

PATENT SPECIFICATION

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(54) AROMATIC DIURETHANES AND THEIR USE AS HERBICIDES

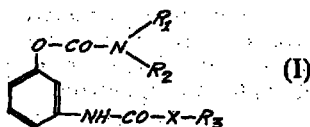
(71) We, SCHERING AKTIENGESELLSCHAFT, a Body Corporate organised according to the laws of Germany, of Berlin and Bergkamen, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention is concerned with new diurethanes having selective herbicidal properties, which herbicides are especially suitable for combating weeds in cotton crops, with processes for their manufacture, with herbicidal preparations containing them and with their use.

The herbicidal action of diurethanes has been known (cf. German Offenlegungsschriften Nos. 1,567,151 and 1,568,621). These Specifications are, however, preponderantly concerned with selective herbicides for use in beet crops. Cotton plants however, do not exhibit a particularly high tolerance to these known compounds.

The problem upon which the present invention is based has been to provide a weed combating agent that can be used especially in cotton crops, which problem has now been solved by the present invention.

The present invention provides compounds of the general formula I



in which

R₁ represents an allyl group, an unsubstituted alkyl group containing 1 to 4 carbon atoms or a halogen-substituted alkyl group containing 1 to 4 carbon atoms,

R₂ represents an allyl group, an alkynyl group, an unsubstituted alkyl group containing 1 to 4 carbon atoms, a halogen-substituted alkyl group containing 1 to 4 carbon atoms, or a cyclohexyl, benzyl or phenylethyl group,

R₃ represents a methyl or ethyl group, and

X represents an oxygen or sulphur atom, and in which, when R₁ and R₂ are the same alkyl groups, R₃ is not an ethyl group.

As alkyl groups containing 1 to 4 carbon atoms which may be halogen-substituted there are included, for example, the groups methyl, ethyl, chloroethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec.-butyl and tert.-butyl. There may be mentioned, more especially, compounds in which R₁ and R₂ each represent a 2-chloroethyl group.

The compounds of the present invention are distinguished by the fact that cotton in all stages of development exhibits a high tolerance to them, the crop plants not being damaged by the use of the compounds even in the embryo stage. The surprisingly good tolerability of crops to the new compounds represents an extraordinary technical advance, because weeds can be combated in these crops

independently of the crops' stage of development at any time when an optimum attack by the weeds present can be expected. This is not possible with the herbicides based on phenyl-ureas hitherto used for combating weeds in cotton, which can be used only in a stage of growth of the cotton at which the weeds have also strongly developed and are therefore difficult to combat.

The compounds of the present invention exhibit the most favourable action when they are applied by the post-emergence method. In this case the compounds act not only against the weeds that have sprouted, but also, owing to their residual action in the soil, attack germinating weed plants, so that a long lasting attack is ensured.

Apart from their use in cotton crops, the compounds of the present invention can also be used for combating weeds in carrot crops, ground-nut crops and rice crops, which also have a surprisingly high tolerance to the compounds.

The herbicidal action of the compounds of the general formula I extends to many kinds of weeds, among which, for example, there may be mentioned the following:

Setaria verticillata, *Amarantus spinosus*, *Datura stramonium*, *Portulaca oleracea*, *Xanthium pensylvanicum*, *Eleusine indica*, *Rottboellia exaltata*, *Sinapis sp.*, *Solanum sp.*, *Stellaria media*, *Senecio vulgaris*, *Lamium amplexicaule*, *Centaurea cyanus*, *Amarantus retroflexus*, *Galium aparine*, *Chrysanthemum segetum*, *Echinochloa crus galli*, *Setaria italica*, *Ipomoea purpurea*, *Polygonum lapathifolium*, *Digitaria sanguinalis* and *Setaria faberi*.

The rates of application for a selective weed control are approximately 1 to 3 kg of active substance per hectare; when two or more compounds of the general formula I are used the range of approximately 1 to 3 kg refers of course to the total amount applied of these compounds. These rates of application may even be increased to 10 kg of active substance per hectare without substantially injuriously affecting the crop plants.

The present invention accordingly also provides a herbicidal preparation which comprises a compound of the general formula I, in admixture or conjunction with a suitable carrier. The preparation may of course contain one or more compounds of the general formula I.

The present invention further provides a method of protecting a living plant against weeds, wherein the area in the vicinity of a living plant is treated with a compound of the general formula I.

The present invention further provides a method of protecting a crop area against weeds, wherein a crop area is treated with a compound of the general formula I.

The compounds of the present invention can be used either singly as the sole active substance, or in admixture with one another or in admixture with other active substances. If desired, there may be added other plant protecting agents or pest combating agents, for example fungicides, nematocides or other agents, depending on the purpose desired. Fertilizers may also be added.

When it is intended to widen the spectrum of action there may also be added other herbicides, but, of course, the selectivity is not always maintained. As herbicidally active co-components of such mixtures there are suitable, for example, active substances belonging to the groups of carbamic acid esters and thiocarbamic acid esters, substituted anilines and anilides, triazines, amino-triazoles, diazines, for example uracils, for example 3-cyclohexyl-5,6-trimethylene-uracil, 1-phenyl-4-amino-5-chloropyridazine-(6), aliphatic carboxylic acids and halogen-carboxylic acids, halogenated benzoic acids and phenyl-acetic acids, aryloxy-carboxylic acids, hydrazides, amides, nitriles, halogen-carboxylic acids, for example 2,2-dichloropropionic acid or salts thereof and tetrafluoropropionic acid or salts thereof, esters of such carboxylic acids, ureas, N-phenyl-ureas, 2,3,6-trichlorobenzoyloxy-propanol, agents containing thiocyanogen and other compounds.

Depending on the purpose of use other substances may be added, among which there are also to be understood, for example, non-phytotoxic additions, which in combination with herbicides may give a synergistic increase in action, for example wetting agents, emulsifiers, solvents and oily additions.

The active compounds of the present invention are advantageously used in the form of herbicidal preparations, for example powders, spraying or dusting agents, granules, solutions, emulsions or suspensions, with the addition of liquid and/or solid vehicles or diluents, and, if desired, of surface-active agents, for example wetting, adherent, emulsifying and/or dispersing agents.

