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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 5 : A01N 57/26 — **A1**

(11) International Publication Number: WO 92/00673

(43) International Publication Date: 23 January 1992 (23.01.92)

(21) International Application Number: PCT/US91/04450

(22) International Filing Date: 28 June 1991 (28.06.91)

(30) Priority data:
551,121 — 11 July 1990 (11.07.90) — US

(60) Parent Application or Grant
(63) Related by Continuation
US — 551,121 (CIP)
Filed on — 11 July 1990 (11.07.90)

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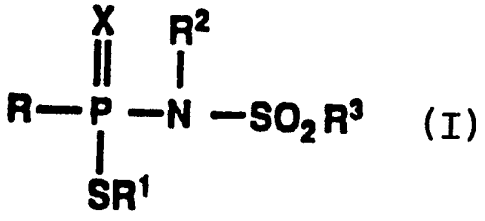
(81) Designated States: AT (European patent), BE (European patent), CH (European patent), DE (European patent), DK (European patent), ES (European patent), FR (European patent), GB (European patent), GR (European patent), IT (European patent), JP, LU (European patent), NL (European patent), SE (European patent), US.

Published
With international search report.
Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.

(54) Title: NEMATICIDAL N-(SULFONYL)PHOSPHONAMIDOTHIOATES AND DITHIOATES



(I)

(57) Abstract

A method for controlling nematodes by application of an effective amount of a compound of formula (I), wherein X, R, R^1, R^2 and R^3 are defined in the text.

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TITLE

NEMATICIDAL N-(SULFONYL)PHOSPHON-AMIDOTHIOATES AND DITHIOATES

BACKGROUND OF THE INVENTION

Field of the Invention

This invention relates to a method for controlling nematodes by the application of an effective amount of certain N-(R-sulfonyl) phosphonamidothioates and dithioates.

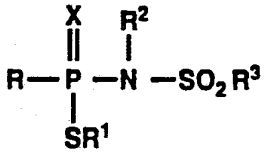
State of the Art

Plant parasitic nematodes are microscopic worms found in virtually all soils in which plants will grow. Consequently, every crop is a potential target of these pathogens. The roots of infected plants display a stunted appearance, numerous galls, knots, lesions or various other manifestations depending upon the plant and the specific nematode. The entire plant often has the appearance of being nutritionally deficient. There is great commercial interest in finding compounds that control nematodes while exhibiting little or no plant phytotoxicity.

U.S. Patent 4,683,224 discloses certain N-formyl phosphonamidothioates and their use as insecticides, miticides and/or nematicides. U.S. Patent No. 4,804,654 discloses the N-(R-sulfonyl) phosphonamidothioates and dithioates employed in this invention and their use as insecticides and miticides. There is, however, no disclosure of the use of such compounds as nematicides.

SUMMARY OF THE INVENTION

This invention pertains to a method for controlling nematodes by applying to them or to their environment (the soil) an effective amount of a compound of Formula I:



I

wherein:

X is oxygen or sulfur;

R is alkyl or alkenyl of up to twenty carbon atoms, phenyl, or phenylalkyl of up to ten carbon atoms;

R^1 is alkyl, or alkenyl of up to six carbon atoms, or phenalkyl of up to ten carbon atoms;

R^2 is alkyl, alkenyl, alkynyl, haloalkyl or alkylthioalkyl of up to ten carbon atoms; naphthyl, pyridyl, or thienyl; phenyl, or phenalkyl or phenalkenyl of up to ten carbon atoms, which may be substituted on the ring by from one to three substituents selected from halogen, methyl, methoxy, nitro, amino, mono- and dialkylamino, and mono- and dialkylaminocarbonyl wherein each alkyl moiety contains from one to four carbon atoms;

R^3 is R^2 or NR^4R^5;

R^4 is alkyl of one to four carbon atoms, phenyl, or phenalkyl of up to ten carbon atoms;

R^5 is hydrogen or R^4; or

R^4 and R^5 together with the nitrogen atom to which they are attached form 1-piperidino, 2-(ethoxycarbonyl)-1-piperidino, or 4-morpholino.

