1,035,092

PATENT SPECIFICATION

NO DRAWINGS

1,035,092

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Application made in United States of America (No. 232,311) on Oct. 22, 1962.
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Int. Cl.:—C 07 d 51/28 // A 01 n

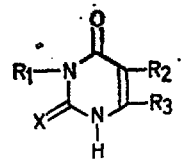
COMPLETE SPECIFICATION

Improvements relating to Herbicides and novel 3-Substituted Uracils

We, E. I. DU PONT DE NEMOURS AND COMPANY, a Corporation organised and existing under the laws of the State of Delaware, United States of America, of Wilmington 98, Delaware, United States of America, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

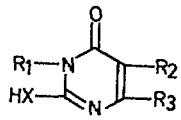
This invention relates to 3-substituted uracils and is an improvement in or modification of the invention described in our Patent No. 968,661.

Our Specification No. 968,661 discloses that certain 3-substituted uracils have herbicidal activity. Those 3-substituted uracils are compounds of the general formula:



I

or the corresponding isomeric formula:



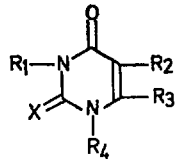
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wherein R_1 is a covalent organic radical of molecular weight 15 to 250 which is an alkyl group of up to 10 carbon atoms, a substituted alkyl group in which the alkyl group contains up to 3 carbon atoms, a phenyl group, a substituted phenyl group, an aralkyl group of 7 or 8 carbon atoms, a substituted aralkyl group of 7 to 12 carbon atoms, an alkenyl group of 3 to 6 carbon atoms, a propynyl group, a butynyl group, a cycloalkyl group of 5 to 8 carbon atoms, a cycloalkenyl group of 5 to 8 carbon atoms or a cyano group; R_2 is a hydrogen or halogen atom or an alkyl group of up to 4 carbon atoms, an alkoxy group of up to 3 carbon atoms, a nitro group, an alkoxymethyl group of 2 to 4 carbon atoms, a hydroxy-methyl group, a hydroxyethyl group, an allyl group, a thiocyanato group, a cyano group, a halogenomethyl group or a methyl thiomethyl group; R_3 is a hydrogen or chlorine

[*Price 4s. 6d.*]

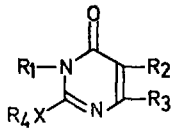
atom or an alkyl group of up to 5 carbon atoms, a methoxy group or an ethoxy group; or R_2 and R_3 together form a divalent alkylene bridge of formula $(CH_2)_n$, wherein n is 3, 4 or 5; and X is oxygen or sulphur, preferably oxygen; with the proviso that if R_2 is a hydrogen atom, then either R_3 is an alkyl group of 1 to 5 carbon atoms and R_1 is an alkyl group of 3 to 6 carbon atoms, an allyl group, a propynyl group or a cycloalkyl or cycloalkenyl group of 5 to 8 carbon atoms, or R_3 is a hydrogen atom and R_1 is a cycloalkyl or cycloalkenyl group of 5 to 8 carbon atoms; and metal salts of such uracils.

Within the broad class of 3-substituted uracils defined in Specification No. 968,661 are four particular classes which are the subject of our Patents Nos. 968,662; 968,663; 968,664 and 968,665, which are patents of addition to Patent No. 968,661. Certain other 3-substituted uracils having herbicidal properties are disclosed in our Patent No. 968,666, which is also a patent of addition to our Patent No. 968,661. These uracils, all of which have a substituent in the 1- or 2-position are compounds of the general formula:



I

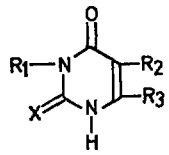
or the corresponding isomeric formula:



II

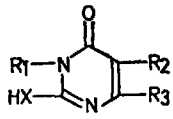
wherein R_1 is a covalent organic radical of molecular weight 15 to 250 which is an alkyl group of up to 10 carbon atoms, a substituted alkyl group in which the alkyl group contains up to 3 carbon atoms, a phenyl group, a substituted phenyl group, an aralkyl group of 7 or 8 carbon atoms, a substituted aralkyl group of 7 to 12 carbon atoms, an alkenyl group of 3 to 6 carbon atoms, a propynyl group, a butynyl group, a cycloalkyl group of 5 to 8 carbon atoms, a cycloalkenyl group of 5 to 8 carbon atoms or a cyano group; R_2 is a hydrogen or halogen atom or an alkyl group of up to 4 carbon atoms, an alkoxy group of up to 3 carbon atoms, a nitro group, an alkoxymethyl group of 2 to 4 carbon atoms, a hydroxymethyl group, a hydroxyethyl group, an allyl group, a thiocyanato group, a cyano group, a halogenomethyl group, or a methyl thiomethyl group; R_3 is a hydrogen or chlorine atom or an alkyl group of up to 5 carbon atoms, a methoxy group, or an ethoxy group; or R_2 and R_3 together form a divalent alkylene bridge of formula $(CH_2)_n$, wherein n is 3, 4 or 5; R_4 is a covalent radical of molecular weight up to 250 which is an alkyl group of up to 3 carbon atoms, a hydroxymethyl group, a hydroxyethyl group, a carboxyethyl group, a substituted carbamyl group, a substituted thiocarbamyl group, a diethylphosphoryl group, a chloromethylthio group, a phenylthio group, a halogenophenylthio group, a nitrophenylthio group, a dinitrophenylthio group, a tolylthio group or an acyl group; and X is oxygen or sulphur; with the proviso that if R_2 is a hydrogen atom and R_4 is a methyl group, then R_1 is a cycloalkyl or cycloalkenyl group of 5 to 8 carbon atoms.

The present invention relates to another group of 3-substituted uracils which have herbicidal activity and are believed to be new compounds, namely compounds of the general formula:



I

or the corresponding isomeric formula:



II

wherein R_1 represents a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms, a (substituted cycloalkyl)alkyl radical containing 5 to 14 carbon atoms or a (substituted cycloalkenyl)alkyl radical containing 5 to 14 carbon atoms; R_2 represents a hydrogen, chlorine, fluorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 6 carbon atoms, a hydroxy alkyl radical containing 1 to 6 carbon atoms, an alkenyl radical containing 3 to 6 carbon atoms, a thiocyanato radical, a cyano radical, a mercaptomethyl (alternatively named methylthiol) radical, a methylthiomethyl radical, a phenylthiomethyl radical, an alkylthio radical containing 1 to 4 carbon atoms, a bromomethyl radical, a fluoromethyl radical, a chloromethyl radical or a carboxymethylthiomethyl radical; R_3 represents a hydrogen, chlorine or bromine atom, an alkyl radical containing 1 to 5 carbon atoms, a chloroalkyl radical containing 1 to 4 carbon atoms, a bromoalkyl radical containing 1 to 4 carbon atoms, or an alkoxy radical containing 1 to 5 carbon atoms; or R_2 and R_3 together represent a divalent alkylene radical of formula $(CH_2)_n$ where n is

3, 4 or 5; with the proviso that when R_2 represents a hydrogen atom, R_3 represents hydrogen or an alkyl radical containing 1 to 5 carbon atoms, and X represents an oxygen or sulphur atom, and metal salts thereof. X preferably represents an oxygen atom.

If R_1 represents a (substituted cycloalkyl) alkyl or (substituted cycloalkenyl)alkyl radical, the or each substituent is preferably an alkyl radical containing 1 to 4 carbon atoms, a methoxy radical or a chlorine or bromine atom.

The cycloalkyl or cycloalkenyl radical in R_1 may for example be a cyclohexyl or cyclohexenyl radical, as is particularly preferred, or a cyclopentyl, cyclopentenyl, norbornyl, norbornenyl, bicyclo(2,2,2)octyl,bicyclo(2,2,2)-octenyl, cyclobutyl, cyclobutenyl, hexahydroindenyl, hexahydroindanyl, tetrahydroindanyl, hexahydro-4,7-methanoindenyl, hexahydro-4,7-methanoindanyl, tetrahydro - 4,7 - methanoindanyl, decahydronaphthyl, tetrahydronaphthyl, decahydro-1,4-methanonaphthyl, octahydro - 1,4 - methanonaphthyl, decahydro - 1,4,5,8 - dimethanonaphthyl or octahydro - 1,4,5,8 - dimethanonaphthyl radical.

If the 3-substituted uracil is a metal salt, it is preferably a sodium, potassium or lithium salt, or it may for example be a calcium, magnesium, barium, strontium or manganese salt.

In one class of the 3-substituted uracils, R_1 represents a cycloalkyl alkyl radical containing 4 to 13 carbon atoms or a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents an alkyl radical containing 1 to 5 carbon atoms.

In a second class, R_1 is a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms or a (substituted cycloalkyl) alkyl or (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms and R_2 and R_3 each represents a hydrogen atom.

In a third class, preferred because of the generally excellent herbicidal properties of the compounds within it, X represents an oxygen atom; R_1 represents a cycloalkyl alkyl radical containing 4 to 13 carbon atoms or a substituted cycloalkyl alkyl radical containing 5 to 13 carbon atoms; R_2 represents a chlorine, bromine or iodine atom, a methyl radical, a hydroxymethyl radical, a methoxymethyl radical or a nitro radical; and R_3 represents a hydrogen atom or a methyl radical.

In a fourth class, R_1 is a substituted or unsubstituted cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms or a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms; R_2 represents a chlorine, fluorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 6 carbon atoms, a hydroxy alkyl radical containing 1 to 6 carbon atoms, an alkenyl radical containing 3 to 6 carbon atoms, a thiocyanato radical, a cyano radical, a mercaptomethyl radical, a methylthiomethyl radical, phenylthiomethyl radical, a chloromethyl radical or a carboxymethylthiomethyl radical; and R_3 represents a chlorine or bromine atom, an alkyl radical containing 1 to 5 carbon atoms, a chloroalkyl radical containing 1 to 4 carbon atoms, a bromoalkyl radical containing 1 to 4 carbon atoms or an alkoxy radical containing 1 to 5 carbon atoms.

