DDT AND PCB IN SOUTH SWEDISH STREAMS

A. Södergren, Bj. Svensson & S. Ulfstrand

Department of Animal Ecology, Ecology Building, S-223 62 Lund, Sweden

ABSTRACT

Gammarus pulex (*Amphipoda*) *was used as an indicator organism in a regional study of the distribution of chlorinated hydrocarbon residues in different streams in southernmost Sweden. Levels of* Σ *DDT and PCB residues covaried regionally in spite of the different mechanisms involved in the dispersal of the substances. Seasonal changes seemed to be correlated with run-off conditions. Only diffuse relationships between residue levels and the age and trophic levels of organisms in a stream ecosystem were found. This suggests that transport routes and accumulation processes are much more complicated and inter-specifically variable phenomena than often assumed.*

INTRODUCTION

The widespread distribution of chlorinated hydrocarbon residues as contaminants in the global ecosystem is now an established fact (*e.g.* Tatton & Ruzicka, 1967; Risebrough *et al.*, 1968; Jensen *et al.*, 1969). However, patterns of regional and local variation, as well as of temporal fluctuations, have been only superficially investigated, and the detailed pathways of residues within trophic networks are poorly understood. The physiological and ecological effects of chlorinated hydrocarbon residues at sublethal levels are also, to a large extent, unknown, particularly as far as invertebrates are concerned.

The present report supplies some information about the regional distribution of DDT and its metabolites as well as PCB (polychlorinated biphenyls) in populations of the amphipod *Gammarus pulex* L. within the province of Skane in southern Sweden. Further, it describes and discusses seasonal fluctuations of contaminant levels and the accumulation of residues in different components of a stream ecosystem. The reason why *G. pulex* was chosen as an indicator organism is that the species is abundant in a variety of habitats, including moderately polluted ones.

Environ. Pollut. (3) (1972) pp. 25–36—

STUDY AREA

The province of Skane (approx. 55°30′ to 56°30′N, 12°45′ to 14°25′E) is a region with growing industry, dense human population and, generally speaking, intense exploitation of the landscape, particularly in comparison with the rest of Scandinavia. The largest towns and industries of the province are situated along its west coast, that is the coast of Öresund (the Sound). Across Öresund, at a distance of approx. 25 km to the west, is Copenhagen with its halo of suburbs and industries. Malmö, the biggest town of Skane, and Copenhagen are probably responsible for a good portion of the airborne pollution in the region, but some of the largest industrial areas of Europe, such as the Ruhr district of Western Germany, are not very distant and, moreover, lie in the dominant wind direction (SW to W). Long-range dispersal of contaminants from this and other similar areas, is far from improbable.

Rural Skane is agricultural, but managed woodland occupies about half its area and is entirely dominant in the northern part of the province.

The exploitation of southern Scandinavia for industrial and agricultural purposes has resulted in large-scale environmental changes, and many undesirable side-effects present problems difficult to solve. Many streams are reduced to effluent canals, and the previous use of mercury compounds as fungicides led to serious depletion of certain wildlife populations. These are but two examples of the development going on here as in many other parts of the world.

The most detailed study was carried out in a small stream, Stampenbäcken, in south-central Skane (approx. 55°35′N, 13°30′E). For information about the environmental features of this stream, *see* Hultin (in press). In addition, the accumulation of chlorinated hydrocarbon residues in populations of *Gammarus pulex* was studied in nine streams in Skane, the names and situations of which are given in Fig. 1.

ANALYTICAL METHODS

Among the chlorinated hydrocarbon residues, lindane, p,p′-DDE, p,p′-DDT and PCB have been found in most samples analysed; aldrin and dieldrin would also have been detected if present above their limits of detection.