These compounds have been found to be useful for controlling a variety of nematodes that cause damage to growing plants. The compounds of Formula I are well-

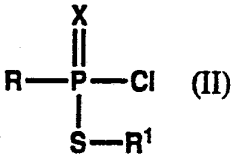
suited for the control of most plant parasitic nematodes, including the following:

Awl nematode	-	Dolichodorus heterocephatus
Banana nematode	-	Pratylenchus musicola
Bud and leaf nematode	-	Aphelenchoides spp.
Burrowing nematode	-	Radopholus similus
Carrot root nematode	-	Heterodera carotae
Coffee root-knot nematode	-	Meloidogyne exigua
Corn nematode	-	Praylenchus zeae
Dagger nematodes	-	Xiphinema spp.
Golden Nematode	-	Globodera rostochiensis
Grass nematode	-	Anguina agrosis
Lance nematodes	-	Hoplolaimus spp.
Lesion nematodes	-	Pratylenchus spp.
Northern Root-knot nematode	-	Meloidogyne hapla
Pea root nematode	-	Heterodera gottingiana
Peanut root-knot nematode	-	Meloidogyne arenaria
Pin nematodes	-	Paratylenchus spp.
Potato rot nematode	-	Ditylenchus destructor
Reinform nematode	-	Rotylenchulus reinformus
Rice nematode	-	Ditylenchus angustus
Ring nematodes	-	Criconemoides spp.
Smooth-headed lesion nematode	-	Pratlyenchus brachyurus
Southern root-knot nematode	-	Meloidogyne incognita
Soybean cyst nematode	-	Heterodera glycines
Spiral nematodes	-	Helicotylenchus spp.
Stem and bulb nematode	-	Ditylenchus dipsaci
Sting nematodes	-	Belonolaimus spp.
Stubby-root nematodes	-	Trichodorus spp.
Sugar beet nematode	-	Heterodera schachtii
Tobacco cyst nematode	-	Heterodera tabacum
Tobacco stunt nematode	-	Tylenchorhynchus claytoni
Wheat nematode	-	Anguina tritici

Application is generally made to the soil of a nematicidally effective amount of a compound of Formula I. Preferred are those compounds wherein R is ethyl, R^1 is 1-methylpropyl, R^2 is methyl and R^3 is methyl.

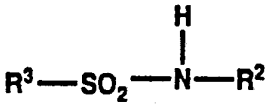
DETAILS OF THE INVENTION

Compounds of Formula I can be prepared by the processes disclosed in U.S. Patent No. 4,804,654. Specifically, S-(1-methylpropyl) P-ethyl-N-methyl-N-(methylsulfonyl)-phosphonamidothioates of Formula I can be prepared by treating a phosphonothioic or phosphonodithioic chloride of the formula:



(II)

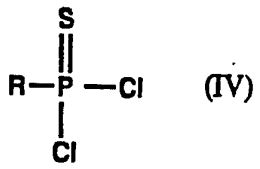
with an alkali metal salt of a sulfonamide of the formula



(III)

The treatment of the chloride (II) with a salt of the sulfonamide (III) is effected by adding the chloride at a controlled rate to a solution of the salt in an inert solvent, at a low temperature -- for example, 0°C-5°C -- moisture being excluded, then warming the mixture to room temperature. Preferably, the chloride is added as a solution in the same solvent in which the salt is dissolved. Suitable as the solvent are organic materials such as ether and tetrahydrofuran, or acetonitrile. The product is isolated and purified by conventional procedures.

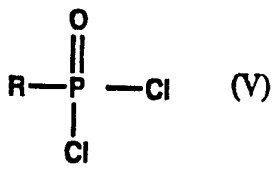
As shown in U.S. Patent No. 4,390,929, and in U.S. Patent No. 4,190,652, the phosphonodithioic chloride precursor (*II, X is* sulfur) can be prepared by treating a phosphonothioic dichloride of the formula:



(IV)

with an appropriate thiol, R^1-SH, in the presence of a solvent and an amine base, as hydrogen halide acceptor. Aromatic hydrocarbons, such as toluene, are suitable as the solvent. Any tertiary amine base is suitable, but the trialkylamines appear to be most suitable. Water should be excluded from the reaction mixture -- as by using anhydrous reagents and conducting the treatment under nitrogen. Isolation of the product is effected by conventional techniques.

