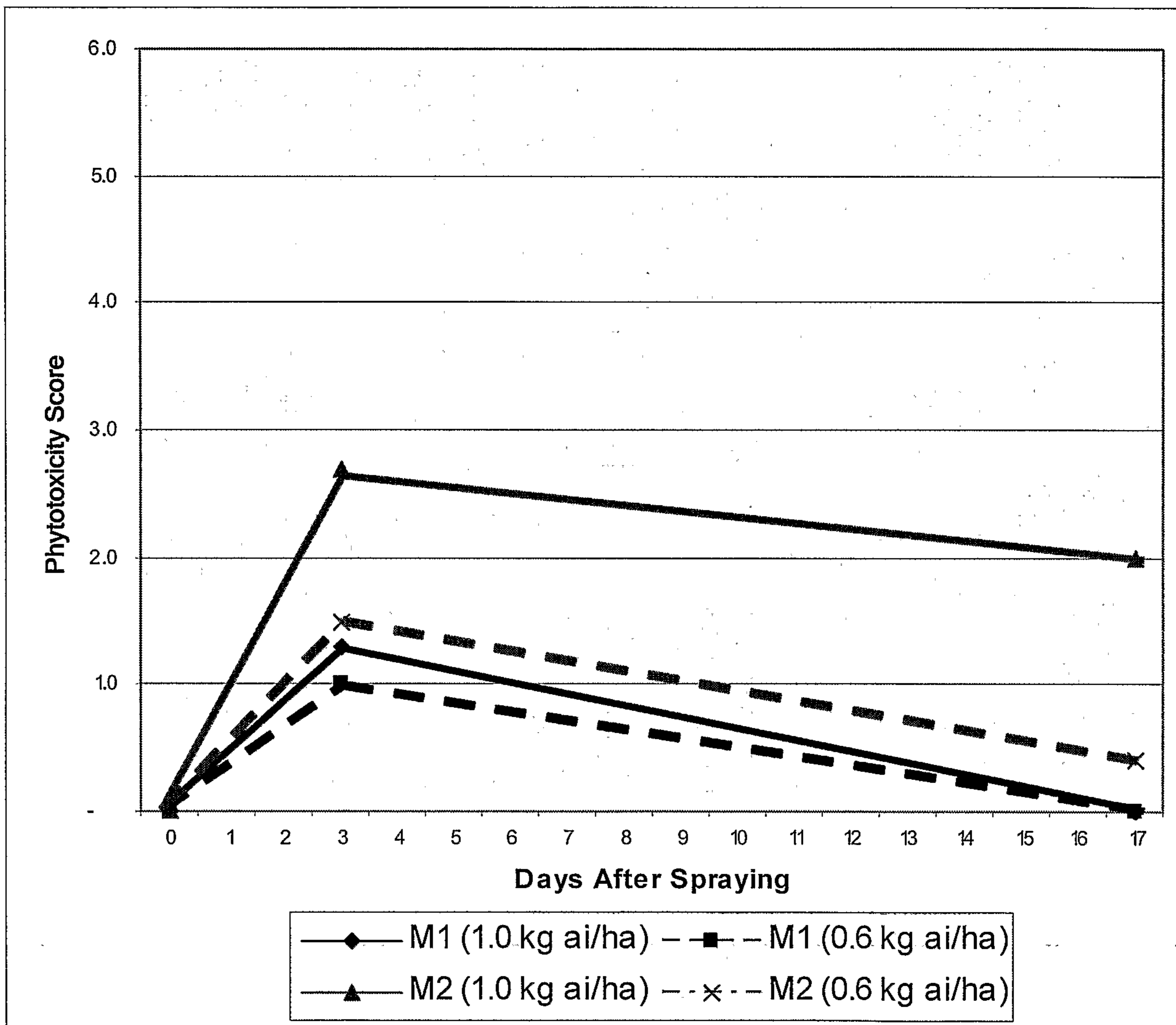
15 49. A method of controlling insects or fungi which comprises applying an insecticidal or fungicidal spray composition to a target plant, wherein  
said composition comprises a non-aqueous, oil-continuous microemulsion,  
and

20 said applying an insecticidal or fungicidal spray composition to a target plant is performed by either conventional mechanical, non-electrostatic or by electrostatic low-volume or ultra-low volume techniques, which deliver a droplet size of between about 5 and about 30 microns to said target plant.

25 50. A method of controlling weeds which comprises applying a post-emergence herbicidal spray composition to a target plant, wherein  
said composition comprises a non-aqueous, oil-continuous microemulsion,  
and

30 said applying a post-emergence herbicidal spray composition to a target plant is performed by either conventional mechanical, non-electrostatic or by electrostatic techniques, which deliver a volume of formulated product to said target plant, at a volume rate that is less than about 500 lts/ha.

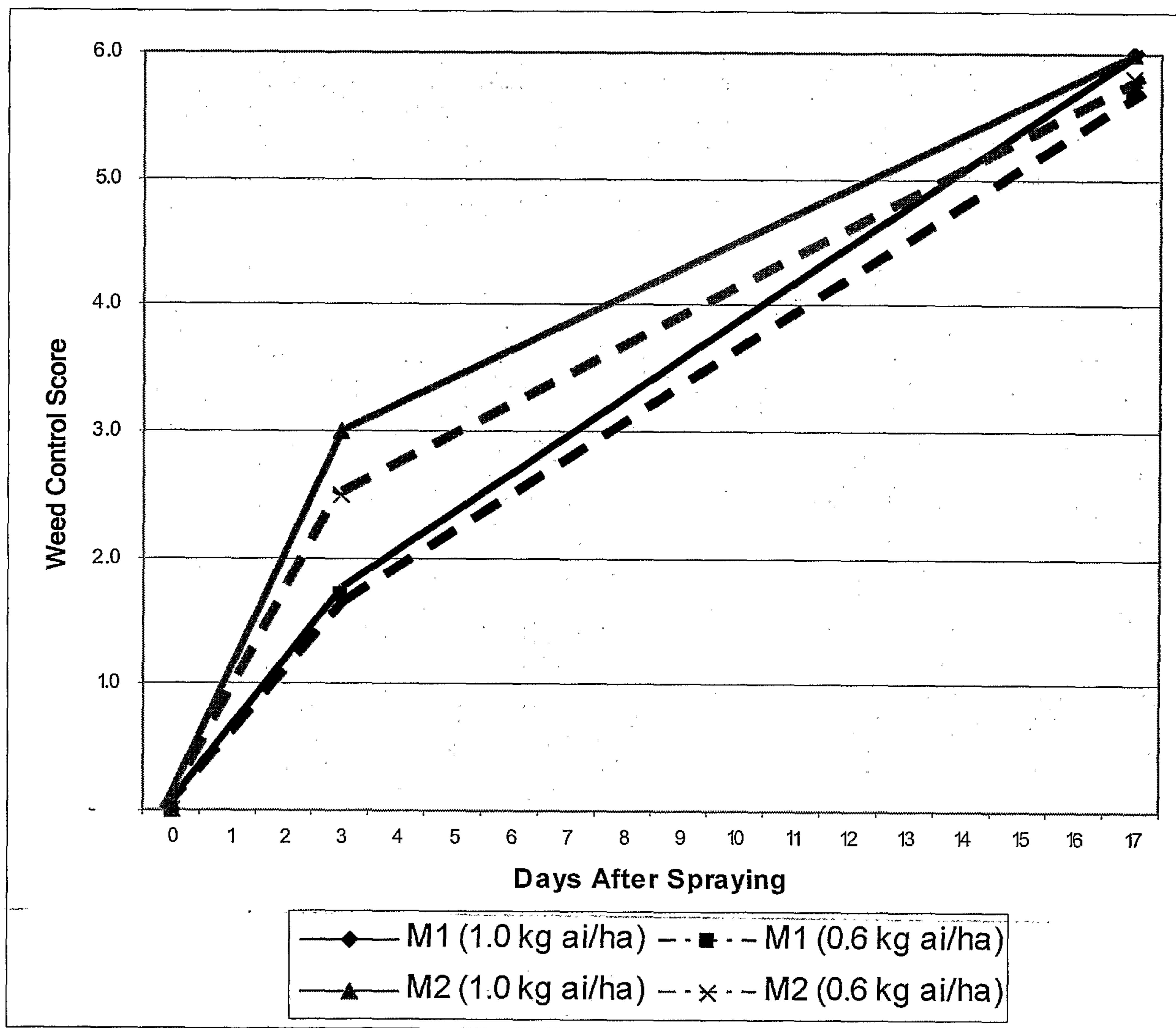
Figure 1. Soybean Phytotoxicity T2: A Comparison of M1 and M2



Note: The values for M2 at 0.6 kg ai/ha are the average values for the 0.67 and 0.56 kg ai/ha rates actually achieved in the field.

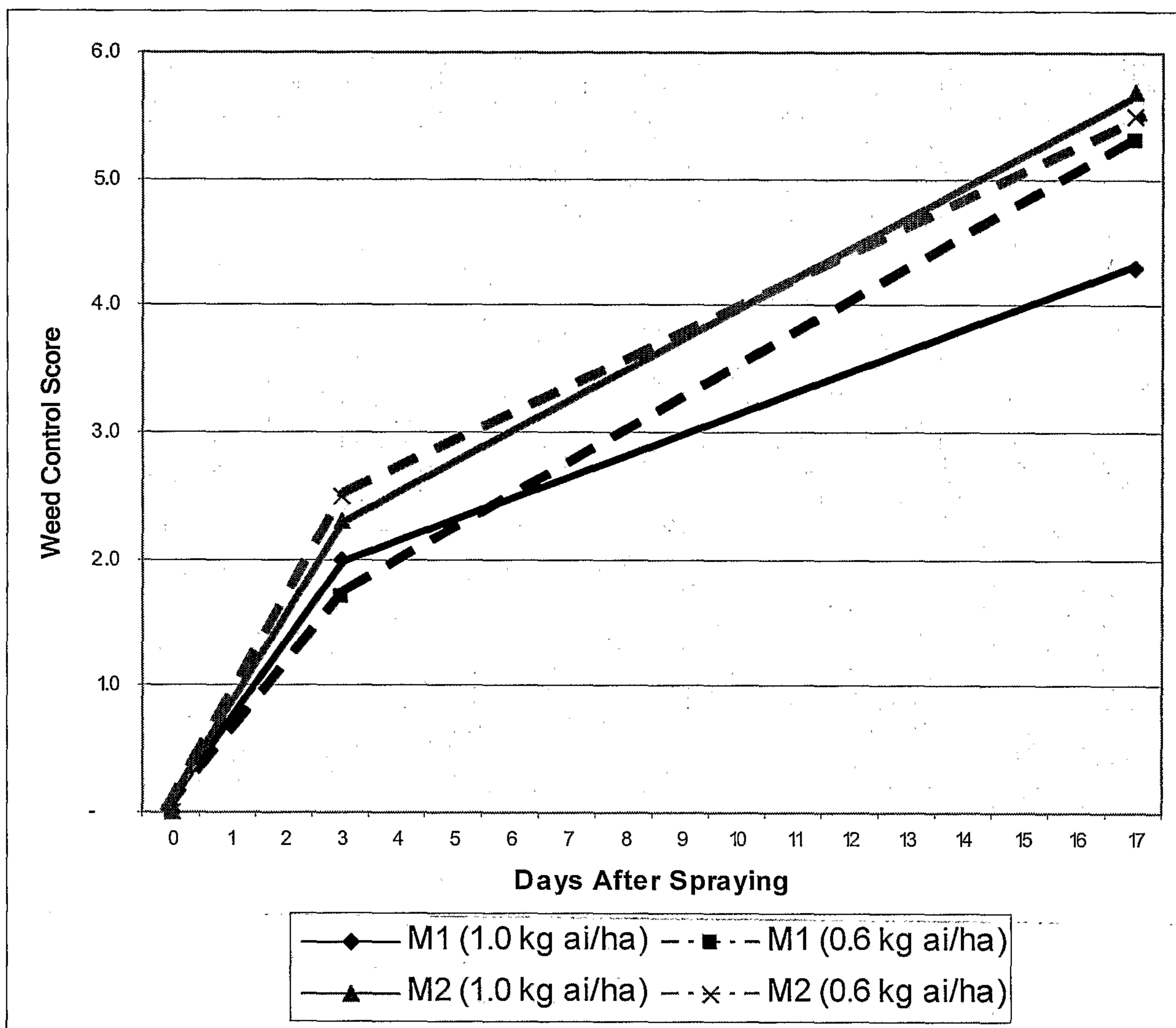


Figure 2. *Setaria* Control T2: A Comparison of M1 and M2



Note: The values for M2 at 0.6 kg ai/ha are the average values for the 0.67 and 0.56 kg ai/ha rates actually applied.

