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Transport of mecoprop from agricultural soils to an adjacent salt marsh

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Abstract

Salt marshes are important ecological areas and play a significant role in coastal flood defence schemes. In many areas of the UK they are adjacent to agricultural areas utilised for the growth of cereal crops, for which mecoprop is used as a selective herbicide in the control of broad-leafed weeds. This study measured concentrations of mecoprop in soils, drainage ditch waters and sediments and salt marsh sediments over a period of 138 days following spring application. Soil concentrations of up to 1827 μg/g were recorded after application, which demonstrated a half life for mecoprop of from 9 to 12 days, with first order kinetics. However, a major rainfall event 9 days after application resulted in significant transport of herbicide to the salt marsh via subsurface field drains, drainage ditches and discharge sluice. Mecoprop concentrations of up to 386 μg/l observed in water samples were above UK guidelines.
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Keywords: Herbicide; Runoff; Sediment; Contamination; Salt marsh; Mecoprop

1. Introduction

Salt marshes constitute 20% of the United Kingdom coastline, mainly in or close to estuaries (Brampton, 1992). A dominant feature along the East Anglian coastline, salt marshes are especially important in Essex where 264 km of the sea defences rely on these natural barriers for seawall stability and coastal protection (Dixon et al., 1998). Salt marsh disappearance is widely documented and attributed to factors such as erosion, changes in climate and tidal movements and flora disease. Within the last few years concern has been expressed over the rise in anthropogenic compounds such as herbicides and insecticides that may enter the marshes and increase their rate of decline (Leggett et al., 1995).

Along the Essex coast many farm sites drain directly onto the salt marshes via sluices, therefore there is a potential for the contamination of the Essex salt marshes due to their proximity to intensively managed agricultural areas. Mecoprop is the most commonly used selective acid herbicide in the UK for the control of broad-leafed weeds in cereal crops, predominantly due to its use with the production of winter cereals as herbicide applications are now required in autumn and in spring.

Following application of mecoprop, several factors influence the degree of transport through the environment. These include herbicide properties (solubility, affinity for complexation or adsorption and persistence), soil characteristics (soil type, moisture and organic carbon content) and climatic conditions (rainfall and temperature) (Frank et al., 1982). The timing of a rainfall event is critical for herbicide transport due to their relatively short half lives, with most herbicide transport occurring within the first few weeks after application and particularly following heavy rainfall (Wauchope, 1978). The contamination in runoff depends upon the residual herbicide concentrations in the soil, the moisture content of the catchment area before rainfall and on the characteristics of the rainfall. The extent of leaching is dependent on vegetative cover, soil type and climatic conditions. Prolonged periods of dry weather before the first significant rainfall for example allows herbicides to degrade in soil before runoff occurs.

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The partitioning of herbicides between the aqueous and solid phases during runoff events is important with respect to herbicide mobility, transport and potential availability to non-target organisms away from the point of application. Acid herbicides are generally relatively polar and ionisable, with high water solubilities and low affinities of adsorption to sediments or soils (Wauchope et al., 1992). Their low sorption potential indicates that they are likely to be leached from soils if not decomposed or biodegraded on surface layers (Helweg, 1993). Mecoprop is very mobile with leaching from the soil column occurring immediately after application and a rain event (MAFF, 1994). Organic micropollutants such as acid herbicides may degrade through biological, chemical and physical degradation processes, however, in the environment, biological degradation is the most common pathway. Half lives in soils have been reported as ranging from 10 to 25 days (Wauchope et al., 1992).

The aim of this study was to trace mecoprop leaching from fields of application to the sediment and water in an adjacent salt marsh, taking into consideration the environmental influences, and to quantify the inputs of mecoprop onto the salt marsh in relation to the field spray.