The phosphonothioic chloride precursor (II, X is oxygen) can be prepared by a method analogous to that described in U.S. Patent No. 4,190,652 for preparing the corresponding phosphonodithioic chloride -- i.e., by treating a phosphonic dichloride of the formula:



(V)

with an appropriate thiol, R^1-SH, in the presence of an inert solvent and an amine base as hydrogen chloride acceptor. Aromatic hydrocarbons, such as toluene, are suitable as the solvent. Any tertiary amine base is suitable, but the trialkylamines appear to be most suitable. Water should be excluded from the reaction

mixture -- as by using anhydrous reagents and conducting the treatment under nitrogen. Isolation of the product is effected by conventional techniques.

The sulfonamide precursors (III) as a class are known compounds, and the alkali metal salts thereof are prepared by conventional methods and techniques such as by treating the appropriate sulfonyl halide, R^3-SO_2 halogen, with the appropriate amine, R_2NH_2. Compounds of Formula III wherein $R^3=NR^4R^5$ are prepared as described by Weisz and Schulze, Annalen Der Chemic, volume 729, pages 40-51 (1969).

Formulations

A compound of Formula I is ordinarily applied most effectively by formulating it with a suitable inert carrier. The invention, therefore, also includes compositions suitable for combating nematodes, such compositions comprising an inert carrier as well as the active ingredient of Formula I. The term "inert carrier" includes use of a surfactant as well. The invention also provides a method of combating pests at a locus, which comprises applying to that locus a compound of Formula I or a pesticidal composition according to the invention.

The term "carrier" as used herein means an inert solid or liquid material, which may be inorganic or organic and of synthetic or natural origin, with which the active compound is mixed or formulated to facilitate its application to the plant, seed, soil or other object to be treated, or its storage, transport and/or handling. Any of the materials customarily employed in formulating pesticides -- i.e., horticulturally acceptable adjuvants -- are suitable.

Representative solid carriers are natural and synthetic clays and silicates, for example, natural

silicas such as diatomaceous earths; magnesium silicates, for example, talcs; magnesium aluminum silicates, for example, attapulgites are vermiculites; aluminum silicates, for example, kaolinites, montmorillonites and micas; calcium carbonate; calcium sulfate, synthetic hydrated silicon oxides and synthetic calcium or aluminum silicates; elements such as, for example, carbon and sulfur, natural and synthetic resins such as, for example, coumarone resins, polyvinyl chloride and styrene polymers and copolymers; bitumen; waxes such as, for example, beeswax, paraffin wax, and chlorinated mineral waxes; solid fertilizers, for example, superphosphates; and ground, naturally-occurring, fibrous materials, such as ground corncobs.

Examples of suitable liquid carriers are water, alcohols such as isopropyl alcohol and glycols; ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone and cyclohexanone; ethers such as cellosolves; aromatic hydrocarbons such as benzene, toluene and xylene; petroleum fractions such as kerosene, light mineral oils; chlorinated hydrocarbons such as carbon tetrachloride, perchloroethylene and trichloromethane. Also suitable are liquified, normally vaporous and gaseous compounds. Mixtures of different liquids are often suitable.

The surface-active agent can be an emulsifying agent or a dispersing agent or a wetting agent; it can be nonionic or ionic. Any of the surface-active agents usually applied in formulating herbicides or insecticides can be used. Examples of suitable surface-active agents are the sodium and calcium salts of polyacrylic acids and lignin sulfonic acids; the condensation products of fatty acids or aliphatic amines or amides containing at least 12 carbon atoms in the molecule with ethylene oxide and/or propylene oxide;

fatty acid esters of glycerol, sorbitan, sucrose or pentaerythritol; condensates of these with ethylene oxide and/or propylene oxide; condensation products of fatty alcohols or alkyl phenols, for example, p-octylphenol or p-octylcrfesol, with ethylene oxide and/or propylene oxide; sulfates or sufonates of these condensation products, alakali or alkaline earth metal salts, preferably sodium slats, of sulfuric or sulfonic acid esters containing at least 10 carbon atoms in the molecule, for example, sodium lauryl sulfate, sodium secondary alkyl sulfates, sodium salts of sulfonated castor oil, and sodium alkylaryl sulfonates such as sodium dodecylbenzene sulfonate; and polymers of ethylene oxide and copolymers of ethylene oxide and propylene oxides.