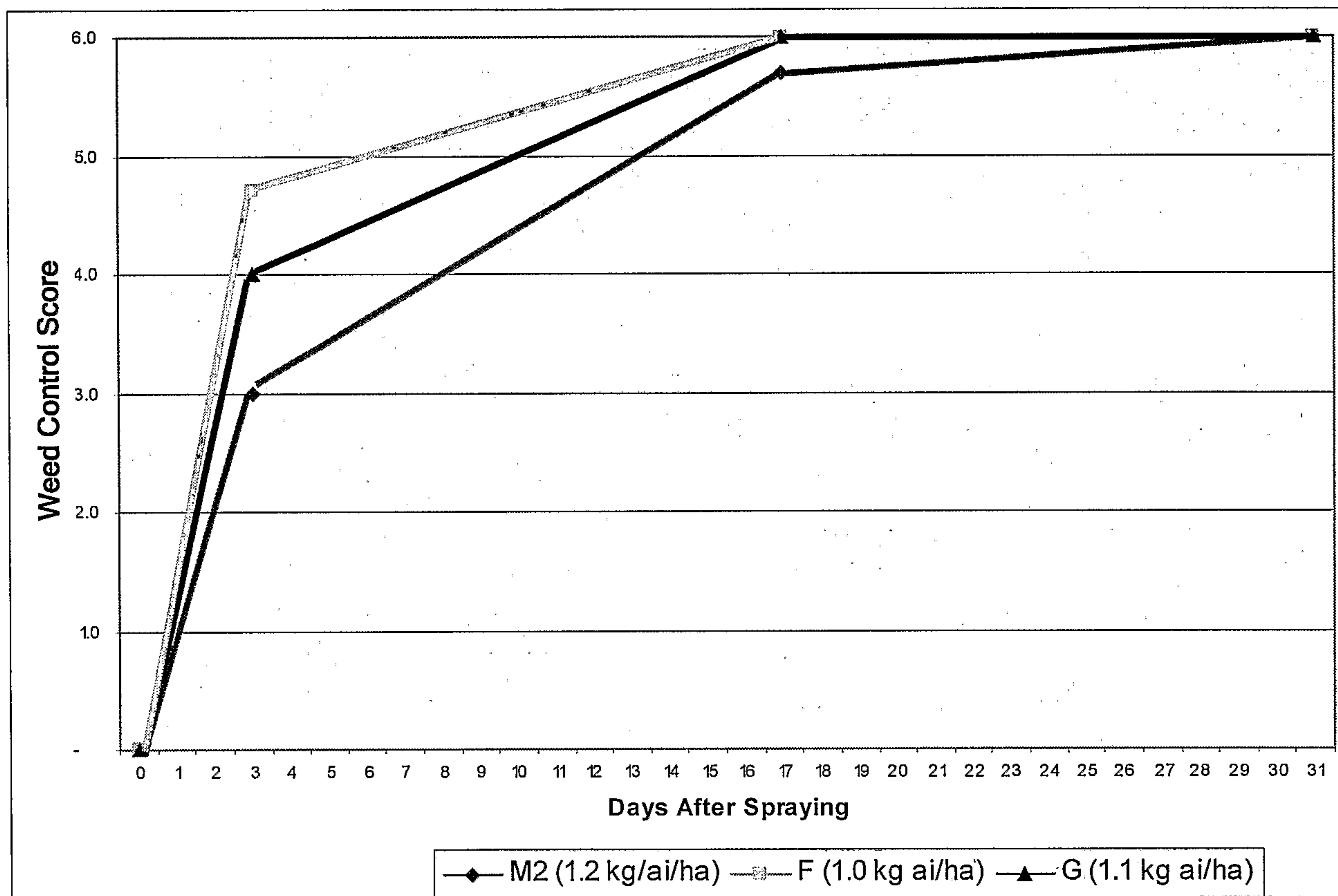
Figure 3. Broad-Leaved Weed Control T2: A Comparison of M1 and M2



Note: The values for M2 at 0.6 kg ai/ha are the average values for the 0.67 and 0.56 kg ai/ha rates actually applied.

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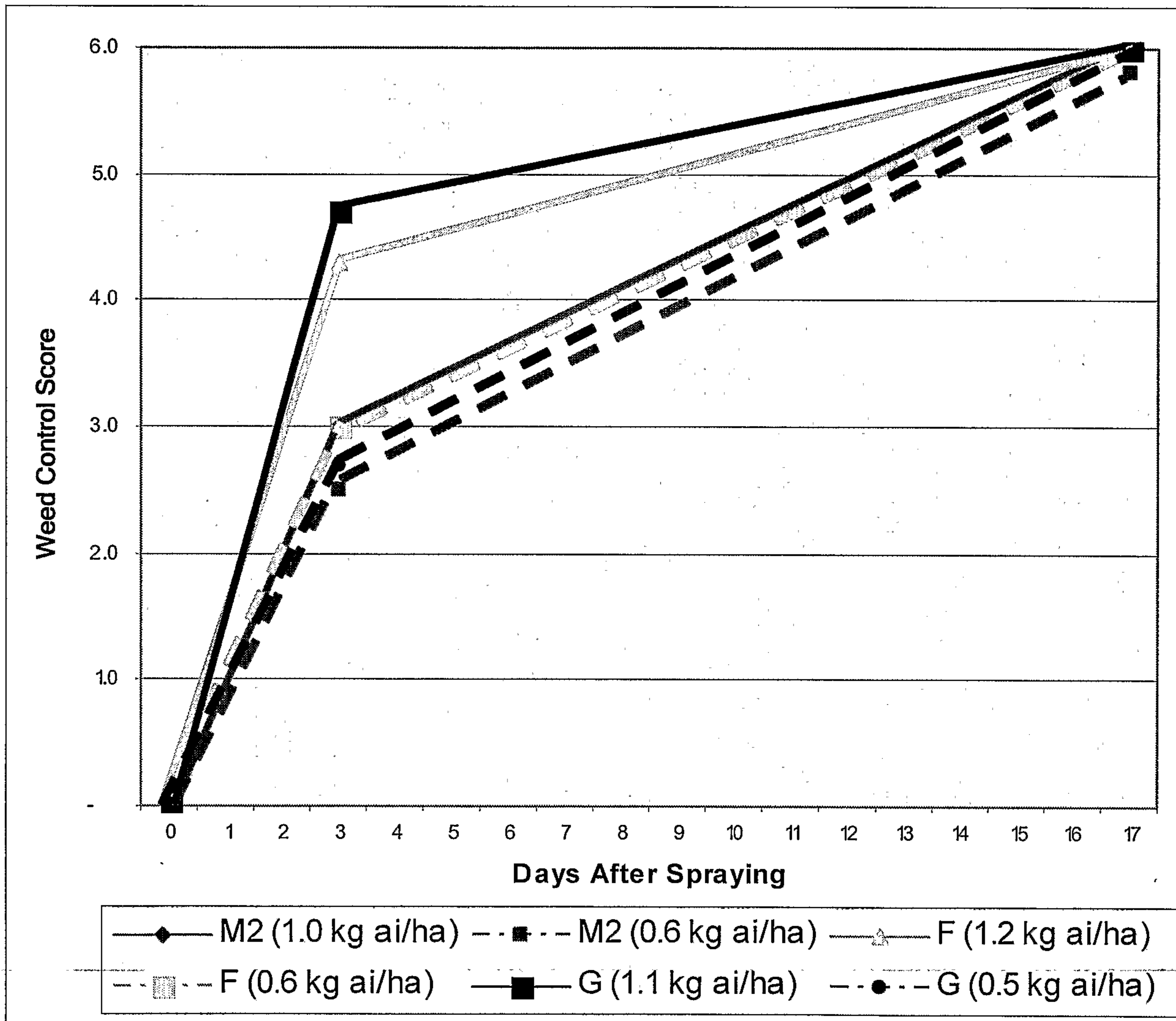
Figure 4. *Setaria* Control T1: A Comparison of M2 with F and G



Note: The actual field-applied dose of M2 was 1.2 kg ai/ha compared to 1.0 and 1.1 kg ai/ha for F and G respectively.

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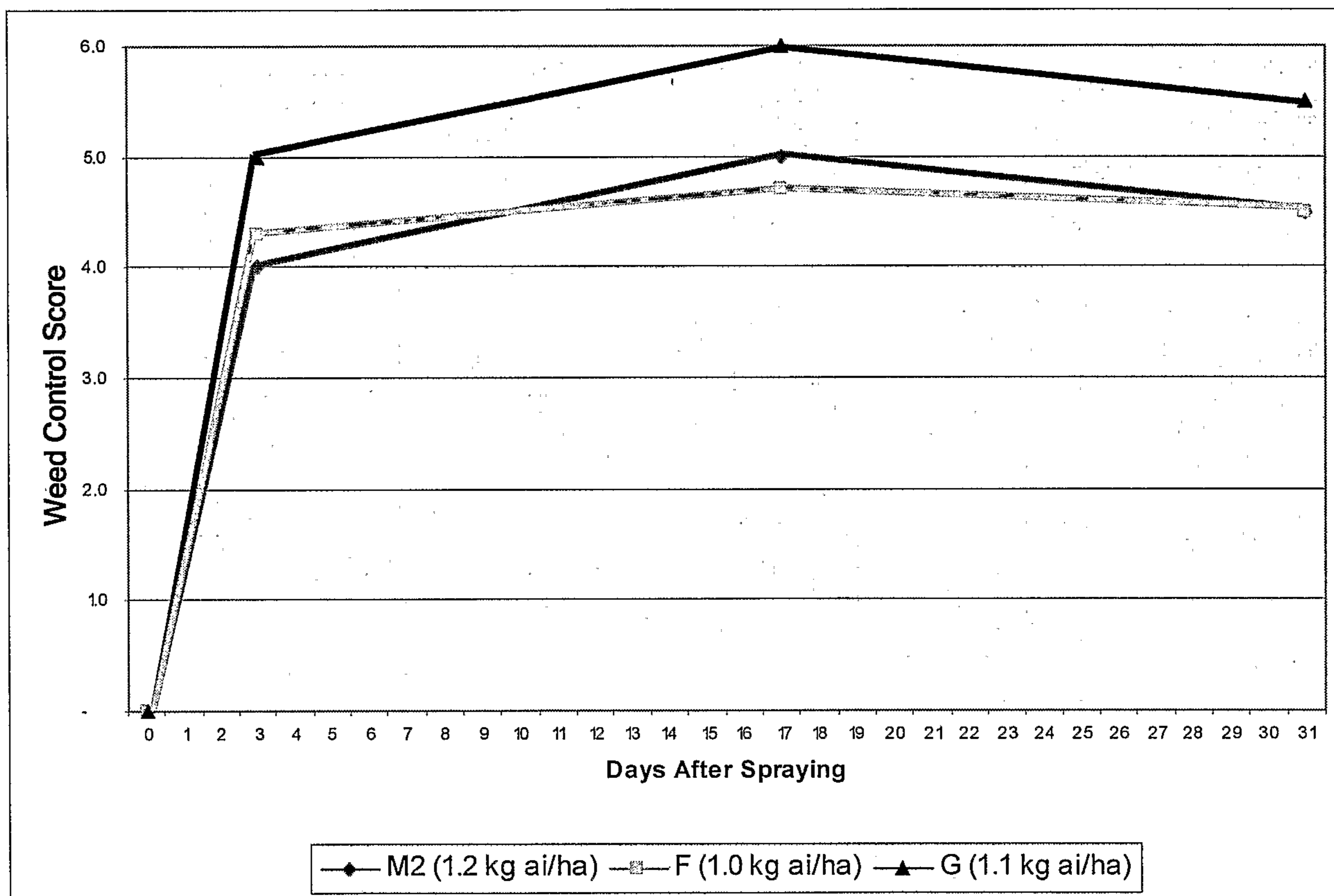
Figure 5. *Setaria* Control T2: A Comparison of M2 with F and G



Note: The values for M2 at 0.6 kg ai/ha are the average values for the 0.67 and 0.56 kg ai/ha rates actually applied.