To remove excess water, organisms and detritus collected were carefully dried on filter pads before weighing. A sample of the lateral muscle was taken from fish. A quantity of 2 to 3 g was homogenised and extracted in a mixture of acetone/hexane (1 : 1). After discarding the acetone, the hexane solution was evaporated to a small volume, cleaned up by treatment with concentrated sulphuric acid and dried with anhydrous sodium sulphate and silica gel. An aliquot of the extract was then injected on a Varian 204 gas chromatograph equipped with electron capture detectors, and the relative retention times estimated and compared with retention

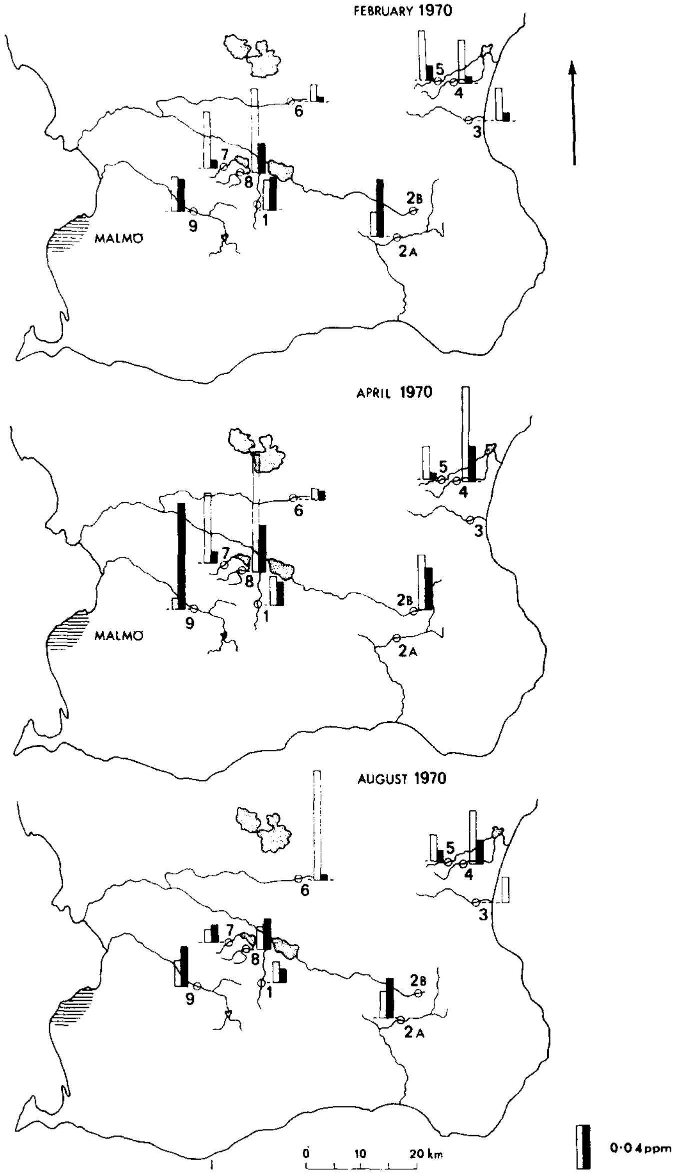

Fig. 1. Maps of southern Scania indicating streams sampled and levels of Σ DDT (white columns) and PCB (black columns) in *Gammarus pulex* populations on three occasions (ppm, fresh weight). Numbers refer to the following streams: (1) Stampenbäcken, (2A) Trydean, (2B) Tolangaan, (3) Julebodaan, (4) Segesholmsän, (5) Forsakarsbäcken, (6) Braan, (7) Skrivaremöllan, (8) Silvakraan, (9) Höje a.

times of known substances. As a reference for PCB, a commercial polychlorobiphenyl (Clophen A50) was used.

The columns were packed with SF 96 (4%) and a mixture of SF 96/QF 1 (3:1) as the stationary phases on GasChrom P (100/120 mesh). The temperature of the injection block, column and detector, was 225°, 175°, and 195°C respectively, and nitrogen was used as carrier gas.

The identity of the more prominent peaks was confirmed by chemical methods. By boiling with alkali solutions, p,p'-DDT and p,p'-DDD were hydrolysed to p,p'-DDE and 1-chloro-2,2-bis (p-chlorophenyl) ethylene, and these products were estimated with the aid of gas chromatography. With this procedure oxygen-containing products (*e.g.* dieldrin) lost in the sulphuric acid treatment could also be estimated.