Suitable liquid carriers are, for example, water, aliphatic and aromatic hydrocarbons, for example benzene, toluene and xylene, cyclohexanone and isophorone, and also mineral oil fractions.

As solid carriers there are suitable mineral earths, for example tonsil, silica gel, talcum, kaolin, attaclay, limestone and silicic acid, and vegetable products, for example meals.

As surface-active agents there may be mentioned, for example, calcium lignin sulphonate, polyoxyethylene-octyl-phenol ethers, naphthalene sulphonic acids, phenol sulphonic acids, formaldehyde condensates, fatty alcohol sulphates and alkali metal and alkaline earth metal salts of fatty acids.

It has been surprisingly found that the herbicidal action and the selectivity of the preparations can be increased, when they contain the surface-active agents in proportions that exceed the usual quantities.

The total amount of the active compound or compounds of the present invention in the various herbicidal preparations may vary within wide limits. The preparations may contain, for example, approximately 20 to 80 per cent by weight of active compound(s), approximately 80 to 20 per cent by weight of liquid or solid carrier, and also, if desired, up to 30 per cent by weight of surface-active agent(s).

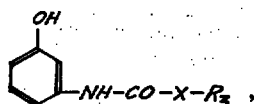
The active compounds may be applied in the usual manner, for example, with water as carrier in quantities of spray liquor from 100 to 1,000 litres per hectare. For the total destruction of weeds it may be necessary in some cases to apply quantities of spray liquor exceeding 1,000 litres per hectare. It is also possible to use the active compounds in the so-called "Ultra-low-Volume method", and also to apply them in the form of the so-called microgranules.

These preparations may be prepared in a manner known *per se*, for example, by mixing or grinding methods. If desired, the individual components may be mixed together only shortly before being used, for example, in the so-called tank mix method carried out in practice.

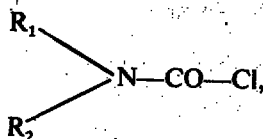
The new compounds of the present invention may be prepared by methods known *per se*, for example by one of the following processes which are also included within the scope of the present invention.

The present invention accordingly further provides a process for the manufacture of a compound of the general formula I, wherein

a) a compound of the general formula

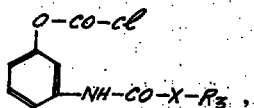


in which R_3 and X have the meanings given above, or an alkali metal salt thereof is reacted with a carbamoyl chloride of the general formula

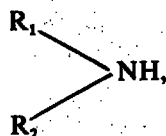


in which R_1 and R_2 have the meanings given above, if desired in the presence of a tertiary organic base, for example triethylamine or pyridine, at a temperature of 0°C to 100°C , or

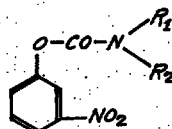
b) a compound of the general formula



in which R_3 and X have the meanings given above, is reacted in the presence of an acid-binding agent, for example an inorganic base, for example sodium hydroxide solution, sodium carbonate or potassium carbonate, or a tertiary organic base, for example triethylamine, with an amine of the general formula



in which R_1 and R_2 have the meanings given above, or
c) a nitro compound of the general formula



5 in which R_1 and R_2 have the meanings given above, is catalytically hydrogenated,
for example, with the use of nickel in methanol, to form the corresponding amine
which is then reacted in the presence of an acid-binding agent, for example an
inorganic base, for example sodium hydroxide solution, sodium carbonate or
10 potassium carbonate, or an organic base, for example triethylamine, with a
compound of the general formula



in which R_3 and X have the meanings given above.

The products of the process of the present invention are isolated in the usual
manner.

15 Some compounds of the present invention are listed in the following Table.

TABLE I.

Compound No.	Name of the compound	Physical constant
1	Ethyl-N-[3-(N-ethyl-N-isopropylcarbamoyloxy)-phenyl]-carbamate	M.p.: 65– 66°C
2	Ethyl-N-[3-(N,N-dipropyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 82– 84°C
3	Methyl-N-[3-(N-ethyl-N-benzylcarbamoyloxy)-phenyl]-carbamate	M.p.: 97– 98°C
4	S-Methyl-N-[3-(N-ethyl-N-benzylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 108–111°C
5	Ethyl-N-[3-(N-ethyl-N-benzylcarbamoyloxy)-phenyl]-carbamate	M.p.: 93– 94°C
6	Methyl-N-[3-[N-methyl-N-(2-phenylethyl)-carbamoyloxy]-phenyl]-carbamate	M.p.: 88– 90°C
7	S-Methyl-N-[3-(N-butyl-N-benzylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 82– 84°C
8	Ethyl-N-[3-(N-methyl-N-benzylcarbamoyloxy)-phenyl]-carbamate	M.p.: 83– 85°C
9	S-Methyl-N-[3-(N-methyl-N-benzylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 83– 85°C
10	Methyl-N-[3-(N-butyl-N-benzylcarbamoyloxy)-phenyl]-carbamate	M.p.: 57– 61°C

TABLE 1 (Continued)

