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Pesticides in the environment

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ABSTRACT. **The most important studies on the behaviour of six classes of pesticides in objects within the environment are described systematically and discussed. General schemes for the metabolism of certain pesticides in animal organisms, plants, and the soil are presented. The principal requirements which must be met by modern pesticides and the conditions for their safe use are characterised.**

The bibliography includes 250 references.

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I. Introduction

Nature began to be acted upon by human society in the period when human activity changed from food gathering to farming and cattle rearing. As a consequence of the vigorous development of industry in the XXth century, the pollution of the environment proceeded most vigorously. In a simplified way, the term environment is understood to imply the upper part of the lithosphere (in the main soil) together with the biosphere and cosmic space surrounding the terrestrial globe and acting upon vital processes. The biosphere comprises all living organisms, including animals, plants, microorganisms, as well as the residues from these organisms at different stages of their decomposition and conversion into the simplest organic and inorganic compounds. The atmosphere, the hydrosphere, and the soil are included here. An enormous number of organisms live in these spheres. For example, according to the data of different investigators, one hectare of soil contains 1000–7000 kg of bacteria, 100–1000 kg of microfungi, 10–300 kg of algae, 5–10 kg of protozoa, 1000 kg of arthropods, and 300–1000 kg of earthworms.[1]

In connection with the considerable pollution due to man's economic activity, the problem of the protection of the environment has become urgent in our time.[2–5]

The scale and nature of the pollution of the environment by various products of anthropogenic activity can be inferred even from the following examples: as a result of the combustion of coal and brown coal, about 3000 tonnes of mercury enter the atmosphere annually and, when account is taken of the waste from different branches of industry and medical services about 10000 tonnes of mercury enter the environment.[6–10] Since mercury is able to accumulate in living organisms and can be converted into the ethylmercury ion, which is highly toxic and carcinogenic, it constitutes a serious danger to man.[10–12]

Many marine and river organisms already contain considerable amounts of mercury and its concentration increases every year. In view of this, the use of mercury compounds as pesticides in economically developed countries has virtually ceased despite their considerable value.

A second important pollutant of the environment is lead, which is known to exhibit a psychotropic activity. Lead compounds enter the environment in large amounts with the exhaust gases of automobiles employing petrol containing lead compounds as anti-knock additives.[13] In addition, polyhalodibenzofurans and polyhalodibenzo-*p*-dioxins, which exhibit teratogenic and blastomogenic activities, affect the skin, and are acutely toxic,[14] as well as carcinogens such as benzopyrene and benzanthracenes[18] enter the environment with exhaust gases. According to the results of some investigators,[15] the amount of the above compounds formed per kilometre of highways is 1.5 μg. We may note that the use of lead anti-knock agents has continued in many countries to this day, as a result of which lead salts and many organochlorine compounds accumulate in the environment.

One should also note that the accumulation of heavy metals such as cadmium[8,15] and chromium[17] in the environment has likewise increased.

Nitrogen and sulphur oxides, which are the products of the combustion of various types of fuel containing sulphur, are fairly serious pollutants.[19] Polycyclic hydrocarbons with blastomogenic activity are known to be formed on combustion of wood, coal, and brown coal. We may note that tobacco smoke is also a pollutant, since it contains about 800 different substances, including extremely toxic aromatic and heterocyclic compounds.

The pollution of the environment by arsenic compounds is significant.[10,20,20a] Some of them were used until the end of the 1940's as agents combating agricultural pests and also for the protection of wood against rotting. Organic arsenic compounds also continue to be used as herbicides at the present time.[21] Arsenic compounds can penetrate into plants and can migrate via their vascular systems, which frequently leads to the pollution of the products of the cultivation of plants. The soil of many countries as well as the world ocean contain nowadays very large amounts of arsenic compounds.[20,22] Up to 8 million tonnes of petroleum and petroleum products enter the world ocean annually.[22]

Polychlorobiphenyls, used in the electrical industry, as plasticisers, and in other branches of industry, polychlorodibenzofurans and polychlorodibenzo-*p*-dioxins, formed mainly on chlorination of various products and on combustion of refuse and industrial waste are also pollutants.[23] The latter compounds are so stable that they withstand heating above 600 °C.[14] Certain polychlorodibenzo-*p*-dioxins are extremely toxic and exhibit teratogenic and carcinogenic

activities and affect the skin. Thus LD_{50} for 2,3,7,8-tetrachlorodibenzo-*p*-dioxin in relation to monkeys is only 70 μg kg^{-1}.[14] The high solubility of the above compounds in lipids leads to their accumulation in human and animal fatty tissue and also in human milk.[23,24] Polychloro-derivatives have been detected in human milk in many countries. All these compounds are extremely stable in objects within the environment and can persist for more than 10 years even under tropical conditions.

We may point out that about 55000 chemical compounds have now been detected in objects within the environment. According to our calculations, about 500 million tonnes of different products of anthropogenic activity are discharged into the environment annually.

Pesticides are physiologically active substances and therefore special requirements ensuring adequate safety in their practical application must be imposed on them. Altogether a little more than 1 million tonnes of pesticides are used annually. As the relevant data accumulate, the requirements which must be met by pesticides become more stringent. The fraction of pesticides in all the pollutants introduced by man into the environment is ~0.2%, which is less almost by a factor of 5 than the fraction of detergents.

Standards which must be satisfied by modern pesticides have been established in terms of the following parameters: persistence characterised by the half-life (or the total decomposition into simplest chemical compounds safe to man and to useful organisms); minimum chronic toxicity for man and animals; the absence of mutagenic, carcinogenic, and teratogenic activities;[25,26] the absence of negative effects when small doses of the preparations are consumed with food products by man and animals; the absence of cumulative activity; selectivity of action in relation to useful organisms; low toxicity for useful hydrobionts and many others.[27]

Together with acute and chromic toxicity and the existence of other remote effects resulting from the action of the preparation on man, animals, birds, and other useful organisms, the persistence of pesticides, which depends on their chemical stability and the rate of their metabolism in objects within the environment, is very important. The type of metabolism is also significant, since it determines the safety of the preparation.[28] The following formula has been proposed to compare the ecological stresses exerted by different preparations:[29]

$$ES = (C/LD_{50})H,$$

where ES is the ecological stress per hectare, C the norm set for the consumption of the preparation (g ha^{-1}), H the half-life (persistence) of the preparation in weeks, and LD_{50} the acute toxicity for the given organism. Naturally the above formula does not yield an absolute value, but it makes it possible to obtain comparative characteristics of individual preparations.

Despite the vigorous propaganda by the "greens" against the use of pesticides, the scale of their practical employment has continuously expanded, which is indicated by the data in Table 1. However, by virtue of the systematic improvement of the range of preparations, their tonnage has significantly decreased. This is associated with the fact that the new preparations, developed in recent years, have extremely low consumption norms which are tens of times and in certain cases hundreds of times lower than the consumption norms of the pesticides used previously. There has been not only a growth of the requirement for pesticides but also a decrease in the employment of obsolete preparations and preparations with large consumption norms and a high persistence. Thus there has been a sharp decrease in the requirement for all organochlorine insecticides and in the economically developed countries their use has ceased altogether. For example, DDT is used only in China, India, and in certain countries of Asia and Africa. The use of herbicides such as sodium trichloroacetate and dalapon has been reduced.

Table 1. The amounts of pesticides sold in 1972 and 1989 and the forecast for 1995 in millions of US dollars.[30]

Group of pesticides	1972	1989	1995 (forecast)
Herbicides	4160	9500	10940
Insecticides	4090	6200	7110
Fungicides	2525	4490	5055
Plant growth regulators	375	800	1080
Nematocides and fumigants	410	510	575
Total	11560	21500	24760

New insecticides with consumption norms up to 20 g ha^{-1},[31,32] herbicides with consumption norms of 10–50 g ha^{-1},[33–35] and fungicides with consumption norms up to 150 g ha^{-1} [36–38] have appeared and the use of insect pheromones has also accelerated,[39] which makes it possible to reduce significantly the consumption of insecticides.

Data on individual most important classes of compounds are presented below.

II. Chlorohydrocarbons

Among organochlorine insecticides, preparations such as DDT, hexachlorocyclohexane, aldrin, dieldrin, heptachlor, endrin, chlordan, mirex, kepone, endosulfan, and certain others have found practical applications in agriculture, industry, and the health service. DDT has been used on the largest scale. During 50 years (since the discovery of its insecticidal properties), approximately 5 million tonnes have been used. Hexachlorocyclohexane is employed on a somewhat smaller scale and preparations obtained by diene synthesis are used in still smaller amounts. Their accumulation in the environment is therefore significantly smaller than that of DDT.

Despite the negative attitude of many specialists to DDT, one cannot fail to note its positive role in health protection and in agriculture. In agriculture, DDT replaced extremely toxic arsenic compounds and in health protection measures it saved more than 15 million people from malaria and prevented epidemics of typhus, encephalitis, sleeping sickness, elephantiasis, the plague, tularemia, and others. However, one cannot close one's eyes also to the negative properties of this preparation, which include primarily a high persistence in a wide variety of objects in the environment and in living organisms. DDT can accumulate in the tissues of man, animals, birds, and fish.[50] In particular, DDT has been detected in human blood and fatty tissues,[40–45] in human[46,47] and cows'[48] milk, and in food products.[49]

The behaviour of organochlorine insecticides in aqueous ecosystems[51-54] and in the eggs and flesh of wild and domestic birds[54-56] has been studied. Table 2 presents data on the contents of DDT and its first metabolite—dichlorodiphenylethylene (DDE)—in the fatty tissues of inhabitants of different countries.[40]

Table 2 shows that the greatest amounts of DDT and DDE are present in the fatty tissues of the inhabitants of those countries where DDT is widely used. We may note that the content of DDT has diminished in the tissues of the inhabitants of those countries where its use has been stopped.