In order to decompose p,p'-DDE, an aliquot of the extract was oxidised with chromic acid, and PCB peaks, not separated from p,p'-DDE, could be estimated. The chemical procedures did not affect PCB (Westöö & Norén, 1970).

REGIONAL AND SEASONAL DIFFERENCES OF RESIDUE LEVELS

On three occasions, *viz.* February, April, and August 1970, samples of the amphipod *Gammarus pulex* were taken in nine different streams in Skane (Fig. 1). No distinct regional differences were found, neither for Σ DDT(=p,p'-DDT + p,p'-DDD + p,p'-DDE) nor for PCB. However, there is a clear tendency of covariation with regard to levels of the two substances. This is remarkable considering their different origin; DDT being used, up to 1970, for spraying crops, and PCB being passively distributed from unknown sources, which, however, are obviously not agricultural.

The information presented in Fig. 1 also demonstrates the existence of seasonal fluctuations in residue levels. When the samples were taken in February, the ground was frozen and snow-covered. The water level was very low in all streams from which samples were drawn. In fact, some of these streams were at the time fed largely by underground water. Residue levels were generally low on this occasion.

In April, flow volumes were at a peak because of the melting of the snow after an exceptionally long winter. Large quantities of surface water containing abundant amounts of organic and inorganic material were swept into the streams. Residue levels were considerably higher in April than in February.

A closer examination of residue levels in a *Gammarus pulex* population in Stampenbäcken yielded partly divergent results (Fig. 2). From October 1969, to April 1970, inclusive, a steady decline of residue level was established. Thus, there was no trace of an increase at the period of snow melting. However, this pattern holds true only as long as residue levels are expressed in relation to fresh weight. The extractable lipid contents of the animals were decreasing throughout the winter, and for this reason residue levels were rising in relation to lipid weight.

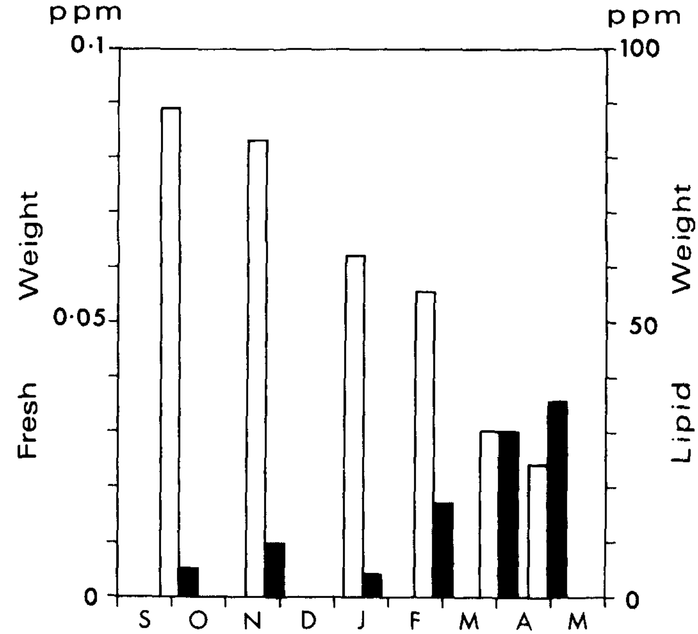

Fig. 2. Levels of Σ DDT + PCB in a *Gammarus pulex* population in Stampenbäcken (loc. 1 in Fig. 1) expressed as ppm fresh weight (white columns, left ordinate) and lipid weight (black columns, right ordinate).

RESIDUE DISTRIBUTION IN A STREAM ECOSYSTEM

In Stampenbäcken, samples of detritus and of the quantitatively dominant animal populations were taken in April and August 1970. The material was analysed for chlorinated hydrocarbon residues, as described above.

Residue levels in April

The analytical results from the April samples are presented in Fig. 3, in which the organisms have been arranged from left to right in order of increasing contents of chlorinated hydrocarbon residues.