In a fifth class R_1 represents a cycloalkyl alkyl radical containing 4 to 13 carbon atoms or a (substituted cycloalkyl)alkyl radical containing 5 to 13 carbon atoms, R_2 represents a chlorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 5 carbon atoms, a hydroxymethyl radical, a hydroxyethyl radical, a cyano radical or a methylthiomethyl radical; and R_3 represents a chlorine or bromine atom or a methyl, ethyl, propyl, chloromethyl or bromomethyl radical.

In a sixth class R_1 represents a cycloalkyl alkyl radical containing 6 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 6 to 13 carbon atoms, a (substituted cycloalkyl)alkyl radical containing 6 to 14 carbon atoms or a (substituted cycloalkenyl)alkyl radical containing 6 to 14 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents an alkyl radical containing 1 to 5 carbon atoms.

In a seventh class, R_1 represents a substituted or unsubstituted cycloalkyl alkyl or cycloalkenyl alkyl radical as defined in the second class above; R_2 is as defined in the fourth class above or represents an alkyl thio radical containing 1 to 4 carbon atoms, a bromomethyl radical or a fluoromethyl radical; and R_3 represents a chlorine or bromine atom, an alkyl radical containing 1 to 5 carbon atoms, a chloroalkyl radical containing 1 to 4 carbon atoms, a bromoalkyl radical containing 1 to 4 carbon atoms, or an alkoxy radical containing 1 to 5 carbon atoms.

In an eighth class R_1 represents a substituted or unsubstituted cycloalkyl alkyl or cycloalkenyl alkyl radical; R_2 represents a hydrogen atom and R_3 represents an alkyl radical containing 1 to 5 carbon atoms.

The 3-substituted uracils to which the present invention relates can be prepared by methods known for the preparation of related compounds, including those disclosed in Specifications Nos. 968,661; 968,662; 968,663;

968,664 and 968,665. Examples of specific methods are disclosed in the Examples given later in this specification.

It will be noted that the definition above refers to the isomeric enol form of the uracil, as well as the usual keto form. It is thought that there is a tautomeric equilibrium between the two forms. However, for the sake of convenience the keto form is generally referred to herein. It may also be noted that since "uracil" is the commonly accepted name for 2,4-dihydroxypyrimidine, or, in its keto form, 1,2,3,4-tetra-hydropyrimidine-2,4-dione, the compounds can also be named as pyrimidines.

The compounds to which the present invention relates (referred to herein as "the said compounds") can be formulated as herbicidal compositions comprising at least one of the said compounds associated ~~with~~ a compatible diluent or surface-active agent, or both. The term "herbicidal composition" is used herein to include both compositions which are ready for use as herbicides and concentrates which require dilution before use as herbicides. The compositions may contain for example 0.5 to 95% by weight of the 3-substituted uracil. (Parts, proportions and percentages are by weight throughout this specification except where otherwise noted.)

The diluent may be a liquid other than an oil, especially water, or an oil, or a solid carrier. The term "oil" is used herein to denote a liquid which is immiscible with water and soluble in ether, and which has a boiling point not lower than 125°C. Two or more different types of diluent may be present, as for example in compositions obtained by dispersing a wettable powder in a liquid diluent. Liquid compositions may be in the form of solutions or dispersions.

Powder and dust preparations can be made by mixing the said compounds with finely-divided solids, preferably talcs, natural clays, pyrophyllite, diatomaceous earth, flours such as walnut shell, wheat, redwood, soya bean and cotton seed flours, or inorganic substances such as magnesium carbonate, calcium carbonate, calcium phosphate, sulphur and lime. These preparations are made by thoroughly blending the active ingredient and the solid. The particles in such preparations are preferably less than 50 microns in average diameter.

Water-soluble preparations can be prepared by mixing a said compound with an alkaline solubilising agent. Solid bases having a pH of at least 9.5 in a 1% aqueous solution, such as sodium or potassium phosphates, silicates, carbonates, borates, oxides or hydroxides are suitable. The preparations can contain from 0.5 to 80% of the said compound and from 5 to 99.5% of the solubilising agent. Water-soluble diluents such as urea and dextrose may also be used.

Granules and pellets can be made by mixing a said compound in finely-divided form with a suitable clay, moistening this mixture with from 15 to 20% by weight of water, and for example extruding the mass through a suitable die under pressure. The extrusions are cut into pellets of pre-determined lengths and then dried. These pellets can be granulated if desired.

Granules or pellets can also be prepared by spraying a suspension or solution of a said compound onto the surface of a preformed granule of clay, vermiculite or othere suitable granular material. If the said compound is in solution, it will penetrate into the pores of the granule and so will adhere without the aid of a binding agent. When the said compound is insoluble in the liquid and is carried as a suspension, it is preferable that a binding agent such as goulac, dextrin, swollen starch, glue or polyvinyl alcohol be added. In either case, the granule is then dried and ready for use.

The said compounds can also be formulated with non-aqueous liquids. Aliphatic and aromatic hydrocarbons, especially those derived from petroleum and having boiling points of from 125°C. to 400°C. are preferred. Hydrocarbons having lower boiling points should not be used because of their undesirable volatilisation characteristics and inflammability. In the case of those compounds having inadequate solubility in the carrier, it is convenient to prepare liquid compositions by milling the components in a mill such as a pebble mill until the particles have average diameters of from 1 to 50 microns, preferably 5 to 20 microns.

The herbicidal compositions, whatever physical form they take, can also contain a surface-active agent. The surfactant renders the preparations readily dispersible in liquids and improves their action on waxy leaves and the like. For general application, surface-active agents are used in the compositions at concentrations of from 1 to 10% by weight. Levels of from 0.5 to 6 parts of surfactant for each part of uracil, however, sometimes give unusual and unexpected results. Preparations having these higher levels of surfactants sometimes show greater herbicidal effectiveness than can be expected from a consideration of activity of the components used separately.

The term "surface-active agent" is used herein to include wetting agents, dispersing agents, suspending agents and emulsifying agents. Surface-active agents suitable for use are set forth in "Detergents and Emulsifiers . . . up-to-date", 1962, John W. McCutcheon, Inc., Morristown, New Jersey. Other surface-active agents which can be used in these preparations are listed in U.S. Patent Specifications 2,412,510; 2,426,417 and 2,655,447, British Specification No. 492,742 and Bulletin E-607 of the Bureau of Entomology and Plant

Quarantine of the U.S. Department of Agriculture.

The compositions can also optionally contain adhesives such as gelatin, blood albumin and such resins as rosin alkyd resins. These increase retention and tenacity of deposits following application.

The preparations can also contain corrosion inhibitors, antifoam agents, dyes and pigments, anticaking agents, and hard water stabilisers.

The herbicidal compositions can be formulated to contain two or more of the said compounds. They can also be formulated to contain other herbicides in addition to the said compounds to give compositions which have advantages over compositions containing only the individual herbicides. The other herbicide may be a 3-substituted uracil or salt or nitrogenous base or phenol addition compound thereof as disclosed in our copending application Nos. 46342/62, 1,035,091; 6192/66, 1,035,093; 6193/66, 1,035,094; 6194/66, 1,035,095; 6195/66, 1,035,096; 6196/66, 1,035,097, and 8987/66, 1,035,098.

Among the known herbicides which can be combined with the said compounds are:

Substituted Ureas

3-(3,4-dichlorophenyl)-1,1-dimethylurea
3-(4-chlorophenyl)-1,1-dimethylurea
3 - phenyl - 1,1 - dimethylurea
3 - (3,4 - dichlorophenyl) - 3 - methoxy - 1,1 - dimethylurea
3 - (4 - chlorophenyl) - 3 - methoxy - 1,1 - dimethylurea
3 - (3,4 - dichlorophenyl) - 1 - *n* - butyl - 1 - methylurea
3 - (3,4 - dichlorophenyl) - 1 - methoxy - 1 - methylurea
3 - (4 - chlorophenyl) - 1 - methoxy - 1 - methylurea
3 - (3,4 - dichlorophenyl) - 1,1,3 - trimethylurea
3 - (3,4 - dichlorophenyl) - 1,1 - diethylurea
3 - (*p* - chlorophenoxyphenyl) - 1,1 - dimethylurea

These ureas can be mixed with the said compounds in proportions of from 1:4 to 4:1, respectively, the preferred ratio being 1:2 to 2:1.

Substituted Triazines

2-chloro-4,6-bis(ethylamino)-*s*-triazine
2 - chloro - 4 - ethylamino - 6 - isopropylamino - *s* - triazine
2 - chloro - 4,6 - bis(methoxypropylamino) - *s* - triazine
2 - methoxy - 4,6 - bis(isopropylamino) - *s* - triazine
2 - diethylamino - 4 - isopropylacetamido - 6 - methoxy - *s* - triazine
2 - isopropylamino - 4 - methoxyethylamino - 6 - methylmercapto - *s* - triazine
2 - methylmercapto - 4,6 - bis(isopropylamino) - *s* - triazine
2 - methylmercapto - 4,6 - bis(ethylamino) - *s* - triazine
2 - methylmercapto - 4 - ethylamino - 6 - isopropylamino - *s* - triazine
2 - methoxy - 4,6 - bis(ethylamino) - *s* - triazine
2 - methoxy - 4 - ethylamino - 6 - isopropylamino - *s* - triazine
2 - chloro - 4,6 - bis(isopropylamino) - *s* - triazine

These triazines can be mixed with the said compounds in proportions of from 1:4 to 4:1 respectively, the preferred ratio being 1:2 to 2:1.