2. Materials and methods

The study site, which was typical of the area, was located on the Essex coast of England, south of Harwich. The focus of the study was an 11 ha plot, within a larger, controlled catchment of approximately 200 ha consisting primarily of arable fields, where a total of 92.4 kg of mecoprop were applied to a total of 77 ha, a loading of 120 mg/m^3, equating to 13.2 kg on the 11 ha plot. This 11 ha plot was selected as it had a hydrologically isolated network of subsurface drainage which discharged through a single field drain (Fig. 1). The edge of the field farthest from the seawall followed the 10 m contour above sea level. Drainage pipes were located 0.5 m below the field surface, and the field drain discharged directly to the main drainage ditch system, which subsequently discharged to sea via a sluice. The site discharged into Hamford Waters, an area of embayment salt marsh with no direct fluvial inputs. The field soil cover consisted of sandy loam (organic matter content of 3–3.8% in the top 2.5 cm) with underlying bedrock throughout the area being predominantly London clay.

2.1. Sampling

Field soil samples were collected from a square sampling site (20 m × 20 m) towards the lower edge of the gently sloping, flat field (Fig. 1), to a depth of 2.5 cm over a period of 5 months. The area was assumed to be representative of the site and selected to give data on temporal, rather than spatial variation. Samples were collected on 18 dates totalling 72 soil samples. The first date was day 0 (15th April 1993) before spraying, the second was on day 1 (directly after spraying). Samples were then taken daily for 7 days followed by fortnightly for 8 weeks until day 64. The frequency was then further reduced and samples were taken at 4 week intervals until day 138.

Analysis of water from the field drain, which collected all the rainwater percolating through the 11 ha plot, allowed the measurement of the total flux of mecoprop discharged via this route to the drainage ditch. Two water samples were taken from the drain before the application of mecoprop to evaluate background concentrations. Subsequently, samples were taken twice daily when the field drain was discharging. Flow rates from the field drain were measured at the discharge point.

Water samples were taken from the main drainage ditch on the landward side at the entrance to the sluice. Background samples were taken fortnightly for a period of 12 weeks prior to the April application of mecoprop. Sampling frequency was then increased for a further 18 weeks to the end of the salt marsh flowering season. Samples were taken manually and using an EPIC EPS 1011 portable automatic effluent sampler (EPIC Products Ltd., Salford, UK). Following the first significant rise in the drainage ditch water level, the autosampler was programmed to run continuously and take one 500 ml sample every hour for 24 h. Manual samples were collected twice daily for the first 40 days and then daily for the remainder of the test period.

All water samples remained unfiltered after collection to represent the total mecoprop input in the aqueous and particulate phases. They were collected in 2.5 l amber glass bottles, with the addition of 250 ml of dichloromethane and 5 ml of concentrated sulphuric acid, stored in the dark at 4 °C and analysed within 72 h of collection.

Sixteen days after mecoprop spraying and 7 days following the main rain event, sediment core samples were collected at three locations with a Jenkins corer. One core was located within the drainage ditch, directly adjacent to the field drain discharge pipe, another at the sluice entrance (landward side) and a third in the drainage channel seaward of the sluice exit (Fig. 1). Sediment cores ranged from 10 to 25 cm deep.

Salt marsh sediment core samples were taken using a device developed for the study (Fletcher et al., 1994a), 16 days and 97 days after spraying, on each occasion two core samples were taken. One core was located on the mudflat, 3.2 m from the sluice drainage channel and 20 m from the sluice, the core depth was at least 40 cm. The second core was located on the vegetated marsh (predominately *Puccinillia maritima*), 23.4 m from the sluice drainage channel and 40 m from the sluice, the core depth was 55 cm (Fig. 1).