The pulmonate gastropod *Ancylus* (*Theodoxus*) *fluviatilis* O. F. Müll. and larvae of Tipulidae contained only small quantities of Σ DDT and PCB. *Gammarus pulex* showed moderate values. Trichoptera and the mayfly *Baetis rhodani* Pict. also contained moderate quantities of Σ DDT, but no PCB was detected in these samples. Plecoptera, the mayfly *Ephemera danica* O. F. Müll. and trout *Salmo trutta* L. contained comparatively large amounts of both substances.

Both Σ DDT and PCB were detected in the detritus samples, although levels were

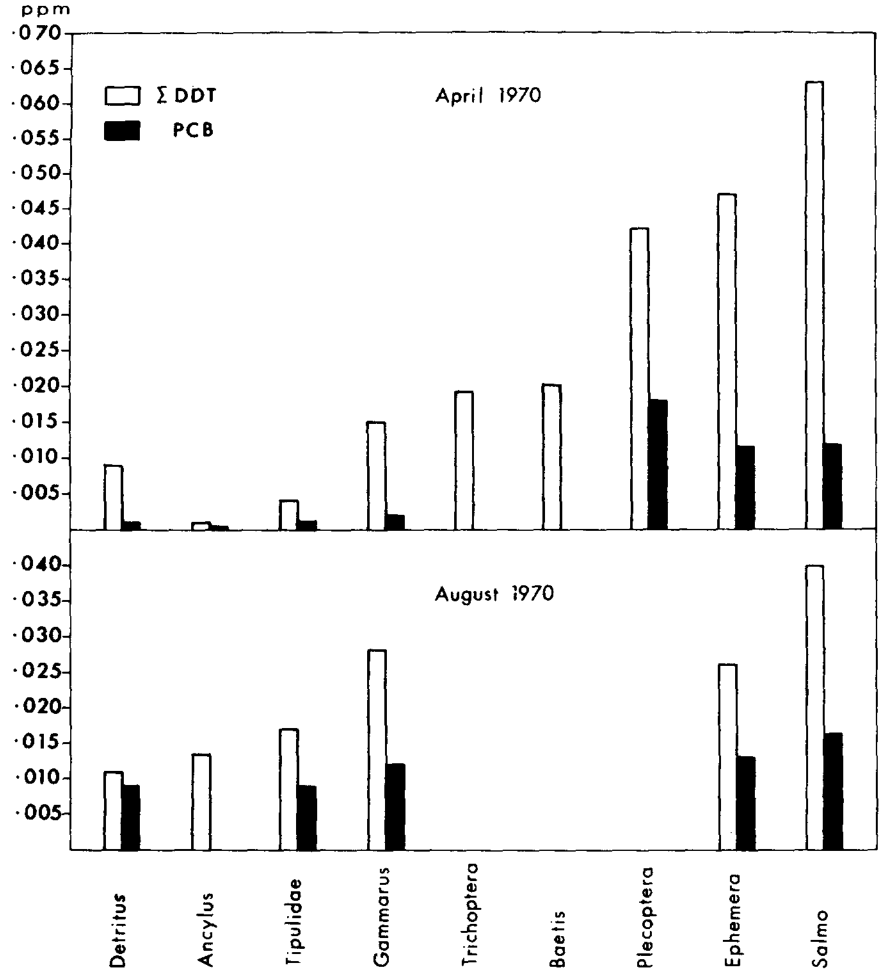

Fig. 3. Levels of Σ DDT (white columns) and PCB (black columns) in different components of a stream ecosystem (loc. 1 in Fig. 1) in southern Sweden (ppm, fresh weight).

low. It should be kept in mind that detritus of course contains an extremely small proportion of lipids. Therefore, in terms of lipid weight, the detritus presumably has a high residue level.

Residue levels in August

Certain of the taxa sampled in April were unavailable in adequate quantities in August (Fig. 3). However, broadly speaking, the differences between the residue contents of different components of the ecosystem were smaller in August than in April but tended in the same direction. Thus, for example, the level in *Gammarus* *pulex* was almost as high as in *Ephemera danica*. Levels in *Ancylus* and Tipulidae had also risen comparatively. In all samples except *Ancylus*, PCB was found in addition to Σ DDT.

