

I do not feel qualified to judge whether the usual soil classifications like sandy loam, silt loam, etc. already offer an adequate possibility for grouping. But if we had such a grouping of soils, the pesticides could be assigned to these groups according to their behaviour, in the way Harris (1970) did so for a sandy loam although on the basis of laboratory experiments.

Many gaps in the information on the behaviour and persistence of pesticides in soil have meanwhile been filled (cf. Kearney, 1970). Adsorption, volatilization, photodecomposition, humidity, water flux and microbial activities have been thoroughly investigated together with the chemical and physical properties of both the substrate and the compound as "standard" factors which always must be held responsible for the behaviour of a chemical in a biological substrate. Some of these parameters hold analogously for plants and even for animals (e.g. monogastric versus ruminant or avian species). The complex influences which the factors "indigenous" (Suett, 1975a) to the soil exert on pesticide behaviour in soil have been discussed intensively in the literature. The most important properties seem to be organic matter and clay content. Persistence tends to increase with increasing organic content, but biological availability and accumulation by plants decline. Volatilization, mobility and metabolism also decline with increasing organic content. This agrees with the fact based on experience that higher application rates are required on soils containing high amounts of organic matter to ensure that a sufficient concentration of insecticide remains available to maintain bioactivity. However, higher application rates may not always compensate fully for reduced availability to the plant so that residues may be lower in plants grown in organic than in mineral soil. However, in many instances it has also been noted that the organic fraction cannot be used as the factor to predict the adsorptive capacity of soils because the interaction between the organic matter and mineral colloids (White and Cruz, 1972) ultimately defines the nature of the available adsorptive surfaces. Other factors having an influence on persistence in soil range from such obvious parameters as climatic variables, pH and microbial population to those like mineral-ion content which are not at first expected to be of importance. On the other hand, other information of a more pragmatic nature is scanty. Meant by this are quantitative data on the behaviour in winter of residues which have outlasted the season; the influence (physical, chemical and microbiological) of soil cultivations on residues, and influences of vegetation on the level and degradation rate of residues in soil.

Data on the persistence of pesticidal compounds in soil usually provide little indication of the seasonal variations of degradation. When residues are preserved as it were in the soil during winter, physical factors like adsorption phenomena may meanwhile have become active, which positively or negatively influence later degradation or uptake by plants. It would be interesting to make a comparative and quantitative study of the kinetics of the degradation of persisting compounds in successive years (example for herbicides: Smith, 1975; for insecticides: Suett, 1975b). It would also be important to know which plants, apart from the famous carrot, are especially suitable for studies of this kind.

MODELS FOR PERSISTENCE

It is the clear-cut policy of pesticide manufacturers, registration authorities and official crop protection organisations to avoid pesticides which persist in a negative sense, and furthermore to evaluate those products already on the market according to whether and where they might be potentially persistent. The need for such evaluations has arisen out of the development of new evaluation criteria during the last few years. It is necessary to establish which criteria should be applied or which models should be used in order to obtain such information as quickly and as reliably as possible. Monitoring cannot supply the needed information but at the most support it. The need for models which can be used for simulating field

conditions or which permit a prediction to be made of behaviour in field conditions is as great as the unsatisfactoriness of the proposals so far made.

a) Soil

In order to be able to conduct comparable studies on soil, agreement should be reached on the use of particular soil types. The EPA (1975a, b) in the United States and the BBA (1973a, b) in the Federal Republic of Germany operate by this procedure. However, there is a certain risk that one day many countries might require these studies to be conducted on yet other soil types. A risk is also involved in the limited interpretability of the results.