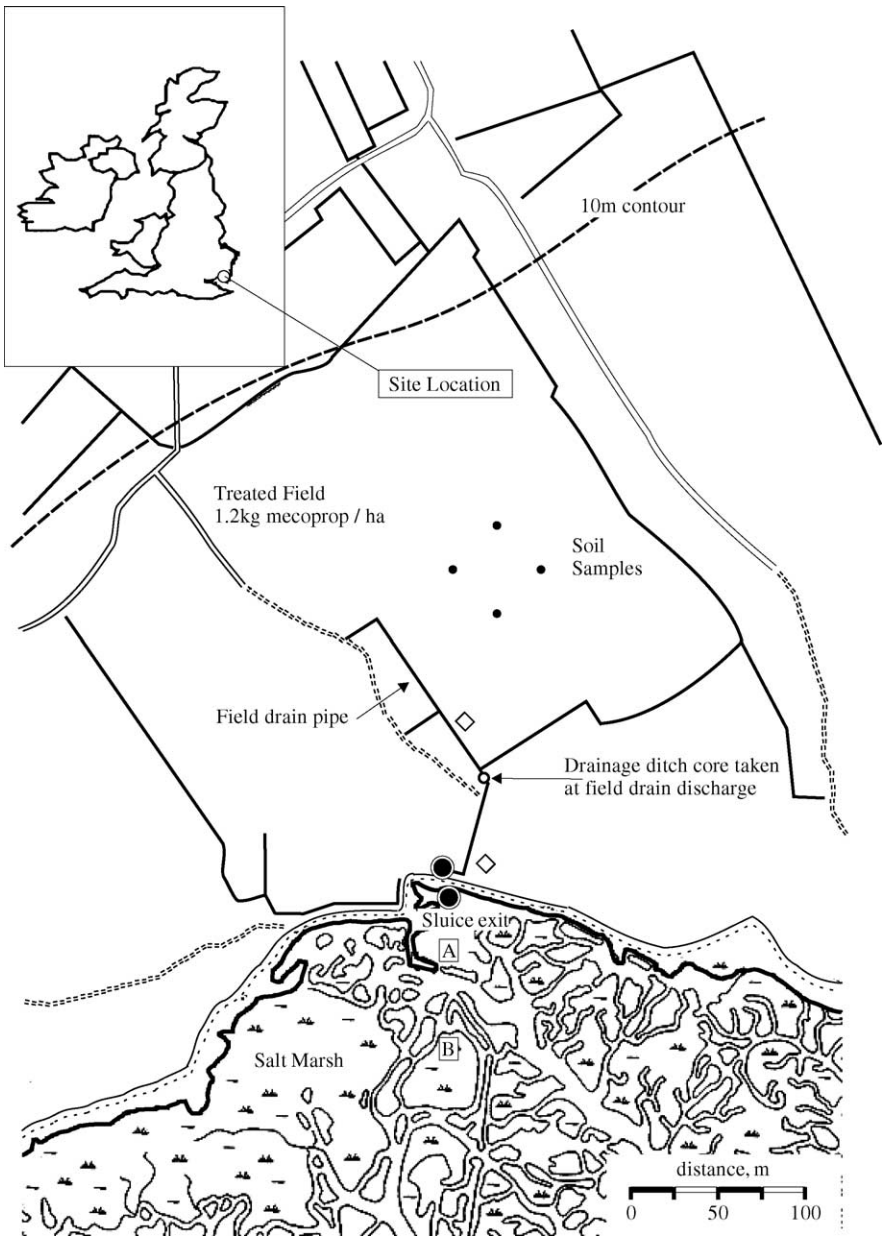

Fig. 1. Location of sampling site, with detail of 11 ha study plot. (⎓) Seawall; (▬) drainage ditches; (=) roads; (⎓) tracks; (◇) rainwater collection points; (◉) sampling points at sluice, landward and seaward; A and B locations of sediment cores taken from salt marsh.

All sediment cores were sectioned at 5 cm intervals, sediment physico-chemical analyses were undertaken on site and samples were stored at −18 °C for subsequent mecoprop analysis. On the salt marsh, sediment type and consistency, plant detritus, vegetation cover and the presence or absence of vertebrates were recorded.