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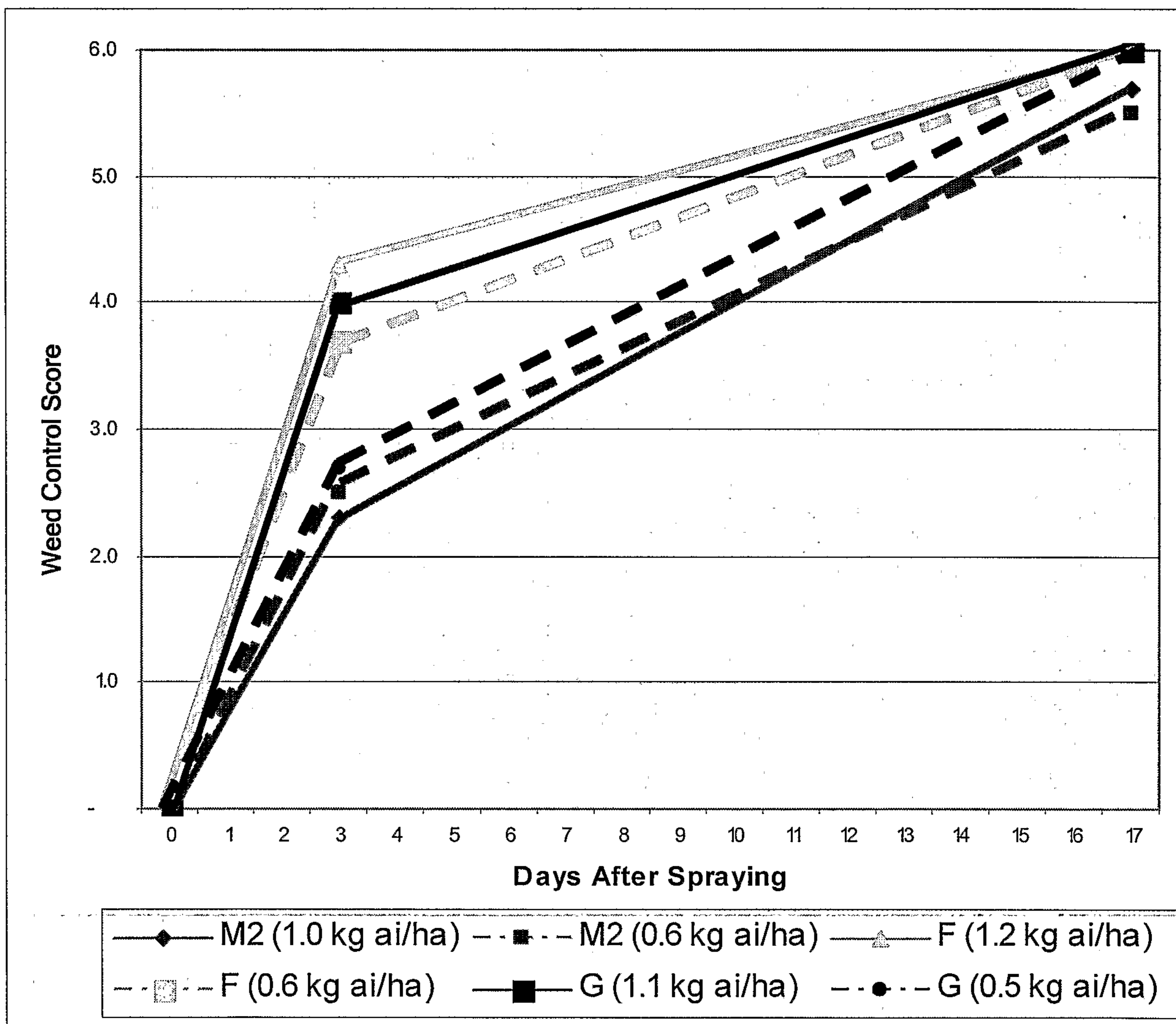
Figure 6. Broad-Leaved Weed Control T1: A Comparison of M2 with F and G



Note: The actual field-applied dose of M2 was 1.2 kg ai/ha compared to 1.0 and 1.1 kg ai/ha for F and G respectively.

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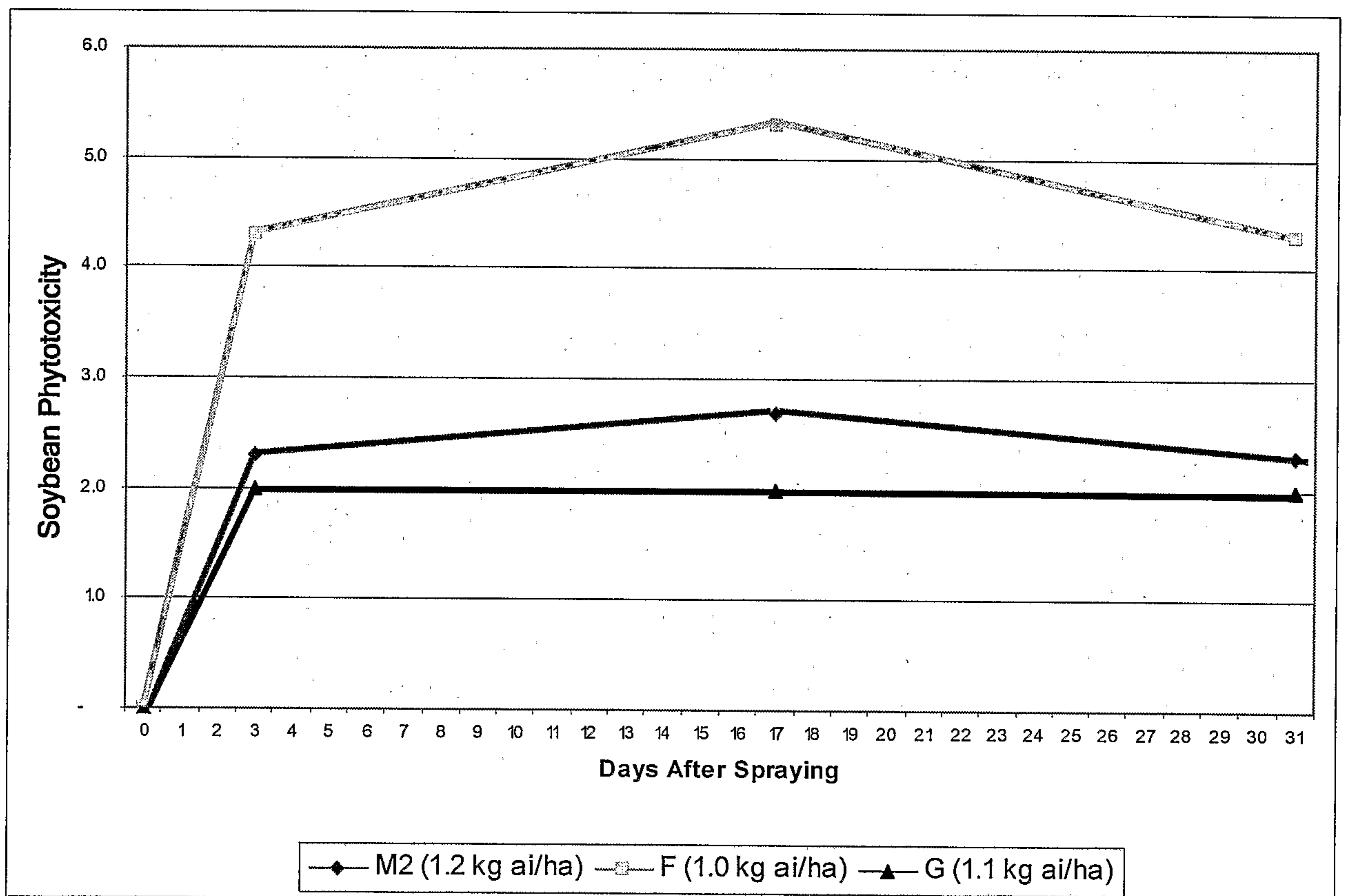
Figure 7. Broad-Leaved Weed Control T2: A Comparison of M2 with F and G



Note: The values for M2 at 0.6 kg ai/ha are the average values for the 0.67 and 0.56 kg ai/ha rates actually applied in the field.

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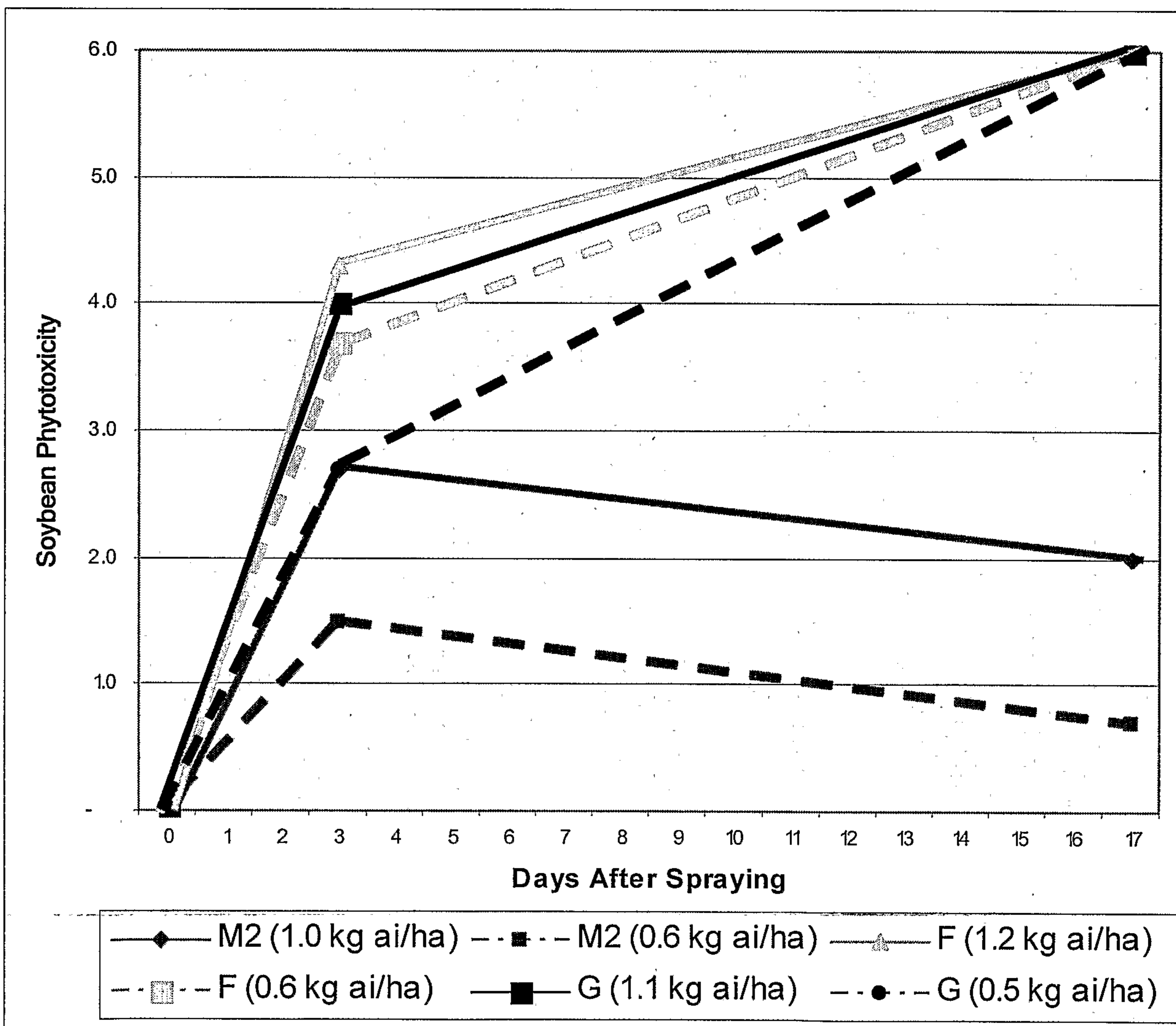
Figure 8. Soybean Phytotoxicity T1: A Comparison of M2 with F and G



Note: The actual field applied dose of M2 was 1.2 kg ai/ha compared to 1.0 and 1.1 kg ai/ha for F and G - respectively.