Compound No.	Name of the compound	Physical constant
11	Methyl-N-[3-(N-methyl-N-benzylcarbamoyloxy)-phenyl]-carbamate	M.p.: 79–82°C
12	Ethyl-N-[3-[N-methyl-N-(2-phenylethyl)-carbamoyloxy]-phenyl]-carbamate	M.p.: 101–103°C
13	S-Methyl-N-[3-[N-methyl-N-(2-phenylethyl)-carbamoyloxy]-phenyl]-thiocarbamate	M.p.: 69–72°C
14	Methyl-N-[3-[N,N-bis-(2-chlorethyl)-carbamoyloxy]-phenyl]-carbamate	M.p.: 58–61°C
15	Ethyl-N-[3-[N,N-bis-(2-chlorethyl)-carbamoyloxy]-phenyl]-carbamate	M.p.: 69–72°C
16	S-Methyl-N-[3-(N,N-dipropylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 92–93°C
17	S-Methyl-N-[3-(N-ethyl-N-butylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 98–100°C
18	S-Methyl-N-[3-(N,N-di-isobutylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 117–118°C
19	S-Methyl-N-[3-(N,N-dibutylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 57–58°C
20	S-Methyl-N-[3-(N-ethyl-N-isopropylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 124–126°C
21	S-Methyl-N-[3-(N-methyl-N-cyclohexylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 110–113°C
22	S-Methyl-N-[3-(N-ethyl-N-cyclohexylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 155–156°C
23	S-Methyl-N-[3-[N,N-bis-(2-chlorethyl)-carbamoyloxy]-phenyl]-thiocarbamate	M.p.: 104–106°C
24	S-Methyl-N-[3-(N,N-diallylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 67–70°C
25	S-Methyl-N-[3-(N,N-di-isopropylcarbamoyloxy)-phenyl]-thiocarbamate	M.p.: 177–178°C
26	Methyl-N-[3-(N-propyl-N-cyclohexylcarbamoyloxy)-phenyl]-carbamate	M.p.: 102–103.5°C
27	Methyl-N-[3-(N-isobutyl-N-cyclohexylcarbamoyloxy)-phenyl]-carbamate	M.p.: 105.5–107°C
28	Methyl-N-[3-(N,N-dipropylcarbamoyloxy)-phenyl]-carbamate	M.p.: 68–69°C
29	Methyl-N-[3-(N-ethyl-N-butylcarbamoyloxy)-phenyl]-carbamate	M.p.: 69–71°C
30	Methyl-N-[3-(N-ethyl-N-isopropylcarbamoyloxy)-phenyl]-carbamate	M.p.: 93–95°C

TABLE 1 (Continued)

Compound No.	Name of the compound	Physical constant
31	Methyl-N-[3-(N,N-diallyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 46–48°C
32	Ethyl-N-[3-(N-ethyl-N-butylcarbamoyloxy)-phenyl]-carbamate	M.p.: 57–59°C
33	Ethyl-N-[3-(N,N-diallyl-carbamoyloxy)-phenyl]-carbamate	$n_D^{20}=1.5250$
34	Methyl-N-[3-(N-methyl-N-cyclohexyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 120–121°C
35	Ethyl-N-[3-(N-methyl-N-cyclohexyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 77–79°C
36	Methyl-N-[3-(N-cyclohexyl-N-isobutyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 105.7–107°C
37	Ethyl-N-[3-(N-cyclohexyl-N-propyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 91–95°C
38	Ethyl-N-[3-(N-cyclohexyl-N-isopropyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 134–137°C
39	Methyl-N-[3-(N-cyclohexyl-N-isopropyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 128–130°C
40	S-Methyl-N-[3-(N-cyclohexyl-N-propyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 116–119°C
41	Ethyl-N-[3-(N-butyl-N-cyclohexylcarbamoyloxy)-phenyl]-carbamate	M.p.: 89–93°C
42	Methyl-N-[3-(N-n-butyl-N-cyclohexyl-carbamoyloxy)-phenyl]-carbamate	$n_D^{20}=1.5140$
43	S-Methyl-N-[3-(N-cyclohexyl-N-isobutyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 117–119°C
44	Ethyl-N-[3-(N-cyclohexyl-N-sec.-butyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 98–100°C
45	Methyl-N-[3-(N-cyclohexyl-N-sec.-butyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 106–107°C
46	Methyl-N-[3-[N-(1-methyl-2-propynyl)-N-methyl-carbamoyloxy]-phenyl]-carbamate	M.p.: 115–117°C
47	Ethyl-N-[3-(N-isobutyl-N-cyclohexyl-carbamoyloxy)-phenyl]-carbamate	$n_D^{20}=1.5210$
48	S-Ethyl-N-[3-(N-ethyl-N-benzyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 86–89°C
49	S-Ethyl-N-[3-(N-cyclohexyl-N-methyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 144–146°C
50	S-Ethyl-N-[3-(N,N-diallyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 48–52°C

TABLE 1 (Continued)

Compound No.	Name of the compound	Physical constant
51	S-Ethyl-N-[3-(N-ethyl-N-isopropyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 95–96°C
52	S-Ethyl-N-[3-(N-ethyl-N-butyl-carbamoyloxy)-phenyl]-thiocarbamate	$n_D^{20}=1.5445$
53	S-Ethyl-N-[3-[N,N-bis-(2-chlorethyl)-carbamoyloxy]-phenyl]-thiocarbamate	M.p.: 100–102°C
54	S-Ethyl-N-[3-(N-butyl-N-cyclohexyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 122–124°C
55	S-Ethyl-N-[3-(N-cyclohexyl-N-isobutyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 140–142°C
56	S-Ethyl-N-[3-(N-sec.-butyl-N-cyclohexyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 130–132°C
57	Methyl-N-[3-[N-ethyl-N-(2-chlorethyl)-carbamoyloxy]-phenyl]-carbamate	M.p.: 100–102°C
58	Ethyl-N-[3-[N-ethyl-N-(2-chlorethyl)-carbamoyloxy]-phenyl]-carbamate	M.p.: 96–98°C
59	Ethyl-N-[3-[N-methyl-N-(2-chlorethyl)-carbamoyloxy]-phenyl]-carbamate	M.p.: 86–88°C
60	S-Ethyl-N-[3-(N-benzyl-N-methyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 68–70°C
61	S-Ethyl-N-[3-[N-methyl-N-(2-phenylethyl)-carbamoyloxy]-phenyl]-thiocarbamate	M.p.: 84–87°C
62	S-Ethyl-N-[3-(N,N-diethyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 83–84°C
63	S-Methyl-N-[3-[N-methyl-N-(2-chlorethyl)-carbamoyloxy]-phenyl]-thiocarbamate	M.p.: 80–82°C
64	S-Methyl-N-[3-[N-ethyl-N-(2-chlorethyl)-carbamoyloxy]-phenyl]-thiocarbamate	M.p.: 83–85°C
65	S-Ethyl-N-[3-[N-methyl-N-(2-chlorethyl)-carbamoyloxy]-phenyl]-thiocarbamate	M.p.: 86–88°C
66	S-Ethyl-N-[3-(N,N-dibutyl-carbamoyloxy)-phenyl]-thiocarbamate	$n_D^{20}=1.5250$
67	Methyl-N-[3-(N-allyl-N-cyclohexyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 99–101°C
68	Methyl-N-[3-[N-ethyl-N-(2-bromethyl)-carbamoyloxy]-phenyl]-carbamate	M.p.: 117°C
69	Methyl-N-[3-(N-butyl-N-tert.-butyl-carbamoyloxy)-phenyl]-carbamate	M.p.: 59–62°C
70	Ethyl-N-[3-[N-ethyl-N-(2-bromethyl)-carbamoyloxy]-phenyl]-carbamate	M.p.: 110°C