Table 2. The contents of DDT and DDE in the fatty tissues of the inhabitants of different countries (mg/kg of fat).

Country	Year	DDT	DDE
Belgium	1975	8.29	6.50
Great Britain	1976–77	2.6	2.1
Holland	1969	4.0	1.26
Denmark	1972–73	4.8	—
Zaire	1983	62.45	—
India	1982	5.43	5.48
Iran	1974–76	8.13	5.26
Italy	1981–82	0.8	7.35
Canada	1979–81	3.69	2.56
Mexico	1975	8.31–21.47	18.36
New Zealand	1973	6.36	5.34
Pakistan	1971	25.0	13.6
Poland	—	4.08	7.28
Romania	1983	3.33	—
USA	1972	6.88	9.58
USA	1970	7.88	5.03
Turkey	1984–85	7.12	5.83
Finland	1983	0.33	1.58
France	—	3.27	—
Federal Republic of Germany	—	1.1	4.4
Czechoslovakia	1980–81	12.33	8.74
Japan	1984	1.99	1.85

The bioconcentration of DDT in various objects in the environment has also been investigated in both natural and model experiments.[57,58] The bioconcentration of DDT in biological objects is to a large extent associated with the type of its metabolism,[59] in which hydrophobic metabolites, accumulating in lipids, are formed in the first place.

Hexachlorocyclohexane accumulates in much smaller amounts in the fatty tissues of man and in milk, which is associated not only with the smaller scale of its use but also with its significantly smaller persistence.[40] Among all the isomers of hexachlorocyclohexane, the β-isomer accumulates in greatest amounts. It is distinguished not only by a lower volatility but also by a greater chemical stability.

Endosulfan,[60] toxaphene,[61] and other organochlorine insecticides[40] accumulate in objects in the environment in much smaller amounts. In view of the sharp decrease in the use of organochlorine insecticides, their content in the environment is being gradually reduced.

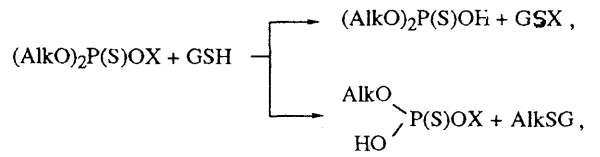
The effect of organochlorine pesticides on the soil microflora has been studied.[62-64] It has been established that, when aldrin and certain other insecticides are systematically applied, microorganisms are suppressed and the pesticide is gradually degraded.[64]

Despite the appreciable contamination of food products by organochlorine insecticides, in most cases their content in milk and other food products is within the limits of the norms established by the World Health Organisation.[65,66]

It is of interest that methoxychlor (I), whose metabolism leads to the formation of hydrophilic products, does not accumulate in the organisms of mammals and hardly contaminates milk.[67] Furthermore, methoxychlor is less toxic by an order of magnitude than DDT (for many arthropods, it is less toxic by only a factor of 1.5 than DDT).

$$CH_3OC_6H_4CHC_6H_4OCH_3$$
$$\quad\quad\quad

formation of diesters from triesters, interaction with glutathione transferase, for example[68]



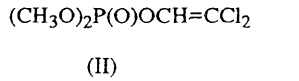
degradation of the ester part of the molecule containing the carboxy-group, and the formation of conjugates with carbohydrates in plants, with glucuronic and humic acids in the soil, and with sulphates in mammalian organisms.

Some of the reactions listed above are illustrated below by individual examples. We may note that the reactions of organophosphorus compounds depend not only on the structure of the phosphorus-containing part of the molecule but also on the structure of the organic groups linked to the phosphorus atom.[68]

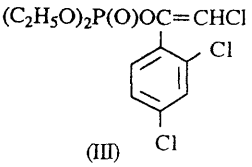
The methyl esters of phosphorus acids are comparatively effective alkylating agents, which also leads to their degradation and conversion into compounds with an extremely low toxicity.

About 100 organophosphorus pesticides are used nowadays on different scales.[69] In the present review, it is impossible to characterise the behaviour in the environment of all the organophosphorus compounds employed, so that only substances used on a fairly wide scale will be described.

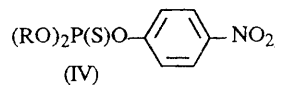
Among phosphoric acid esters these are dichlorophos (II) and chlorfenvinphos (III); among phosphorothioic acid esters these are metaphos (methylparathion) and thiophos (parathion) (IV), chlorpyrifos (V), diazinon (VI), fenitrothion (VII), fenthion (VIII); among phosphorodithioic acid derivatives these are phorate and terbufos (IX), phosalone (X), azinphos (XI), dimethoate (XII), thiometon (XIII), carbofos (malathion) (XIV), ninosan (XV), and also kitazin P (XVI), and acephate (XVII); phosphonates include chlorophos (XVIII) and glyphosate (XIX).



(II)

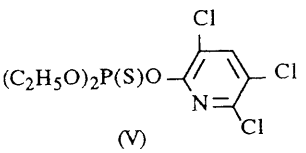


(III)

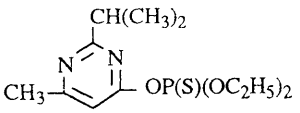


(IV)

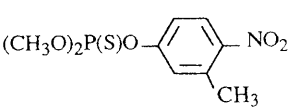
R = CH_3, C_2H_5



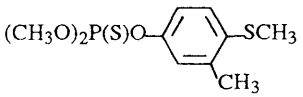
(V)



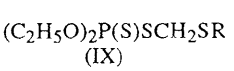
(VI)



(VII)

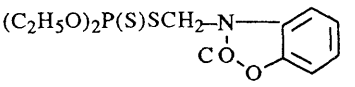


(VIII)

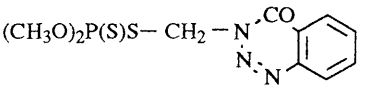


(IX)

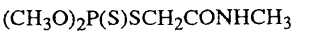
R = C_2H_5, $C(CH_3)_3$.



(X)



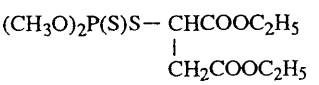
(XI)



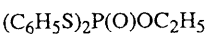
(XII)



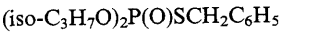
(XIII)



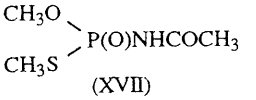
(XIV)



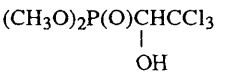
(XV)



(XVI)



(XVII)



(XVIII)



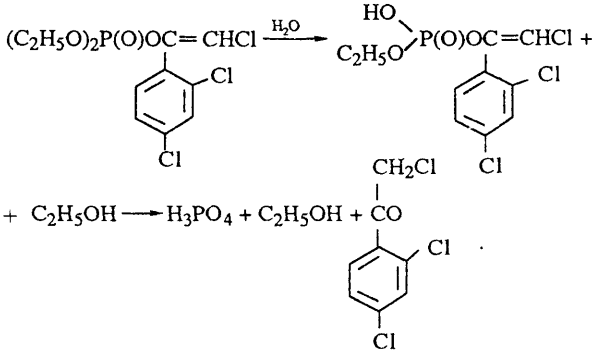
(XIX)

As stated above, the behaviour of pesticides in objects in the environment is influenced by numerous factors, including chemical and biological ones; the most important are the hydrolytic and oxidation reactions as well as the metabolism in various species of living organisms.

The first representative of the organophosphorus compounds listed above, namely dichlorophos, is a fairly labile substance and is readily hydrolysed by water even at comparatively low temperatures.[68,69] Thus in the tissues of the Atlantic salmon dichlorophos is not detected at all one day after entry at a water temperature of 12 °C, whilst at 4 °C only traces are found.[70] A similar picture has been observed also in milk: the preparation introduced into milk at 4–6 °C is not detected after 24 h.

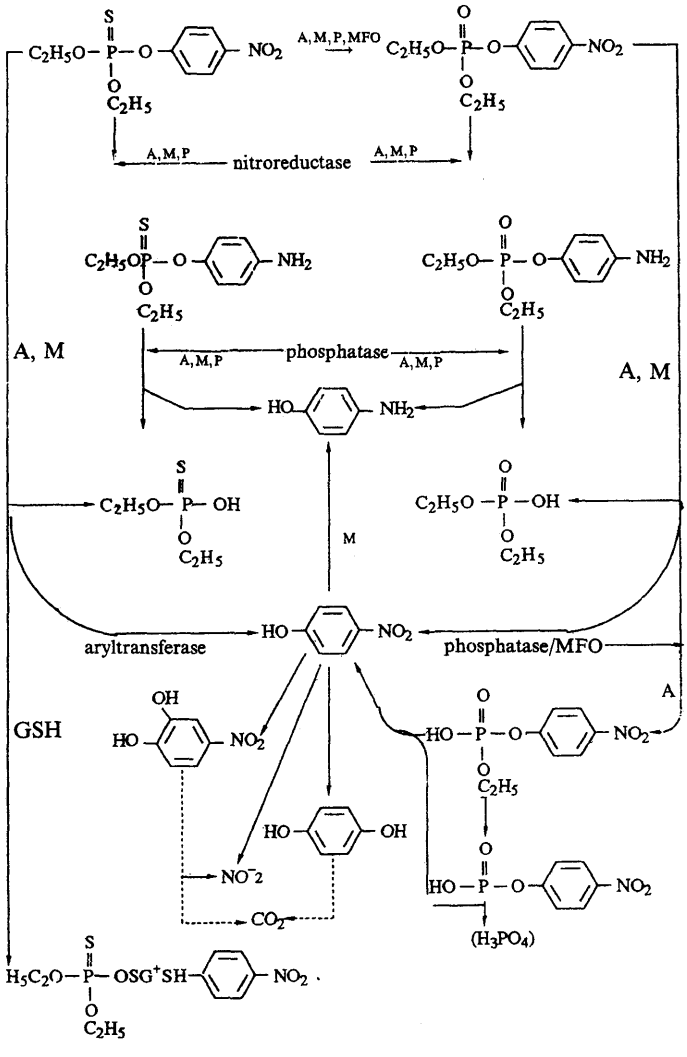
Dichlorophos decomposes to phosphoric acid and dichloroacetaldehyde and the latter is converted as a result of hydrolysis and oxidation into hydrochloric and oxalic acids.