The compounds of Formula I can be prepared as wettable powders, dusts, granules, solutions, emulsifiable concentrates, emulsions, suspension concentrates and aerosols. Wettable powders are usually compounded to contain 25-75% by weight of active compound and usually contain, in addition to the solid carrier, 3-10% by weight of a dispersing agent, 2-15% of a surface-active agent and, where necessary, 0-10% by weight of stabilizer(s) and/or other additives such as penetrants or stickers. Dusts are usually formulated as a dust concentrate having a similar composition to that of a wettable powder but without a dispersant or surface-active agent, and are diluted in the field with further solid carrier to give a composition usually containing 0.5-10% by weight of the active compound. Granules are usually prepared to have a size between 10 and 100 BSA mesh (1.676-0.152 mm), and can be manufactured by agglomeration or impregnation techniques. Generally, granules will contain about 0.5-25% by weight of the active compound, 0-1% by weight of

additives such as stabilizers, slow release modifiers and binding agents. Emulsifiable concentrates usually contain, in addition to the solvent and, when necessary, cosolvent, about 10-50% weight per volume of the active compound, 2-20% weight per volume emulsifiers and 0-20% weight per volume of appropriate additives such as stabilizers, penetrants and corrosion inhibitors. Suspension concentrates are compounded so as to obtain a stable, non-sedimenting, flowable product and usually contain about 10-75% weight of the active compound, 0.5-5% weight of dispersing agents, 1-5% of surface-active agent, 0.1-10% weight of suspending agents, such as defoamers, corrosion inhibitors, stabilizers, penetrants and stickers, and as carrier, water or an organic liquid in which the active compound is substantially insoluble; certain organic solids or inorganic salts can be dissolved in the carrier to assist in preventing sedimentation or as antifreeze agents for water.

Of particular interest in current practice are the water-dispersible granular formulations. These are in the form of dry, hard granules that are essentially dust-free, and are resistant to attrition on handling, thus minimizing the formation of dust. On contact with water, the granules readily disintegrate to form stable suspensions of the particles of active material. Such formulations contain 90% or more by weight of finely divided active material, 3-7% by weight of a blend of surfactants, which act as wetting, dispersing, suspending and binding agents, and 1-3% by weight of a finely divided carrier, which acts as a resuspending agent.

Aqueous dispersions and emulsions, for example, compositions obtained by diluting a wettable powder or a concentrate according to the invention with water, also lie within the scope of the present invention. The

emulsions can be of the water-in-oil or of the oil-in-water type, and can have thick, mayonnaise-like consistency. It is evident from the foregoing that this invention contemplates compositions containing as little as about 0.0001% by weight to as much as about 95% by weight of a compound of Formula I as the active ingredient.

Also of interest are controlled release granular formulations to effect prolonged activity and reduce phytotoxicity. Slow release formulations are well known in the art and can be prepared as disclosed in U.S. 4,223,070 and Japanese Kokai 59-206302.

EXAMPLE A

Controlled Release Granules

S-(1-methylpropyl) P-ethyl-N-methyl-N-(methylsulfonyl)-phosphonamidothioate (1.5 g) is added to a solution of 30 g of cellulose acetate in acetone, and the resultant solution is added slowly to 25.5 g of agitated inert mineral granules (25/50 mesh Agsorb® made by Oil Dri Corporation). The acetone is removed by exposing the granules to a flow of nitrogen at room temperature. The product contains 5% active ingredient, entrapped in the polymeric barrier.

Useful nematicidal compositions can also contain other ingredients, for example, other compounds possessing pesticidal, especially insecticidal properties, as are appropriate to the intended purpose.

The method of applying a compound of Formula I to control pests comprises applying the compound, ordinarily in a composition of one of the aforementioned types, to the soil to be protected from the nematodes. The compound, of course, is applied in an amount sufficient to effect the desired action. This dosage is dependent upon many factors, including the carrier employed, the method and the conditions of the

application. Proper consideration and resolution of these factors to provide the necessary dosage of the active compound to the environment to be protected are within the skill of those versed in the art. In general, however, the effective dosage of the compound of Formula I to the environment to be protected, i.e., the dosage which the nematode contacts, is of the order of 0.001 to 0.5% based on the total weight of the formulation, though under some circumstances the effective concentration will be as little as 0.0001% or as much as 2%, on the same basis.