Phenols

Dinitro-o-*sec.*-butylphenol and its salts
Pentachlorophenol and its salts

These phenols can be mixed with the said compounds in the proportions of 1:10 to 20:1, respectively, the preferred ratio being 1:5 to 5:1.

Carboxylic Acids and Derivatives

The following carboxylic acids and derivatives can be mixed with the said compounds in the listed proportions:

A. 2,3,6-trichlorbenzoic acid and its salts
2,3,5,6-tetrachlorobenzoic acid and its salts
2-methoxy-3,5,6-trichlorobenzoic acid and its salts
2-methoxy-3,6-dichlorobenzoic acid and its salts
3-amino-2,5-dichlorbenzoic acid and its salts
3-nitro-2,5-dichlorobenzoic acid and its salts
2-methyl-3,6-dichlorobenzoic acid and its salts
2,4-dichlorophenoxyacetic acid and its salts and esters
2,4,5-trichlorophenoxyacetic acid and its salts and esters
(2-methyl-4-chlorophenoxy)acetic acid and its salts and esters
2-(2,4,5-trichlorophenoxy)propionic acid and its salts and esters
2 - (2,4,5 - trichlorophenoxy)ethyl 2,2 - dichloropropionate
4-(2,4-dichlorophenoxy)butyric acid and its salts and esters.
4-(2-methyl-4-chlorophenoxy)butyric acid and its salts and esters
2,3,6-trichlorobenzoyloxypropanol
Mixed in a 1:16 to 8:1 ratio, preferably a 1:4 to 4:1 ratio.

B. 2,6-dichlorobenzonitrile
Mixed in a 1:4 to 4:1 ratio, preferably a 1:3 to 3:1 ratio.

C. Trichloroacetic acid and its salts
Mixed in a 1:2 to 25:1 ratio, preferably a 1:1 to 8:1 ratio.

D. 2,2-dichloropropionic acid and its salts
Mixed in a 1:4 to 8:1 ratio, preferably a 1:2 to 4:1 ratio.

E. N,N-di(*n*-propyl)thiolcarbamic acid, ethyl ester
N,N-di(*n*-propyl)thiolcarbamic acid, *n*-propyl ester
N - ethyl - N - (*n* - butyl)thiolcarbamic acid, ethyl ester
N - ethyl - N - (*n* - butyl)thiolcarbamic acid, n-propyl ester
Mixed in a 1:2 to 24:1 ratio, preferably a 1:1 to 12:1 ratio.

F. N-phenylcarbamic acid, isopropyl ester
N-(*m*-chlorophenyl)carbamic acid, isopropyl ester
N-(*m*-chlorophenyl)carbamic acid, 4-chloro-2-butynyl ester
Mixed in a 1:2 to 24:1 ratio, preferably a 1:1 to 12:1 ratio.

G. 2,3,6-trichlorophenylacetic acid and its salts
Mixed in a 1:12 to 8:1 ratio, preferably a 1:4 to 4:1 ratio.

H. 2-chloro-N,N-diallylacetamide
Maleic hydrazide
Mixed in a 1:2 to 10:1 ratio, preferably a 1:1 to 5:1 ratio.

Inorganic and Mixed Inorganic-Organic Salts

The following salts can be mixed with the said compounds in the listed proportions:

A. Calcium propylarsonate
Disodium monomethylarsonate
Octyl-dodecylammoniummethylarsonate
Dimethylarsinic acid
Mixed in a 1:4 to 4:1 ratio, preferably a 1:2 to 2:1 ratio.

B. Sodium arsenite
Mixed in a 1:5 to 40:1 ratio, preferably a 1:4 to 25:1 ratio.

C. Lead arsenate
Calcium arsenate
Mixed in a 100:1 to 600:1 ratio, preferably a 150:1 to 400:1 ratio.

D. Sodium tetraborate hydrated, granulated
Sodium metaborate
Sodium pentaborate
Polyborchlorate
Unrefined borate ore such as borascu
Mixed in a 3:1 to 1500:1 ratio, preferably a 6:1 to 1000:1 ratio.

E. Ammonium thiocyanate
Mixed in a 1:10 to 20:1 ratio, preferably a 1:5 to 5:1 ratio.

F. Sodium chlorate
Mixed in a 1:1 to 40:1 ratio, preferably a 2:1 to 20:1 ratio.

G. Ammonium sulphamate
Mixed in a 1:1 to 100:1 ratio, preferably a 1:1 to 50:1 ratio.

Other Organic Herbicides

These organic herbicides can be mixed with the said compounds in the listed proportions:

A. 5,6 - dihydro - (4A,6A) - dipyrido - (1,2-A,2',1'-C) pyrazinium dibromide
Mixed in a 1:20 to 16:1 ratio, preferably a 1:5 to 5:1 ratio.

B. 3-amino-1,2,4-triazole
Mixed in a 1:20 to 20:1 ratio, preferably a 1:5 to 5:1 ratio.

C. 3,6-endoxohexahydrophthalic acid
Mixed in a 1:3 to 20:1 ratio, preferably a 1:2 to 10:1 ratio.

D. Hexachloroacetone
Mixed in a 1:2 to 16:1 ratio, preferably a 1:1 to 8:1 ratio.

E. Diphenylacetonitrile
N,N-dimethyl-α,α-diphenylacetamide
N,N - di - *n* - propyl - 2,6 - dinitro - 4 - trifluoromethylaniline
N,N - di - *n* - propyl - 2,6 - dinitro - 4 - methylaniline
Mixed in a 1:10 to 30:1 ratio, preferably a 1:5 to 20:1 ratio.

F. O - (2,4 - dichlorophenyl) - O - methyl - isopropylphosphoramidothioate
2,3,5,6-tetrachloroterephthalic acid, dimethyl ester
Mixed in a 1:4 to 20:1 ratio, preferably a 1:3 to 15:1 ratio.

G. 2,4-dichloro-4'-nitrodiphenyl ether
Mixed in a 1:10 to 30:1 ratio, preferably a 1:5 to 20:1 ratio.

In a further aspect the invention provides a method of controlling the growth of weeds at a locus which comprises applying to the locus a herbicidally effective quantity of at least one of the said compounds. The amount of active compound used will depend on the weeds to be controlled, the formulation used, the mode of application, the climate, season of the year, rainfall and other variables, and because of this it is not possible to state generally a particular rate of application which will be suitable for all situations.

Some of the said compounds are especially useful in controlling the growth of weeds on uncultivated areas. Thus in suitable circumstances some of the said compounds can be used as soil sterilants and/or in soil foliage applications.

It has been found that strikingly similar herbicidal activity can be obtained with seemingly unrelated substituents round the uracil ring. It has also been found that changes in useful selective phytotoxicity (applicable in certain weed-crop combinations) may result when a relatively minor structural modification of R_1 (for example a normal alkyl chain altered to a branched chain) has been made. The 5-substituents (described for R_2 above) for example, have been found to have a minimum range of herbicidal effectiveness and/or species selectivity dependent on the nature of the substituent (if any) in the 3- or 6-position. There has been found little continuity of herbicidal effect which could be ascribed to a particular substituent independently of the other two substituents in the 3,5,6-positions.

Some of the said compounds are particularly useful ~~aagainst~~ against both broadleaf and grass weeds. Others are particularly effective against

hard-to-kill nutsedge and perennial grasses, such as quack grass, Johnson grass, and Bermuda grass. Others are particularly effective on highly adsorptive substrates such as railroad ballast, heavy clay soil, and soils high in organic matter. For example, a pre-emergence treatment has been used to control such germinating broadleaf weeds as pigweed, lamb's quarter, mustard, chickweed and ragweed, and such grass weeds as crab grass and foxtails. Some of the said compounds are particularly effective in soil-foliage applications for the control of such broadleaves as pigweed, lamb's quarter, mustard, chickweed and ragweed and such grass weeds as crab grass, foxtails, quack grass and seedling Johnson grass.

Some of the said compounds are particularly useful as soil sterilants and for general weed control, e.g. on industrial areas, railroad rights-of-way, and areas adjacent to croplands in agricultural areas.

Certain of the said compounds also exhibit selective herbicidal action in crops. By properly selecting a said compound and a rate and time of application, annual grass and broadleaf seedlings in such crops as asparagus, corn, flax, sugar cane, safflower, peanuts, citrus, alfalfa, strawberries, gladiolus, stone fruits, sorghum, sugar beets, red beets, mangels, spinach, potatoes, pineapple, cotton and cucurbits can be controlled.

By proper selection of rate and time of application, certain of the said compounds can also be used to control weeds growing in dormant crops.

This selective activity and activity on weeds growing in dormant crops is described in more detail in the Examples.

Many of the compounds have unusually high oil solubility, and so are useful for application in oils such as herbicidal oils, diesel oil, kerosene, xylene, and other commercially available spray oils. High oil solubility makes shipment of many uracil/oil "concentrates" practical. Such concentrates can be diluted with low-cost diesel oil and herbicidal oils at the site of application.

Broadly speaking, the compounds are used at levels of about $\frac{1}{4}$ pound per acre to about 80 pounds per acre for general weed control. For selective weed control in crops, rates of $\frac{1}{4}$ or $\frac{1}{2}$ to 3 or 8 pounds per acre will generally be used, and in pre-emergence treatments from $\frac{1}{4}$ to 5, preferably $\frac{1}{2}$ to 2 or 4 pounds per acre, may for example be used. More of the active material can be used to control difficult-to-kill species growing under adverse conditions. Economic factors, such as inaccessibility of the area to be treated, e.g. fire breaks in forests, may also favour higher rates, with less frequent treatments.