2.2. Analysis

Mecoprop concentrations were determined with a method based on that used by Hill et al. (1983), and has been described in detail elsewhere (Fletcher et al., 1994b). It involved extraction, derivatisation with pentafluorobenzyl bromide and final analysis by gas chromatography with electron capture detection. Total solids were determined by gravimetric analysis according to standard methods at 105 °C (Standing Committee of Analysts, 1980).

3. Results

3.1. Concentration of mecoprop in the soil surface

Before spraying, all sample sites exhibited mecoprop concentrations below the limit of detection (0.2 μg/kg).

The spray application rate of 1.2 kg/ha of active ingredient was equal to 120 mg/m^2 of mecoprop. The average concentration determined in the top 2.5 cm of soil within approximately 2 h of spraying was 1827 μg/kg, equivalent to a concentration of approximately 45.0 mg/m^2, or approximately 38% of the applied mecoprop, with the remainder probably being associated with vegetation. After the initial rapid rise in concentrations of mecoprop, they were observed to decrease to 580 μg/kg by day 9. As a result of the rainfall event on day 9, the concentration fell by almost an order of magnitude to 72 μg/kg by day 10 (Table 2). Subsequently, concentrations declined to below detection limits by 93 days after spraying although an increase in soil concentrations in the area sampled between days 65 and 79 (Table 2 and Fig. 2) may have been the result of transport over the site, influenced by local topography, due to the rainfall event on day 62 (Table 1).

The degradation of the mecoprop applied can be demonstrated by plotting the concentration on a logarithmic scale against time (Fig. 2). The degradation is demonstrated to be essentially first order, with a significant step appearing in the plot between days 9 and 10, due to a rapid reduction in concentrations as a result of the rainfall event. Between days 1 and 9, a half life of 12.5 days was observed, calculated from the slope of the best-fit line and the equation:

$$\text{Half life}(t_{1/2}) = -K/\ln 2$$

where $t_{1/2}$ is the half life in days and K is the slope of the line. From days 10 to 37, degradation in the soils continued, with a half life of 9.9 days, however, after this there was little change until day 93 when concentrations of mecoprop decreased and subsequently remained below the method detection limits (Table 2). This may have been related to rainfall events between days 85 and 89 (Table 1) resulting in further leaching or the increase in soil moisture concentrations (Table 2) allowing for greater microbial activity. Calculation of the half lives is based on the slopes of lines from Fig. 2, and the validity of the data may be demonstrated through the significance of the regression lines. From days 1 to 9, the regression was not significant ($r^2 = 0.58$), however, between days 10 and 37, the regression was significant ($r^2 = 0.91$; 99.9% confidence limit) which would indicate that a half life for mecoprop of 9.9 days is the most reliable figure.

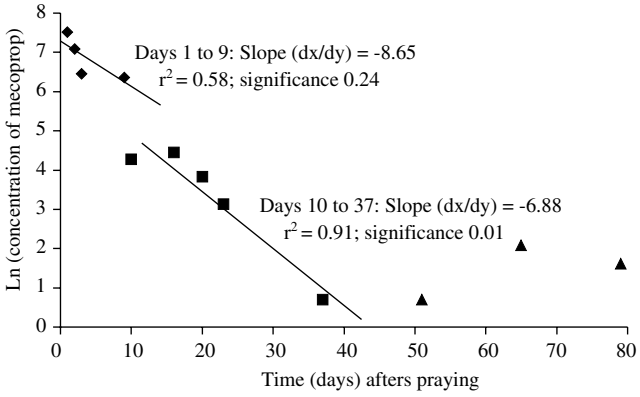

Fig. 2. Natural log (ln) of soil mecoprop concentrations against time, with slope of regression lines through days 1–9 and days 10–37. The step indicates loss through the rainfall event.