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Figure 9. Soybean Phytotoxicity T2: A Comparison of M2 with F and G

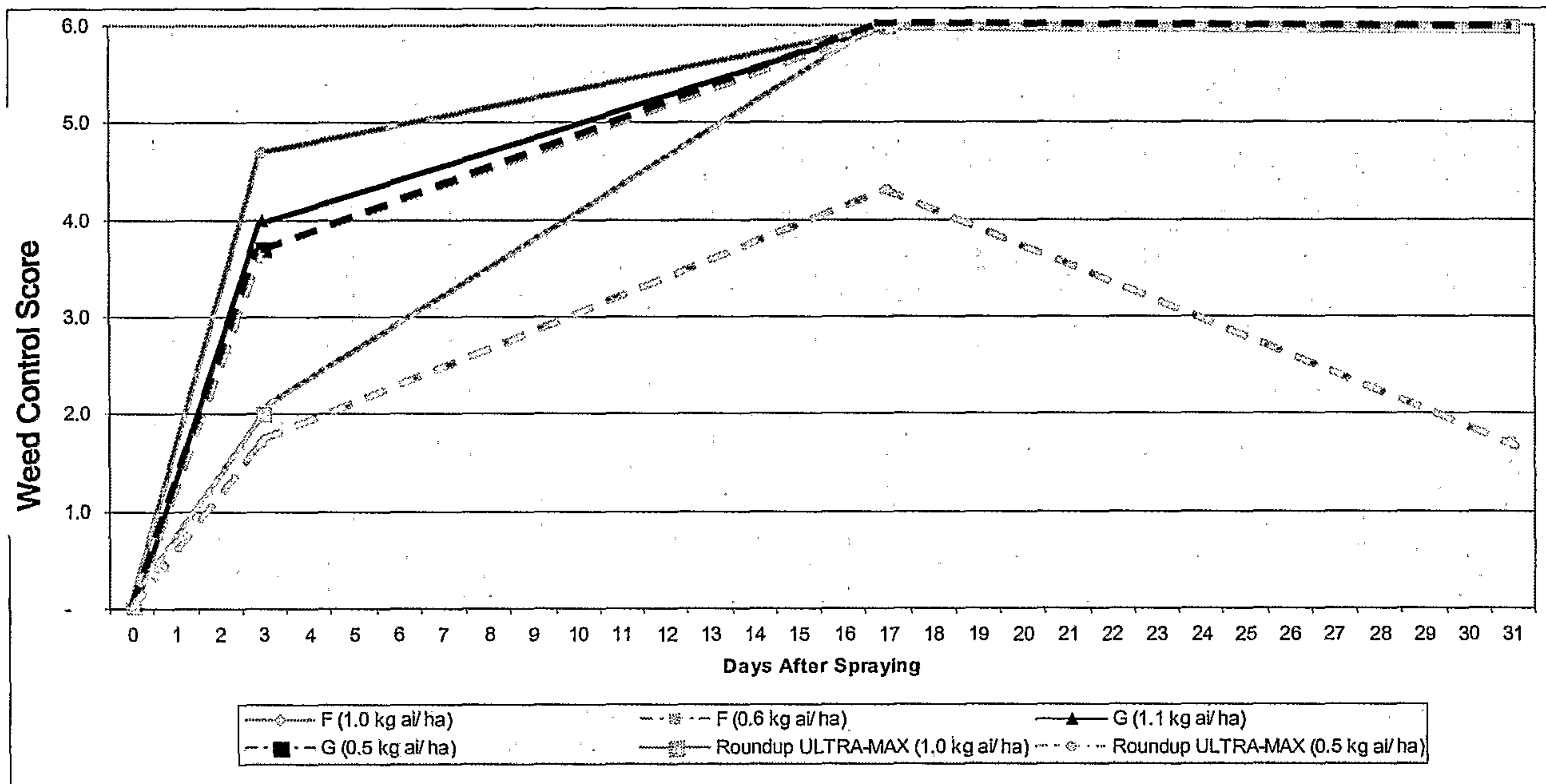


Note: The values for M2 at 0.6 kg ai/ha are the average values for the 0.67 and 0.56 kg ai/ha rates actually achieved in the field.



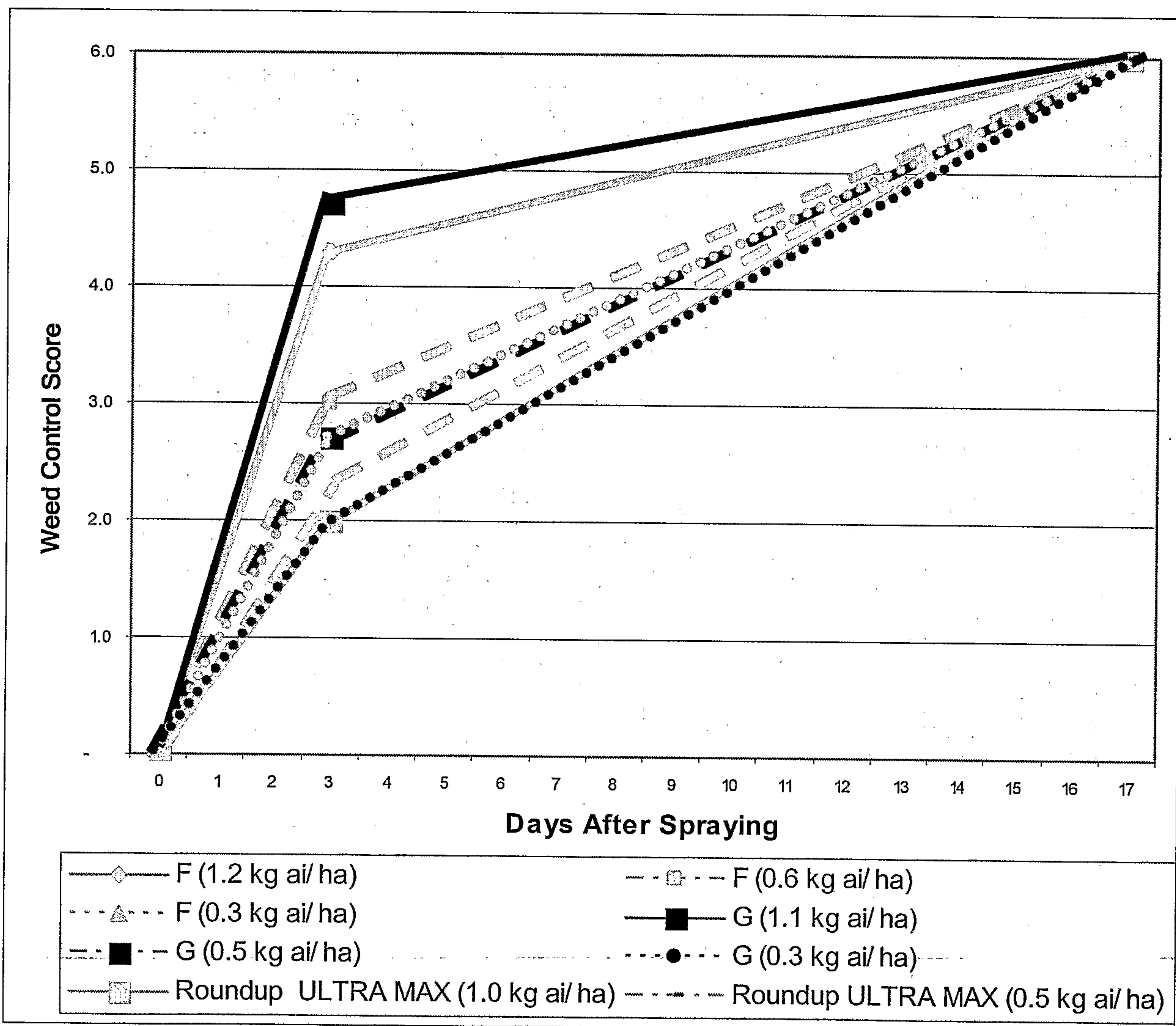
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Figure 10. *Setaria* Control T1: A Comparison of F and G (the most efficacious EFET formulations) with Roundup ULTRA MAX



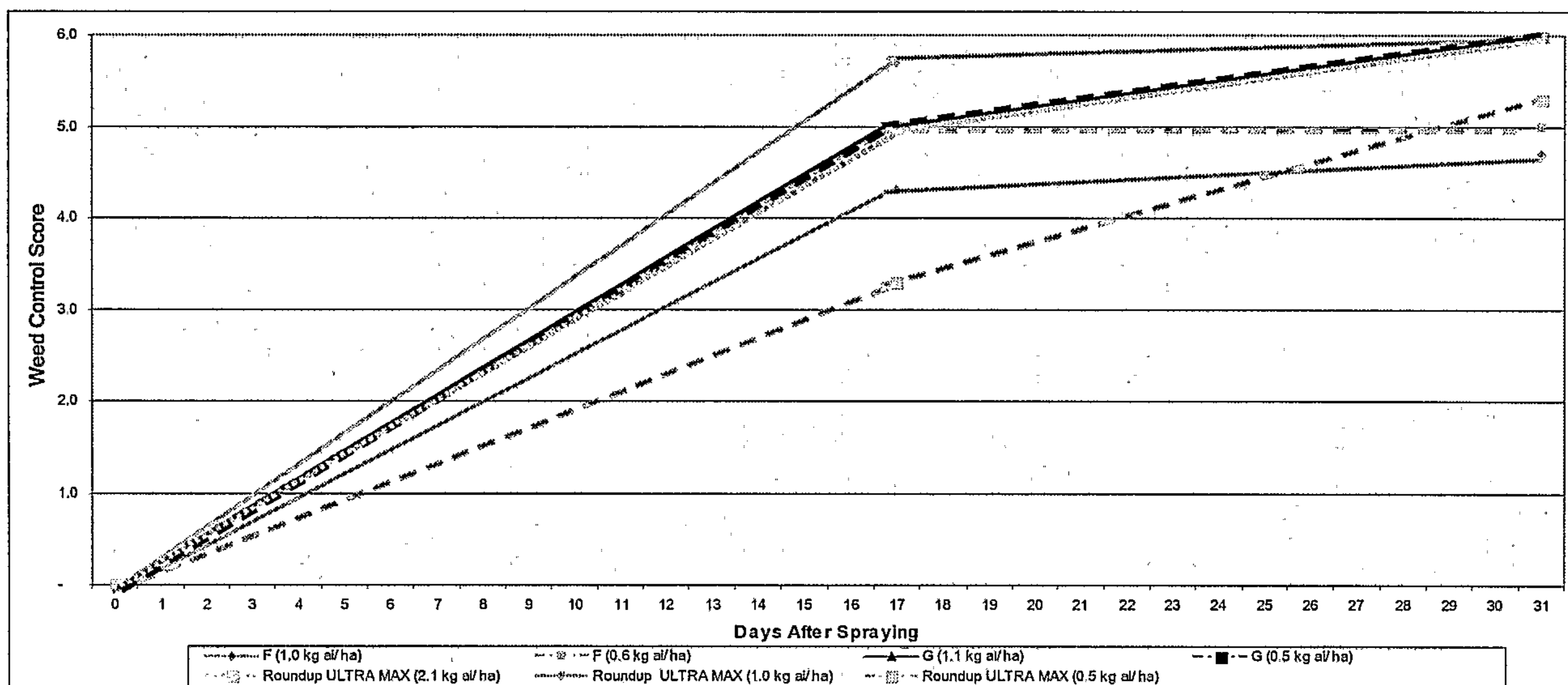
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Figure 11. *Setaria* Control T2: A Comparison of F and G with Roundup ULTRA MAX



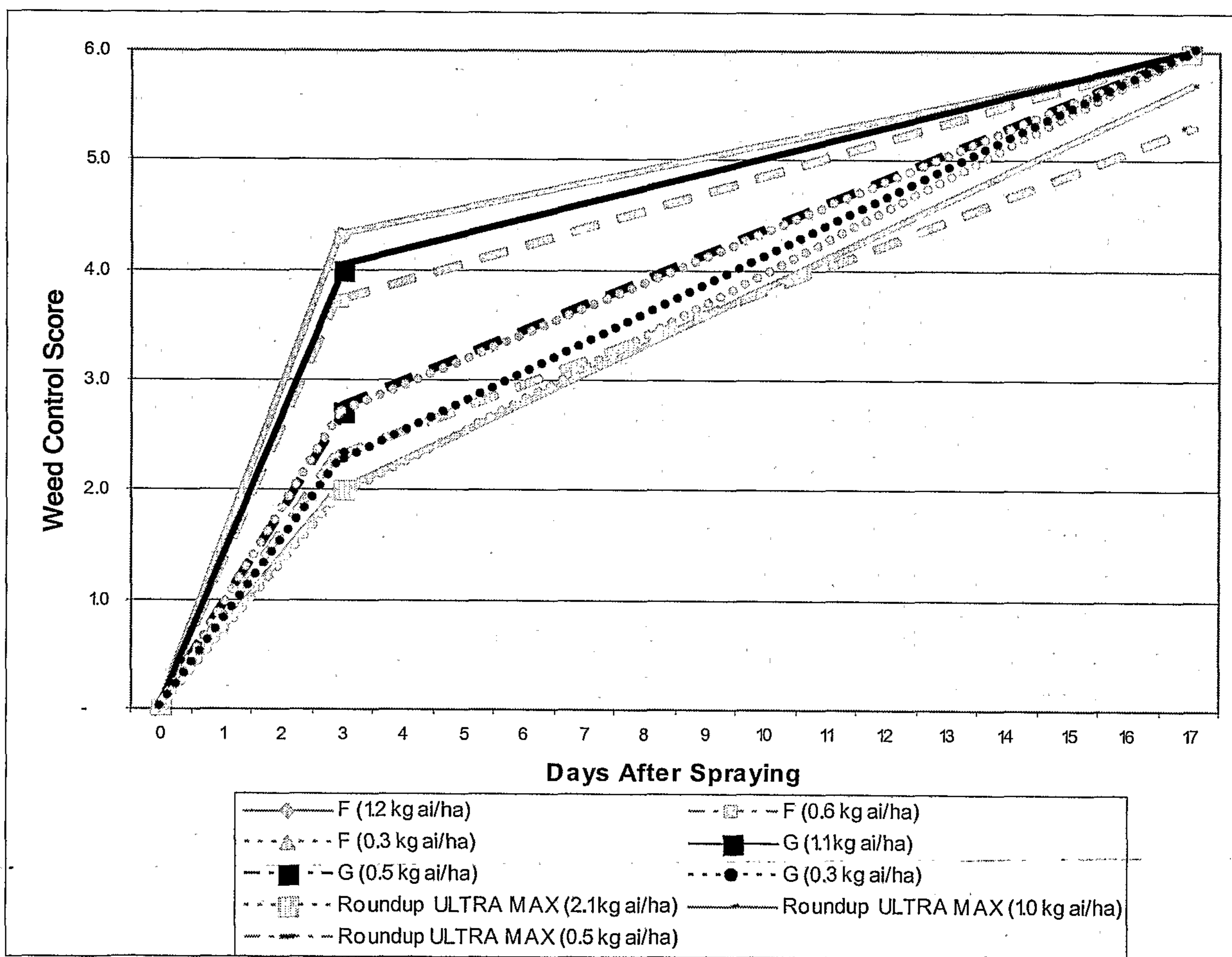
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Figure 12. Broad-Leaved Weed Control T1: A Comparison of F and G with Roundup ULTRA MAX