The invention is illustrated by the following Examples.

EXAMPLE 1

Preparation of 3-norbornylmethyl-5-chloro-6-methyluracil

B 8.0 parts of chlorine are added to a stirred solution of 23.4 parts of 3-norbornylmethyl-6-methyluracil in 100 parts of glacial acetic acid. The temperature is maintained below 30°C. during this addition. The solution is stirred 1/2 hour longer at room temperature, then poured into ice water, whereupon a solid separates. This solid is collected by filtration and washed well with water to give 21 parts of crude 3-norbornylmethyl-5-chloro-6-methyluracil.

C The following compounds of this invention may be prepared as in Example 1, by substituting equivalent amounts of the substituted uracils and halogen set forth in the following table for the uracils and halogens in Example 1.

	Uracil Starting Reactant	Parts by Weight	Halogen	Parts by Weight	Uracil Product
D	3-norbornylmethyl-6-methyluracil	23.4	Br	17.0	3-norbornylmethyl-5-bromo-6-methyluracil
D	3-cyclopropylmethyl-6-methyluracil	18.0	Br	17.0	3-cyclopropylmethyl-5-bromo-6-methyluracil
E	3-(1,4,5,6-tetrachloro-7,7-dimethoxybicyclo [2,2,1]hept-5-en-2-ylmethyl) uracil	30.5	Cl	8.0	3-(1,4,5,6-tetrachloro-7,7-dimethoxybicyclo-[2,2,1]hept-5-en-2-ylmethyl)-5-chlorouracil

EXAMPLE 2

Preparation of 3-(3-cyclohexen-1-yl methyl)-5-chloro-6-methyluracil

A mixture of 218 parts of 3-(3-cyclohexen-1-yl methyl)-6-methyluracil, 500 parts of glacial acetic acid, and 82 parts of sodium acetate was stirred at 20—25°C. while 150 parts of sulfuryl chloride were added over a one-hour period. Stirring was continued for one hour longer, after which the reaction mixture was evaporated and the resulting solid recrystallized from an ethanol-water mixture to give 3-(3-cyclohexen-1-yl methyl)-5-chloro-6-methyluracil.

The following 5-chloro uracils are prepared in a similar fashion by substituting the designated amounts of the listed uracil starting reactants for the 3-(3-cyclohexen-1-yl methyl)-6-methyluracil:

Uracil Starting Reactant	Parts by Weight	Uracil Product
3-(cyclohexylmethyl)-6-methyluracil	222	3-(cyclohexylmethyl)-5-chloro-6-methyluracil
3-(4-methoxy-3-cyclohexene-1-methyl)-6-methyluracil	251	3-(4-methoxy-3-cyclohexene-1-methyl)-5-chloro-6-methyluracil

EXAMPLE 3

Preparation of 3-(α-methyl-1-cyclopentyl-1-ethyl)-5-bromo-6-methyluracil

A solution of 234 parts of 3-(α-methyl-1-cyclopentyl-1-ethyl)-6-methyluracil in 700 parts of acetic acid containing 82 parts of sodium acetate was treated with 160 parts of bromine. After standing overnight, the mixture, which contained some solid, was evaporated to a solid under reduced pressure. The solid was recrystallized from an ethanol-water mixture to give 3-(α-methyl-1-cyclopentyl-1-ethyl)-5-bromo-6-methyluracil.

The following 5-bromo uracils are similarly prepared by substituting the designated amounts of the listed uracil starting reactants for the 3-(α-methyl-1-cyclopentyl-1-ethyl)-6-methyluracil:

Uracil Starting Reactant	Parts by Weight	Uracil Product
3-(bicyclo [2,2,2]oct-2-yl methyl)-6-methyluracil	262	3-(bicyclo[2.2.2]oct-2-yl methyl)-5-bromo-6-methyluracil
3-(norbornenylmethyl)-6-methyluracil	232	3-(norbornenylmethyl)-5-bromo-6-methyluracil

EXAMPLE 4
Preparation of 3-[2-(5-indanyl)ethyl]-5-fluoro-6-methyluracil

B A mixture of 38.0 parts of 2-(5-indanyl)ethylurea, 29.4 parts of ethyl 2-fluoro-3-ketobutyrate, 0.5 part of 85% orthophosphoric acid, and 200 parts of benzene was heated at reflux. Water was collected in a Dean Stark trap. After 24 hours, 2.5 parts of water had been collected and there appeared to be no more separating. The solution was decanted and evaporated to a mixture of viscous oil and solid, which was taken up in 200 parts of ethanol containing 10.5 parts of sodium methoxide and heated at reflux for 10 minutes. Most of the ethanol was removed under reduced pressure and the residue was taken up in just sufficient water to dissolve it. Concentrated hydrochloric acid was added to lower the pH to 4 and the resulting white solid was collected by filtration. This was recrystallized from an ethanol-water mixture to give pure 3 - [2 - (5 - indanyl)ethyl] - 5 - fluoro - 6 - methyluracil.

C The following uracils are prepared as in Example 4 by substituting equivalent amounts of the listed substituted urea reactants for the 2-(5-indanyl)ethylurea and using ethylacetoacetate in place of the ethyl 2-fluoro-3-ketobutylate:

	Urea Reactant	Uracil Products
	norbornylmethylurea	3-norbornylmethyl-6-methyluracil
	norbornenylmethylurea	3-norbornenylmethyl-6-methyluracil
	cyclohexylmethylurea	3-cyclohexylmethyl-6-methyluracil
	3-cyclohexene-1-methylurea	3-(3-cyclohexen-1-methyl)-6-methyluracil
	4-methoxy-3-cyclohexen-1-methylurea	3-(4-methoxy-3-cyclohexen-1-methyl)-6-methyluracil
	α-methylcyclopenten-1-yl-1-ethylurea	3-(α-methyl-1-cyclopenten-1-yl-1-ethyl)-6-methyluracil
D	bicyclo[2,2,2]oct-2-yl-methylurea	3-(bicyclo[2.2.2]oct-2-ylmethyl)-6-methyluracil
	2-(3a,4,5,6,7,7a-hexahydro-5-indanyl)ethylurea	3-[2-(3a,4,5,6,7,7a-hexahydro-5-indanyl)ethyl]-6-methyluracil
	hexahydro-5-indanylmethylurea	3-(hexahydro-5-indanylmethyl)-6-methyluracil
	cyclohexenylethylurea	3-cyclohexenylethyl-6-methyluracil
	(5,6,7,8,9,9-hexachloro-1,2,3,4,4a,5,8,8a-octahydro-5,8-methano-2-naphthylmethyl urea	3-(5,6,7,8,9,9-hexachloro-1,2,3,4,4a,5,8,8a-octahydro-5,8-methano-2-naphthylmethyl)-6-methyluracil
	(cyclopropylmethyl)urea	3-(cyclopropylmethyl)-6-methyluracil
	cyclohexylethylurea	3-cyclohexylethyl-6-methyluracil
E	[cyclobutylmethylurea	3-cyclobutylmethyl-6-methyluracil
D	[α-ethylbicyclo[2,2,2]oct-5-en-yl methyl urea	3-(α-ethylbicyclo[2,2,2]oct-5-en-yl-methyl)-6-methyluracil

EXAMPLE 5

Preparation of 3 - (1,4,5,6 - tertachloro - 7,7 - dimethoxy bicyclo[2,2,1]hept - 5 - en - 2 - yl - methyl) - 5 - hydroxymethyl - 6 - methyluracil

A mixture of 208 parts by weight of 3 - (1,4,5,6 - tetrachloro - 7,7 - dimethoxy bicyclo[2,2,1]hept - 5 - en - 2 - yl - methyl) - 6 - methyluracil, 1400 parts by weight of water, 315 parts by weight of ethyl alcohol, 66 parts by weight of paraformaldehyde, and 20 parts by weight of barium hydroxide is heated until the components are completely dissolved. The solution is then cooled and the oil which precipitates is extracted with ether. This ether extract is dried ~~with~~ with magnesium sulfate, filtered, and concentrated at reduced pressure to give 3 - (1,4,5,6 - tetrachloro - 7,7 - dimethoxy bicyclo[2,2,1]hept - 5 - en - 2 - yl - methyl) - 5 - hydroxymethyl - 6 - methyluracil.

The following compounds are prepared as in Example 5 by substituting equivalent amounts of the designated uracil starting reagents set forth below for the 3 - (1,4,5,6 - tetrachloro - 7, 7- dimethoxy bicyclo[2,2,1]-hept - 5 - en - 2 - yl - methyl) - 6 - methyluracil in Example 5.

Uracil Starting Reactant	5-Hydroxymethyluracil Product
3-(α-ethylbicyclo[2,2,2]oct-5-en-2-yl-methyl)-6-methyluracil	3-(α-ethylbicyclo[2,2,2]oct-5-en-2-yl-methyl)-5-hydroxymethyl-6-methyluracil
3-(hexahydro-5-indenyl-methyl)-6-methyluracil	3-(hexahydro-5-indenylmethyl)-5-hydroxymethyl-6-methyluracil

EXAMPLE 6

Preparation of 3-cyclopropylmethyl-5-bromo-6-methyluracil, sodium salt

A solution of 4 parts of sodium hydroxide in 100 parts of water is treated with 25 parts of 3 - cyclopropylmethyl - 5 - bromo - 6 - methyluracil. Stirring and warming is employed to effect solution. The water is removed from the solution under reduced pressure, leaving 3 - cyclopropylmethyl - 5 - bromo - 6 - methyluracil, sodium salt as a white solid.