TABLE 1 (Continued)

Compound No.	Name of the compound	Physical constant
71	S-Methyl-N-[3-[N-ethyl-N-(2-chlorethyl)-carbamoyloxy]-phenyl]-thiocarbamate	M.p.: 98–100°C
72	Methyl-N-[3-[N-methyl-N-(2-chlorethyl)-carbamoyloxy]-phenyl]-carbamate	M.p.: 103–104°C
73	S-Ethyl-N-[3-(N,N-dipropyl-carbamoyloxy)-phenyl]-thiocarbamate	M.p.: 73–74°C

These compounds are readily soluble in acetone, cyclohexanone, ethyl acetate, isophorone, ether and tetrahydrofuran and are practically insoluble in water and light gasoline.

Among the compounds of the invention, the compounds numbered 2, 14, 15, 16, 17, 21, 23, 28, 30 and 34 in the above Table are especially suitable as selective herbicides in cotton, carrot, ground-nut and rice crops.

Attention is directed under section 9 to our U.K. Patent Specification No. 1,385,802.

The following Examples illustrate the invention. Examples 1 and 2 illustrate the manufacture of compounds of the present invention and Examples 3 to 5 illustrate the herbicidal activity of compounds of the present invention.

Example 1.

Ethyl-N-[3-(N-ethyl-N-isopropyl-carbamoyloxy)-phenyl]-carbamate
(Compound No. 1).

Into a mixture of 5.3 grams of N-ethyl-isopropylamine, 50 ml of water and 30 ml of ethyl acetate were introduced dropwise while stirring at 10 to 15°C a solution of 14.6 grams of chloroformic acid-3-(N-carbethoxyamino)-phenyl ester in 30 ml of ethyl acetate and simultaneously a solution of 8.3 grams of potassium carbonate in 30 ml of water. The mixture was further stirred for 30 minutes while cooling with ice, and then the organic phase was separated and washed at 0°C with a small amount of a dilute aqueous solution of sodium hydroxide. After drying the organic phase with magnesium sulphate, it was concentrated under reduced pressure, pentane was added to the evaporation residue and cooled with ice, whereupon the reaction product crystallized out.

Yield: 14.1 grams = 80% of the theoretical yield.

M.p.: 65–66°C.

Example 2.

Ethyl-N-[3-(N,N-dipropyl-carbamoyloxy)-phenyl]-carbamate
(Compound No. 2).

A solution of 16.7 grams of 3-hydroxy-carbanilic acid methyl ester and 19.6 grams of N,N-dipropyl-carbamoyl chloride in about 50 ml of dry pyridine was heated for 45 minutes in a water bath at 90°C. After evaporating the solution *in vacuo*, the residue was dissolved in ethyl acetate, the solution was washed at 0°C with a small amount of a dilute aqueous solution of sodium hydroxide, dilute hydrochloric acid, and the solution was dried with magnesium sulphate and concentrated under reduced pressure. The evaporation residue was crystallized by the addition of pentane, and was recrystallized from tetrahydrofuran/pentane.

Yield: 21.0 grams = 68% of the theoretical yield.

M.p.: 82–84°C.

The other compounds listed in Table I above can be prepared in an analogous manner.

Example 3.

In a greenhouse the plants listed in the following Table were treated by the post-emergence method in a series of tests with preparations used in such amounts that 3 kg of the active substance being tested was applied per hectare. The active substances were in each case applied in the form of an aqueous emulsion using 500 litres of water per hectare at an early stage in the development of the plants. Evaluation of the results was carried out 14 days after the treatment by classification according to a scale of values extending from 0 representing "totally destroyed" to 10 representing "not damaged".

The results show the much better selectivity and antiweed action of the compounds of the present invention as compared with those of the known agents.

[illegible]

TABLE (Continued)

Compound of the present invention in the preparation	Rate of application in kg of active substance per hectare	Carrots	Cotton	Ground-nut	Rice	<i>Polygonum lapathifolium</i>	<i>Stellaria media</i>	<i>Lamium amplexicaule</i>	<i>Centaurea cyanus</i>	<i>Amarantus retroflexus</i>	<i>Ipomoea purpurea</i>	<i>Echinochloa crus galli</i>	<i>Setaria italica</i>	<i>Digitaria sanguinalis</i>	<i>Setaria faberi</i>
Ethyl-N-[3-(N-methyl-N-cyclohexyl)-carbamoyloxy]-phenyl]-carbamate	3	—	9	10	9	0	0	0	0	0	0	0	0	3	0
Methyl-N-[3-(N-propyl-N-cyclohexyl)-carbamoyloxy]-phenyl]-carbamate	3	—	10	10	—	0	0	0	0	0	0	0	0	0	0
Methyl-N-[3-(N-isobutyl-N-cyclohexyl)-carbamoyloxy]-phenyl]-carbamate	3	10	10	10	10	0	0	0	0	0	0	1	0	1	1
Methyl-N-[3-(N-ethyl-N-benzyl)-carbamoyloxy]-phenyl]-carbamate	3	10	10	0	0	—	1	3	0	—	0	1	0	—	—
S-Methyl-N-[3-(N-ethyl-N-benzyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	10	10	0	0	2	0	0	0	0	0	0	0	—	0
Ethyl-N-[3-(N-ethyl-N-benzyl)-carbamoyloxy]-phenyl]-carbamate	3	—	10	10	—	1	0	1	—	1	—	3	3	—	—
Methyl-N-[3-(N-methyl-N-(2-phenylethyl)-carbamoyloxy)-phenyl]-carbamate	3	—	10	10	10	0	0	0	—	0	1	1	1	1	1
S-Methyl-N-[3-(N-butyl-N-benzyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	—	10	10	10	0	0	0	0	0	0	0	0	0	0
Ethyl-N-[3-(N-methyl-N-benzyl)-carbamoyloxy]-phenyl]-carbamate	3	10	10	10	10	0	0	0	3	0	0	0	2	2	—