Determination of adsorption constants or leaching behaviour in a laboratory by means of standardized experimental techniques is easy and informative as long as the results are evaluated in relation to each other. But apparently there is some temptation for registration authorities so to interpret these results in an absolute sense that pesticidal compounds which exhibit a certain leaching behaviour are regarded as presenting a "hazard to groundwater". This influence is not permissible. Compounds which display significant tendency towards leaching in laboratory studies should merely be regarded as candidates to be tested for a potential groundwater hazard, for two reasons:

- 1) The physical leaching behaviour is only one basic parameter for the behaviour of a compound in soil. Under natural conditions, it is inevitably accompanied by the parameter of metabolism to which it is subjected during downward movement through the soil. This constellation could perhaps be expressed as the difference between two vectors (Fig. 11). The vector L represents the leaching potential during time t and vector m represents "metabolizability" during this time. The difference between the two vectors gives the vector of the "real" leaching potential L of the metabolizing compound, in which the dimension of time is no longer contained.
- 2) Attempts to use the leaching behaviour in the laboratory for predicting the behaviour of pesticides in the field seem to have failed so far (cf. Burkhard et al., 1975). Not only is the relative leaching behaviour different in the field; additional factors like the direction of the water flux are also superimposed. There can be a considerable upward movement of water in the soil as a result of water evaporation from the soil surface. Such movement will cause upward movement of any pesticide, obscuring its "real" leaching potential.

The similarity of leaching behaviour in soil columns with chromatographic phenomena, which allows for the calculation of relative mobility factors or leaching indices (e.g. Harris, 1969; Haque and Freed, 1974, Table 6), has promoted attempts to investigate correlations between pesticide mobilities, soil properties and adsorption constants. The limited information we have does not yet permit a more generalized conclusion (cf. Guth, 1972; Gerber and Guth, 1973). Perhaps we shall learn more about this during this Symposium.

The meanwhile standardized model experiments with small columns in the laboratory are not adequate for the purpose of evaluating a compound for its leaching potential, particularly because in such experiments columns filled with air-dried and screened soils are used, which do not exactly represent the natural conditions of undisturbed soils. Therefore, we began a year ago (Jarczyk, 1976) to work with soil columns which correspond to the conditions of undisturbed soils. Working in

co-operation with the Agricultural Science Department of Bonn University, we obtained soil cores measuring 1.30 to 1.50 metres by 10 centimetres in diameter from our experimental stations, and transferred them in an undisturbed condition in coolable glass columns to the laboratory. Fig. 12 shows the special corer type used for this work, Fig. 13 illustrates the transfer of the cores, and Fig. 14 shows the battery of columns in our greenhouse. Soil structure and composition as well as the micro-organisms remain unchanged. In keeping with the amounts of rainfall at the sites of origin, the columns are irregularly sprinkled with water simulating rainfall for many months using time-controlled pumps. The leachates are analyzed at 14-day intervals for residues of the applied pesticidal compounds. The object of these experiments is to conduct what might be called "field experiments in the laboratory". The experiences we have so far accumulated with 14 radioactive-labelled compounds indicate that we must reckon partly with results different from those expected to be given by standardized laboratory leaching experiments.

In this context, lysimeter experiments will of course always retain their value especially as they permit a better study to be made particularly of the influence of plant growth on soil residues. However, lysimeters also have their problems. They cannot be filled in such manner for the soil they contain to retain its natural structure. To overcome this difficulty, we are currently preparing lysimeters to be filled with undisturbed "monoliths". We are quite hopeful that they will provide us with an indication of possibilities for comparing the results of lysimeter experiments with those obtained from small and large columns in the laboratory.

Considering the usefulness of model experiments of this kind as a means of facilitating early estimates to be made of a potential groundwater hazard of newly developed pesticides, it is astonishing that there is so much theorization on the hazard of groundwater contamination by pesticides. Instead of lamenting that groundwater might be contaminated with pesticides, surely it could be established with relatively little outlay whether it actually is. Experiments we have been conducting for two years with groundwater sampled from an area where agriculture is intensively practised, have as yet yielded only negative results. The analysis covered approximately 60 parent compounds and metabolites.