The DDE/DDT quotients

Figure 4 illustrates the DDE/DDT quotients found in the different taxa and in the detritus. The most striking feature is the completely deviating quotient in

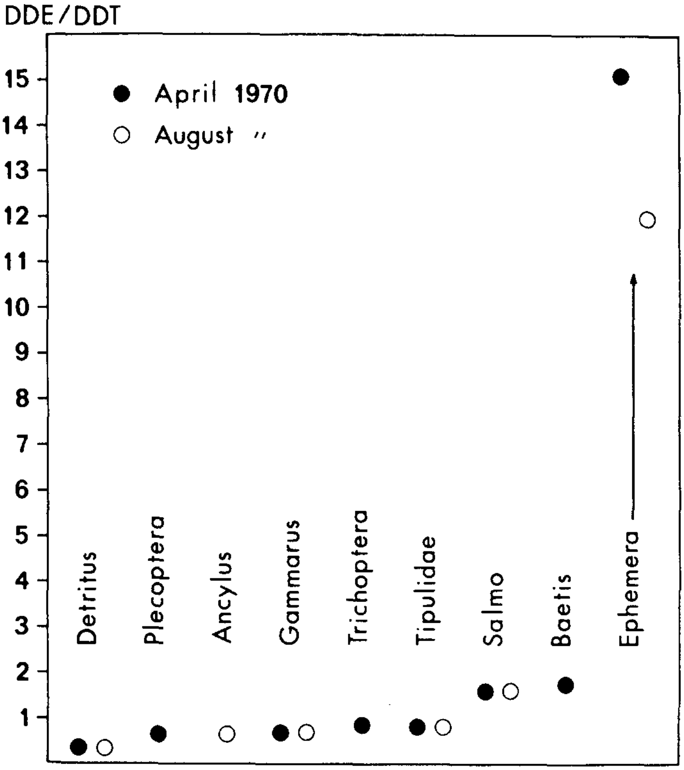

Fig. 4. DDE/DDT quotients in different components of the ecosystem as in Fig. 3.

Ephemera, both in April and in August. *Baetis rhodani* and *Salmo trutta* have much lower values but still the quotients were above unity. All other analyses yielded values below unity. The lowest quotient of all was found in the detritus. In every case there is close agreement between the quotients found on the two sampling occasions.

Food ecology and life cycles

Neumann (1961) has shown that *Ancylus fluviatilis* is a grazer on epilithic algae and requires a substrate of a very special surface roughness to thrive. Moreover, *Ancylus* is limited to sites where the processes of erosion are perpetually at work, since sedimentation reduces its food supply seriously, or at least makes it unavailable. According to Lambret (quoted from Schwoerbel, 1969), these molluscs live for no more than one year.

Little can be said about the tipulids, since they could not be identified to species. Quite likely the samples consisted of mixtures of more than one species. Furthermore, the larval ecology of this group seems to be extremely poorly known. Tipulid (including limnobiid) larvae are said sometimes to be predatory (Wesenberg-Lund, 1943).

Gammarus is a so-called detritivore, ingesting plant litter with its microorganisms. Evidence is accumulating that the microorganisms, rather than the dead organic material, provide the essential food source for the detritivore macroorganisms (Marzolf, 1966; Hargrave, 1970). According to information from Dr L. Hultin, *Gammarus* specimens in Stampenbäcken rarely exceed one year of age; those analysed in April probably were a good deal less than that.

The caddis larvae belonged to three species, *viz. Rhyacophila fasciata* Hag., *Plectrocnemia conspersa* Curt. and (a single specimen of) *Hydropsyche* sp. Various *Rhyacophila* species have been shown to be exclusively carnivorous (*e.g.* Jones, 1950). *Plectrocnemia* builds a net and filters the water. Edington (1968) states that the larvae selectively feed upon small animals and reject the vegetable matter. *Hydropsyche* is also a filter-feeder, and in a recent study Schuhmacher (1970) demonstrated that the larvae of this genus mainly consume vegetable matter, partly gnawed from the stones, partly caught in the nets. However, since only one *Hydropsyche* specimen was included in the sample, it is legitimate to regard the Trichoptera as representative of a mainly carnivorous component of the stream ecosystem.