TABLE (Continued)

Compound of the present invention in the preparation	Rate of application in kg of active substance per hectare	Carrots	Cotton	Ground-nut	Rice	<i>Polygonum lapathifolium</i>	<i>Stellaria media</i>	<i>Lamium amplexicaule</i>	<i>Centaurea cyanus</i>	<i>Amarantus retroflexus</i>	<i>Ipomoea purpurea</i>	<i>Echinochloa crus galli</i>	<i>Setaria italica</i>	<i>Digitaria sanguinalis</i>	<i>Setaria faberi</i>
S-Methyl-N-[3-(N-methyl-N-benzyl-carbamoyloxy)-phenyl]-thiocarbamate	3	—	—	10	10	0	0	0	0	0	0	0	0	0	0
Methyl-N-[3-(N-butyl-N-benzyl-carbamoyloxy)-phenyl]-carbamate	3	10	10	10	10	1	1	1	—	0	—	—	—	3	3
Methyl-N-[3-(N-methyl-N-benzyl-carbamoyloxy)-phenyl]-carbamate	3	10	10	10	—	0	0	0	0	0	0	0	0	0	0
Ethyl-N-[3-(N-methyl-N-(2-phenylethyl)-carbamoyloxy)-phenyl]-carbamate	3	—	10	10	10	0	1	0	—	1	1	1	2	2	2
S-Methyl-N-[3-(N-methyl-N-(2-phenylethyl)-carbamoyloxy)-phenyl]-thiocarbamate	3	10	10	10	10	0	0	0	0	0	0	0	0	0	0
Methyl-N-[3-(N,N-bis-(2-chloroethyl)-carbamoyloxy)-phenyl]-carbamate	3	—	10	10	—	0	0	0	0	0	0	0	0	0	2
Ethyl-N-[3-(N,N-bis-(2-chloroethyl)-carbamoyloxy)-phenyl]-carbamate	3	—	10	10	—	0	0	3	0	0	0	0	0	0	0
S-Methyl-N-[3-(N,N-dipropyl-carbamoyloxy)-phenyl]-thiocarbamate	3	—	10	10	—	0	0	0	0	0	1	0	0	0	0

TABLE (Continued)

Compound of the present invention in the preparation	Rate of application in kg of active substance per hectare	Carrots	Cotton	Ground-nut	Rice	<i>Polygonum lapathifolium</i>	<i>Stellaria media</i>	<i>Lamium amplexicaule</i>	<i>Centaurea cyanus</i>	<i>Amarantus retroflexus</i>	<i>Ipomoea purpurea</i>	<i>Echinochloa crus galli</i>	<i>Setaria italica</i>	<i>Digitaria sanguinalis</i>	<i>Setaria faberi</i>
S-Methyl-N-[3-(N-ethyl-N-butyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	-	10	10	10	0	0	0	0	0	0	0	0	0	0
S-Methyl-N-[3-(N,N-di-isobutyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	-	10	10	10	0	1	0	0	0	0	0	0	0	0
S-Methyl-N-[3-(N,N-di-butyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	-	10	10	10	0	0	0	0	0	0	0	0	0	0
S-Methyl-N-[3-(N-ethyl-N-isopropyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	10	10	10	10	0	0	0	0	0	2	0	0	0	0
S-Methyl-N-[3-(N-methyl-N-cyclohexyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	-	-	10	10	0	0	0	0	0	0	0	0	0	0
S-Methyl-N-[3-(N-ethyl-N-cyclohexyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	-	-	10	10	0	0	0	0	0	0	0	0	0	0
S-Methyl-N-[3-(N,N-bis-(2-chlorethyl))-carbamoyloxy]-phenyl]-thiocarbamate	3	-	10	10	10	0	0	0	0	0	0	0	0	-	0
S-Methyl-N-[3-(N,N-diallyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	10	10	10	-	0	0	0	0	0	1	0	0	0	0

TABLE (Continued)

Compound of the present invention in the preparation	Rate of application in kg of active substance per hectare	Carrots	Cotton	Ground-nut	Rice	<i>Polygonum lapathifolium</i>	<i>Stellaria media</i>	<i>Lamium amplexicaule</i>	<i>Centaurea cyanus</i>	<i>Amarantus retroflexus</i>	<i>Ipomoea purpurea</i>	<i>Echinochloa crus galli</i>	<i>Setaria italica</i>	<i>Digitaria sanguinalis</i>	<i>Setaria faberi</i>
S-Methyl-N-[3-(N,N-di-isopropyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	—	—	10	10	0	—	4	—	—	—	—	2	—	2
Methyl-N-[3-(N-cyclohexyl-N-isobutyl)-carbamoyloxy]-phenyl]-carbamate	3	—	10	10	10	0	1	0	0	0	0	—	0	0	3
Ethyl-N-[3-(N-cyclohexyl-N-propyl)-carbamoyloxy]-phenyl]-carbamate	3	—	—	10	10	0	0	0	0	0	0	0	0	0	0
Ethyl-N-[3-(N-cyclohexyl-N-isopropyl)-carbamoyloxy]-phenyl]-carbamate	3	—	—	10	10	—	—	0	3	0	0	—	—	—	—
Methyl-N-[3-(N-cyclohexyl-N-isopropyl)-carbamoyloxy]-phenyl]-carbamate	3	—	—	10	10	1	—	0	1	0	0	—	—	—	—
S-Methyl-N-[3-(N-cyclohexyl-N-propyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	—	—	10	10	0	0	0	0	0	0	0	0	—	—
Ethyl-N-[3-(N-butyl-N-cyclohexyl)-carbamoyloxy]-phenyl]-carbamate	3	—	10	10	10	0	0	0	0	0	0	0	0	0	0

Rate of application in kg of
active substance per hectare

[illegible]

TABLE (Continued)