Table 1
Rainfall history at the study site

Datum time (days)	Date	Rainfall (mm)	Rainwater sampling
0	April 15th	0.0	
1	16th	2.0	
2	17th	1.0	Collected
8	23rd	3.0	
9	24th	20.5	Collected
23	May 7th	1.0	
33	18th	6.0	
34	19th	5.5	
35	20th	9.0	Collected
41	26th	3.5	
44	29th	5.0	
48	June 2nd	1.5	Collected
56	10th	1.5	
57	11th	3.0	
58	12th	5.5	
59	13th	6.0	
60	14th	5.0	
62	16th	15.0	
63	17th	1.0	Collected
69	23rd	1.5	
85	July 9th	7.0	
89	13th	12.0	
91	15th	4.0	Collected
94	18th	1.5	
95	19th	5.5	
100	24th	6.0	
101	25th	6.0	
102	26th	2.0	
103	27th	1.5	
104	28th	1.0	
105	29th	1.5	Collected
111	August 4th	1.0	
116	9th	1.0	
118	11th	12.5	
119	12th	4.0	Collected
121	14th	3.0	

3.2. Mecoprop runoff from the plot

Discharge from the field drain which collected from the 11 ha plot was monitored along with the concentration of mecoprop (Fig. 3). This discharge accounted for leaching through the soil column, and excluded surface runoff. The maximum concentration of meco-

Table 2
Soil mecoprop concentrations and moisture content throughout the study period

Date	Days after spraying	Mecoprop concentration (μg/kg)	Moisture (%)
April 15th	0	ND	16.3
16th	1	1827	12.5
17th	2	1188	15.0
18th	3	634	15.2
24th	9	580	16.1
25th	10	72	18.3
May 1st	16	86	20.7
5th	20	46	18.6
8th	23	23	18.9
22nd	37	2	19.4
June 5th	51	2	21.4
19th	65	8	20.8
July 3rd	79	5	26.6
17th	93	ND	23.8
31st	107	ND	19.6
August 12th	119	ND	21.7
27th	134	ND	21.1

ND denotes concentration below the limit of detection (0.2 μg/kg).

prop (386 μg/l) coincided with the peak flow rate of 474 l/h within 12 h of the rainfall on day 9, implying that transport of mecoprop through the soil layer and into the drainage pipes, located at a depth of 50 cm below the field surface was rapid. During the first 20 days after application, the mass of mecoprop discharged through the field drain was determined to be 5.7 g, or approximately 0.04% of active ingredient applied. After day 20 no further flow was observed from the field drain, despite the further rainfall that occurred over the period (Table 1).

The discharge from the drainage ditch, to sea, which encompassed all water from the whole 77 ha farm site followed a similar temporal pattern to that from the study plot, with flows through the sluice increasing from 156 m^3 per day before rainfall to over 500 m^3 per day

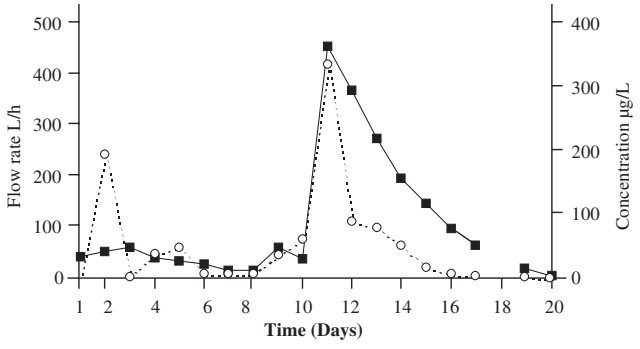

Fig. 3. Water flows (■) and mecoprop concentration (○) from the field drain observed over the first 20 days after spraying.