Chlorfenvinphos decomposes in a more complex manner, forming on hydrolysis phosphoric acid and the lipophilic trichloro-acetophenone, which can accumulate in living organisms like other organochlorine compounds. For this reason, the application of this preparation is limited.



Parathion and methylparathion, whose behaviour in the environment has been studied in detail,[68,69,71–77] have found the greatest application in agriculture. The metabolism of parathion is different in different organisms, which can be clearly seen from Scheme 1.[68] We may note that the decomposition of methylparathion in the soil and other objects is significantly faster than that of parathion. Furthermore, it is less toxic and for this reason it has been successfully replacing parathion in recent years. The rate of decomposition of parathion and methylparathion depends not only on the nature of the soil, its moisture content, its content of organic substances, and the concentration of the insecticide, but also on the microflora, since different species of microorganisms will react with the preparations at different rates.[71,78]

Scheme 1

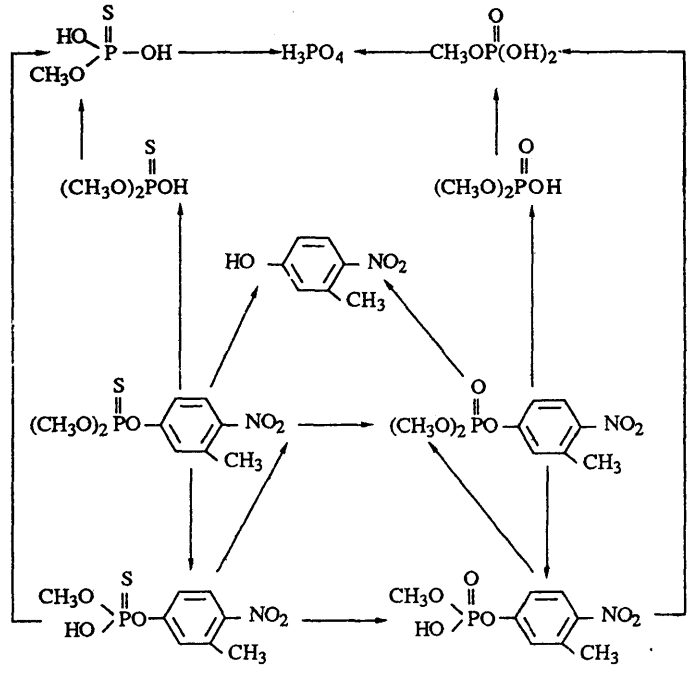


Scheme 1. Metabolism of parathion.
A = animals, M = microorganisms, P = plants, MFO = mixed function oxidases.

It is of interest to note that, in combination with toxaphene, polychlorcamphene, and oils, the duration of action of methylparathion on the cotton plant increases significantly,[77,79,80] which is apparently associated with the decrease in the rate of its hydrolysis in solutions in hydrophobic solvents such as the substances listed above. There are grounds for the assumption that a similar phenomenon may occur also in the soil. However, methylparathion decomposes in the soil in the course of 15–20 days, whilst parathion decomposes in 30–45 days, being fully converted into simple compounds (CO_2 and H_3PO_4 [71]).

We may point out that the accumulation of parathion and methylparathion in the organisms of mammals or hydrobionts has not been observed.[10,71] Special experiments on cows have shown that parathion administered in sublethal doses is almost completely eliminated from the organism in the course of 48 h.[10] The preparation does not remain in milk either.

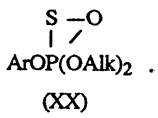
The closest homologue of methylparathion is fenitrothion, which exhibits a significantly smaller toxicity for animals compared with methylparathion.[81–83] The persistence of fenitrothion in ponds and bottom deposits,[84,85] in running water,[85] on leaves of the apple tree,[87] the poplar, and the birch,[86] and also its toxicity for fish[88] have been investigated. It has been shown that the persistence of fenitrothion is greater than that of methylparathion. The photolysis of fenitrothion[89,90] as well as its metabolism in vertebrates (mammals),[91] in the blue crab,[92] in the house fly which is resistant to the preparation,[93] and in the wheat plant,[94] as well as its oxidative dealkylation when acted upon by cytochrome P450 [95] have been studied. The general scheme illustrating the metabolism of phenitrothion in mammals is presented below (Scheme 2).



Scheme 2. Metabolism of fenitrothion in mammals.

In contrast to metabolism in mammals (rats), in which demethylation and dearylation take place, in the organisms of insects (the resistant house flies), the methyl group bound to the benzene ring is oxidised.[93] It may be that cytochrome P450 participates in this process. Many enzymes, including mixed function monoxidases, are involved in the degradation of insecticides of this type.

It has been suggested that the oxidation to oxones proceeds via a stage involving the formation of an unstable intermediate:



(XX)

We may note that soil microorganisms utilise in most cases the products of the metabolism of organophosphorus compounds as a source of carbon and phosphorus and that ultimately the majority of organophosphorus compounds decompose to CO_2 and H_3PO_4. The thione sulphur is converted into sulphuric acid, but the amount of the latter is so small that it has no practical importance as regards the acidification of the soil; in most cases the amount is only several tens of grammes per hectare.

The metabolism of fenthion is similar to that of fenitrothion,[68] but, in view of the presence of the sulphide group in the fenthion molecule, it is oxidised to a sulphoxide and a sulphone by the successive decomposition of the molecule by soil microorganisms. Fenthion is safe to a number of microorganisms and small invertebrates,[96] is moderately toxic for fish,[97] is moderately persistent in aqueous ecosystems,[98] and its toxicity is comparatively low for fungi and nitrogen-fixing bacteria and also for the little brown bat (*Myotis lucifugus*).[99]

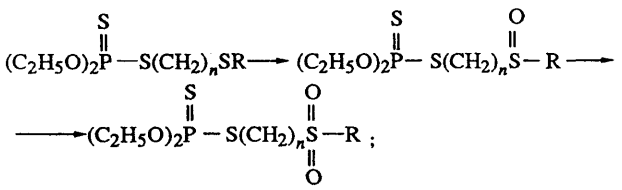
The persistence of diazinon is somewhat greater than that of the preparations described above.[100–106] Thus diazinon can be retained in organic soils for more than a year.[100] As a result of being washed out of the surface of the soil, it contaminates water bearing strata. It has been noted that diazinon is capable of being bioconcentrated by the aqueous fauna[101] but it is relatively rapidly eliminated from the organisms of fish. After 8 days, it cannot be found in the fish tissues. The metabolism of diazinon is even faster in warm blooded animals. It takes place in accordance with the general mechanism of the metabolism of phosphorothioates.[68,102,105] The principal reactions are oxidation and hydrolysis. Together with the formation of diazinonoxone derivatives, the side chains of the pyrimidine residue are oxidised at both the methyl and the isopropyl groups.[10,68,105] The main products of its metabolism in the soil are CO_2, H_3PO_4, and nitrogen. In plants diazinon also undergoes degradation. Thus, after cranberries have been treated with diazinon, the latter cannot be dectected in them after 42 days.[106]

Chlorpyrifos has similar properties.[107–118] In terms of its stability in the soil and in water, it approaches diazinon and in certain cases is superior to the latter.[107,108] The metabolism of chlorpyrifos involves, together with the hydrolysis of the ester linkages, the replacement of one or two chlorine atoms in the pyridine ring by the methylthio- and hydroxy-groups.[68,112] The hydrolysis of chlorpyrifos is fastest in an alkaline medium.[110]

It is of interest to note that phosphorothiolates (kitazin and hinosan) are hydroxylated in the aromatic ring in the course of their metabolism in plants (rice).[68]

In most cases the first product of the metabolism of organophosphorus insecticides forms conjugates with plant substances, which are sometimes fairly stable.[68] Such conjugates are obtained with carbohydrates and aminoacids. Acid sulphates are formed in animal organisms, being eliminated from them with products of their vital activity.