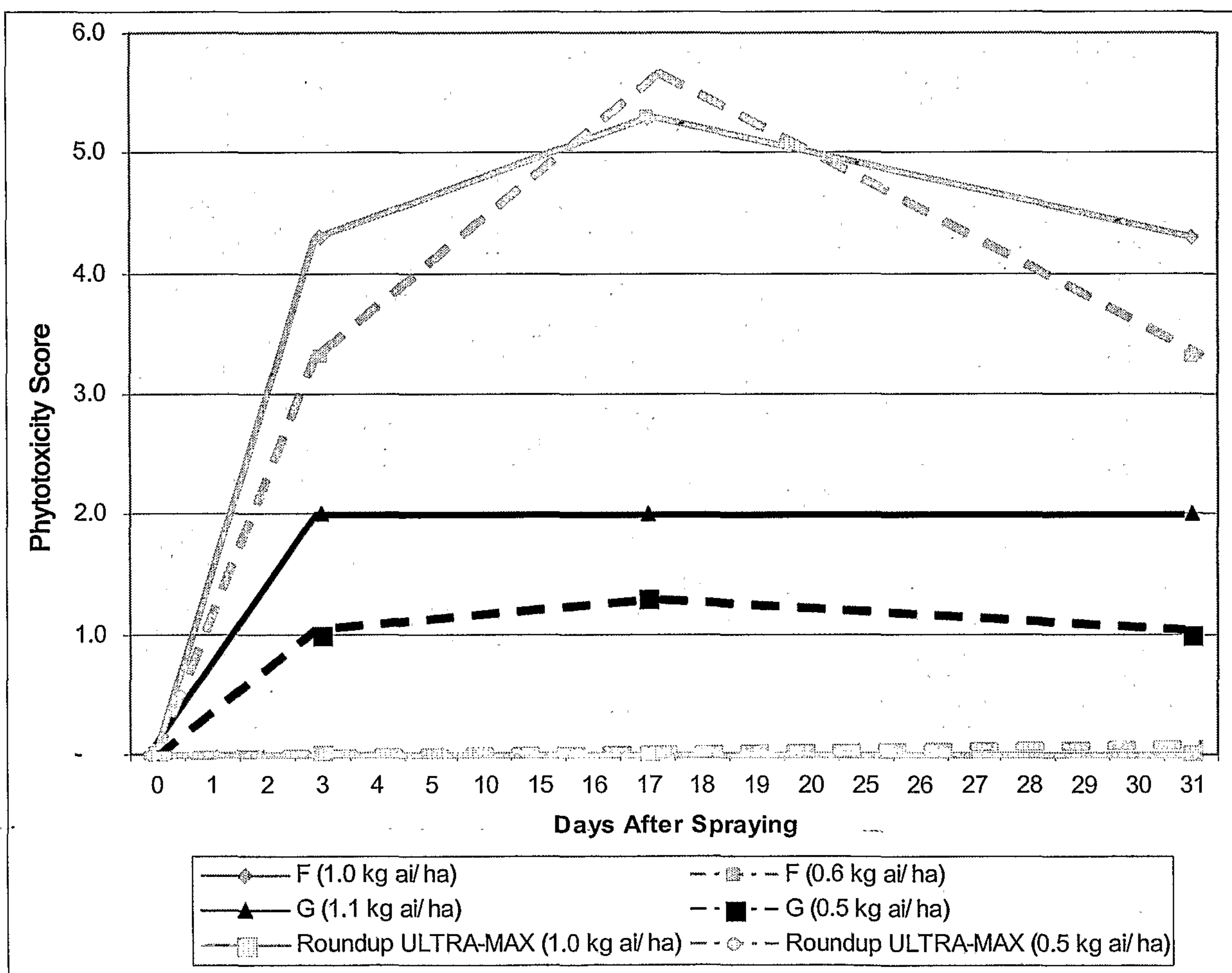
Note: No assessment of broad-leaved weeds was attempted 3 DAT due to apparent weed sparsity.

Figure 13. Broad-Leaved Weed Control T2: A Comparison of F and G with Roundup ULTRA MAX



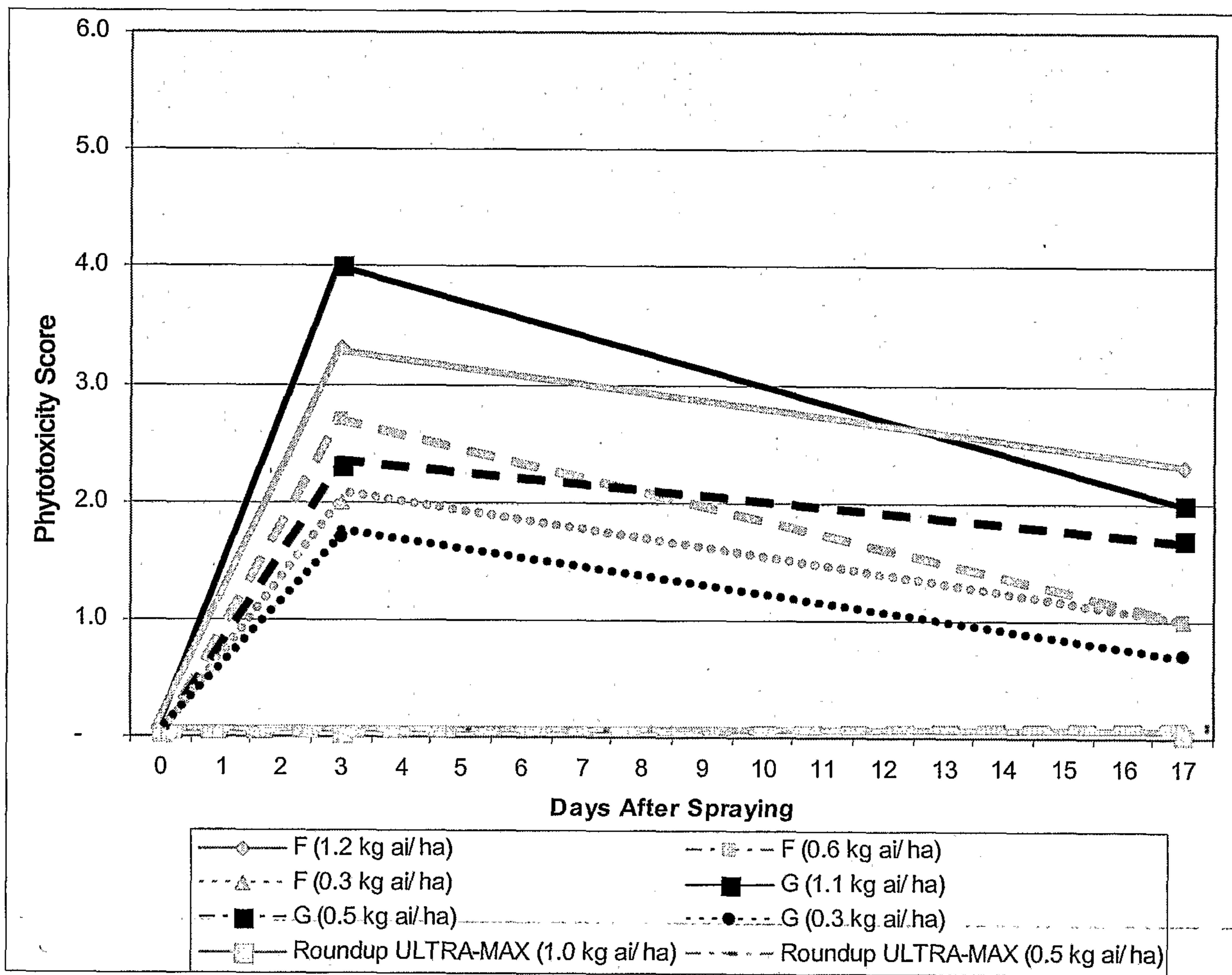
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Figure 14. Soybean Phytotoxicity T1: A Comparison of F and G with Roundup ULTRA MAX



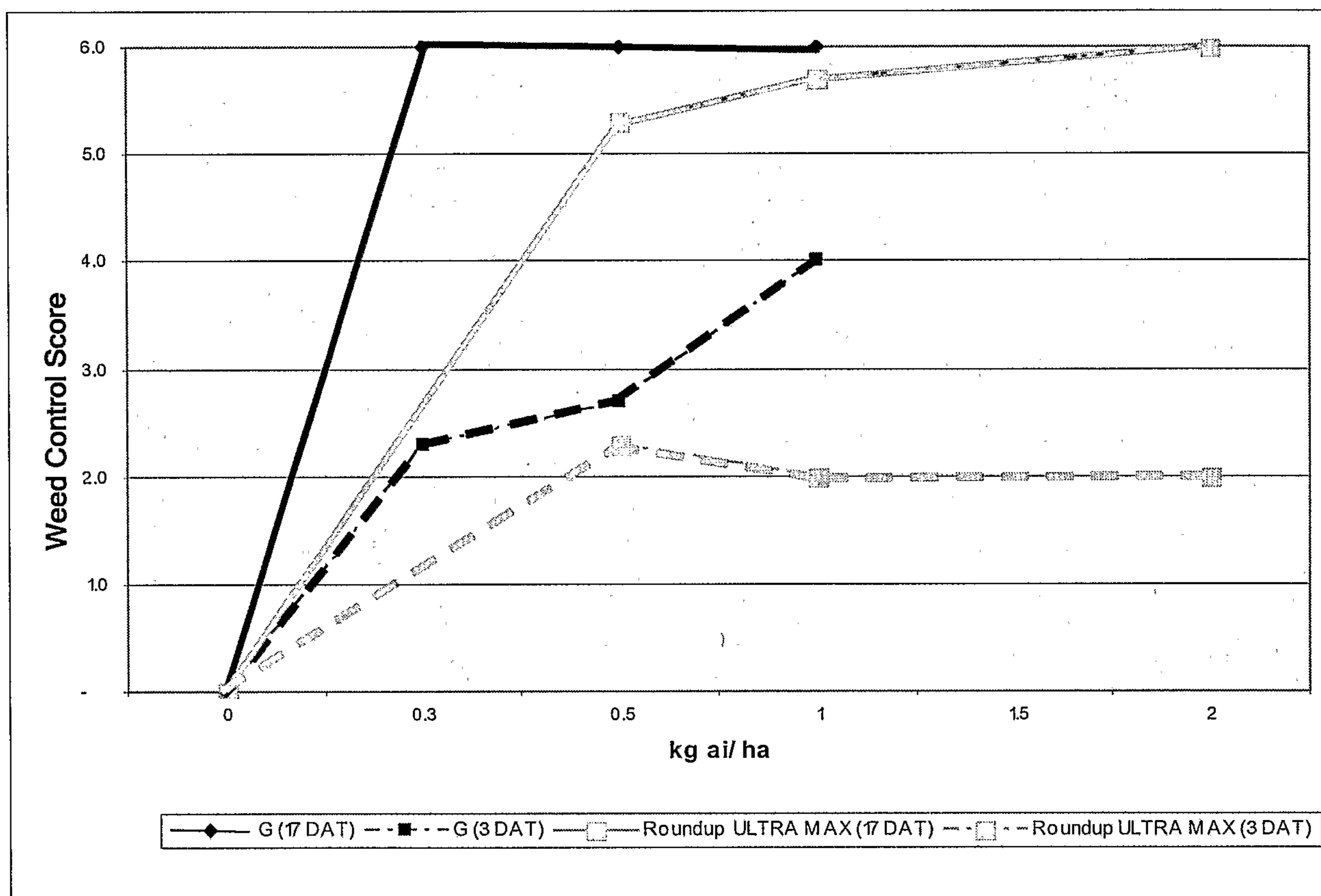
Note: Roundup ULTRA MAX did not produce any phytotoxicity at any rate of application up to 2.1 kg ai/ha at any time of assessment.