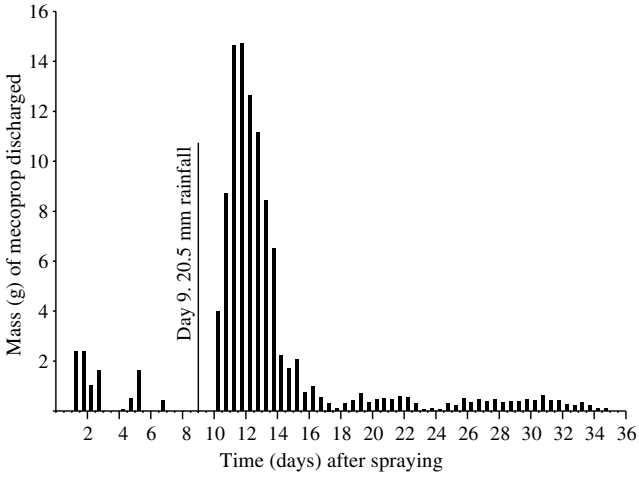

Fig. 4. Mass of mecoprop discharged to sea over 12 h periods via the sluice up to day 36.

during day 10 (one day after the rainfall). Concentrations of mecoprop increased rapidly in discharge, from <5 μg/l to approximately 25 μg/l over a period of 4 days after the rainfall event. This resulted in a total discharge of 114 g of mecoprop to the surrounding salt marsh throughout (0.12% of the total applied to the site) over the first 37 days of the study, with 77% of this occurring within 6 days of the rainfall event (Fig. 4).

3.3. Drainage ditch core samples

The mecoprop concentrations measured in the three core samples along the drainage ditch route are shown in Table 3. The concentrations varied from less than the detection limit to a maximum of 5.4 μg/kg. The highest concentrations were in the sluice exit core and no herbicide was detected in the field drain core. It is possible that high flow rates may have contributed to the transport of sediments with bound residues of herbicides to the seaward side of the sluice.

3.4. Mecoprop contamination in salt marsh sediment core sediments

Prior to spraying, in core samples taken in November 1992 and March 1993, mecoprop was not detected in sediments from non-vegetated marsh adjacent to the sluice drainage channel and only in the upper sediments of the vegetated marsh at mean concentrations of 0.7 and 0.4 μg/kg. On the 16th day after spraying with mecoprop (7 days after the main rain event) the results demonstrated increased concentrations of mecoprop in sediment samples, with maximum concentrations observed in sediments from the mudflat by the drainage channel. Concentrations had increased up to 9.9 μg/kg and mecoprop was found at depths of up to 40 cm in the sediment (Fig. 5a). Eighty three days after spraying,

Table 3
Sediment mecoprop concentrations (μg/kg dry weight) in cores from the drainage ditch adjacent to discharge of the field drain and from the land and seaward sides of the discharge sluice

Datum time (days)	Core location	Core depth (cm)	Mecoprop concentration (μg/kg)	Moisture (%)
Day 16	Drainage ditch field drain	2.5	ND	51.1
		5	ND	50.2
		10	ND	48.2
Day 16	Sluice entrance (landside)	2.5	ND	53.1
		5	0.5	54.4
		10	1.3	66.4
		15	ND	56.5
		22	ND	52.1
Day 16	Sluice entrance (seaward)	2.5	4.1	60.7
		5	5.4	59.0
		10	2.0	57.7
		15	ND	55.8
		20	ND	55.5
		25	1.4	54.4