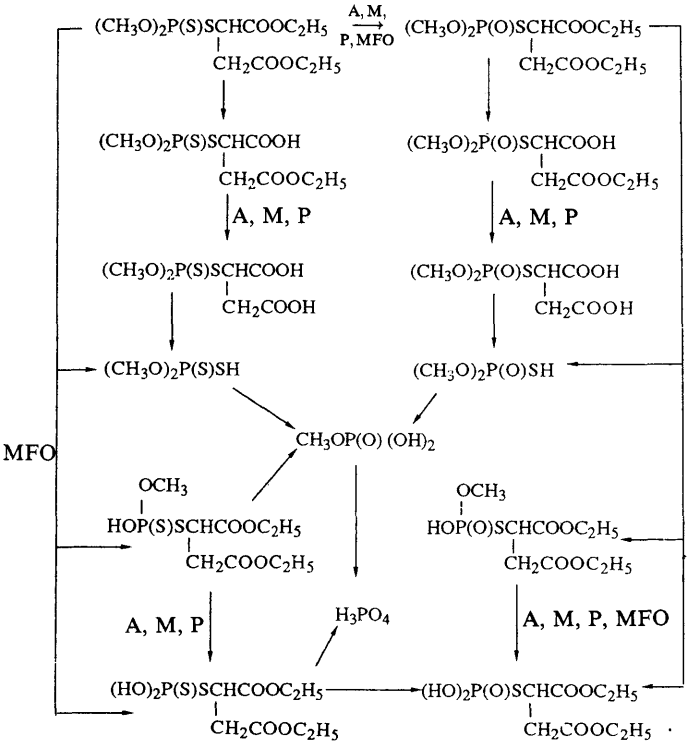
The behaviour of phosphorodithioic acid derivatives in objects in the environment depends to a large extent on the structure of the ester groups in the phosphorodithioate.[68] In this respect, the preparations phorate, terbufos, and disulfoton behave similarly.[119–125] Hydrolysis and oxidation of sulphide sulphur to the corresponding sulphoxides and sulphones are characteristic of these substances.



$R = C_2H_5$, $C(CH_3)_3$, $n = 1.2$.

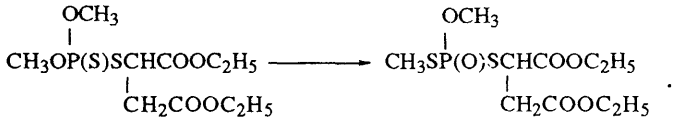
Together with the reactions described, oxidation of the thione sulphur and hydrolysis to phosphoric acid take place.[68,119] These preparations are used as soil insecticides.[121–123] Their complete degradation in the soil takes place in the course of 65–120 days, which depends to a large extent on the temperature and the nature of the soil. We may note that the preparations have a weak effect on the soil microflora.

Among phosphorodithioic acid derivatives, malathion (carbofos) has found the most extensive application as a contact insecticide.[71,126–135] A positive property of malathion is its low toxicity for mammals and its comparatively rapid decomposition in objects in the environment.[71] The metabolism of malathion is illustrated in Scheme 3.



Scheme 3. Metabolism of malathion.

Under the influence of certain inorganic substances and after prolonged heating, malathion may be converted into isomalathion, which is extremely toxic for mammals. It is sufficient to state that an admixture of 4% of isomalathion in malathion increases the toxicity of the preparation by an order of magnitude. The toxicity of malathion is also increased by an order of magnitude by an admixture of a side product of its manufacture—trimethyl phosphorodithioate.[126,131,132] The isomerisation of malathion to its toxic isomer led to the poisoning of humans in the course of its widespread application in the fight against the malaria mosquito in Pakistan.[71] The purity of the preparation used is therefore very important.

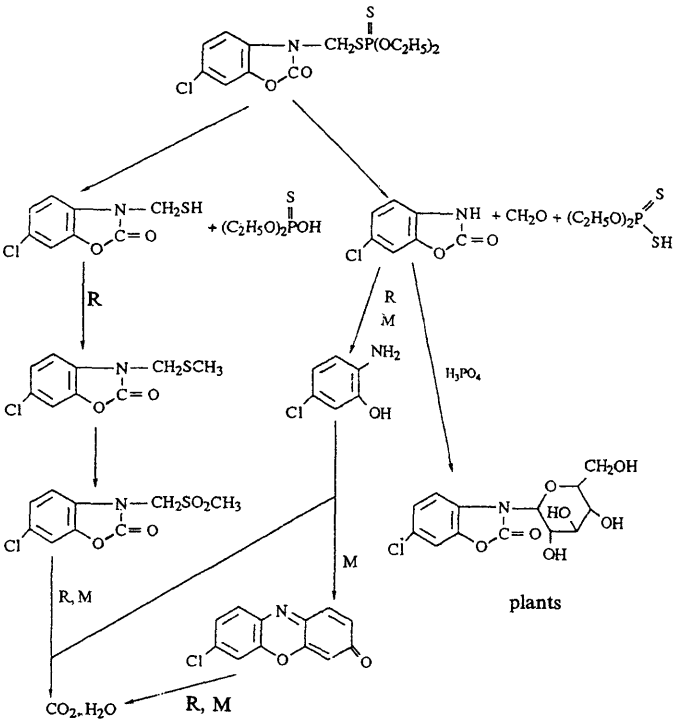


The rate of decomposition of malathion in objects in the environment is high and it vanishes rapidly from plants and animal organisms.[71,135] In the soil, malathion is also unstable and decomposes fairly rapidly. For example, in cherries, in red and black currants, and in other plants, malathion is hardly detected only 6–8 days after treatment with the preparation.[71]

Table 3. Toxicity of pyrethroid metabolites (LC_{50}) for hydrobionts (mg litre^{-1}).

Metabolite	LC_{50} (daphnia)	LC_{50} (fish)
3-(2,2-Dichlorovinyl)-2,2-dimethylcyclopropane-carboxylic acid	130	3
3-(2,2-Dibromovinyl)-2,2-dimethylcyclopropane-carboxylic acid	> 0.05	—
3-(2-Chloro-3,3,3-trifluoroprophenyl)-2,2-dimethyl-cyclopropanecarboxylic acid	100	16
3-Phenoxybenzyl alcohol	200	3–7
3-Phenoxybenzoic acid	85	13–36
2-(4-Chlorophenyl)-3-methylbutyric acid	—	10
3-Phenoxybenzaldehyde	> 0.025	—

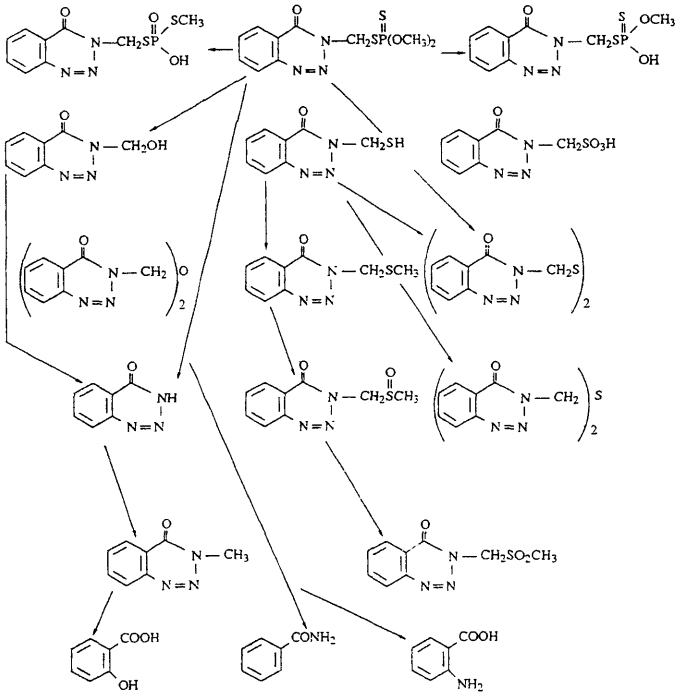
Phosalone persists in the environment for a longer time.[10] Thus it disappears from oak leaves on the twentieth day after spraying. During this period, it also decomposes in turf. Naturally, the phosalone decomposition time depends on the temperature and humidity. The higher the temperature, the faster the decomposition of the preparation. Phosalone decomposes in the soil over approximately the same period. The rate of its decomposition depends greatly on the composition and nature of the soil. However, in most cases part of the pesticide is bound firmly to the soil colloids and is included in the metabolism of organic substances.[136,137] The general direction of the metabolism of phosalone is illustrated in Scheme 4.[10,27,138]



Scheme 4. Metabolism of phosalone.
R = rats; M = microorganisms.

The final products of its metabolism in the soil under the influence of microorganisms are CO_2, H_3PO_4, N_2, etc.

Azinphos-methyl and especially azinphos-ethyl are more stable in objects in the environment. They have found fairly extensive applications in the fight against many plant pests. As can be seen from Scheme 5, even a relatively stable insecticide, such as azinphos-methyl, is almost completely decomposed by soil microorganisms.[139]



Scheme 5. Metabolism of azinphos-methyl.[139–143]

Other organophosphorus insecticides are less stable and are rapidly degraded to simple compounds.[10,27,71,68]

Among phosphonates, glyphosate, which is used nowadays in the fight against weeds, has found the most extensive application in agriculture. Numerous communications have been published on the properties of glyphosate and its behaviour in objects in the environment.[144,145]

Like all organophosphorus compounds, glyphosate decomposes almost completely in objects in the enviornment in the course of one vegetation period.[144] The decomposition products are phosphoric acid, CO_2, and other simple compounds.

It is seen from the data presented that among organophosphorus pesticides there are substances with different persistences, and from the necessary range of preparations available, it is possible to select the best substances for use under specific conditions.

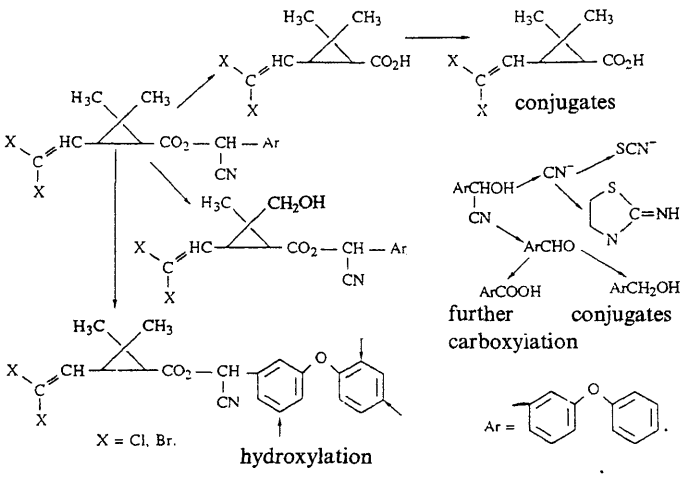
IV. Synthetic pyrethroids

The manufacture and use in agriculture of synthetic pyrethroids began only in the 1970's but at the present time they occupy a firm position in the range of modern insecticides. More than 15 preparations have found practical applications, the most important of which are permethrin, cypermethrin, and decis.