Compound of the present invention in the preparation	Rate of application in kg of active substance per hectare	Carrots	Cotton	Ground-nut	Rice	<i>Polygonum lapathifolium</i>	<i>Stellaria media</i>	<i>Lamium amplexicaule</i>	<i>Centaurea cyanus</i>	<i>Amarantus retroflexus</i>	<i>Ipomoea purpurea</i>	<i>Echinochloa crus galli</i>	<i>Setaria italica</i>	<i>Digitaria sanguinalis</i>	<i>Setaria faberi</i>
Ethyl-N-[3-[N-ethyl-N-(2-chloroethyl)-carbamoyloxy]-phenyl]-carbamate	3	10	10	10	10	0	0	0	0	0	0	3	2	1	0
Ethyl-N-[3-[N-methyl-N-(2-chloroethyl)-carbamoyloxy]-phenyl]-carbamate	3	10	10	10	10	0	0	0	0	0	0	1	1	1	1
S-Ethyl-N-[3-(N-benzyl)-N-methyl-carbamoyloxy]-phenyl]-thiocarbamate	3	10	10	10	10	1	0	0	2	0	0	1	1	1	1
S-Ethyl-N-[3-(N-ethyl-N-benzyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	10	9	10	10	0	5	0	1	0	0	1	1	1	1
S-Ethyl-N-[3-(N-cyclohexyl)-N-methyl-carbamoyloxy]-phenyl]-thiocarbamate	3	10	10	10	10	0	0	0	0	0	0	4	0	1	1
S-Ethyl-N-[3-(N,N-diallyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	10	10	10	10	0	0	0	3	0	3	0	0	1	1
S-Ethyl-N-[3-(N-ethyl-N-isopropyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	10	10	10	10	0	0	4	5	0	0	1	1	1	1
S-Ethyl-N-[3-(N-ethyl-N-butyl)-carbamoyloxy]-phenyl]-thiocarbamate	3	10	10	10	10	0	1	5	1	0	0	4	2	1	1

TABLE (Continued)

Compound of the present invention in the preparation	Rate of application in kg of active substance per hectare				Carrots	Cotton	Ground-nut	Rice	<i>Polygonum lapathifolium</i>	<i>Stellaria media</i>	<i>Lamium amplexicaule</i>	<i>Centarea cyanus</i>	<i>Amarantus retroflexus</i>	<i>Ipomoea purpurea</i>	<i>Echinochloa crus galli</i>	<i>Setaria italica</i>	<i>Digitaria sanguinalis</i>	<i>Setaria faberi</i>
S-Ethyl-N-[3-(N,N-diethyl-carbamoyloxy)-phenyl]-thiocarbamate	3	10	9	10	10	9	10	-	0	0	0	3	0	2	0	0	0	1
S-Methyl-N-[3-(N-methyl-N-(2-chlorethyl)-carbamoyloxy)-phenyl]-thiocarbamate	3	-	-	9	9	-	9	9	0	0	0	0	0	0	0	0	0	0
S-Ethyl-N-[3-(N-ethyl-N-(2-chlorethyl)-carbamoyloxy)-phenyl]-thiocarbamate	3	10	9	9	9	9	9	8	0	0	0	0	1	1	1	0	0	0
S-Ethyl-N-[3-(N-methyl-N-(2-chlorethyl)-carbamoyloxy)-phenyl]-thiocarbamate	3	9	10	10	10	10	10	10	0	0	0	0	1	1	3	3	0	1
S-Ethyl-N-[3-(N,N-dibutyl-carbamoyloxy)-phenyl]-thiocarbamate	3	-	10	10	10	10	10	10	1	2	-	0	0	0	2	0	0	0
Methyl-N-[3-(N-allyl-N-cyclohexyl-carbamoyloxy)-phenyl]-carbamate	3	-	-	9	9	-	9	9	0	0	0	0	0	0	3	0	0	0
Methyl-N-[3-(N-ethyl-N-(2-bromoethyl)-carbamoyloxy)-phenyl]-carbamate	3	-	8	10	10	10	10	10	2	0	0	0	0	0	3	0	0	0
Methyl-N-[3-(N-butyl-N-tert.-butyl-carbamoyloxy)-phenyl]-carbamate	3	9	10	10	10	10	10	10	-	1	3	-	0	5	-	0	0	-

TABLE (Continued)

Compound of the present invention in the preparation	Rate of application in kg of active substance per hectare									
	Carrots	Cotton	Ground-nut	Rice	Polygonum lapathifolium	Stellaria media	Lamium amplexicaule	Centaurea cyanus	Amarantus retroflexus	Ipomoea purpurea
Ethyl-N-[3-[N-ethyl-N-(2-bromethyl)-carbamoyloxy]-phenyl]-carbamate	3 10	10	10	10	-	1	0	0	0	2
S-Methyl-N-[3-[N-ethyl-N-(2-chlorethyl)-carbamoyloxy]-phenyl]-thiocarbamate	3 8	-	10	-	0	0	0	0	0	0
Methyl-N-[3-[N-methyl-N-(2-chlorethyl)-carbamoyloxy]-phenyl]-carbamate	3 -	-	10	-	0	0	0	0	0	0
S-Ethyl-N-[3-(N,N-diptypyl-carbamoyloxy)-phenyl]-thiocarbamate	3 8	10	10	10	0	3	3	-	3	0
Compound in preparation used for comparison										
N-(3-Trifluoromethylphenyl)-N,N-dimethyl-urea	3 0	1	1	0	0	0	0	0	0	0
Methyl-N-[3-[N-(3-methylphenyl)-carbamoyloxy]-phenyl]-carbamate	3 5	2	8	-	2	0	0	0	5	0

0=totally destroyed

10=not damaged

Example 4.

In a greenhouse, the plants listed in the following Table were uniformly sprayed in a series of tests before emergence (that is by the pre-emergence method) with preparations used in such amounts that 3 kg of the active substance being tested was applied per hectare. The active substances were in each case used in the form of an aqueous emulsion using 500 litres per hectare. After 14 days, the results were evaluated according to the scale given in Example 3.