Laboratory studies to investigate pesticide degradation or metabolism in soil of the kind likewise required in the United States (EPA, 1975b) and in the Federal Republic of Germany (BBA, 1973b) are perhaps still more problematic than leaching studies. From the experience we have so far gained clear relationships between the degradation rates in the field and those measured in the laboratory are still not recognizable. Table 4 underlines this by showing the variations of what I propose to call the Acceleration Quotient, in other words the ratio of the times required for 90% "disappearance". Where $q_s < 1$, degradation in the laboratory is faster than in the field. For the present, these data represent only very rough estimates. However, we are inclined to think that these experiences do not as yet provide much encouragement to undertake an attempt to use such laboratory models as a basis for predicting behaviour in the field.

b) Water

Among the present-day aspects of environmental protection, increasing interest is being focussed also on water. However, in order to tackle these questions experimentally we need to know what is understood to be meant by "water" in this context.

Flowing waters cannot be simulated by a model. Stagnant waters have the variable parameters of biological factors, of pH and of adsorption phenomena (Pionke and Chesters, 1973) in common with soil, and to a still greater degree temperature influences. Storage of "biological" model water is impossible. Attempts to reach

agreement on a method of preparing standardized "biological" water, as far as I am aware, have not yet materialized. Here, too, laboratory models would not have any relevance for practical conditions without inclusion of the most essential "natural" factors. Table 5 provides a few examples to illustrate how much faster a pesticidal compound degrades in pond water than in buffer solutions at pH 7. These data which I likewise call Acceleration Quotients can also be no more than indications. In some cases, the influence of temperature had to be extrapolated in order to be able to compare laboratory and field results. But it is obvious that a somewhat constant acceleration quotient between buffer solutions and natural water cannot be expected.

Results of field experiments so far available are scanty. A review summarizing work reported in the literature over the 25-year period from 1945 to 1971 in respect of the aquatic degradation of 9 selected pesticides (atrazine, captan, carbaryl, diazinon, butoxyethyl-2,4-D, malathion, methoxychlor, parathion, toxaphene) and PCBs (Paris and Lewis, 1973) revealed large gaps in the information on this particular subject.

c) Animals and plants

I shall only briefly discuss this aspect. In the choice of suitable objects for degradation, disappearance and balance studies, it is common for handiness and availability to the experimenter to outweigh other criteria.

Often the principal agricultural use of a product dictates the choice of the model plant. For those instances in which the choice of the model plant cannot be made in accordance with this criterion, proposals should be put forward expressing which plants should be chosen as representative ones. Exploratory experiments very soon show that considerable differences may be expected from one plant to another, as illustrated, for example in a model experiment with trichlorfon (Unterstenhöfer and Frehse, 1963). For the purpose of carrying out pharmacokinetic studies as the first major source of information on a potential persistence or accumulation, it would be extremely desirable for experts to prepare guidelines on experimental techniques and the scope of animal experiments. From the standpoint of registration, agreement ought to be reached as to whether animal experiments are at all necessary in such cases where no residues can be ingested with food or feed.

d) "Micro-ecosystems"

In a paper on "The usefulness of model ecosystems in insecticide development", Booth (1975) states, among other things,

"Pesticides may be classed as purposeful microcontaminants since they are intentionally placed into the environment. This has often resulted in their ubiquitous distribution in our global ecosystem. The term ecological magnification or biomagnification was coined to describe microcontaminant accumulation in food chains. Today, there is increased pressure to provide biomagnification information on all new candidate pesticides. Model ecosystems such as described by Metcalf (e.g. 1974) can be used to monitor the fate of insecticides in the laboratory prior to their use in the field"

However, this very confident statement leaves some questions unanswered:

To what extent can model ecosystems really be used as models for the environmental and persistence behaviour of pesticides? What share does the thus simulated water/land community have in the entirety of the landscape types treated with pesticides? Can they be a substitute for separate studies on soil, on water? Which classes of compounds are particularly suitable for this purpose? Does not the inevitable distribution over so many different, simultaneously present biota raise more questions than it is possible to obtain answers? Do the experiences hitherto

accumulated permit an especially proven model to be proposed for standardization? Are such models at all useful for routine tests?