A positive property of this group of compounds is their relatively moderate toxicity for mammals and birds and also their low consumption norms, not exceeding 10–20 g ha^{-1} for a number of preparations.[146,147] Their comparatively high toxicity for hydrobionts, and in the first place for fish, must be included among the disadvantages of this group of substances.[148] Since synthetic pyrethroids are esters, they are relatively readily hydrolysed. The hydrolysis is faster in an alkaline medium. Synthetic pyrethroids decompose even more rapidly under the influence of soil microorganisms and also in animal organisms.[149,150] The rates of decomposition (%) of certain synthetic pyrethroids over a period of 8 weeks in mineral and organic soils are given below.[150]

	Permethrin	Cypermethrin	Fenvalerate	Decis
Mineral soil	94	96	88	48
Organic soil	84	84	42	26

Naturally not only the soil composition but also temperature and humidity are important for the decomposition of pyrethroids. The higher the temperature, the faster the decomposition of the preparations. The metabolism of cypermethrin and decis is indicated in Scheme 6.



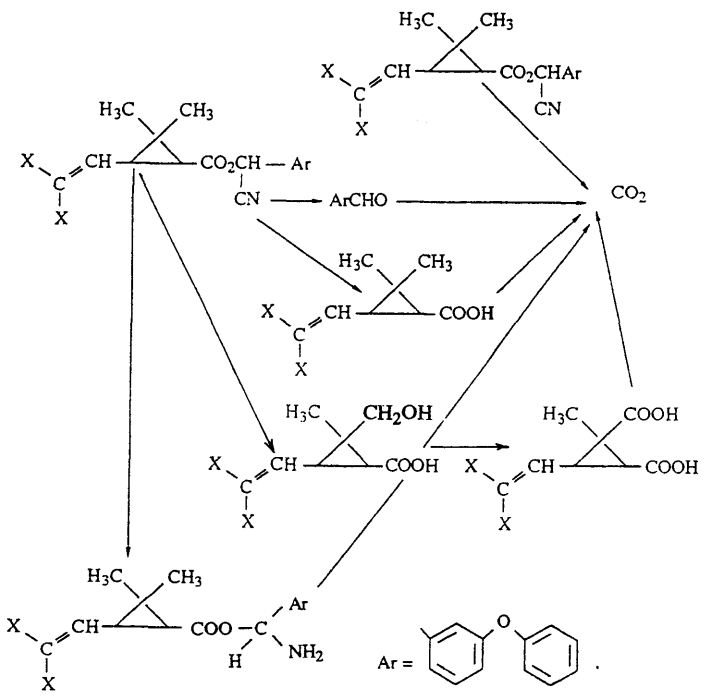
Scheme 6. Metabolism of cypermethrin and decis in animal organisms.

Since the products of the metabolism of pyrethroids are readily soluble in water, they do not accumulate in animal organisms, are rapidly eliminated with the products of their vital activity, and synthetic pyrethroids do not accumulate in milk.[148] The products of the metabolism of synthetic pyrethroids are much less toxic than the initial compounds for the majority of organisms, including hydrobionts.[151,152] Table 3 indicates the toxicity of certain metabolites of synthetic pyrethroids for hydrobionts.

For comparison we may point out that in the case of fish LC_{50} after exposure for 96 h is 0.0004–0.0007 mg $litre^{-1}$ for permethrin, 0.0004–0.002 mg $litre^{-1}$ for cypermethrin, 0.002 mg $litre^{-1}$ for fenvalerate, and 0.001 mg $litre^{-1}$ for decis.[151]

The metabolism of synthetic pyrethroids in plants also yields conjugates with glucose and other carbohydrates whose toxicity for mammals is low. However, these conjugates are incorporated in the general plant metabolism and their amount gradually diminishes.

In the soil, pyrethroids are fully decomposed to simple compounds, as shown in Scheme 7.[151]



Scheme 7. Metabolism of cypermethrin and decis in the soil.

It must also be pointed out that pyrethroids undergo photochemical transformations in the atmosphere and in an aqueous medium with almost complete degradation of the molecule.[149]

Analogues of cypermethrin and other pyrethroids which do not contain the phenoxybenzoyl fragment have now been synthesised. The study of the metabolism of the new groups of pyrethroids is being continued.[153]

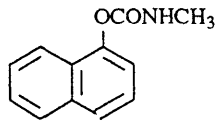
It is seen from the foregoing that synthetic pyrethroids are of great practical interest, particularly bearing in mind their extremely low consumption norms.

V. Carbamic acid derivatives

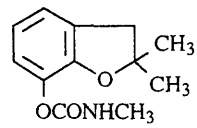
More than 40 carbamic acid derivatives have now found applications as pesticides in agriculture. They include mainly derivatives of *N*-methylcarbamic acid as insecticides[69] and arylcarbamic acid esters as herbicides.

Carbamic acid derivatives include compounds with different acute toxicities and different persistence. Despite their high toxicity, many such preparations have found fairly extensive applications in agriculture. Not being able to consider in detail all the preparations which have found practical applications, we shall examine certain typical examples.

The behaviour of carbamic acid derivatives in objects in the environment depends not only on the specific compound but also on the object in which the preparation is present. The metabolism of carbamic derivatives is most complete when the latter are acted upon by soil microorganisms.[154–165] Soil microorganisms also decompose stable preparations such as carbaryl (XXI) [154–156] and carbofuran (XXII).[157–165]

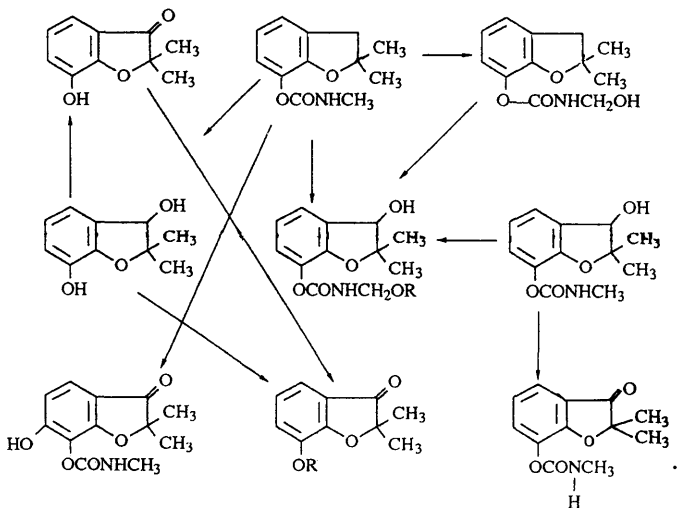


(XXI)



(XXII)

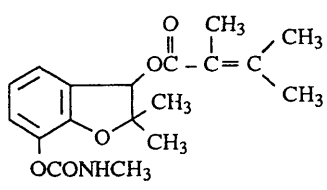
The main metabolic reactions of the carbamate insecticides are hydrolysis and oxidation.[155] The hydrolysis of the aryl esters of methylcarbamic acid leads to the formation of the corresponding phenol or other hydroxy-compound and methylcarbamic acid, which decomposes to an amine and CO_2.[155] This is accompanied by simultaneous oxidation processes which involve the methyl group and the aromatic or heterocyclic ring. Hydroxy-compounds form conjugates with organic substances (for example, humates) in the soil, with glucose in plants, and with sulphates and glucuronic acid in animal tissues. Scheme 8 presents the metabolism of carbofuran as an example.



R = glucose, glucuronic acid, or sulphate.

Scheme 8. Metabolism of carbofuran.

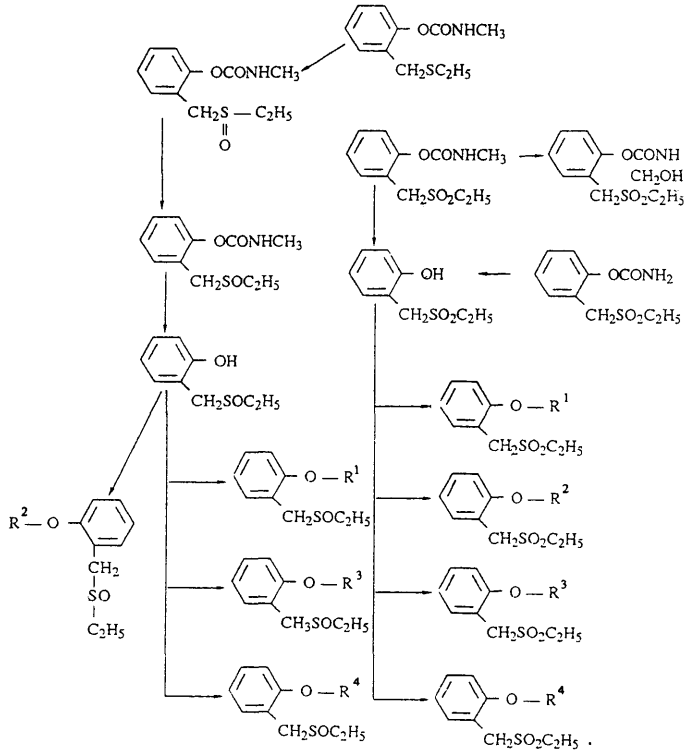
We may note that after the first stages of the metabolism shown in Scheme 8 the aromatic ring is hydroxylated and this is followed by further conversion into simple compounds. Conjugation with glucose in the 3-position takes place in plants. The carbofuran metabolites have been found in peppermint,[166] carrots,[167] ground nuts, and the cotton plant.[168] The 3-ester between carbofuran and angelic acid (XXIII) has been detected in the carrot.[167]



(XXIII)

It is of interest that the carbofuran 3-glucoside, obtained from carbofuran in plants, gives rise to glucuronides and the sulphates of the hydroxylated carbofuran metabolites in the rat organism.[169]

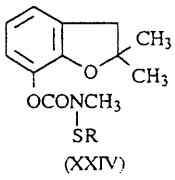
The metabolism of croneton may serve as a second example.[155] Like other preparations, croneton is fully decomposed in the soil, while in other objects it affords the products illustrated in Scheme 9.