EXAMPLE 7

Preparation of 3-cyclopropylmethyl-5,6-pentamethyleneuracil

A mixture of 115 parts by weight of cyclopropylmethylurea, 202 parts by weight of ethyl 2-cycloheptanone-1-carboxylate, 40 parts by weight of phosphoric acid, 1000 parts by weight of dioxane, and 879 parts by weight of benzene was stirred at reflux for 4 hours. During this time, the water given off by the reaction was trapped out. The solvent was stripped at reduced pressure and a portion of the resulting solid was recrystallized from cyclohexane to give ethyl 2-(3-cyclopropylureido)-1-cycloheptene-1-carboxylate.

The remaining unrecrystallized solid was dissolved in 2360 parts by weight of absolute alcohol containing 248 parts by weight of sodium methoxide, and was refluxed for 10 minutes. The solvent was stripped at reduced pressure and the residue dissolved in water, cooled, and acidified. The solid was filtered off, dried, and recrystallized from ethanol to give the desired 3-cyclopropylmethyl-5,6-pentamethyleneuracil.

EXAMPLE 8

Preparation of 3-(4-methoxycyclohexylmethyl)-5-methyluracil

A mixture of 70.8 parts of 1-(4-methoxycyclohexylmethyl) urea, 73 parts of diethyl 2-methyloxalacetate, and 4 parts of 85% orthophosphoric acid in 400 parts of benzene is heated at reflux for 8 hours, during which time 7.8 parts of water are collected. The solvent is removed and the residual oil taken up in 400 parts of water containing 24 parts of sodium hydroxide. The solution is heated on a steam bath with agitation for 2 hours. Acidification with concentrated hydrochloric acid causes precipitation of 6-carboxy-3-(4-methoxycyclohexyl)-5-methyluracil. This is collected by filtration, air-dried, and taken up as a suspension in 100 parts of a eutectic mixture of diphenyl ether and diphenyl. This mixture is stirred and heated at reflux, whereupon CO_2 is evolved. Upon completion of the CO_2 evolution, the mixture is cooled, filtered, and the solvent washed out with benzene. Crystalline 3-(4-methoxycyclohexylmethyl)-5-methyluracil remains.

The following compounds are prepared as in Example 8 by substituting equivalent amounts of mono substituted ureas and thioureas and equivalent amounts of oxalacetates set forth in the following table for the 1-(4-methoxycyclohexylmethyl) urea and diethyl 2-methyloxalacetate in Example 8.

Oxalacetate	Parts by Weight	Urea	Parts by Weight	Substituted Uracil Product
diethyl 2-methyloxalacetate	73	1-(decahydro-1,4-5,8-dimethanonaphth-2-yl methyl)urea	88.4	3-(decahydro-1,4-5,8-dimethano-naphth-2-ylmethyl)-5-methyluracil
diethyl 2-methyloxalacetate	73	1-(norbornylmethyl) urea	67.2	3-(norbornylmethyl)-5-methyluracil
diethyl 2-methyloxalacetate	73	cyclopropylmethylurea	42	3-cyclopropylmethyl-5-methyluracil
diethyl 2-~~methoxalacetate~~ methyloxalacetate	73	cyclohexylmethylurea	58.8	3-cyclohexylmethyl-5-methyluracil
diethyl 2-methyloxalacetate	73	1-(4-methylcyclohexyl-methyl)urea	64.4	3-(4-methylcyclohexylmethyl)-5-methyluracil
diethyl 2-methyloxalacetate	73	(hexahydroindan-5-ylmethyl)urea	78.5	3-(hexahydroindan-5-ylmethyl)-5-methyluracil
diethyl 2-methyloxalacetate	73	4-bromocyclohexyl-methyl-urea	94.0	3-(4-bromocyclohexylmethyl)-5-methyluracil
diethyloxalacetate	75.2	3-(3-cyclohexen-1-ylmethyl)urea	61.6	3-(3-cyclohexen-1-ylmethyl)uracil
diethyloxalacetate	75.2	3-(4-methoxy-3-cyclo-hexene-1-methyl)urea	73.6	3-(4-methoxy-3-cyclohexene-1-methyl)uracil

EXAMPLE 9

Preparation of 3-norbornenylmethyl-5-cyanouracil

A mixture of 166 parts of norbornenylmethylurea, 66 parts of malononitrile and 444 parts of triethyl orthoformate is heated at 70—100°C. for 3 hours with stirring. The volatile constituents are distilled off at reduced pressure at steam-bath temperature The resulting solid, [(3-norbornenylmethylureido)-methylene]malononitrile is purified by recrystallizing from a mixture of alcohol and water.

242 parts of [(3-norbornenylmethylureido)-methylene]malononitrile, 54 parts of sodium methoxide and 1000 parts of methanol are stirred, protected from atmospheric moisture, and allowed to stand for 5 days at room temperature. The alcohol is removed by distillation at reduced pressure, and the resulting solid is dissolved in 2000 parts of cold water. The solution is made slightly acidic by the gradual addition of acetic acid, and the 3-norbornenylmethyl - 5 - cyanocytosine which separates is filtered off, washed with 1000 parts of cold water and dried.

A solution of 242 parts of 3-norbornenylmethyl-5-cyanocytosine in 1000 parts of 3N hydrochloric acid is refluxed for 3 hours and then cooled. The 3-norbornenylmethyl-5-cyanouracil which separates is filtered off and recrystallized from a mixture of alcohol and water.

EXAMPLE 10

Preparation of 3-norbornenylmethyl-5-methoxymethyluracil

Five hundred parts of methanol, ten parts of chloroacetic acid, and 262 parts of 3-norbornenylmethyl-5-hydroxymethyluracil are charged into a bomb and heated at 125°C. for 5 hours. The reaction mixture is then concentrated to an oil, which is taken up in 400 parts of acetonitrile, refluxed for a short time with 5 parts of decolorizing carbon, filtered hot and distilled under reduced pressure to yield solvent-free 3-norbornenylmethyl-5-methoxymethyluracil.

EXAMPLE 11

3 - (4 - chlorocyclohexylmethyl) - 5,6 - dichlorouracil	25%
Alkyl naphthalene sulfonic acid, sodium salt	1%
Lignin sulfonic acid, sodium salt	1%
Attapulgite clay	73%

These ingredients are blended and then charged to a grinding mill where they are moist ground and granulated. The resulting granules are then dried and screened.

The following uracil can be formulated in the same fashion:

3 - (decahydro - 1,4 - 5,8 - dimethanonaphthylmethyl) - 5 - hydroxymethyl - 6 - methyluracil.

These compositions control cheat and wild oats growing along roadsides when applied

EXAMPLE 24

Pellets

3 - (decahydro - 1,4 - 5,8 - dimethanonaphth - 2 - ylmethyl) - 5,6 - trimethyleneuracil	25%
Anhydrous sodium sulfate	10%
Sodium lignin sulfonate	10%
Ca, Mg bentonite	55%

The components are blended and micropulverized, then moistened with 18—20% water and extruded through small die holes. The extrusions are cut as formed to give small pellets which are then dried.

These pellets control both annual and perennial weeds along highway guard rails, around bridges, cyclone fences, and highway signs when applied by hand at levels of 15 to 30 pounds of active ingredient per acre.

EXAMPLE 25

Oil Suspension

The following ingredients are used to prepare an oil suspension:

3 - (4 - isopropylcyclohexylmethyl) - 5 - bromouracil	15%
Diesel oil	80%
Polyoxyethylene sorbitan esters of mixed rosin and fatty acids	5%

The ingredients are ground together in a ball or roller mill until the solids are finely dispersed in the oil and the average particle size of the active ingredient is less than 5 microns. This oil suspension is diluted with water to form a water emulsion for application to plants.

3 - (4 - methylcyclohexylmethyl) - 5 - methyluracil is prepared as an oil suspension in a similar manner.

The emulsifiable oils of this Example are useful for weed control in railroad yards and cattle yards. When either of these compositions are diluted with water at a rate of 160 gallons per acre and sprayed from a raidroad spray car at a level of about 25 pounds of active material per acre, vegetation such as quack grass, crab grass, seedling Johnson grass, seedling Bermuda grass, brome grass, ragweed, cockleburr, lamb's quarter, and mares-tail is controlled for an extended period.

These same oil suspension compositions, when applied to a drainage ditch infested with mixed annual and perennial broadleaf and grass weeds at concentrations of 25 pounds of active material per acre in 150 gallons of water, also give excellent control of vegetation. The ditch remains bare for an extended period.

EXAMPLE 26

Pellets

3 - (1,4,5,6 - tetrachloro - 7,7 - dimethoxybicyclo[2,2,1]hept-5-ene - 2 - ylmethyl) - 5 - chlorouracil	25.0%
Anhydrous sodium sulfate	10.0%
Alkyl naphthalene sulfonic acid, Na salt	1.0%
Non-swelling Ca, Mg bentonite	64.0%

These components are prepared as pellets by first blending and grinding them in a micropulverizer, moistening them with 18—22% water and then extruding them through a die having $\frac{1}{8}$-inch holes. The extrusions are cut at the die face into $\frac{1}{8}$-inch lengths and dried.

Another compound suitable for formulation in a similar manner is

3 - (decahydro - 1,4 - 5,8 - dimethanonaphth - 2 - ylmethyl) - 5 - methyluracil

Either of the pellet compositions of this Example can be applied by hand at a level of 25 pounds of active ingredient per acre for control of annual and perennial weeds along guard rails, safety fences, and highway markers and dividers.

The same compositions, when applied by airplane at concentrations of 12—18 pounds of active material per acre, give effective control of willow and oak.