The mayfly *Baetis rhodani* seems to have a mixed but definitely vegetarian diet, subsisting upon dead, as well as living, plant material (Jones, 1950; *see* Minckley, 1963 for information about a closely related species). The life cycle of the species in Stampenbäcken has not been studied, but there is no reason to suspect anything but a univoltine cycle. The specimens analysed were probably considerably less than one year old.

Like the Trichoptera, the Plecoptera were represented by three species, *viz. Brachyptera risi* Mort. (the most numerous species), *Capnia bifrons* Newm. and *Capnia nigra* Pict. All belong to the Filipalpia and ingest vegetable matter (Brinck, 1949; Hynes 1967).

Ephemera danica is a large, burrowing, mayfly generally stated to have a two-year life cycle (*e.g.* Gleiss, 1954). According to Wesenberg-Lund (1943) the burrows are not permanent, but the animals perpetually dig their way through the substrate, perhaps ingesting organic material in the process. With the use of their gills they create a current sweeping along their body (Eastham, 1939; Eriksen, 1966). This may, to some extent, also enrich their food supply, but probably not to an appreciable extent, for the water has been sieved through the substrate before reaching the animal. However, the sieving effect of the substrate must vary with its grain size composition. Also it is clear that certain other burrowing mayflies, such as species of the genus *Tortopus*, ecologically comparable with *Ephemera* though placed in the family Polymitarcidae, may obtain a considerable portion of their food in the form of particles filtered from the water. The larvae of *Tortopus* live in permanent burrows excavated in clayey river banks (Scott *et al.*, 1959). As Brinck (1961) concludes, it is most likely that *Ephemera* nymphs chiefly subsist upon 'organic debris'. The specimens obtained in April were of uniform size and approaching emergence, hence they were probably about 20 months of age, whilst those obtained in August, likewise of uniform size, were 12 to 14 months old.

Trout, of course, differs from the rest in being a highly mobile predatory vertebrate. The specimens examined were roughly two years old and had mainly *Gammarus* in their stomachs.

DISCUSSION

Certain principles for the relationship between residue accumulation in a given organism and its ecological properties have been accepted by many workers. In general, there seem to be two ways of attaining a particular level of a persistent substance in an organism, *viz.* (1) by progressive accumulation, the rates of excretion and metabolism being more or less negligible, and (2) by reaching an equilibrium between uptake and excretion (with or without preceding metabolic breakdown) at a certain level.

In the first case, the concentration of the residue in question will rise continuously with age, while, in the second case, no further rise will occur after equilibrium has been reached. The level at which equilibrium is attained is a function of the rates of uptake and elimination of the substance.

There is some evidence that both these processes may be operating at the same time. Moreover, for DDT the pattern of accumulation has been found to be dose-dependent (Grzenda *et al.*, 1970; Macek *et al.*, 1970).

Due to the pronounced lipophilic character of the chlorinated hydrocarbon residues, the concentration of these substances in different components of the food chains is usually much higher than in the water. At the lowest trophic level, *Chlorella* has been found to have great capacity for accumulating DDT (Södergren, 1968) and dieldrin (Wheeler, 1970), and it is assumed that these substances are transferred along the food chains to higher trophic levels. Macek & Korn (1970) have shown that fish, in this case *Salvelinus fontinalis* Mitch., accumulated approx. 10 times more DDT from the food chain than directly from the water. Therefore it is to be expected that as a rule high levels should be found in organisms at high trophic levels. It is an established fact that there is a change in the relative proportions of DDT and DDE from lower to higher trophic levels (Woodwell *et al.*, 1967). Whether this phenomenon is due to metabolic changes within the organisms or to selective transport mechanisms is unknown.