Ecological studies are always accompanied by data on concentration factors (e.g. Kenega, 1972) intended to express by how many times a compound has become concentrated in an organism in relation to the surrounding substrates (e.g. ppm in fish/ppm in water). I find it difficult to recognize the significance of these factors. If Greater London has a population density of 10,000 per square mile, and a department store in Oxford Street has an extrapolated customer density of 200,000 per square mile, do they exert a concentration factor of 20? Does the use of such a factor, except perhaps in some special cases, attach any general practical value as regards information on persistence?

e) Microbial degradation

One of the more important factors in determining the persistence of a pesticide in soil is its resistance to microbial degradation. In the early days of pesticide research, this finding was more of an empirical nature. Further evidence came from experiments in which the inhibition of microbial activity resulted in much reduced rates of degradation. Indeed the capacity to grow on pesticides as sole carbon, energy and sometimes nitrogen sources seems to be widespread amongst microbial genera (cf. Bollag, 1972; Edwards, 1973; Higgins and Burns, 1975). However, the isolation of a "responsible" organism and its growth in pure culture using the pesticide as the sole source of carbon or nitrogen is perhaps not too conclusive. Mixed cultures of microbes may degrade a compound whereas no single species is able to. Microbes can also bring about chemical alterations of pesticides without deriving energy from these reactions. This process is known as co-metabolism and for it to occur the microbes must obtain the bulk or all of their carbon and energy from other substrates. This phenomenon is fortuitous in relation to persistence problems because it presents the possibility that a series of co-metabolic reactions involving several microorganisms can lead to the total degradation of a pesticide. In the light of these findings, it seems difficult to decide whether and how we could arrive at an agreed model combining all these factors, with the object of being able to conduct laboratory experiments with "soilless soil".

f) Individual parameters

For all practical purposes, degradation behaviour and persistence behaviour can be regarded as an interaction of all involved parameters. Naturally, it is always of academic interest to study what share is contributed by single parameters. Often such single studies provide valuable indications of the chemism of the degradation processes. Photodecomposition and volatilization are among the most frequently studied individual parameters, although many of these studies are lacking similarity with natural conditions. Very few data are available on the relationship between vapour pressure and the fate of residues on plants, nor have photochemistry techniques been elaborated which could suitably simulate plant surfaces.

More information seems to be available concerning the relationship between volatility of pesticides and their interaction with soil (cf. Spencer et al., 1973). It is interesting to note that evaporation losses of single pesticides from soil can amount to several kg./ha./year. Perhaps it is due to volatilization that residues are able to form in plants also when they are not absorbed from the soil. Haque and Freed (1974) have attempted to classify pesticides in 4 groups on the basis of a vaporization index (Table 6), in other words according to their evaporation loss expressed as kg./ha./year (indices ranging from 1 to 4, with 1 = <0.1 kg.; 4 = 7 - 14 kg.). Considering that pesticides losses from volatilization during and after application may range from a few percent to more than 50% (Guenzi and Beard, 1975), I wonder how this impact on persistence could ever be studied quantitatively in a model system.

BALANCES

Obviously one ought not to speak of persistence in a quantitative sense unless all substrates of the environment are considered equally. Valid statements on the behaviour of a pesticide in a target substrate and its surroundings can only be made on the basis of balance studies.

Figure 15 illustrates a lysimeter model which is currently in use at the Nuclear Research Centre at Jülich (Federal Republic of Germany; see e.g. Führ et al., 1976). The lysimeters are either cylindrical with a surface area of 0.76 m² or rectangular with a surface area of 1 m², and packed with 40 cm. of surface soil. A 5-cm. layer of quartz gravel at the base prevents impoundment of the percolate. In the centre, a separated control vessel is installed containing untreated soil and plants. Percolates from the treated portion and the control vessel can be collected separately. Such a system permits, for example, investigation of the following factors under field conditions:

- behaviour of residues in the soil under subsequent croppings
- distribution in the soil profile after cultivation
- rate of mineralization as affected by seasonal variation
- uptake of residues by following crops
- the role of roots in the displacement and degradation of the pesticide in the soil
- influence of moisture, temperature and their fluctuations on the degradation and uptake of pesticides.