EXAMPLE 27

Pellets

3 - (5,6,7,8,9,9 - hexachloro - 1,2,3,-4,4a,5,6,7,8,8a - decahydro - 5,8 - methanonaphth - 2 - ylmethyl) - 5 - hydroxymethyluracil	25%
Methyl cellulose, low viscosity	3%
Kaolin clay	72%

These components are blended and micropulverized, then pug-milled with 15% to 20% water, and extruded through $\frac{1}{8}$-inch die holes. The extrusions are cut into $\frac{1}{8}$-inch segments, then dried.

The following uracils can also be formulated in this way.

3 - (4 - methoxycyclohexylmethyl) - 5 - methoxyuracil

3 - [2 - (4 - bromocyclohexyl)ethyl] - 5 - nitrouracil

These formulations, applied in 60 gallons of water at the rate of 25 pounds (active) per acre, control crab grass, barnyard grass, foxtail, pigweed, and lamb's quarters growing along boardwalks.

EXAMPLE 28

Wettable Powder

3 - norbornenylmethyl - 5 - hydroxymethyluracil	50.0%
Alkyl naphthalene sulfonic acid, sodium salt	1.0%
Sodium salt of polymerized alkyl naphthalene sulfonic acid	0.5%
Calcined, non-swelling Montmorillonoid type clay (Pikes Peak clay)	48.5%

This wettable powder is prepared by blending the components in a ribbon blender, followed by micropulverizing in a hammer mill. The ground product is air-reduced until the particles are below 10 microns in size and is then reblended until homogeneous.

EXAMPLE 29

Wettable Powder

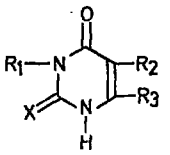
3-cyclopropylmethyl-5-methyl uracil	40.00%
Dioctyl ester of sodium sulphosuccinic acid	0.40%
Low viscosity methyl cellulose	0.30%
Aliphatic substituted butyndiols and octyndiols	0.40%
Kaolin clay	58.90%

These ingredients are blended, ground in a hammer mill until the particle diameters are 50 microns or less, and then reblended.

This formulation, applied as a foliar spray at 25 lb. (active) per acre in 60 gallons of water to emerging weeds, gives excellent control of crab grass, foxtail, chickweed and volunteer small grains growing around loading platforms.

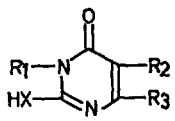
WHAT WE CLAIM IS:—

1. A herbicidal composition comprising a diluent which is at least in part an oil and in association therewith at least one 3-substituted uracil which is a compound of the general formula:



I

or the corresponding isomeric formula:



II

wherein R_1 represents a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms, a (substituted cycloalkyl)alkyl radical containing 5 to 14 carbon atoms or a (substituted cycloalkenyl)alkyl radical containing 5 to 14 carbon atoms; R_2 represents a hydrogen, chlorine, fluorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 6 carbon atoms, a hydroxy alkyl radical containing 1 to 6 carbon atoms, an alkenyl radical containing 3 to 6 carbon atoms, a thiocyanato radical, a cyano radical, a mercaptomethyl radical, a methylthiomethyl radical, a phenylthiomethyl radical, an alkylthio radical containing 1 to 4 carbon atoms, a bromomethyl radical, a fluoromethyl radical, a chloromethyl radical or a carboxymethylthiomethyl radical; R_3 represents a hydrogen, chlorine or bromine atom, an alkyl radical containing 1 to 5 carbon atoms, a chloroalkyl radical containing 1 to 4 carbon atoms, a bromoalkyl radical containing 1 to 4 carbon atoms, or an alkoxy radical containing 1 to 5 carbon atoms; or R_2 and R_3 together represent a divalent alkylene radical of formula $(CH_2)_n$ where n is 3, 4 or 5; with the proviso that if R_2 represents a hydrogen atom, R_3 represents a hydrogen atom or an alkyl radical containing 1 to 5 carbon atoms, and X represents an oxygen or sulphur atom or a metal salt thereof.

2. A herbicidal composition comprising at least one 3-substituted uracil as defined in claim 1 associated with a diluent which is at least in part a solid.

3. A herbicidal composition according to claim 1 which contains a surface-active agent.

4. A herbicidal composition according to claim 2 which contains a surface-active agent.

5. A herbicidal composition comprising at least one 3-substituted uracil as defined in claim 1 associated with a surface-active agent and a diluent which is a liquid other than an oil.

6. A herbicidal composition according to claim 5 wherein the liquid is water.

7. A herbicidal composition comprising at least one 3-substituted uracil as defined in claim 1 associated with a surface-active agent.

8. A herbicidal composition according to any one of claims 3 to 7 which contains 1 to 10% by weight of the surface-active agent.

9. A herbicidal composition according to any one of the preceding claims which contains, in addition to the 3-substituted uracil, another compound having herbicidal properties.

10. A herbicidal composition comprising at least one 3-substituted uracil as defined in claim 1 in association with a diluent and another compound having herbicidal properties.

11. A herbicidal composition according to any one of the preceding claims wherein X represents an oxygen atom.

12. A herbicidal composition according to any one of the preceding claims wherein if R_1 represents a (substituted cycloalkyl) alkyl or (substituted cycloalkenyl)alkyl radical, the or each substituent is an alkyl radical containing 1 to 4 carbon atoms, a methoxy radical or a chlorine or bromine atom.

13. A herbicidal composition according to any one of the preceding claims wherein the cycloalkyl or cycloalkenyl radical in R_1 is a cyclohexyl or cyclohexenyl radical.

14. A herbicidal composition according to any one of claims 1 to 12 wherein the cycloalkyl or cycloalkenyl radical in R_1 is a cyclopentyl, cyclopentenyl, norbornyl, norbornenyl, bicyclo (2,2,2)-octyl, bicyclo (2,2,2) octenyl, cyclobutyl, cyclobutenyl, hexahydroindenyl, hexahydroindanyl, tetrahydroindanyl, hexahydro - 4,7 - methanoindenyl, hexahydro - 4,7 - methanoindanyl, tetrahydro - 4,7 - methanoindanyl, decahydronaphthyl, tetrahydronaphthyl, decahydro - 1,4 - methanonaphthyl, octahydro - 1,4 - methanonaphthyl, deca-

pre-emergency at 12—15 pounds (active) per acre.

EXAMPLE 12

5 - bromo - 6 - methyl - 3 - (2 - norbornylmethyl)uracil (exo - endo mixture)	25%
Low viscosity methyl cellulose	3%
Kaolin clay	72%

These ingredients are mixed and micropulverized, then pug-milled with 16—20% water and extruded through a 9/64 die hole to form rods which are cut as formed by a revolving knife into $\frac{1}{8}$ inch lengths. The resulting pellets are then dried.

Sixty pounds per acre of this formulation gives excellent control of ragweed, volunteer ryegrass, wild mustard, plantain, carpet weed, crab grass, foxtail, cocklebur, and purslane growing around signposts and telephone poles.

EXAMPLE 13

3 - (4 - methoxycyclohex - 3 - enemethyl) - 5,6 - dimethyluracil	80%
Dodecylbenzene sulfonic acid, Na salt	2%
Sodium lignin sulfonate	1%
Attapulgite clay	17%

A wettable powder is prepared by blending these components in a ribbon blender, micropulverizing them in a hammer mill until the particles are substantially all below 50 microns in size and then reblending until homogeneous.

3 - [1 - methyl - 2 - (cyclopenten - 1 - yl)-ethyl] - 5 - methoxymethyl - 6 - methyl uracil can be formulated in the same way.

EXAMPLE 14

3 - cyclopropylmethyl - 5 - bromo - 6 - methyluracil	50.0%
Alkyl naphthalene sulfonic acid, sodium salt	1.0%
Sodium salt of polymerized alkyl naphthalene sulfonic acid	0.5%
Calcined, non-swelling Montmorillonoid type clay (Pikes Peak clay)	48.5%

A wettable powder is prepared by blending the above components in a ribbon blender and then micropulverizing them in a hammer mill. The ground product is air-reductionized until the particle size is substantially below 10 microns, and then is reblended until homogeneous.

Another active material which can be formulated into wettable powders in the same fashion is:

3 - cyclobutenylmethyl - 5 - bromo - 6 - methyluracil

EXAMPLE 15

A wettable powder formulation is prepared by blending the following components in a ribbon blender:

3 - cyclohexylmethyl - 5 - chloro - 6 - methyluracil	50%
Kaolin clay	46%
Diocyl ester of sodium sulfosuccinic acid	1%
Sodium lignin sulfonate	3%

The blended mixture is ground in a hammer mill until the particles are substantially less than 50 microns in size. The mixture is then reblended until homogeneous.

EXAMPLE 16

3 - (4 - methylcyclohexyl methyl) - 5 - bromo - 6 - methyluracil	45.00%
Alkyl naphthalene sulfonic acid, sodium salt	1.50%
Low viscosity methyl cellulose	0.25%
Synthetic calcium silicate	53.25%

These ingredients are blended and micropulverized until the particles are 50 microns or less in diameter. The mixture is then reblended.

The following uracil can be substituted, in eqivalent amounts, for the 3-(4-methylcyclohexyl methyl)-5-bromo-6-methyluracil:

3 - (4 - propylcyclohexyl methyl) - 5 - bromo - 6 - methyluracil

EXAMPLE 17

5 - bromo - 6 - methyl - 3 - (2 - norbornylmethyl)uracil	80.0%
Dioctyl sodium sulfosuccinate concreted 85/15 with sodium benzoate	1.0%
Sodium N-methyl-N-palmitoyltaurate	1.0%
Synthetic fine calcium silicate	18.0%

These components are blended, micropulverized until the particles of uracil have been reduced to about 10 microns in diameter, then reblended.