Figure 15. Soybean Phytotoxicity T2: A Comparison of F and G with Roundup ULTRA MAX



Note: Roundup ULTRA MAX did not produce any phytotoxicity at any rate of application up to 2.1 kg ai/ha at any time of assessment.

Figure 16. Broad-Leaved Weed Control T2: A Comparison of the Rate Response of the Most Efficacious EFET Formulation (G) with Roundup ULTRA MAX



Note: The weed species not fully controlled by Roundup ULTRA MAX at 0.5 and 1.0 kg ai/ha was *Abutilon theophrasti*, the most tolerant weed to glyphosate of the abundant species on the trial site (see Appendix A, Table 11).

Figure 17. *Setaria* Control T1: A Comparison of the Speed-of-Visual Effect of the Most Efficacious EFET Formulation (G) with Roundup ULTRA MAX

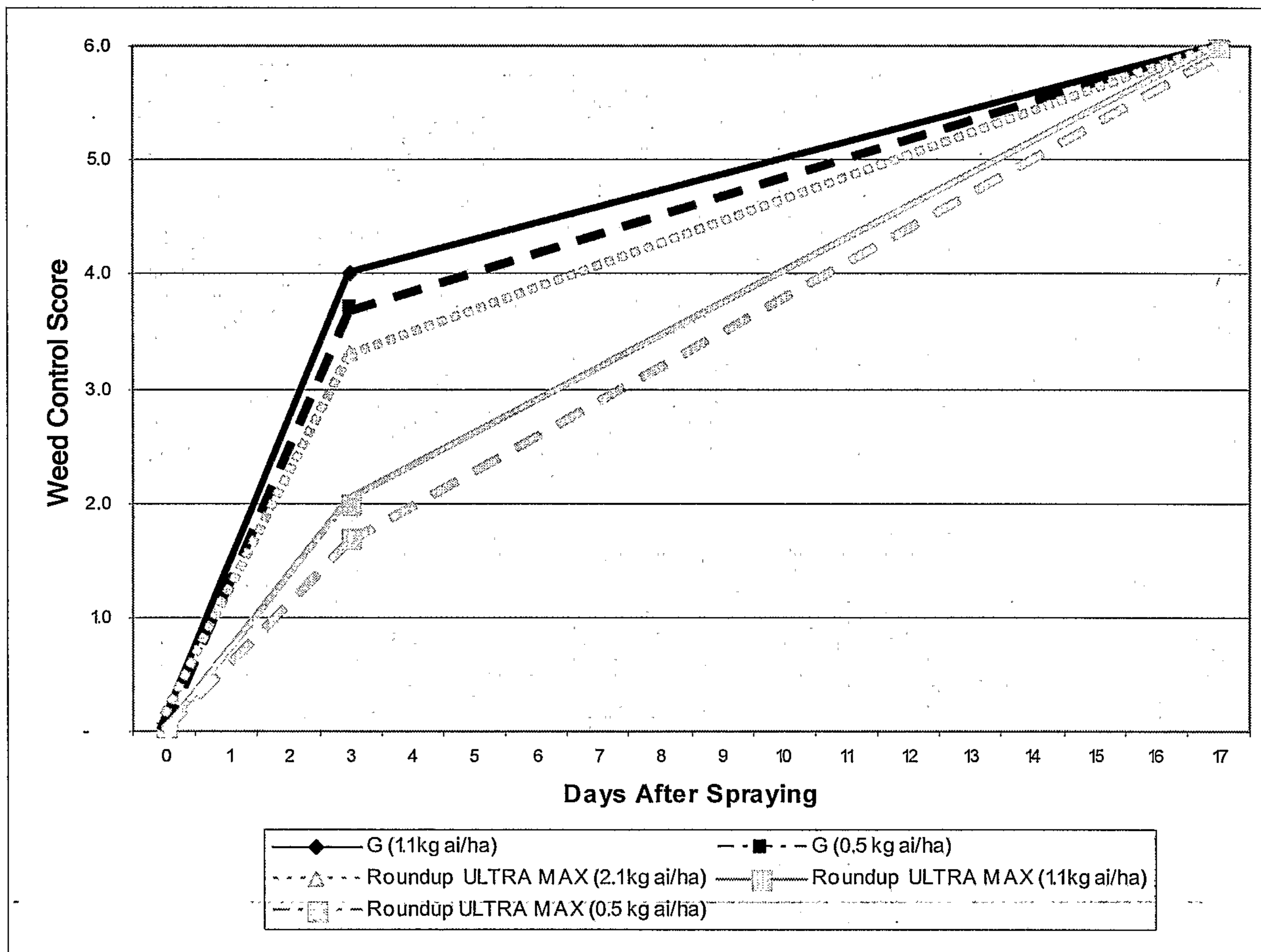
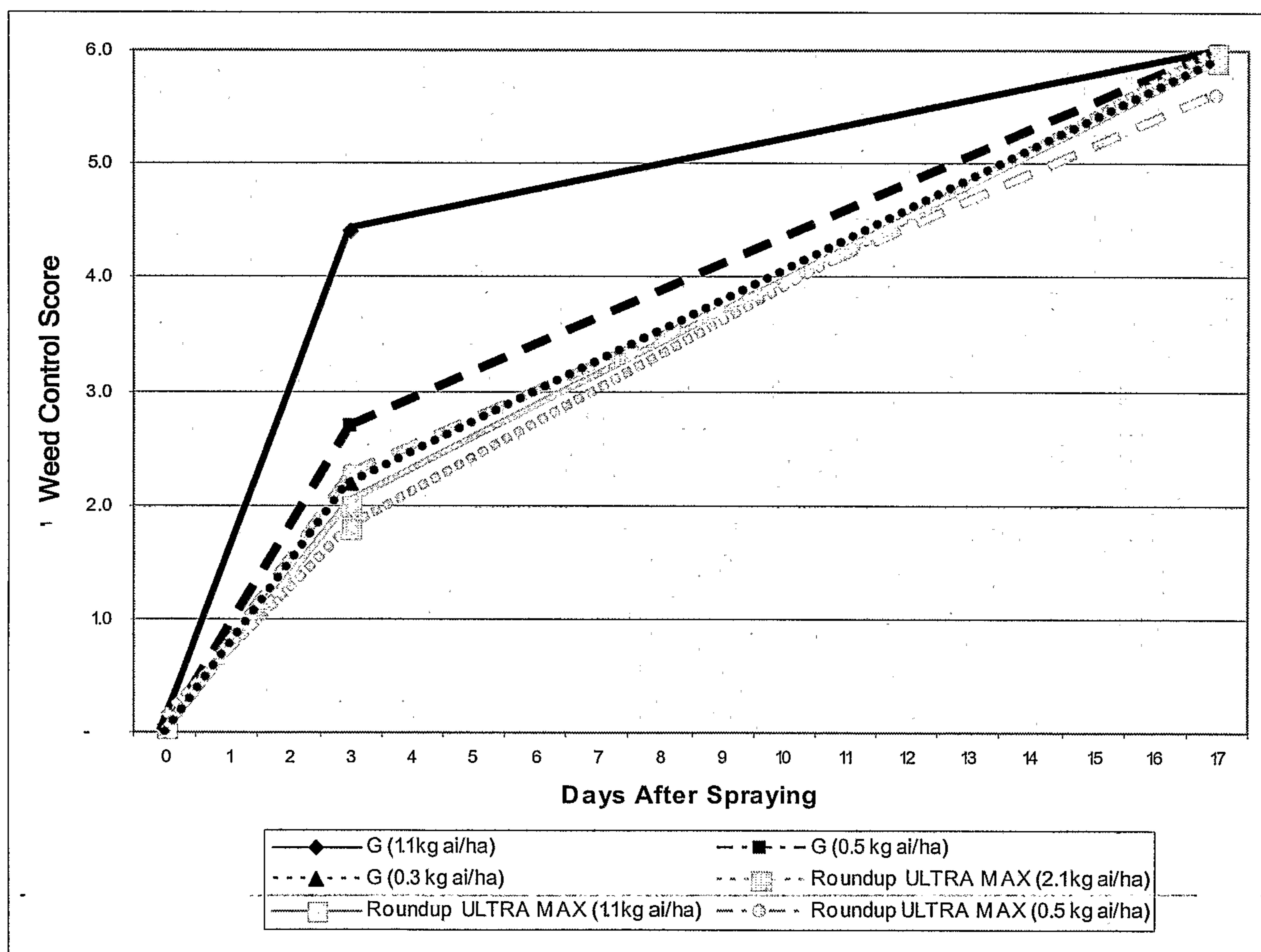




Figure 18. Total Weed Control T2: A Comparison of the Speed-of-Visual Effect of the Most Efficacious EFET Formulation (G) with Roundup ULTRA MAX



Note: The rate response scores for *Setaria* and broad-leaved weeds were very similar hence the data presented are averages for all weeds.

Figure 19. Glasshouse Experiment, *Setaria* Control. A Comparison of the Speed-of-Visual Effect from EFET Formulations (F and G) with Roundup ULTRA MAX