R^1 = glucuronate, R^2 = glucoside, R^3 = condensation product, R^4 = sulphate.

Scheme 9. Metabolism of croneton.

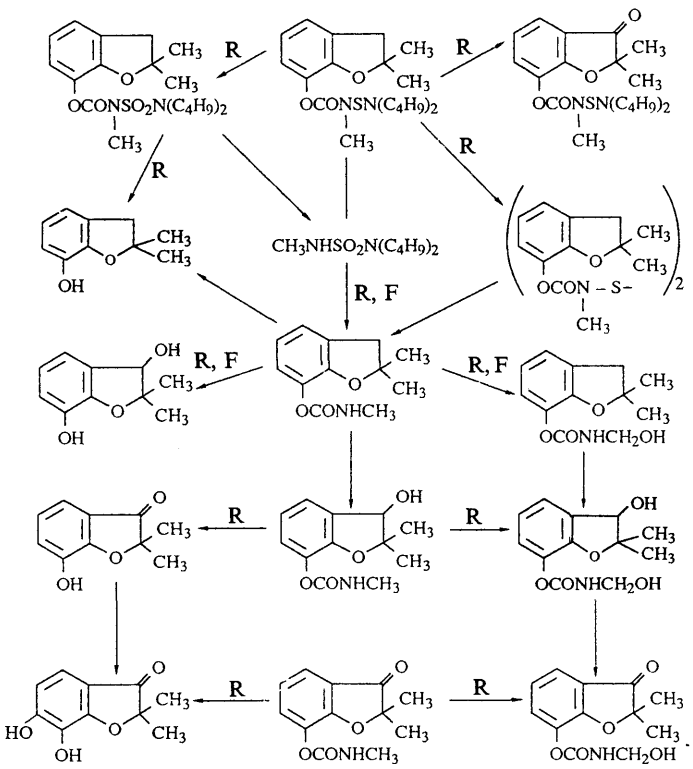
In view of the valuable properties of carbofuran as a pesticide, a new group of preparations with the general formula (XXIV) has been devised. These exhibit moderate insecticidal properties but are converted into carbofuran in the soil or in the insect organism.[170] This group of preparations has been called propesticides. Carbosulfan, promecarb, benfuracarb, and a number of others have found nowadays practical applications as insecticides. The study of the metabolism of this group of compounds showed that their metabolism in insect organisms differs somewhat from that in warm blooded animals.[170–177]



(XXIV)

R = $N(C_4H_9)_2$ (carbosulfan), $N{-}COOC_2H_5$ (promecarb), $N{-}CH(CH_3)_2$, $CH_2CH_2COOC_2H_5$ (benfunacarb).

Thus the metabolism of carbosulfan is different in the rat and fly organisms (Scheme 10). A greater amount of toxic substances is formed in flies, which results in an increase in the selectivity of the action of the preparation. A similar picture is observed also for other compounds of this type. The creation of propesticides is an example of the way in which an approach may be made to the creation of selective preparations on the basis of the study of metabolism in different species of living organisms.



Scheme 10. Metabolism of carbosulfan in the organisms of rats (R) and flies (F).[171]

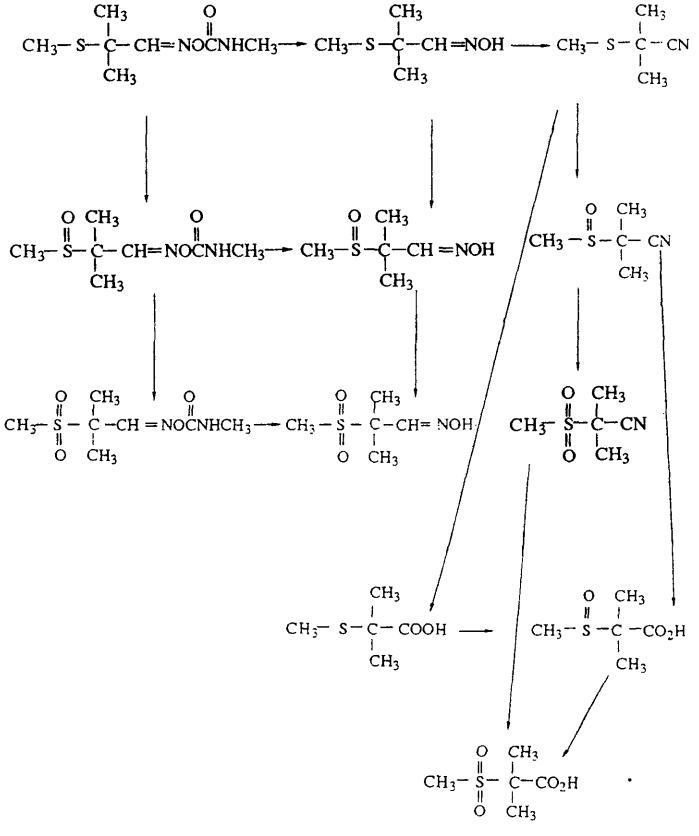
Carbosulfan is one of numerous examples. Such selective action has been observed not only in insecticides but also in herbicides and fungicides.

An important group of carbamate insecticides, which has found fairly extensive application in agriculture, is that of carbamate oximes, the commonest representative of which are aldicarb, aldoxicarb, oxamyl, and methomyl. However, the substances in this group are extremely toxic[178] and fairly stable in objects in the environment.[179–185]

One of the most toxic and stable preparations is aldicarb. In addition, its metabolites—the sulphoxide and the sulphone—are likewise highly toxic. The latter, called aldoxycarb, has found practical application as a soil insecticide. Aldicarb and its sulphoxide and sulphone are fairly stable and, when used systematically, can accumulate in the soil and can enter ground waters.[179,181–184] Even in dilute solutions, their hydrolysis is fairly slow.[186] It is of interest to note that the hydrolysis in chlorine water is faster (by 80% in 20 h).[185] The accumulation of aldicarb and of its toxic metabolites in the radish,[187] in the leaves and roots of sugar beet,[188–190], the potato, the pumpkin,[191,192] and a number of other

cultivated plants has been noted. It has been established that aldicarb and its metabolites persist in soil for up to 400 days after the introduction of the preparation in an amount of 15 kg ha^{-1},[181] but after only 10 days it is converted into the sulphoxide and the sulphone.[181] On the other hand, the sulphoxide is 99% converted into the sulphone after 120 days, the amount of sulphone by that time constituting more than 20% of the aldicarb introduced initially.[181] It has been shown with the aid of the ^{14}C-labelled preparation that ultimately aldicarb decomposes in the soil to CO_2 and other simple compounds.

The general aspects of the metabolism of aldicarb in various objects are presented in Scheme 11.[178,181,182,189] The use of aldicarb and similar preparations requires much caution and the maintenance of techniques and conditions ensuring the safety and protection of the environment. Bearing in mind the possibility of the accumulation of aldicarb and its toxic metabolites in ground waters, the use of this preparation in a number of states of the USA has been restricted. Under aerobic conditions, aldoxycarb was found to decompose in the soil in Florida after 42 days.[193]



Scheme 11. Metabolism of aldicarb.

Aldicarb can decompose photochemically with formation of methylamine, dimethyl disulphide, tetramethylsuccinodinitrile, dimethylurea, 2-methylthioisobutyronitrile, methacrylonitrile, and certain other products.[194] The aldicarb molecule undergoes almost total degradation under the influence of light. Such decomposition is possible, in particular, in an aqueous medium. Carbamate oximes such as methomyl and oxamyl are less stable and decompose faster in the environment.[155,195–201]

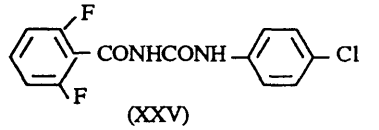
Despite the lower stability of the carbamate oximes enumerated above, work involving them requires much care and caution. This group of compounds is apparently one of the most dangerous among pesticides. However, their complete decomposition in the soil over one year does not give grounds for fear of contamination over any kind of long period.

VI. Urea derivatives

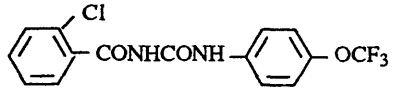
Among urea derivatives, four groups of substances have found practical application as pesticides: aryldimethylureas, arylmethoxymethylureas, benzoylureas, and sulphonylureas.[27,32,35,69]

Since the behaviour of the first groups of urea derivatives in objects within the environment has been fairly adequately described in our literature,[10,202,203] they will not be considered in this review.

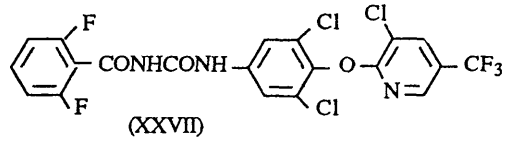
Insecticidal benzoylureas and herbicidal sulphonylureas have been vigorously investigated in recent years.[34,35,204] Among the insecticidal benzoylureas, diflubenzuron (XXV), triflumuron (XXVI), chlorfluazuron (XXVII), triflubenzuron (XXVIII), and certain others[204] have found practical applications.