Significant losses of radioactivity observed in such systems are indicative of volatilization of pesticides or their conversion to gaseous metabolites, which must be separately studied. We have accumulated experience with this experimental technique, and we consider the model to be capable of yielding extremely meaningful results.

OUTLOOK ON PERSISTENCE

The behaviour of pesticides 1 shown in Fig.16 would undoubtedly be termed persistent but that of compound 2 would surely be called "quite persistent". Yet the difficulty is that in both cases one and the same compound is involved and what is more in one and the same substrate, except that in Fig. 17 (1b) the non-extractable portions were also taken into account. In the light of this, one might be tempted to come forward with the provoking thesis that the persistent compounds are perhaps only those which are extractable over long periods while the non-persistent compounds are simply those which are bound faster.

Strictly speaking, all we know about "non-extractability" is that it exists, and that unknown amounts of parent compound or its breakdown products are hiding somewhere or somehow in the substrate. We know a little about the forces which govern and we know in some cases how high its percentage is. We do not know what the non-extractables consist of for the very reason that we cannot extract them. On the other hand, they are by no means definite. Of course they are largely a function of the extraction procedure and the method of analysis. Systematic searches for ideal or at least optimal extraction systems have not yet been conducted. The same basically holds for the more noble form of "hiding", the formation of conjugates (Menzer, 1973). This was also the conclusion reached at the Research Conference on Bound Conjugated Pesticide Residues held in June, 1975 at Vail, Colorado.

The only way out of this situation would seemingly again be by agreeing on a common approach. At all events, we should be cautious when we discuss persistence as long as this aim has not been reached and we cannot know what we are discussing. If non-extractable feeds or foods cannot be mobilized, in other words "extracted", also by the mammalian organism, then it ought to be possible to regard these residues as toxicologically irrelevant. Analogously, non-extractable residues in soil ought to be regarded as irrelevant if they cannot be extracted by plants and soil-borne organisms. In order to make a better appraisal of this, more quantitative details would be needed on the relationships between chemically detectable and biologically (with test plants) detectable residues in soil. The common practice of investigating the carry-over of soil residues to following crops is a diminutive variant of this approach. But also with regard to analytically detectable, in other words chemically extractable soil residues, a distinction should be made between those absorbed by food and feed crop plants and those which are not. There are growing indications that the confused picture we have of residues in soil is very largely due to unsolved problems of extraction.

I have attempted to elucidate some problems linked with the use of the term persistence. The impression might have been gained that the gaps in information are greater than the useful data. What I have intended to stress is the necessity to sort our information according to certain criteria and then to decide where the gaps are in the information. Persistence is only a word which then could be substituted by many other words. No matter what we call the phenomenon, or rather what word we use for describing it, it should be possible to view it objectively. It is my impression that the term persistence has something unnecessarily negative peculiar to it. No one wants to see pesticides at the South Pole, and no one wants to have to store foreign substances in his body fat. Yet for these phenomena to be off-handedly termed persistence really is a mistake that is commonly made. For a compound to reach the South Pole, it must also be "transportable"; for a compound to be stored in adipose tissue, it must be primarily lipophilic. The reverse conclusion that persisting compounds automatically reach the South Pole or are automatically stored in body fat is certainly not admissible. Perhaps the situation of unwarranted elation over results giving way to a mood of pessimism about persistence can be avoided more successfully in future. In this summarization of some perspectives of persistence, I hope to have succeeded in drawing attention to a number of relationships which I consider should not be disregarded in discussions on persistence.

I felt most honoured by the invitation to speak on this subject, and it has been a great pleasure for me to accept it

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The references quoted in the paper serve only to explain or to enlarge upon a number of special points. No attempt has been made to quote, on a broader basis, review articles or monographs pertaining to the various phenomena, parameters, or problems of persistence broached in this paper.

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