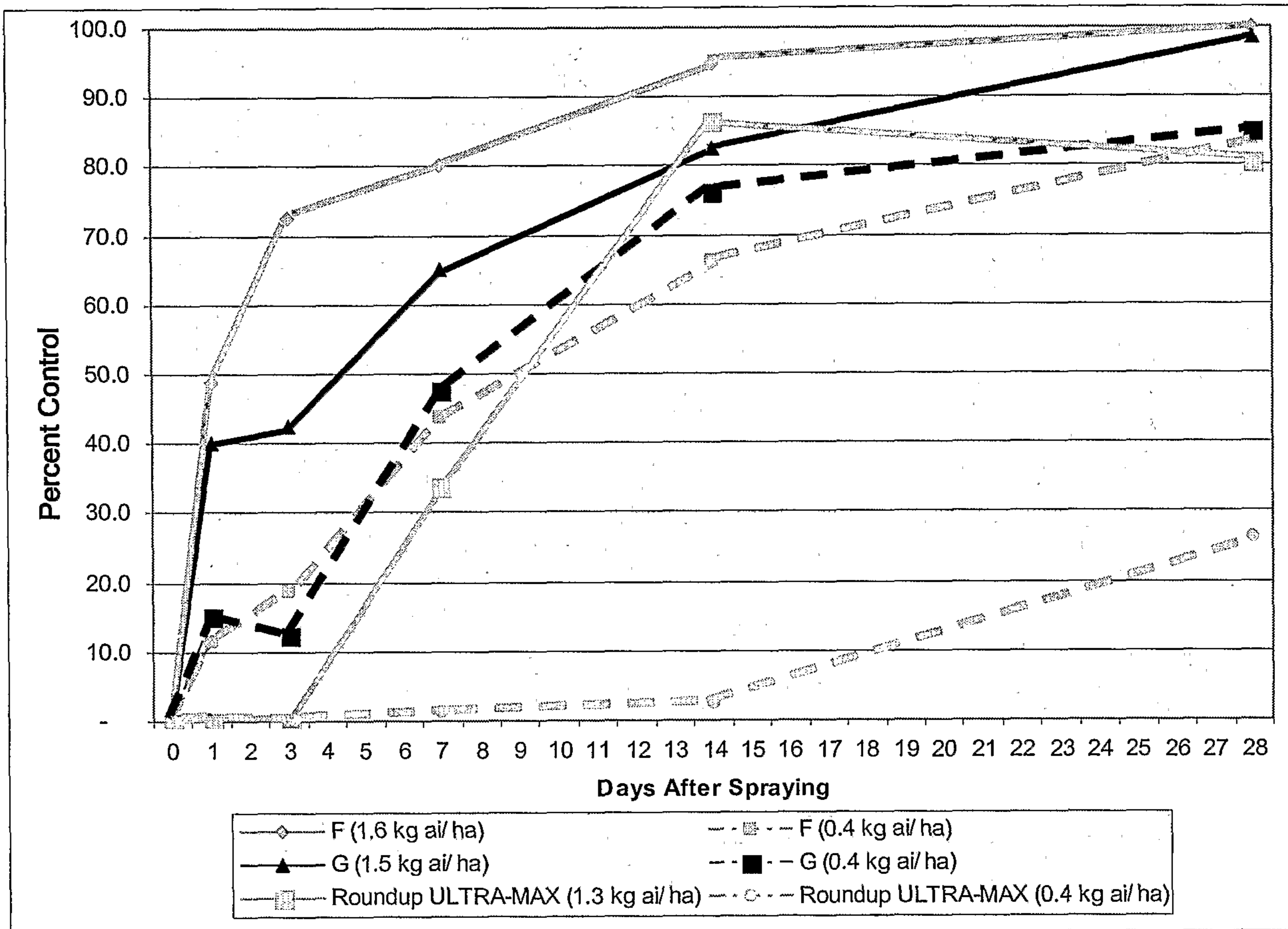
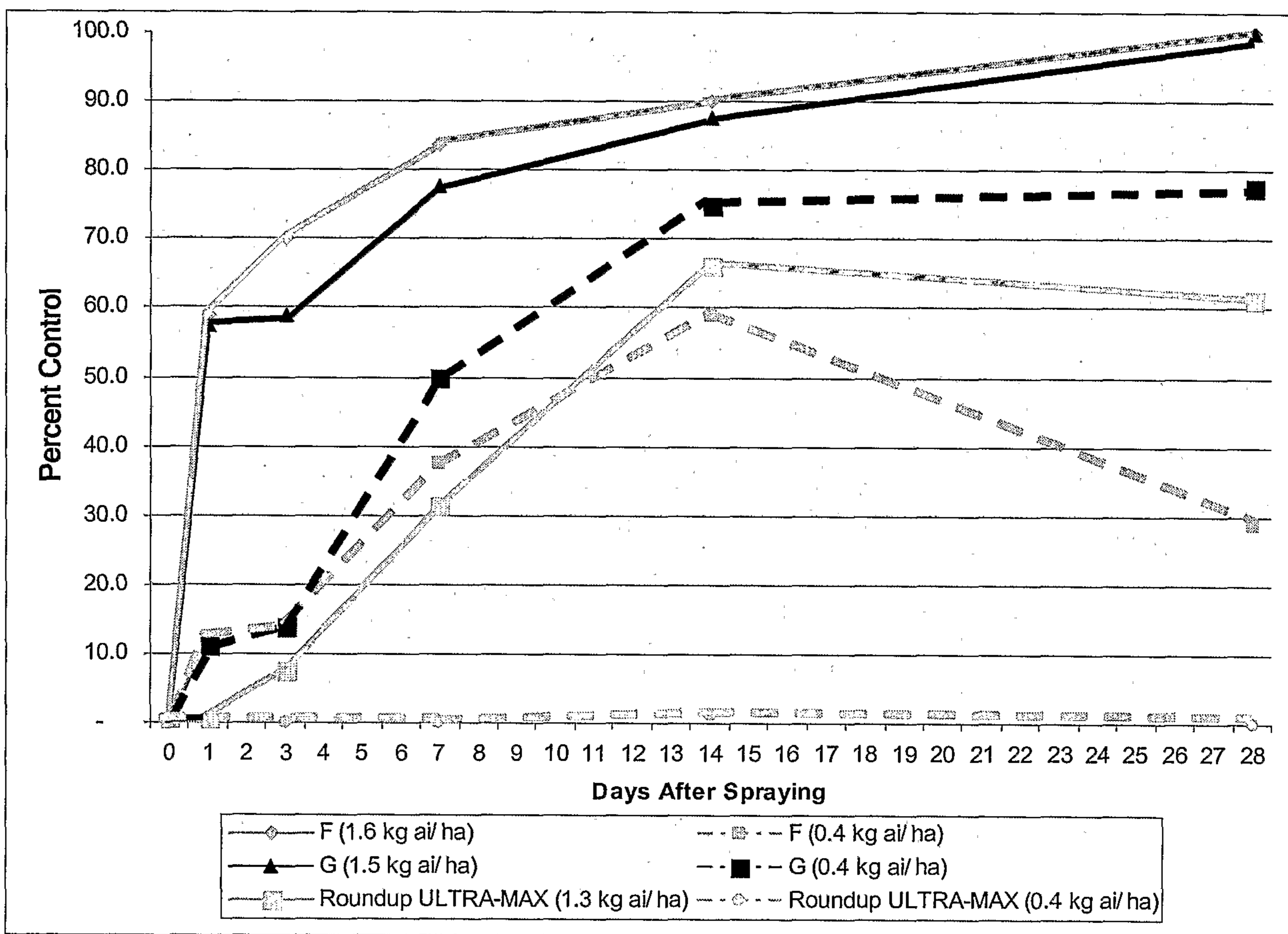
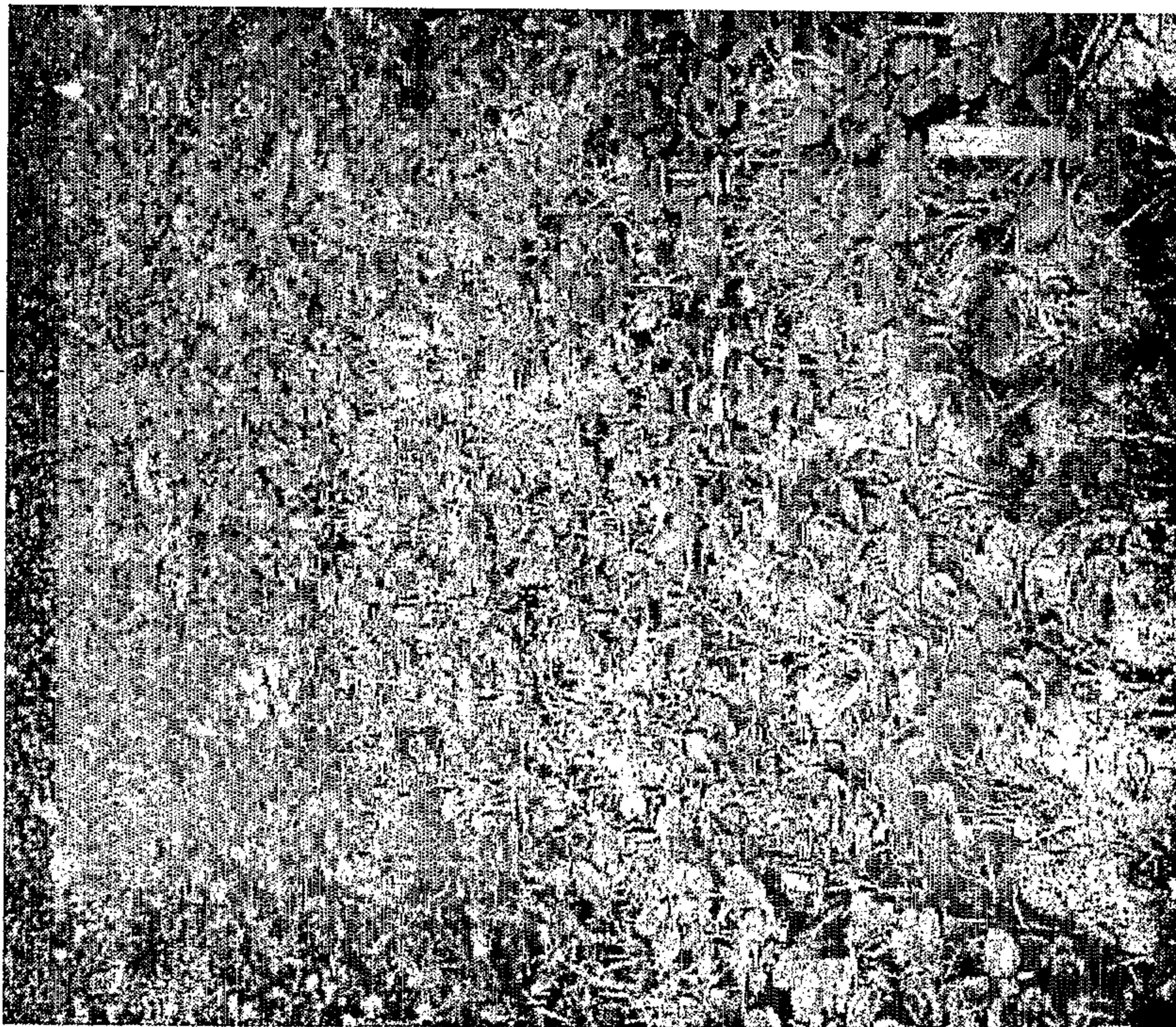


Figure 20. Glasshouse Experiment, *Abutilon* Control: A Comparison of the Speed-of-Visual Effect from EFET Formulations (F and G) with Roundup ULTRA MAX

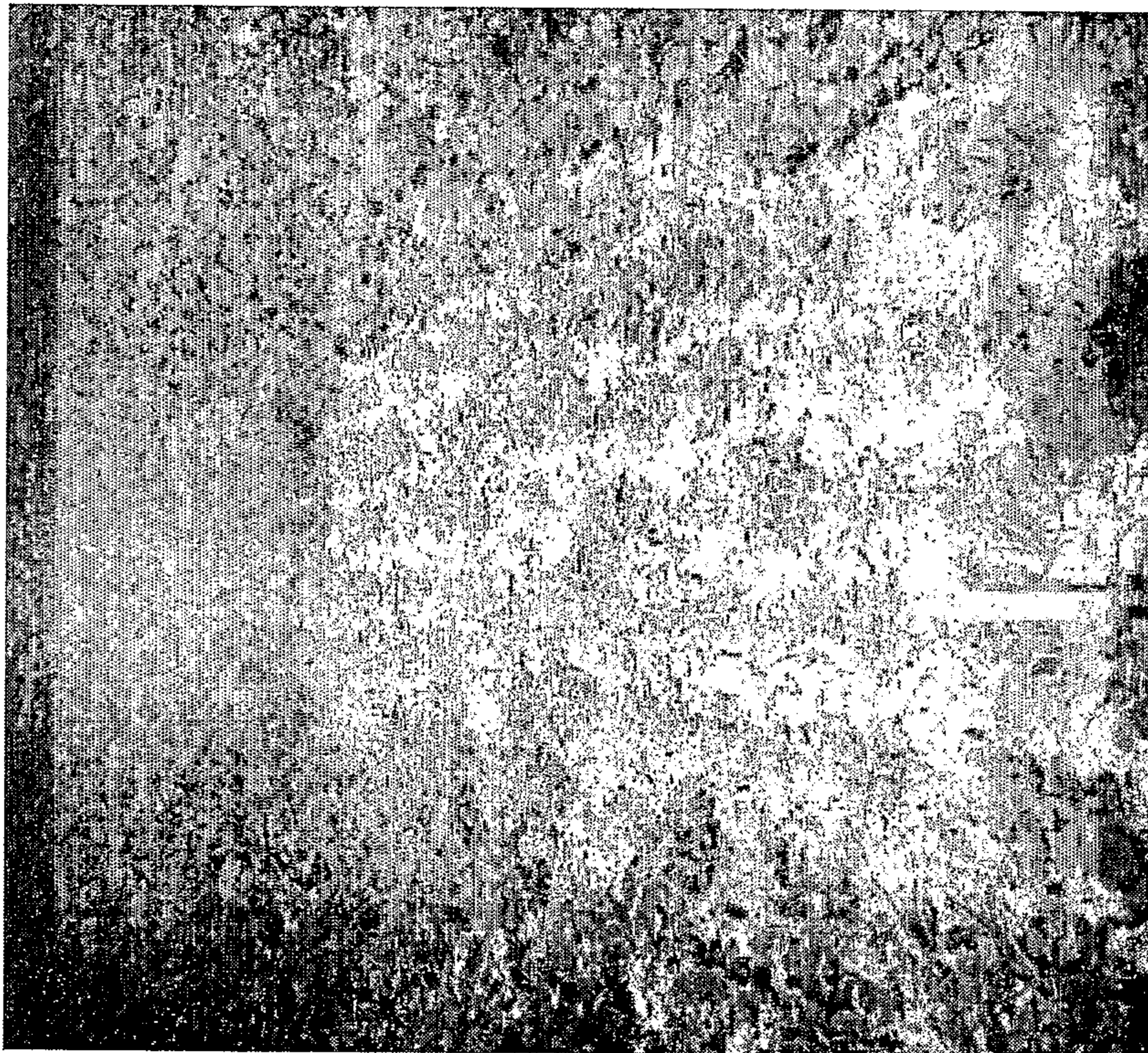


Soybean Phytotoxicity and General Weed Control T1: A Comparison (at 1.0 kg ai/ha) of EFET Formulations M2 with F, Four Days after Spraying