Such results as those acquired in the present study may suggest that both processes of residue accumulation mentioned above are involved in the stream ecosystem.

No clear relationship becomes apparent when residue levels or DDE/DDT quotients are compared with the life cycles and trophic levels of the organisms. It is true that *Ephemera* and trout showed relatively high residue levels in April, and these organisms were older than the rest. However, the Plecoptera had practically the same level as *Ephemera*, although presumably being only half its age, and both the species of Plecoptera and *Ephemera* belong to low trophic levels. Apart from trout, the caddis larvae derived from a higher trophic level than any other taxon, and yet they contained only very modest residue quantities. Again, neither age nor trophic position seem adequately to account for the extraordinary DDE/DDT quotient in *Ephemera* (including the relatively young specimens in August).

In the *Gammarus* population of this stream, a distinctly higher residue level was found in August than in April. This suggests that in summer accumulation of chlorinated hydrocarbon residues prevails over elimination, while in winter the opposite is the case. The comparatively lower level in *Ephemera* nymphs in August may be due to the lower age of these specimens.

The most unexpected finding is the aberrant DDE/DDT quotient in *Ephemera* nymphs. Hypothetically, this may be due to either or both of two factors, *viz.* (1) the ingestion of food containing a high proportion of DDE, or (2) a pronounced metabolic activity resulting in a relatively rapid degradation of DDT to DDE. It is difficult to imagine what food source might be specific to *Ephemera* nymphs and at the same time be characterised by an excessively high proportion of DDE. Regardless of whether *Ephemera* subsists chiefly upon particles filtered from the water, or detritus with its microorganisms, or algae attached to the mineral particles of the bottom substrate, it is a fact that all these food sources are utilised by many other animals as well, so that *Ephemera* could hardly obtain its DDE from them. Probably, therefore, the explanation is to be looked for within *Ephemera* rather than in its environment.

The stream ecosystem, some components of which we have analysed, does not seem to store chlorinated hydrocarbon residues in a very simple way. In order to acquire a detailed picture of the pathways and points of storage for these substances in an ecosystem it is obviously necessary to examine the mechanisms of uptake and elimination for each major ecosystem component. A fuller comprehension of this important problem also seems to require much more detailed information about the food ecology of different animal groups than has been hitherto available. Finally, the results should serve as a warning to avoid generalisations from one ecosystem to another.

ACKNOWLEDGEMENTS

We are grateful to Professor Per Brinck for working facilities at the Department of Animal Ecology and to Dr h.c. Ruben Rausing for generous support, including permission to carry out field work on his property. The work of the Rheoecological Group is supported by a grant from the Swedish Natural Science Research Council. Mrs Astrid Ulfstrand prepared the illustrations, and Mr L. G. Olsson took part in some aspects of the study. For all assistance we feel deeply indebted.

REFERENCES

Brinck, P. (1949). Studies on Swedish stoneflies (Plecoptera). *Opusc. ent. Suppl.*, **11**, 250 pp.

Brinck, P. (1961). Dagsländor, Ephemeroptera, in *Djurens Värld*, ed. B. Hanström, **2**, 359–74.

Eastham, L. E. S. (1939). Gill movements of nymphal *Ephemera danica* (Eph.) and the currents caused by them. *J. exp. Biol.*, **16**, 18–33.

Edington, J. M. (1968). Habitat preferences in net-spinning caddis larvae with special reference to the influence of water velocity. *J. Anim. Ecol.*, **37**, 675–92.

Eriksen, C. H. (1966). Benthic invertebrates and some substrate-current-oxygen interrelationships. *Spec. Publs Pymatuning Lab. Fld Biol.*, **4**, 98–115.

Gleiss, H. (1954). *Die Eintagsfliegen*. Wittenberg-Lutherstadt, A. Ziemsen Verl., Die neue Brehm-Bücherei, **136**, 48 pp.