The results demonstrate the excellent tolerability of cotton, carrots, ground-nut, potatoes and rice towards the compounds of the invention combined with a very good antiweed action.

In contrast therewith, the known agent exhibited no action at the corresponding rate of application.

[illegible]

Example 5.

In a greenhouse, plants in an early stage of development were sprayed in a series of tests with preparations used in such amounts that 2 kg of active substance being tested was applied per hectare, the active substances in each case being used in the form of an aqueous emulsion using 500 litres of water per hectare. The evaluation was carried out 14 days after the treatment by a classification as described in Example 3.

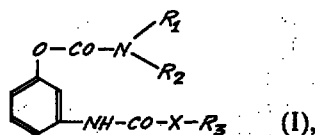
Even at this rate of application the compounds of the present invention still exhibit an excellent herbicidal action.

Compound of the present invention in the preparation	Rate of application in kg of active substance per hectare	<i>Amarantus spinosus</i>	<i>Xanthium pensilvanicum</i>	<i>Portulaca oleracea</i>	<i>Setaria verticillata</i>	<i>Rottboellia exaltata</i>	<i>Eleusine indica</i>
Methyl-N-[3-(N,N-dipropyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	0.5	2
Methyl-N-[3-(N-ethyl-N-butyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	1	4
Methyl-N-[3-(N-ethyl-N-isopropyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	0.5	1
Methyl-N-[3-(N,N-diallyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	1	1
Ethyl-N-[3-(N-ethyl-N-butyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	—	—
Ethyl-N-[3-(N,N-diallyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	—	—
Ethyl-N-[3-(N-ethyl-N-isopropyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	—	—

0=totally destroyed
10=not damaged

WHAT WE CLAIM IS:—

1. A compound of the general formula I



in which

R₁ represents an allyl group, an unsubstituted alkyl group containing 1 to 4 carbon atoms or a halogen-substituted alkyl group containing 1 to 4 carbon atoms,

R₂ represents an allyl group, an alkynyl group, an unsubstituted alkyl group containing 1 to 4 carbon atoms, a halogen-substituted alkyl group containing 1 to 4 carbon atoms, or a cyclohexyl, benzyl or phenylethyl group,

R₃ represents a methyl or ethyl group, and

X represents an oxygen or sulphur atom, and in which, when R₁ and R₂ are the same alkyl groups, R₃ is not an ethyl group.



Example 5.

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In a greenhouse, plants in an early stage of development were sprayed in a series of tests with preparations used in such amounts that 2 kg of active substance being tested was applied per hectare, the active substances in each case being used in the form of an aqueous emulsion using 500 litres of water per hectare. The evaluation was carried out 14 days after the treatment by a classification as described in Example 3.

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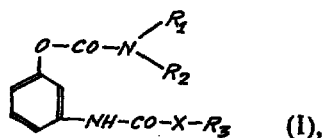
Even at this rate of application the compounds of the present invention still exhibit an excellent herbicidal action.

Compound of the present invention in the preparation	Rate of application in kg of active substance per hectare	<i>Amarantus spinosus</i>	<i>Xanthium pensilvanicum</i>	<i>Portulacae oleracea</i>	<i>Setaria verticillata</i>	<i>Rottboellia exaltata</i>	<i>Eleusine indica</i>
Methyl-N-[3-(N,N-dipropyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	0.5	2
Methyl-N-[3-(N-ethyl-N-butyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	1	4
Methyl-N-[3-(N-ethyl-N-isopropyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	0.5	1
Methyl-N-[3-(N,N-diallyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	1	1
Ethyl-N-[3-(N-ethyl-N-butyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	—	—
Ethyl-N-[3-(N,N-diallyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	—	—
Ethyl-N-[3-(N-ethyl-N-isopropyl-carbamoyloxy)-phenyl]-carbamate	2	0	0	0	0	—	—

0=totally destroyed
10=not damaged

WHAT WE CLAIM IS:—

1. A compound of the general formula I



in which

15

R_1 represents an allyl group, an unsubstituted alkyl group containing 1 to 4 carbon atoms or a halogen-substituted alkyl group containing 1 to 4 carbon atoms,

15

R_2 represents an allyl group, an alkynyl group, an unsubstituted alkyl group containing 1 to 4 carbon atoms, a halogen-substituted alkyl group containing 1 to 4 carbon atoms, or a cyclohexyl, benzyl or phenylethyl group,

20

R_3 represents a methyl or ethyl group, and

20

X represents an oxygen or sulphur atom, and in which, when R_1 and R_2 are the same alkyl groups, R_3 is not an ethyl group.



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2. A compound as claimed in claim 1, wherein each of the halogen-substituted alkyl groups is a 2-chlorethyl group.

3. Ethyl-N-[3-(N,N-dipropyl-carbamoyloxy)-phenyl]-carbamate.

4. Ethyl-N-[3-[N,N-bis-(2-chlorethyl)-carbamoyloxy]-phenyl]-carbamate.

5. Methyl-N-[3-[N,N-bis-(2-chlorethyl)-carbamoyloxy]-phenyl]-carbamate.

6. S-Methyl-N-[3-(N,N-dipropyl-carbamoyloxy)-phenyl]-thiocarbamate.

7. S-Methyl-N-[3-(N-ethyl-N-butyl-carbamoyloxy)-phenyl]-thiocarbamate.

8. S - Methyl - N - [3 - (N - methyl - N - cyclohexyl - carbamoyloxy) - phenyl] - thiocarbamate.

9. S - Methyl - N - [3 - [N,N - bis - (2 - chlorethyl) - carbamoyloxy] - phenyl] - thiocarbamate.

10. Methyl-N-[3-(N,N-dipropyl-carbamoyloxy)-phenyl]-carbamate.

11. Methyl-N-[3-(N-ethyl-N-isopropyl-carbamoyloxy)-phenyl]-carbamate.

12. Methyl-N-[3-(N-methyl-N-cyclohexyl-carbamoyloxy)-phenyl]-carbamate.

13. A herbicidal preparation which comprises a compound of the general formula I given in claim 1, in which R₁, R₂, R₃ and X have the meanings given in claim 1, in admixture or conjunction with a suitable carrier.