EFET Formulation M2

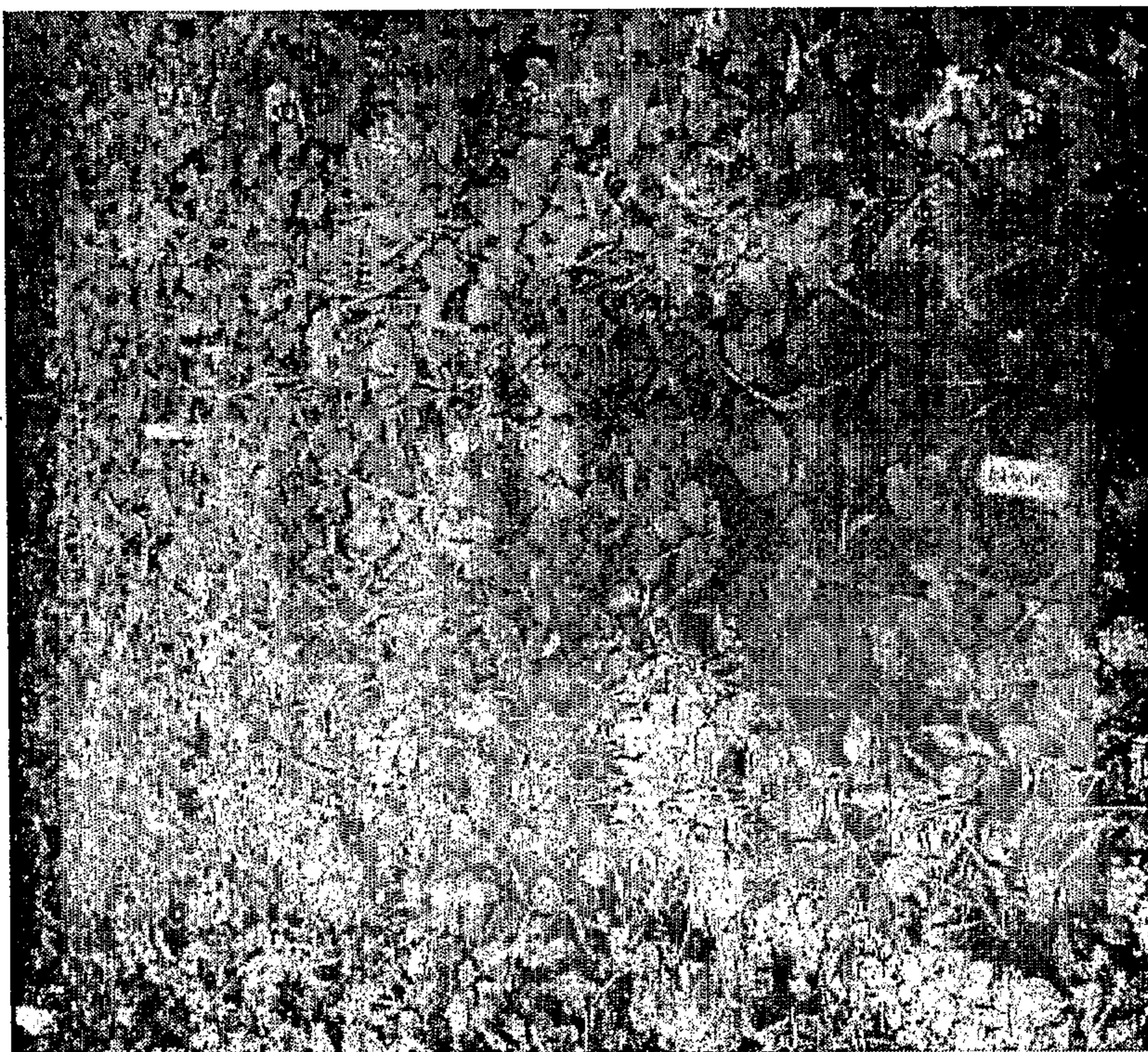


EFET Formulation F



Soybean Phytotoxicity and General Weed Control T1: A Comparison (at 0.5 kg ai/ha) of EFET Formulation and the Untreated Control, 4 Days after Spraying

Untreated Control

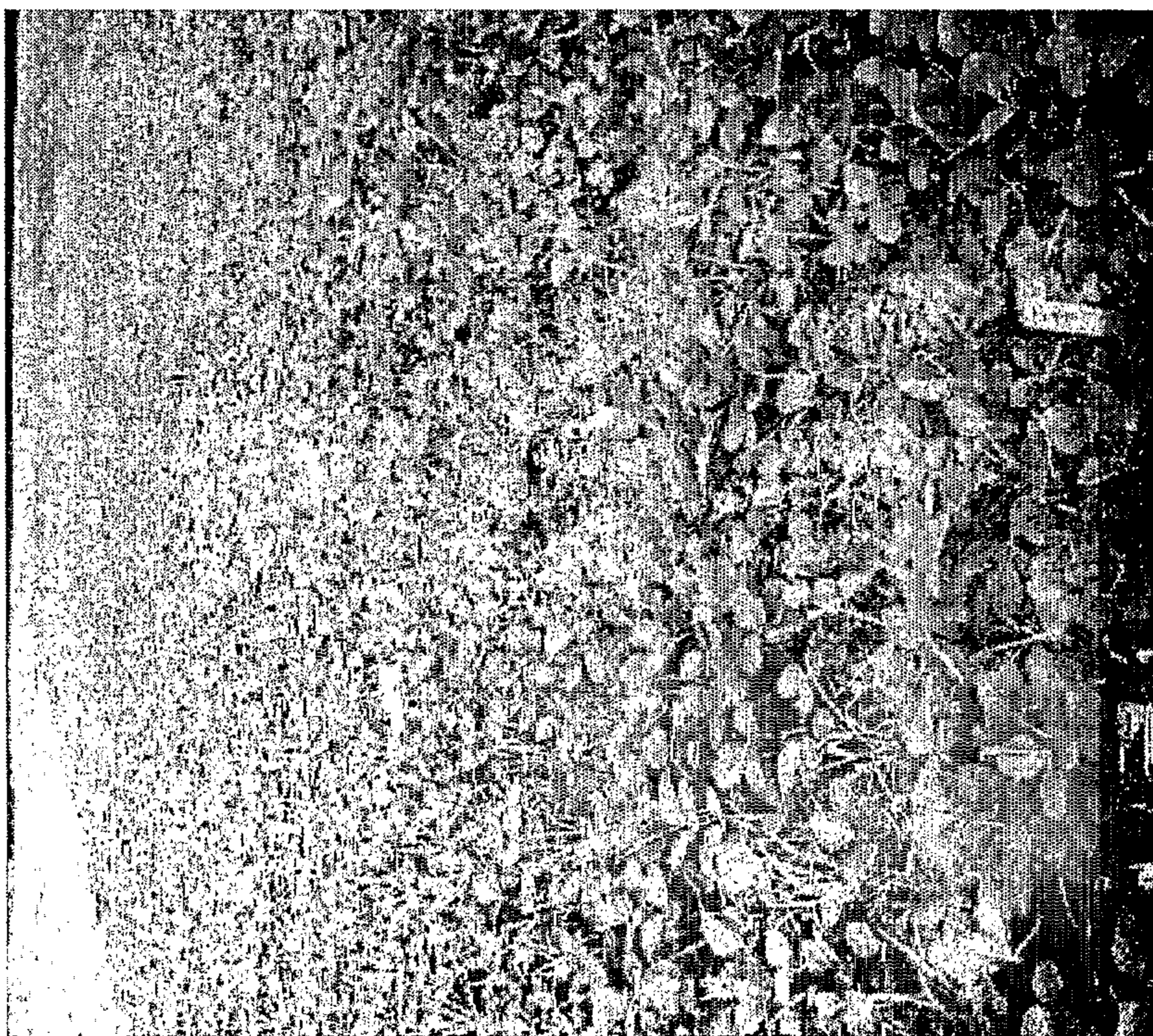


EFET Formulation F

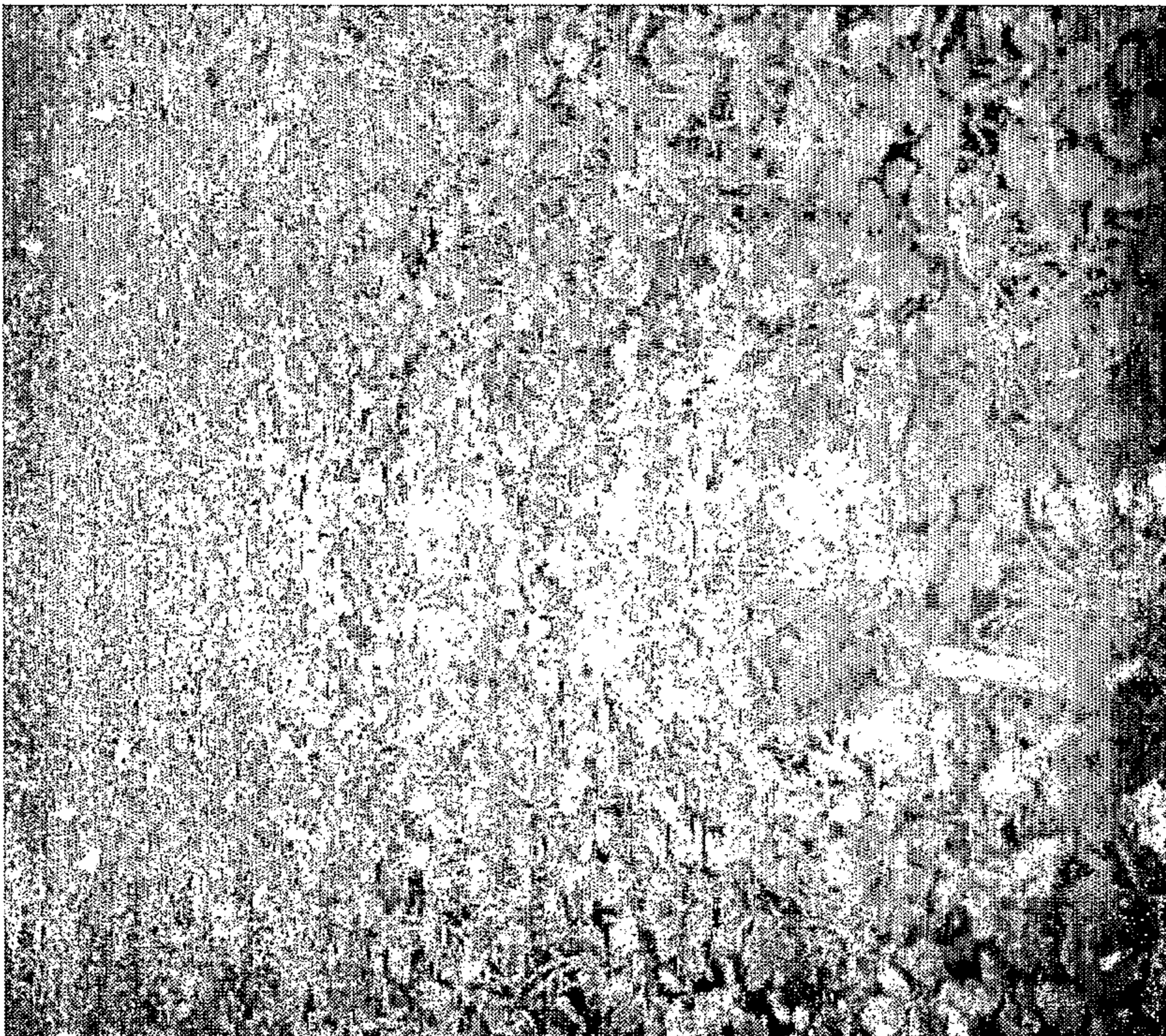


Soybean Phytotoxicity and General Weed Control T1: A Comparison (at 0.5 kg ai/ha) of EFET Formulation:  
Roundup ULTRA MAX and the Untreated Control, 4 Days after Spraying (Continued)

Roundup ULTRA MAX



EFET Formulation G



Soybean Phytotoxicity and General Weed Control T2: A Comparison of EFET Formulation F (at 0.5 kg ai/ha) with Roundup ULTRA MAX (at 1.0 kg ai/ha), 3 Days after Spraying

**Roundup ULTRA MAX**



**EFET Formulation F**



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General Weed Control T1: 100% Weed Control from the Most Efficacious EFET Formulation G (at 0.5 kg ai/ha), 31 Days after Spraying

**EFET Formulation G**





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Glasshouse Trial, Early Symptom Expression: A Comparison of EFET Formulation G (at 1.5 kg ai/ha) with the Untreated Control, within 3 Hours of Spraying

**EFET Formulation G**



**Untreated Control**