This formulation is applied pre-emergence at a rate of from 2—4 pounds (active) per acre in 40 gallons of water to newly-planted fields of cucumbers, watermelons and cantaloupe.

These treatments give excellent control of crab grass, foxtail, barnyard grass, ryegrass, pigweed, lamb's quarters, flower-of-an-hour and wild mustard.

EXAMPLE 18

Aqueous Concentrate

The following aqueous concentrate is prepared by dissolving or suspending the two solid components in water, with agitation. The product can be readily diluted to use levels and sprayed.

3 - norbornylmethyl - 6 - methyluracil, 1/2 calcium salt	10%
Sodium lauryl sulfate	1%
Water	89%

When applied at 20 pounds of active ingredient per acre, this concentrate is useful for general control of annual and perennial weeds on industrial sites and railroad ballast.

EXAMPLE 19

Wettable Powder

3 - (α - ethylbicyclo[2,2,2]oct - 5 - enylmethyl) - 6 - methyluracil	50.00%
Alkylnaphthalene sulfonic acid, sodium salt	1.75%
Partially desulfonated sodium lignin sulfonate	1.00%

Calcined montmorillonite clay 47.25%

These ingredients are blended, micropulverized until all particles are below 50 microns in diameter, then reblended until homogeneous.

This formulation, at 15 pounds (active) per acre, is applied around farm buildings for excellent control of crab grass, foxtail, chickweed, flower-of-an-hour, catnip.

The following compounds are formulated and used in the manner described above with excellent results:

3 - norbornenylmethyl - 6 - methyl - 2 - thiouracil

3 - cyclopentenylmethyl-6-ethyluracil

3 - (4 - methylcyclohexenylmethyl) - 6 - methyluracil

3 - (4 - methoxycyclohexylmethyl) - 6 - ethyluracil

3 - (4 - bromocyclohexylmethyl) - 6 - methyluracil

3 - (1,4,5,6 - tetrachloro - 7,7 - dimethoxybicyclo[2,2,1]hept - 5 - en - 2 - ylmethyl) - 6 - methyluracil

3 - (hexahydroindan - 5 - ylmethyl) - 6 - methyluracil

3 - [2-(5-indanyl)ethyl]-6-methyluracil

3 - (cyclohexene - 1 - methyl) - 6 - methyl - 2 - thiouracil

3 - (3a,4,5,6,7,7a - hexahydroinden - 5 - ylmethyl) - 6 - methyluracil

EXAMPLE 20

Aqueous concentrate

An aqueous concentrate is prepared by dissolving the two solid components in water. The product can be readily diluted to use levels and sprayed.

3 - cyclobutylmethyl - 5,6 - trimethyleneuracil, sodium salt	5%
Sodium lauryl sulfate	1%
Water	94%

This aqueous concentrate is applied with a pressure-type hand sprayer. Three pounds of active ingredient per acre in 40 gallons of water gives excellent pre-emergence control of foxtail, watergrass, Johnson grass seedlings in sugar cane.

This formulation, applied at the rate of 20 pounds (active) in 100 gallons of water to weeds growing around bridge abutments, gives excellent control of wild barley, bedstraw, hedge bindweed, wild carrot, cheat grass, cockleburr, dog fennel and goose grass.

EXAMPLE 21

Oil Suspension

3 - norbornylmethyl - 5,6 - trimethylene - 2 - thiouracil	22%
Blend of polyalcohol carboxylic esters and oil soluble petroleum sulfonates	7%
Xylene	71%

These components are blended, micropulverized, and then sand-ground until the particles have a diameter of 10 microns or less.

This formulation, diluted with Lion Herbicidal Oil No. 6 to a final concentration of 15—25 pounds of active ingredient per 100 gallons of oil, gives excellent control of crab grass, cheatgrass, volunteer wheat and oats growing on railroad sidings.

EXAMPLE 22

Wettable Powder

The following are blended, then micropulverized until the particles are below 50 microns in diameter, and reblended:

3 - (cyclopentenylmethyl) - 5,6 - trimethyleneuracil	25%
Alkylnaphthalene sulfonic acid, sodium salt	1%
Partially desulfonated sodium lignin sulfonate	1%
Attapulgite clay	73%

This wettable powder is suspended in water at a concentration of 15—20 pounds of active ingredient per 100 gallons of water. This suspension is sprayed at the rate of 100 gallons per acre for control of crab grass, water grass and lamb's quarters growing around telephone poles.

Each of the following uracils can be similarly formulated and applied in 60 gallons of herbicidal oil. At rates of 20—30 pounds (active) per acre, they will give good weed control.

3 - (2 - methylcyclohexylmethyl) - 5,6 - trimethyleneuracil

3 - (4 - methoxycyclohexylmethyl) - 5,6 - tetramethyleneuracil

3 - (4 - methylcyclohexenylmethyl) - 5,6 - trimethyleneuracil

EXAMPLE 23

Wettable Powder

3 - (norbornenylmethyl) - 5,6 - trimethyleneuracil	75.0%
Dioctylsodium sulfosuccinate	0.5%
Ditertiary acetylenic glycol	0.5%
Coconut acid ester of sodium isethionate	0.5%
Precipitated sodium silica aluminate	23.5%

These components are mixed and micropulverized until the particles are less than 50 microns in diameter.

An application of this formulation at 25 pounds (active) in 80 gallons water controls beggar tick, wild mustard, lamb's quarters, plantain, velvetleaf, annual bluegrass, Canadian bluegrass, crab grass, foxtail, and seedling Johnson grass growing around radar installations.

The following compounds can be formullated in a similar fashion. When used in the same way, at equivalent rates, they give good weed control.

3 - (1,4,5,6 - tetrachloro - 7,7 - dimethoxybicyclo[2,2,1]hept - 5 - en - 2 - ylmethyl) - 5,6 - trimethyleneuracil

3 - (α - ethylbicyclo[2,2,2]oct - 5 - en - 2 - ylmethyl) - 5,6 - trimethyleneuracil

EXAMPLE 29

Wettable Powder

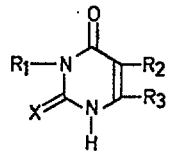
3-cyclopropylmethyl-5-methyl uracil	40.00%
Dioctyl ester of sodium sulphosuccinic acid	0.40%
Low viscosity methyl cellulose	0.30%
Aliphatic substituted butyndiols and octyndiols	0.40%
Kaolin clay	58.90%

These ingredients are blended, ground in a hammer mill until the particle diameters are 50 microns or less, and then reblended.

This formulation, applied as a foliar spray at 25 lb. (active) per acre in 60 gallons of water to emerging weeds, gives excellent control of crab grass, foxtail, chickweed and volunteer small grains growing around loading platforms.

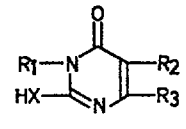
WHAT WE CLAIM IS:—

1. A herbicidal composition comprising a diluent which is at least in part an oil and in association therewith at least one 3-substituted uracil which is a compound of the general formula:



I

or the corresponding isomeric formula:



II

wherein R_1 represents a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms, a (substituted cycloalkyl)alkyl radical containing 5 to 14 carbon atoms or a (substituted cycloalkenyl)alkyl radical containing 5 to 14 carbon atoms; R_2 represents a hydrogen, chlorine, fluorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 6 carbon atoms, a hydroxy alkyl radical containing 1 to 6 carbon atoms, an alkenyl radical containing 3 to 6 carbon atoms, a thiocyanato radical, a cyano radical, a mercaptomethyl radical, a methylthiomethyl radical, a phenylthiomethyl radical, an alkylthio radical containing 1 to 4 carbon atoms, a bromomethyl radical, a fluoromethyl radical, a chloromethyl radical or a carboxymethylthiomethyl radical; R_3 represents a hydrogen, chlorine or bromine atom, an alkyl radical containing 1 to 5 carbon atoms, a chloroalkyl radical containing 1 to 4 carbon atoms, a bromoalkyl radical containing 1 to 4 carbon atoms, or an alkoxy radical containing 1 to 5 carbon atoms; or R_2 and R_3 together represent a divalent alkylene radical of formula $(CH_2)_n$ where n is 3, 4 or 5; with the proviso that if R_2 represents a hydrogen atom, R_3 represents a hydrogen atom or an alkyl radical containing 1 to 5 carbon atoms, and X represents an oxygen or sulphur atom or a metal salt thereof.

2. A herbicidal composition comprising at least one 3-substituted uracil as defined in claim 1 associated with a diluent which is at least in part a solid.

3. A herbicidal composition according to claim 1 which contains a surface-active agent.

4. A herbicidal composition according to claim 2 which contains a surface-active agent.

5. A herbicidal composition comprising at least one 3-substituted uracil as defined in claim 1 associated with a surface-active agent and a diluent which is a liquid other than an oil.

6. A herbicidal composition according to claim 5 wherein the liquid is water.

7. A herbicidal composition comprising at least one 3-substituted uracil as defined in claim 1 associated with a surface-active agent.

8. A herbicidal composition according to any one of claims 3 to 7 which contains 1 to 10% by weight of the surface-active agent.

9. A herbicidal composition according to any one of the preceding claims which contains, in addition to the 3-substituted uracil, another compound having herbicidal properties.

10. A herbicidal composition comprising at least one 3-substituted uracil as defined in claim 1 in association with a diluent and another compound having herbicidal properties.

11. A herbicidal composition according to any one of the preceding claims wherein X represents an oxygen atom.

12. A herbicidal composition according to any one of the preceding claims wherein if R_1 represents a (substituted cycloalkyl) alkyl or (substituted cycloalkenyl)alkyl radical, the or each substituent is an alkyl radical containing 1 to 4 carbon atoms, a methoxy radical or a chlorine or bromine atom.