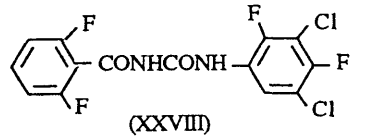
(XXV)



(XXVI)



(XXVII)



(XXVIII)

The insecticidal activity of compounds of this group is based on the inhibition of the formation of chitin during the development of insects and hence their destruction. The decomposition of these substances begins with hydrolysis accompanied by the elimination of the benzoyl group.[204–209] This may be accompanied by the simultaneous hydroxylation of the aniline ring. Next, arylurea decomposes with formation of aniline, which subsequently fully decomposes in the soil.[211]

Experiments with a carbon-labelled preparation showed that the decomposition, for example in the cotton plant, takes place over a period of two weeks.[208] The decomposition is most complete in the soil under the influence of microorganisms. In animal organisms, most of the preparation is eliminated with excrement[205] and virtually none enters milk.

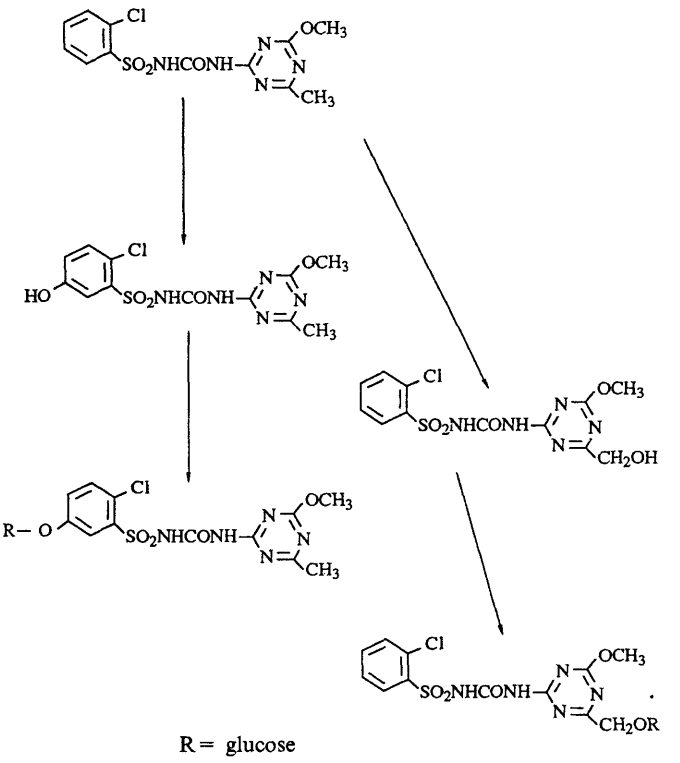
This group of compounds is of great practical interest, since they do not accumulate in objects within the environment and hydrolyse fairly rapidly in water and in the soil. A positive property of these substances is also the high selectivity of their action and low consumption norms (20–50 g ha^{-1}).

The herbicidal properties of sulphonylurea were discovered about 15 years ago and more than 15 preparations have now found practical applications. They are distinguished by a high selectivity of their action and low consumption norms when used to combat weeds, which is associated with their different metabolism in different plant cultures.[210] In plants resistant to sulphonylureas, their metabolism is fairly rapid and terminates after several hours,[210,212–215] whereas in plants sensitive to these compounds the metabolism is extremely slow.

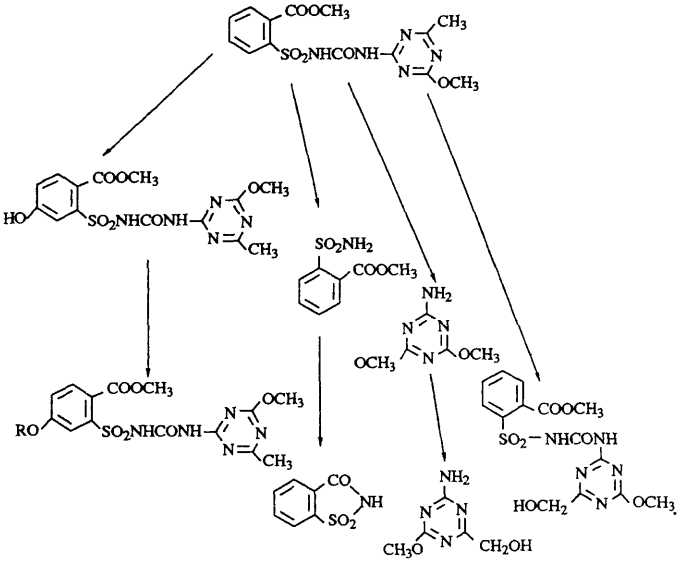
Table 4 presents certain sulphonylureas which are used as herbicides and which have found industrial or experimental industrial applications.

In the series of herbicidal sulphonylureas there are compounds with different persistence. These preparations range from those decomposing in the soil in several hours to those whose decomposition can occur over several months and even more than a year. An example is provided by chlorsulfuron. A plant culture sensitive to this compound may be damaged even a year after the preparation has been used (for a consumption norm up to 20 g ha^{-1}). A plant culture of this kind is, in particular, sugar beet.

The behaviour of a large number of derivatives of sulphonylurea in soil,[225–234] in various plant species,[210,219,235–239] and in animal,[219] bird, and fish organisms has now been studied in detail. It has been shown that the duration of their storage in the soil and their migration along the soil profile depend on the temperature and moisture content of the soil, on the pH of the medium, and on the composition of the soil. Certain sulphonylureas decompose in the soil in several hours. However, such decomposition takes place only as far as the hydrolytic stage and leads to the loss of herbicidal activity of the compound, while the complete decomposition of the majority of preparations in the soil takes place over a period of 15–30 days.[233,234] The complete decomposition of sulphonylureas in the soil by microorganisms takes place with total degradation of the molecule, while in plants a series of products non-toxic to the given plant are formed, as can be seen from Schemes 12 and 13 for the metabolism of chlorsulfuron in wheat and metsulfuron methyl in wheat and barley.[210,219]



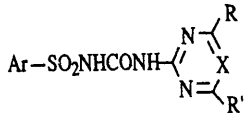
Scheme 12. Metabolism of chlorsulfuron in wheat.



Scheme 13. Metabolism of metsulfuron methyl in the wheat and barley plants.

Scheme 14 presents the metabolism of thifensulfuron in the rat organisms[219] and Scheme 15 illustrates the metabolism of bensulfuron in the rat and goat organisms.[219] We may note that significant amounts of the preparations were not detected in the milk of the animals.

Table 4. Certain herbicidal sulphonylureas



which have found practical applications.

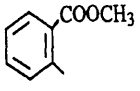
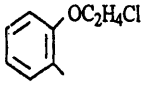
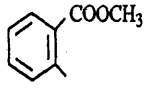
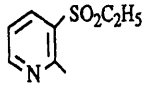
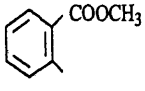
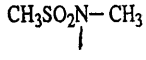
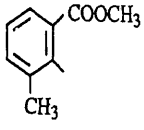
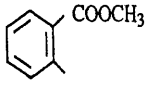
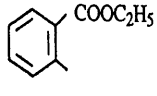
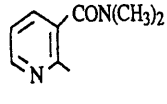
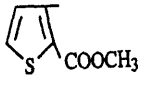
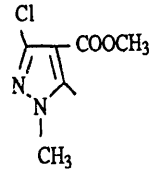
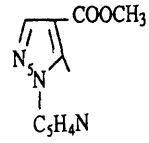
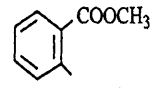
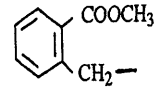
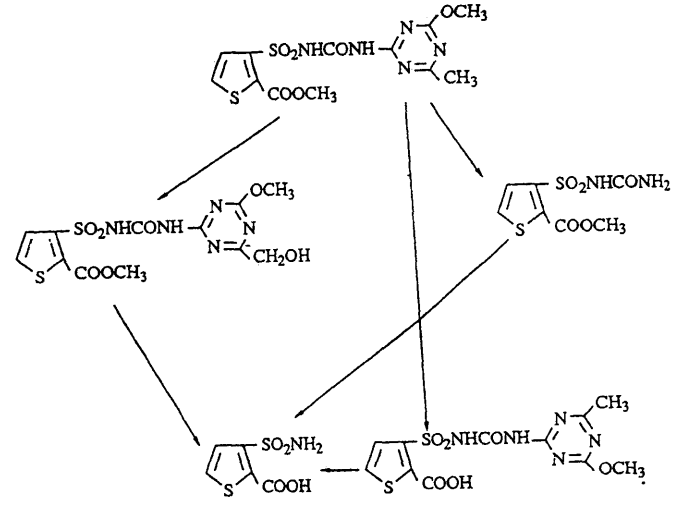
Ar	R	R′	X	Name	LD_{50} for rats, mg kg^{-1}	Consumption norm, g ha^{-1}	Refs.
	CH_3	OCH_3	N	Chlorsulfuron	5000	4–20	[216]
	CH_3	OCH_3	N	Metsulfuron methyl	5000	5–20	[217]
	CH_3	OCH_3	N	Triasulfuron	5000	10–20	[218]
	CH_3	OCH_3	CH	DPX-H6564	5000	20–50	
	OCH_3	OCH_3	CH	DPX E-9636	5000	20–40	[219]
	CH_3	CH_3	CH	Sulometuron methyl	5000	10–20	
	OCH_3	OCH_3	CH	Amidosulfuron	5000	40–60	—
	OCH_2CF_3	$N(CH_3)_2$	N	DPX-66037	5000	10–20	[221]
	$OCHF_2$	$OCHF_2$	CH	Primisulfuron	5000	10–30	—
	OCH_3	Cl	CH	Chlorimuron	4100	10–30	[214]
	OCH_3	OCH_3	CH	Nicosulfuron	5000	15–30	—
	CH_3	OCH_3	N	Thifensulfuron	5000	15–20	[222]