UTILITY

The compounds of Formula I are well suited for the control of plant-parasitic nematodes inhabiting the soil in which the plants are planted or are to be planted. The physical characteristics of the compounds of Formula I permit them to be placed readily at the site of nematode presence or activity without disturbing already established plants. The physical nature of the compounds of Formula I permit their movement deep into undisturbed soil around plant roots, often the site of nematode attack. The safety margin for plants make practical such application as well as application to soil being prepared for planting and application directly to seeds and growing plants at rates well above those required to control the nematodes.

APPLICATION

The compounds of Formula I, formulated for use as granules or powders containing a solid diluent, provide effective control when the formulation is physically lightly mixed with the topsoil. The mixing is concurrent with, preceded or followed by planting seed which germinate into plants.

The compounds can also be applied as a drench in a solution or dispersion in a non-phytotoxic solvent or

liquid diluent, suitably water. Such drenches can be prepared by diluting with water a concentrate containing the compounds, an emulsifying agent, and preferably an organic solvent such as acetone. The active ingredient can be applied as a band, furrow or side dress, either incorporated or not.

The compound is suitably applied to the soil at a rate of about 0.01 to 11 kg/ha. Good control of soil inhabiting nematodes is obtained at rates of about 0.1 to 5 kg/ha, and especially from about 0.125 to 4 kg/ha.

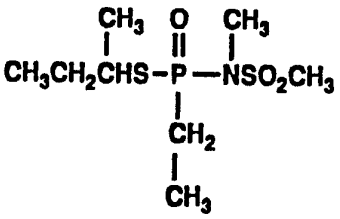
Other methods of applying the compounds of Formula I to prevent the destructive effects of plant-parasitic nematodes include: spraying above-ground parts such as stems, leaves or buds in which nematodes are already present or where later attack is possible; dipping or soaking reproductive parts such as seeds, cane pieces or bulbs (which are already infested or are to be planted in infested soil) in a water suspension, solution or emulsion of the active ingredient; or immersing the root system or the entire plant of nursery stock or transplants in a water system to disinfest them or provide protection against subsequent nematode invasion. The rates of active ingredient in the sprays or dips used as noted just above are 12 grams to 4.8 kilograms per 100 liters of water. Preferred rates are in the range of about 30 grams to 1.2 kilograms per 100 liters.

The Example below illustrates the utility of the compounds of this invention.

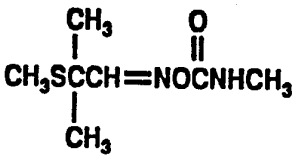
EXAMPLE

S-(1-methylpropyl) P-ethyl-N-methyl-N-(methylsulfonyl)-phosphonamidothioate (Compound 1) and Temik® (used as a standard nematicide - Compound 2) were applied as drench to 2 inch square pots containing Sassafrass soil infested with Southern root-knot nematode (Meloidogyne incognita) and planted cucumber

seeds. The drench solution was prepared by first dissolving an appropriate amount of Compound 1 or Compound 2 in acetone and then diluting with water. The test and check (untreated) pots were placed in a greenhouse at ambient temperature, watered daily, and after two weeks evaluated for nematode control. Table 1 summarizes the results.



Compound 1



Compound 2
Temik®

TABLE 1

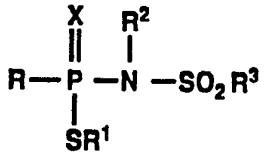
Nematicidal Efficacy of Compound 1 vs. Temik®

Treatment	Rate kg/ha	% Nematode Control
Compound 1	5.0	100
	1.0	100
	0.5	100
	0.25	100
	0.125	100
Compound 2	5.0	100
	1.0	100
	0.5	100
	0.25	70
	0.125	25
Untreated Control	-	0

CLAIMS

What is claimed is:

1. A method for controlling nematodes that comprises contacting the nematodes or their environment with a nematicidally effective amount of a compound of Formula I:



wherein:

X is oxygen or sulfur;

R is alkyl or alkenyl of up to twenty carbon atoms, phenyl, or phenylalkyl of up to ten carbon atoms;

R^1 is alkyl, or alkenyl of up to six carbon atoms, or phenalkyl of up to ten carbon atoms;

R^2 is alkyl, alkenyl, alkynyl, haloalkyl or alkylthioalkyl of up to ten carbon atoms; naphthyl, pyridyl, or thienyl; phenyl, or phenalkyl or phenalkenyl of up to ten carbon atoms, which may be substituted on the ring by from one to three substituents selected from halogen, methyl, methoxy, nitro, amino, mono- and dialkylamino, and mono- and dialkylaminocarbonyl wherein each alkyl moiety contains from one to four carbon atoms;

R^3 is R^2 or NR^4R^5;

R^4 is alkyl of one to four carbon atoms, phenyl, or phenalkyl of up to ten carbon atoms;

R^5 is hydrogen or R^4; or

R^4 and R^5 together with the nitrogen atom to which

they are attached form 1-piperidino, 2-(ethoxy-carbonyl)-1-piperidino, or 4-morpholino.

2. A method according to Claim 1 wherein the compound is S-(1-methylpropyl) P-ethyl-N-methyl-N-(methylsulfonyl)-phosphonamidothioate.

INTERNATIONAL SEARCH REPORT

International Application No PCT/US 91/04450

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all)[6]

According to International Patent Classification (IPC) or to both National Classification and IPC

Int.Cl.5 A 01 N 57/26

II. FIELDS SEARCHED

Minimum Documentation Searched[7]

Classification System	Classification Symbols
Int.Cl.5	A 01 N C 07 F

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched[8]

III. DOCUMENTS CONSIDERED TO BE RELEVANT[9]

Category °	Citation of Document, [11] with indication, where appropriate, of the relevant passages [12]	Relevant to Claim No.[13]
Y	EP,A,0241098 (SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V.) 14 October 1987, see the whole document, & US, A, 4804654 (cited in the application)	1,2
Y	FR,A,2364924 (NIHON TOKUSHU NOYAKU SEIZO K.K.) 14 April 1978, see examples; claims	1,2
Y	US,A,4161524 (BAYER AG) 17 July 1979, see examples; claims	1,2

° Special categories of cited documents : [10]

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

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"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

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IV. CERTIFICATION

Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report
01-10-1991	11. 12. 91
International Searching Authority: EUROPEAN PATENT OFFICE	Signature of Authorized Officer: Natalie Weinberg

Form PCT/ISA/210 (second sheet) (January 1985)

ANNEX TO THE INTERNATIONAL SEARCH REPORT ON INTERNATIONAL PATENT APPLICATION NO. US 9104450 SA 49047

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.
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Patent document cited in search report	Publication date	Patent family member(s)		Publication date
EP-A- 0241098	14-10-87	AU-B-	586550	13-07-89
		AU-A-	7110987	08-10-87
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		US-A-	4804654	14-02-89
FR-A- 2364924	14-04-78	JP-C-	1282252	27-09-85
		JP-A-	53037620	06-04-78
		JP-B-	60004838	06-02-85
		AT-B-	356453	25-04-80
		AU-B-	508774	03-04-80
		AU-A-	2881677	22-03-79
		BE-A-	858835	20-03-78
		DE-A-	2741085	23-03-78
		GB-A-	1544040	11-04-79
		NL-A-	7710259	22-03-78
		SE-A-	7710471	21-03-78
		US-A-	4122174	24-10-78
US-A- 4161524	17-07-79	JP-C-	999231	30-05-80
		JP-A-	52038022	24-03-77
		JP-B-	54036654	10-11-79
		AT-B-	347181	11-12-78
		AU-B-	507454	14-02-80
		AU-A-	1793076	06-04-78
		BE-A-	846423	22-03-77
		CA-A-	1081247	08-07-80
		CH-A-	622678	30-04-81
		DE-A-	2642054	07-04-77
		FR-A,B	2324641	15-04-77
		GB-A-	1527400	04-10-78
		NL-A-	7610487	24-03-77
		SE-A-	7610466	23-03-77
		US-A-	4134979	16-01-79