14. A herbicidal preparation which comprises a compound as claimed in claim 2, in admixture or conjunction with a suitable carrier.

15. A herbicidal preparation which comprises the compound claimed in any one of claims 3 to 12, in admixture or conjunction with a suitable carrier.

16. A herbicidal preparation which comprises any one of the compounds numbered 1, 3 to 13, 18 to 20, 22, 24 to 27, 29, 31 to 33 and 35 in Table I herein, in admixture or conjunction with a suitable carrier.

17. A herbicidal preparation which comprises any one of the compounds numbered 36 to 73 in Table I herein, in admixture or conjunction with a suitable carrier.

18. A preparation as claimed in any one of claims 13 to 17, which is in the form of a powder, a spraying or dusting agent, granules, a solution, an emulsion or a suspension.

19. A preparation as claimed in any one of claims 13 to 18, containing a single compound of the general formula I in an amount of approximately 20 to 80% by weight.

20. A preparation as claimed in any one of claims 13 to 18, containing two or more compounds of the general formula I in a total amount of approximately 20 to 80% by weight.

21. A preparation as claimed in any one of claims 13 to 20, containing a single surface-active agent in an amount of up to 30% by weight.

22. A preparation as claimed in any one of claims 13 to 20, containing two or more surface-active agents in a total amount of up to 30% by weight.

23. Any one of the herbicidal preparations as claimed in claim 13 and substantially as described in Example 3 herein.

24. Any one of the herbicidal preparations as claimed in claim 13 and substantially as described in Examples 4 and 5 herein.

25. A method of protecting a living plant against weeds, wherein the area in the vicinity of a living plant is treated with a compound of the general formula I given in claim 1, in which R₁, R₂, R₃ and X have the meanings given in claim 1.

26. A method as claimed in claim 25, wherein the area is treated with a compound as claimed in claim 2.

27. A method as claimed in claim 25, wherein the area is treated with the compound claimed in any one of claims 3 to 12.

28. A method as claimed in claim 25, wherein the area is treated with any one of the compounds numbered 1, 3 to 13, 18 to 20, 22, 24 to 27, 29, 31 to 33 and 35 in Table I herein.

29. A method as claimed in claim 25, wherein the area is treated with any one of the compounds numbered 36 to 73 in Table I herein.

30. A method as claimed in claim 25, wherein the area is treated with a herbicidal preparation as claimed in any one of claims 13 to 24.

31. A method as claimed in any one of claims 25 to 30, wherein a single compound of the general formula I is used for the treatment in an amount of approximately 1 to 3 kg per hectare.

32. A method as claimed in any one of claims 25 to 30, wherein two or more compounds of the general formula I are used for the treatment in a total amount of approximately 1 to 3 kg per hectare.



33. A method as claimed in any one of claims 25 to 32, wherein the treatment is carried out at the post-emergence stage of the weeds.

34. A method as claimed in claim 25, conducted substantially as described in Example 3 herein.

35. A method as claimed in claim 25, conducted substantially as described in Example 4 herein.

36. A method of protecting a crop area against weeds, wherein a crop area is treated with a compound of the general formula I given in claim 1, in which R_1 , R_2 , R_3 and X have the meanings given in claim 1.

37. A method as claimed in claim 36, wherein the crop area is treated with a compound as claimed in claim 2.

38. A method as claimed in claim 36, wherein the crop area is treated with the compound claimed in any one of claims 3 to 12.

39. A method as claimed in claim 36, wherein the crop area is treated with any one of the compounds numbered 1, 3 to 13, 18 to 20, 22, 24 to 27, 29, 31 to 33 and 35 in Table I herein.

40. A method as claimed in claim 36, wherein the crop area is treated with any one of the compounds numbered 36 to 73 in Table I herein.

41. A method as claimed in claim 36, wherein the crop area is treated with a herbicidal preparation as claimed in any one of claims 13 to 24.

42. A method as claimed in any one of claims 36 to 41, wherein a single compound of the general formula I is used for the treatment in an amount of approximately 1 to 3 kg per hectare.

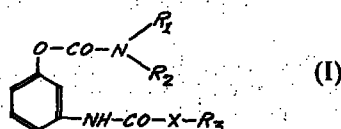
43. A method as claimed in any one of claims 36 to 41, wherein two or more compounds of the general formula I are used for the treatment in a total amount of approximately 1 to 3 kg per hectare.

44. A method as claimed in any one of claims 36 to 43, wherein the treatment is carried out at the post-emergence stage of the weeds.

45. A method as claimed in any one of claims 36 to 44, wherein the crop is a cotton crop.

46. A method as claimed in any one of claims 36 to 44, wherein the crop is a carrot, ground-nut or rice crop.

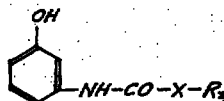
47. A process for the manufacture of a compound of the general formula I



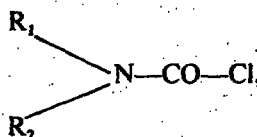
in which

R_1 , R_2 , R_3 and X are as defined in claim 1, wherein

(a) a compound of the general formula

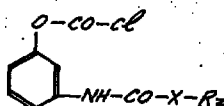


in which R_3 and X have the meanings given above, or an alkali metal salt thereof is reacted with a carbamoyl chloride of the general formula



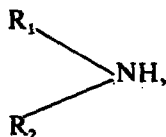
in which R_1 and R_2 have the meanings given above, or

(b) a compound of the general formula

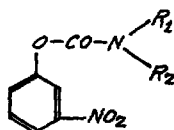




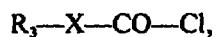
in which R_1 and X have the meanings given above, is reacted in the presence of an acid-binding agent with an amine of the general formula



in which R_1 and R_2 have the meanings given above, or
(c) a nitro compound of the general formula



in which R_1 and R_2 have the meanings given above, is catalytically hydrogenated to form the corresponding amine which is then reacted in the presence of an acid-binding agent with a compound of the general formula



in which R_3 and X have the meanings given above.

48. A process as claimed in claim 47, wherein the reaction with the carbamoyl chloride is carried out in the presence of a tertiary organic base.

49. A process as claimed in claim 47, conducted substantially as described in Example 1 or 2 herein.

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