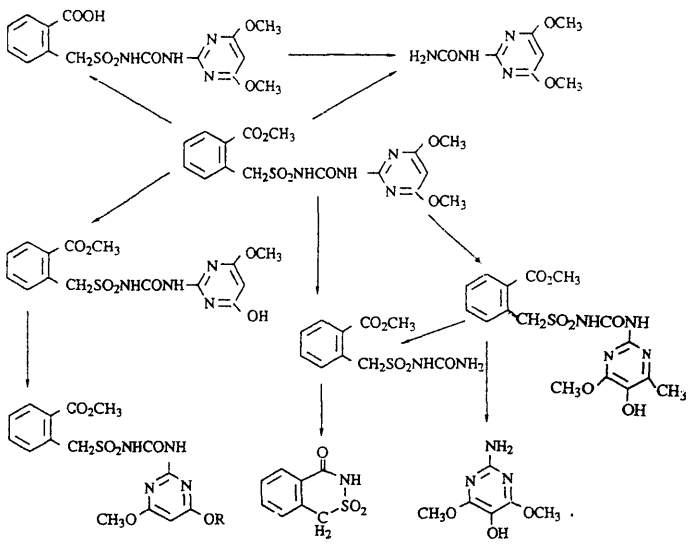
Table 4 (contd.).

Ar	R	R′	X	Name	LD_{50} for rats, mg kg^{-1}	Consumption norm, g ha^{-1}	Refs.
	OCH_3	OCH_3	CH	NC-319	8800	20–30	[223]
	CH_3	CH_3	CH	NC-330	5000	20–30	[224]
	$NHCH_3$	OC_2H_5	N	Cabbadi *	5000	20–40	—
	OCH_3	OCH_3	CH	Bensulfuron-methyl	5000	20–75	—

*Tentative spelling (Ed. of Translation).



Scheme 14. Metabolism of thifensulfuron in the rat organism.



R = glucuronide

Scheme 15. Metabolism of bensulfuron in the rat and goat organisms.

We may note that, together with individual sulphonylureas, their mixtures with other herbicides, including 2,4-D salts, benzoic acid derivatives, and a number of other substances, have found extensive applications. The use of such mixtures makes it possible to reduce significantly the consumption of the preparations and to diminish the amount of herbicide applied per unit treated area, accelerating thereby its total decomposition in the soil. In addition, the use of mixtures makes it possible to reduce the amount of preparations which may enter ground waters. We may note that much attention is being devoted to this problem at the present time because certain preparations do enter ground waters when applied systematically on the same areas (like aldicarb).

The products of the metabolism of sulphonylureas are eliminated from animal organisms in most cases with excrement in the form of glucuronates and sulphates.

The photochemical decomposition of a series of sulphonylureas,[219] including sulphometuron, has been investigated in an aqueous medium.[236,237] It has been established that the main products of the photochemical conversion of sulphometuron are hydrolysis products and two other substances of unknown structure.[237] It is noteworthy that the pyrimidine and triazine rings forming part of the molecule of the majority of sulphonylureas are apparently fairly stable and decompose only slowly. Table 4 shows that virtually all sulphonylureas contain either triazine or pyrimidine rings, whose decomposition has been thoroughly investigated in relation to triazine herbicides.[240–247]

It is seen from the foregoing that sulphonylurea derivatives are of significant interest for use in agriculture and, by virtue of their low toxicity for animals, fish, and birds, they constitute no danger to the environment, particularly since the consumption norms for the majority of these substances are extremely low, as shown by the data in Table 4.

Table 5. The maximum permissible contents of pesticides in drinking water (μg litre^{-1}).

Preparation	CIS (1984)	Australia (1987)	England (1986)	Canada (1987)	USA (1983)
Acephate	—	20	—	—	—
Acifluorfen	—	—	—	—	9
Alachlor	0.3	3	—	—	0.15–1
Aldicarb	—	—	—	9	10
Atrazine	2	—	30	60	3
Benomyl	—	200	—	—	—
Bentazon	25	400	—	—	17.5
Bioresmethrin	—	60	—	—	—
Butilate	—	—	—	—	50
Carbaryl	—	60	—	90	700
BMC	—	200	3	—	—
Carbofuran	—	30	—	90	36
Carboxin	—	—	—	—	700
Cyanazine	—	—	—	10	9
2,4-D	100	100	—	100	70
DDT	1	3	—	30	—
Diazinon	—	10	—	20	—
Dimethoate	—	100	7	20	—
Diuron	—	40	—	150	14
Endosulfan	—	40	—	—	—
Fenitrothion	—	20	—	—	—
Fenvalerate	—	40	—	—	—
Fluometuron	—	100	—	—	90
Glyphosate	—	200	70	280	700
Lindane	3	100	—	4	0.0265
Linuron	—	—	2	—	—
Malathion	—	100	—	190	—
Mancozeb	—	—	1	—	—
Maneb	—	—	1	—	—
Methromyl	—	60	—	—	175
Metalachlor	5	800	—	50	10
Molinat	7	1	—	—	—
Metaphos	—	6	—	7	2
Permethrin	—	300	—	—	—
Propanide	175	1000	—	—	—
Simazine	17	—	30	10	35
TMTD	—	30	—	—	—
Triadimefon	—	—	30	—	—
Trifloralin	170	500	—	—	2
Zineb	—	30	—	—	—

VII. Conclusion

The behaviour of pesticides in the environment has been considered in relation to several classes of chemical compounds. Analysis shows that, having at one's disposal fairly extensive knowledge, it is possible to ensure, by setting standards for the consumption of pesticides in agriculture and other fields, for example in the fight against agents transferring infectious diseases in man and animals, the use of pesticides which are completely safe both to man and a wide variety of objects in the environment can be ensured.

In order to characterise the possible contamination of food products and drinking water by pesticides, the maximum permissible levels of certain pesticides in drinking water and in food products, calculated per kilogram of human body weight per day, are

presented in Tables 5 and 6, which show that pesticides can enter the human organism in insignificant amounts. For example, if one considers diazinon, then it is found that only 0.07 mg of this substance enters the organism of a human being weighing 70 kg in 24 h, which is several times less than the amount of caffeine consumed with one cup of coffee and yet the toxicity of caffeine is approximately twice as high as that of diazinon.

Table 6. The maximum permissible levels of residual amounts of pesticides in food products (mg/kg of weight of a person per day).

Preparation	WHO	USA	CIS
Acephate	0.03	0.004	—
Acifluorfen	—	0.013	—
Alachlor	—	0.01	0.002
Aldicarb	0.005	0.0013	—
Atrazine	—	0.005	0.004
Benomyl	0.02	0.05	0.02
Bentazon	—	0.0025	0.002
Butilate	—	0.05	0.02
Carbaryl	0.01	—	0.01
BMC	0.01	—	0.01
Carbofuran	0.01	0.005	0.01
Carboxin	—	0.1	—
Cartap	0.1	—	—
Chlorthalonil	0.003	0.015	0.03
Cyanazine	—	0.002	—
Cyhalothrin	0.02	0.005	—
Cypermethrin	0.05	—	0.003
2,4-D	0.3	0.01	0
DDT	0.02	0.0005	0.0005
Deltamethrin	0.01	—	0.0003
Diazinon	0.002	—	0.001
Dienzoquat	—	0.08	—
Dimethoate	0.01	0.0002	0.01
Diuron	—	0.002	—
Endosulfan	0.008	0.00005	—
	—	0.00001	—
Fenitrothion	0.005	0.025	0.003
Fenvalerate	0.02	—	0.034
Ferbam	0.02	—	—
Fluometuron	—	0.013	0.03
Folpet	0.01	0.1	0
Fonophos	—	0.002	—
Glyphosate	0.3	0.1	0.01
Lindane	0.01	0.0003	0.01
Linuron	—	0.002	0.01
Malathion	0.02	0.02	0.02
Mancozeb	0.05	—	0.005
Maneb	0.05	0.005	—
Methomyl	0.01	0.025	—
Metalachlor	—	0.15	0.02
Metribuzin	—	0.025	0.004
Molinat	—	0.022	0.01
Metaphos	0.02	—	0.001
Permethrin	0.05	0.05	0.035
Phenmedipham	—	0.04	0.08
Phosalone	0.006	—	0.006
Prochloraz	0.01	0.0075	—
Propanide	—	0.005	0.04
Propiconazole	0.03	0.013	0.04
TMTD	—	0.005	0.005
Triadimefon	0.03	0.025	0.03
Zineb	0.05	0.05	0.02

A similar situation is also observed for the majority of other preparations. Despite the vigorous studies designed to create alternative methods for the protection of plants against pests, diseases, and weeds, the chemical method will undoubtedly occupy a dominant position in the foreseeable future in the integrated system for the protection of plants.[249]

When the modern assortment of pesticides is used intelligently, the latter present no danger either to man or to the environment in view of their moderate persistence and extremely low consumption limits. For example, monitoring of the content of pesticides in California (USA) in 1989 failed to detect pesticides in 80% of the samples, in 19.3% of the samples pesticides were found within permissible limits, and in only 0.7% of the samples was the content of pesticide in excess of the permissible limits.[250]

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