13. A herbicidal composition according to any one of the preceding claims wherein the cycloalkyl or cycloalkenyl radical in R_1 is a cyclohexyl or cyclohexenyl radical.

14. A herbicidal composition according to any one of claims 1 to 12 wherein the cycloalkyl or cycloalkenyl radical in R_1 is a cyclopentyl, cyclopentenyl, norbornyl, norbornenyl, bicyclo (2,2,2)-octyl, bicyclo (2,2,2) octenyl, cyclobutyl, cyclobutenyl, hexahydroindenyl, hexahydroindanyl, tetrahydroindanyl, hexahydro - 4,7 - methanoindenyl, hexahydro - 4,7 - methanoindanyl, tetrahydro - 4,7 - methanoindanyl, decahydronaphthyl, tetrahydronaphthyl, decahydro - 1,4 - methanonaphthyl, octahydro - 1,4 - methanonaphthyl, deca-

* C L M S *

hydro - 1,4,5,8 - dimethanonaphthyl or octahydro - 1,4,5,8 - dimethanonaphthyl radical.

15. A herbicidal composition according to any one of the preceding claims wherein the 3-substituted uracil, if it is a metal salt, is a sodium, potassium or lithium salt.

16. A herbicidal composition according to any one of the preceding claims which contains one of the known herbicides specifically hereinbefore named.

17. A herbicidal composition according to any one of the preceding claims in the form of a concentrate which requires dilution before use as a herbicide.

18. A herbicidal composition according to any one of claims 1 to 17 wherein

R_1 represents a cycloalkyl alkyl radical containing 4 to 13 carbon atoms or a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms;

R_2 represents a hydrogen atom; and

R_3 represents an alkyl radical containing 1 to 5 carbon atoms.

19. A herbicidal composition according to any one of claims 1 to 17 wherein R_2 and R_3 each represents a hydrogen atom.

20. A herbicidal composition according to any one of claims 1 to 17 wherein

X represents an oxygen atom;

R_1 represents a cycloalkyl alkyl radical containing 4 to 13 carbon atoms or a substituted cycloalkyl alkyl radical containing 5 to 13 carbon atoms; and

R_2 represents chlorine, bromine or iodine atom, a methyl radical, a hydroxymethyl radical, a methoxymethyl radical or a nitro radical; and

R_3 represents a hydrogen atom or a methyl radical.

21. A herbicidal composition according to any one of claims 1 to 17 wherein the 3-substituted uracil is one of the compounds specifically named herein.

22. A herbicidal composition according to any one of claims 1 to 15 wherein

R_2 represents a chlorine, fluorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 6 carbon atoms, a hydroxy alkyl radical containing 1 to 6 carbon atoms, an alkenyl radical containing 3 to 6 carbon atoms, a thiocyanato radical, a cyano radical, a mercaptomethyl radical, a methylthiomethyl radical, phenylthiomethyl radical, a chloromethyl radical or a carboxymethylthiomethyl radical; and

R_3 represents a chlorine or bromine atom, an alkyl radical containing 1 to 5 carbon atoms, a chloroalkyl radical containing 1 to 4 carbon atoms, a bromoalkyl radical containing 1 to 4 carbon atoms or an alkoxy radical containing 1 to 5 carbon atoms.

23. A herbicidal composition according to claim 22 wherein

R_1 represents a cycloalkyl alkyl or (substituted cycloalkyl)alkyl radical;

R_2 represents a chlorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 5 ~~carbonatoms~~ carbon atoms, a hydroxymethyl radical, a hydroxyethyl radical, a cyano radical or a methylthiomethyl radical; and

R_3 represents a chlorine or bromine atom or a methyl, ethyl, propyl, chloromethyl or bromomethyl radical.

24. A herbicidal composition according to claim 22 wherein the 3-substituted uracil is one of the compounds specifically named in Examples 1 parts B and D, 2, 3, 4 part B, 5, 12, 14 and 15.

25. A herbicidal composition according to any one of claims 1 to 15 wherein

R_1 represents a cycloalkyl alkyl radical containing 6 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 6 to 13 carbon atoms, a (substituted cycloalkyl)alkyl radical containing 6 to 14 carbon atoms or a (substituted cycloalkenyl)alkyl radical containing 6 to 14 carbon atoms;

R_2 represents a hydrogen atom; and

R_3 represents an alkyl radical containing 1 to 5 carbon atoms.

26. A herbicidal composition according to claim 25 wherein the 3-substituted uracil is one of the compounds specifically named in Example 4 part D.

27. A herbicidal composition according to any one of claims 1 to 15 wherein R_2 is as defined in claim 23 or represents an alkyl thio radical containing 1 to 4 carbon atoms, a bromomethyl radical or a fluoromethyl radical, and R_3 is other than hydrogen.

28. A herbicidal composition according to claim 27 wherein the 3-substituted uracil is one of the compounds specifically named in Example 6, 11, 13, 16 and 17.

29. A herbicidal composition according to any one of claims 1 to 15 wherein R_3 represents a hydrogen atom and R_2 is other than hydrogen.

30. A herbicidal composition according to any one of claims 1 to 15 wherein R_1 represents a cycloalkyl alkyl or cycloalkenyl alkyl radical and R_2 and R_3 each represents a hydrogen atom.

31. A herbicidal composition according to claim 28 or 29 wherein the 3-substituted uracil is one of the compounds specifically named in Examples 1 part E, 8, 9, 25, 26, 27, 28 and 29.

32. A herbicidal composition according to any one of claims 1 to 15 wherein the 3-substituted uracil is one of the compounds specifically named in Examples 7 and 20 to 24.

33. A herbicidal composition according to any one of claims 1 to 15 wherein R_2 represents a hydrogen atom and R_3 represents an alkyl radical containing 1 to 5 carbon atoms.

34. A herbicidal composition according to claim 33 wherein the 3-substituted uracil is one of the compounds specifically named in Examples 2 part E, 18 and 19.

35. A herbicidal composition according to any one of the preceding claims substantially as hereinbefore described.

36. A herbicidal composition according to any one of claims 1 to 34 substantially as described in any one of the foregoing Examples.

37. A method of controlling the growth of weeds at a locus which comprises applying to the locus a herbicidally effective quantity of at least one 3-substituted uracil as defined in any one of the preceding claims.

38. A method according to claim 37 wherein the 3-substituted uracil is applied in the form of a herbicidal composition as claimed in any one of claims 1 to 36, after dilution where appropriate.

39. A method according to claim 37 or 38 wherein the growth of the weeds specified in any one of Examples 11 to 29 is controlled by the application of the 3-substituted uracil specified in that Example.

40. A method according to claim 37 substantially as hereinbefore described.

41. A method according to claim 37 wherein the 3-substituted uracil is as defined in any one of claims 22 to 24.

42. A method according to claim 41 wherein the 3-substituted uracil is applied in the form of a herbicidal composition as claimed in any one of claims 22 to 24, after dilution where appropriate.

43. A method according to claim 41 or 42 wherein the growth of the weeds specified in any one of Examples 12, 14 and 15 is controlled by the application of the 3-substituted uracil specified in that Example.

44. A method according to claim 37 wherein the 3-substituted uracil is as defined in claim 25 or 26.

45. A method according to claim 44 wherein the 3-substituted uracil is applied in the form of a herbicidal composition as claimed in claim 25 or 26, after dilution where appropriate.

46. A method according to claim 37 wherein the 3-substituted uracil is as defined in claim 27 or 28.

47. A method according to claim 46 wherein the 3-substituted uracil is applied in the form of a herbicidal composition as claimed in claim 27 or 28, after dilution where appropriate.

48. A method according to claim 46 or 47 wherein the growth of the weeds specified in any one of Examples 11, 13, 16 and 17 is controlled by the application of the 3-substituted uracil specified in that Example.

49. A method according to claim 37 wherein the 3-substituted uracil is as defined in any one of claims 29 to 31.

50. A method according to claim 49 wherein the 3-substituted uracil is applied in the form of a herbicidal composition as claimed in any one of claims 30 to 32, after dilution where appropriate.

51. A method according to claim 49 or 50 wherein the growth of the weeds specified in any one of Examples 25 to 29 is controlled by the application of the 3-substituted uracil specified in that Example.

52. A method according to claim 37 wherein the 3-substituted uracil is as defined in claim 32.

53. A method according to claim 52 wherein the 3-substituted uracil is applied in the form of a herbicidal composition as claimed in claim 32, after dilution where appropriate.

54. A method according to claim 52 or 53 wherein the growth of the weeds specified in any one of Examples 20 to 24 is controlled by the application of the 3-substituted uracil specified in that Example.

55. A method according to claim 37 wherein the 3-substituted uracil is as defined in claim 33 or 34.

56. A method according to claim 55 wherein the 3-substituted uracil is applied in the form of a herbicidal composition as claimed in claim 33 or 34, after dilution where appropriate.

57. A method according to claim 55 or 56 wherein the growth of the weeds specified in Examples 18 and 19 is controlled by the application of the 3-substituted uracil specified in that Example.

58. As a new compound a 3-substituted uracil as defined in any one of claims 1 to 34.

59. As a new compound a 3-substituted uracil as defined in claim 18.

60. As a new compound a 3-substituted uracil as defined in claim 20.

61. As a new compound a 3-substituted uracil as defined in any one of claims 22 to 24.

62. As a new compound a 3-substituted uracil as defined in claim 25 or 26.

63. As a new compound a 3-substituted uracil as defined in claim 27 or 28.

64. As a new compound a 3-substituted uracil as defined in any one of claims 29 to 31.

65. As a new compound a 3-substituted uracil as defined in claim 32.

66. As a new compound a 3-substituted uracil as defined in claim 33 or 34.

* C L M S *

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