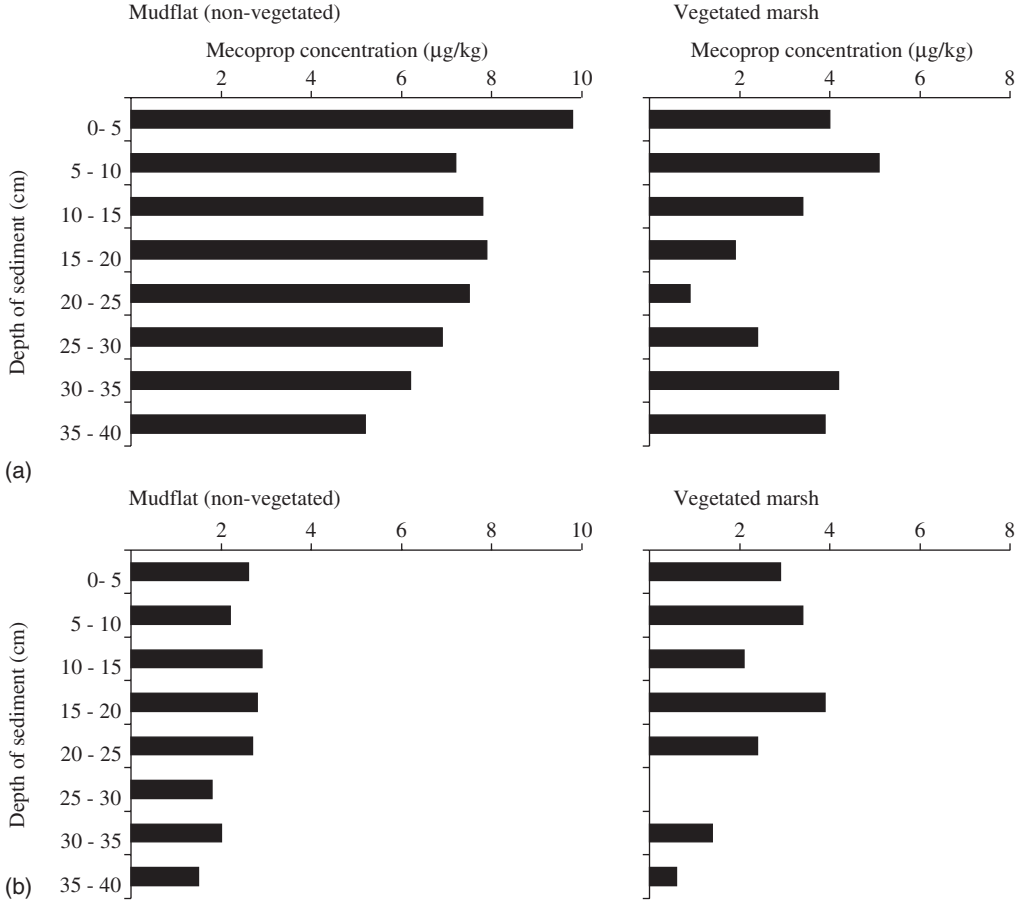

Fig. 5. Mecoprop concentrations in sediment cores from vegetated and non-vegetated (mudflat) areas 16 days after sampling and 83 days after spraying. (a) Samples taken day 16, 7 days after the rainfall event. (b) Samples taken day 83, 74 days after the rainfall event.

concentrations in sediments from the core samples taken (74 days after rainfall) had declined, falling in the range <0.2–3.7 μg/kg and the maximum values in the vegetated core again exceeded those in the non-vegetated (mudflat) sediments (Fig. 5b).

4. Discussion

This study has followed the fate of a herbicide, mecoprop, after application to agricultural land where runoff is discharged to adjacent salt marshes via field

drains, drainage ditches and a sluice system. Losses from the fields were strongly influenced by a single rainfall event, as has been observed by other workers (Wauchope, 1978; Willis and McDowwel, 1982; Klaine et al., 1988). However, losses within the field soils also depend on factors such as soil moisture and organic matter content (Frank et al., 1982; Harris et al., 1991; Albanis, 1992). Site topography may also play a role in determining the fate of contaminants in runoff although studies on this variable have tended to focus on larger catchment areas than covered in this work. Distance from the drainage network of 50–100 m was determined to be critical in transport due to subsurface runoff in inter-storm periods (Colin et al., 2000) although this was not necessarily true for peak runoff, which was observed during the rainfall event in this study. The use of the SoilFug model, which does not include site specific information such as topography, was shown to give good correlation with field data by Williams et al. (1995) on a 150 ha site with "gentle slopes" and total variation in elevation of 39 m, with field drains at 20 m intervals. On a larger, basin scale of 400 km^2 the SoilFug model was used with GIS to account for natural characteristics, however, the surface was described as "almost completely flat". The work demonstrated that in relatively homogenous areas, fugacity modelling alone (with good quality input data) was likely to be a good predictor (Barra et al., 2000). It is therefore probable that in this present study, with the size of site and proximity of drains that topography had little influence on the main runoff event.