Grzenda, A. R., Paris, D. F. & Taylor, W. J. (1970). The uptake, metabolism and elimination of chlorinated hydrocarbon residues by goldfish (*Carassius auratus*) fed a ^{14}C-DDT contaminated diet. *Trans. Am. Fish. Soc.*, **99**, 385–96.

Hargrave, B. T. (1970). The utilisation of benthic microflora by *Hyalella azteca* (Amphipoda). *J. Anim. Ecol.*, **39**, 427–37.

Hultin, L. (in press). Upstream movements of *Gammarus pulex pulex* (Amphipoda) in a South Swedish stream. *Oikos*, **22.**

Hynes, H. B. N. (1967). A key to the adults and nymphs of British stoneflies (Plecoptera). *Scient. Publs Freshwat. biol. Ass.*, No. 17 (rev. ed.), 91 pp.

Jensen, S., Johnels, A. G., Olsson, M. & Otterlind, G. (1969). DDT and PCB in marine animals from Swedish water. *Nature, Lond.*, **224**, 247–50.

Jones, J. R. E. (1950). A further ecological study of the River Rheidol: the food of the common insects of the main stream. *J. Anim. Ecol.*, **19**, 159–74.

Macek, K. J. & Korn, S. (1970). Significance of the food chain in DDT accumulation by fish. *J. Fish Res. Bd Can.*, **27**, 1496–8.

Macek, K. J., Rodgers, C. R., Stalling, D. L. & Korn, S. (1970). The uptake, distribution and elimination of dietary ^{14}C-DDT and ^{14}C-dieldrin in rainbow trout. *Trans. Am. Fish Soc.*, **99**, 689–95.

Marzolf, G. R. (1966). The trophic position of Bacteria and their relation to the distribution of invertebrates. *Spec. Publs Pymatuning Fld Lab.*, **4**, 131–5.

Minckley, W. L. (1963). The ecology of a spring stream, Doe Run, Meade County, Kentucky. *Wildl. Monogr.*, **11**, 124 pp.

Neumann, D. (1961). Die Ernährungsbiologie einer rhipidoglossen Kiemenschnecke. *Hydrobiologia* **17**, 133–50.

Risebrough, R. W., Rieche, P., Peakall, D. B., Herman, S. G. & Kirven, M. N. (1968). Polychlorinated biphenyls in the global ecosystem. *Nature, Lond.*, **220**, 1098–102.

Schumacher, H. (1970). Untersuchungen zur Taxonomie, Biologie und Ökologie einiger Köcherfliegenarten der Gattung *Hydropsyche* Pict. (Insecta, Trichoptera). *Int. Revue ges. Hydrobiol. Hydrogr.*, **55**, 511–57.

Schwoerbel, J. (1969). Ökologie der Süsswassertiere. Fliessgewässer. *Fortschr. Zool.*, **20**, 173–206.

Scott, D. C., Berner, L. & Hirsch, A. (1959). The nymph of the mayfly genus *Tortopus* (Ephemeroptera: Polymitarcidae). *Ann. ent. Soc. Am.*, **52**, 205–13.

Södergren, A. (1968). Uptake and accumulation of C^{14}-DDT by *Chlorella* sp. (Chlorophyceae). *Oikos*, **19**, 126–38.

TATTON, J. O'G. & RUZICKA, J. H. A. (1967). Organochlorine pesticides in Antarctica. *Nature, Lond.*, **215**, 346–8.

WESENBERG-LUND, C. (1943). *Biologie der Süsswasserinsekten.* Kopenhagen, Berlin & Wien, Verl. J. Springer, 682 pp.

WESTÖÖ, G. & NORÉN, K. (1970). Determination of organochlorine pesticides and polychlorinated biphenyls in animal foods. *Acta chem. scand.*, **24**, 1639–44.

WHEELER, W. B. (1970). Experimental absorption of dieldrin by *Chlorella. J. agric. Fd Chem.*, **18** 416–9.

WOODWELL, G. M., WURSTER, C. M. & ISAACSON, P. A. (1967). DDT residues in an East Coast estuary: a case of biological concentration of persistent insecticides. *Science, N.Y.*, **156**, 821–4.