The soil half lives calculated from Fig. 2 agree with the range reported by Helweg (1993), in particular with the data reported for dry and in flooded soil (25% and 200% of total water holding capacity) where values for half lives were estimated at 10 and 15 days. A half life for MCPA, a herbicide from the same family as mecoprop, of 5 days has been reported in a study of herbicide transport from agricultural land, indicating that the acid group (acetic or propionic) may influence the half life of the chlorophenoxy herbicides (Williams et al., 1995). This study also concluded that the total mass of pesticides lost through runoff was a small proportion (<1%) of the total applied, however, high concentration pulses did occur in the field drains and local streams.

The discharge through the field drain accounted for a smaller percentage of the total applied than observed in other studies as the field drain collected only percolated water, and overland flows are reported to account for over 60% of herbicide inputs into watercourses (Frank and Sirons, 1980). High concentrations of herbicides after significant rainfall events have been attributed to point sources such as runoff from hard surfaces as opposed to diffuse runoff sources (Harris et al., 1992). Harris also observed that although lower in concentration, inputs from diffuse sources occurred over longer periods. The occurrence of peak water volumes with peak concentrations was consistent with other studies (Gomme et al., 1991; Harris et al., 1991) and indicates rapid transport mechanisms such as overland flow and, as in this study, routes through the soil drains also providing a pathway to drainage ditches.

The rate of herbicide degradation combined with the timing of rainfall events following mecoprop application appears to be important with respect to leaching and transport from agricultural soils. The mecoprop in the field drain represented that which had percolated through about 50 cm of soil from the hydrologically isolated 11 ha plot. Over the 20 day period of flow from the field drain, the total mass of mecoprop discharged from the plot was 5.7 g. Although this was only 0.04% of the total applied, the concentrations in the field drain waters did exceed the UK Department of Environment Toxicological Guideline maximum for surface water of 10 μg/l by an order of magnitude for several days and herbicide concentrations of the same magnitude were observed by Williams et al. (1995) in drainage water. Atmospheric depositions have previously been reported as a significant source of some herbicides in the environment (Chevreuil and Garmouma, 1993), however, in this study inputs via this pathway were not observed.

Mecoprop residues were not detected in sediments from the drainage ditch by the field drain, however, they were detected both landward and seaward of the sluice. This may have resulted from settling of flushed particulate matter in these areas, with subsequent flushing through the sluice resulting in higher concentrations at the seaward side. A concentration of 1.4 μg/kg mecoprop at 25 cm at the seaward sluice entrance may be due to lack of biodegradation at that depth as mecoprop is persistent in anaerobic conditions (Gintautas et al., 1992). Mecoprop has more affinity for the aqueous phase than the sediment phase, resulting in high mobility, explaining why it can be found at depths through the sediment matrix.

Previous studies (Fletcher et al., 1995) have shown that mecoprop may accumulate in sediments from vegetated marsh over time and persistence of mecoprop residues has also been reported under anoxic conditions which are typical of salt marsh sediments (Babicka and Hubacek, 1988; Buisson et al., 1989; Gintautas et al., 1992). In addition, organic carbon content of natural sediment has been shown to be the primary factor controlling the adsorption of anthropogenic compounds (Karrickhoff et al., 1979) and the lower carbon content typical of non-vegetated sediments may also affect retention. The occurrence of mecoprop to depths of 40 cm within the salt marsh sediment is not clearly understood. However, this distribution pattern of mecoprop may be explained by either the occurrence of preferential water flow pathways arising from cracking in the sediment or by the activities of burrowing fauna, or it may be

attributed to the low affinity of mecoprop adsorption and association with interstitial waters in agricultural soils.

5. Conclusions

Around 0.04% of the applied mecoprop was transported through the soil to the field drain from the 11 ha field studied, the majority of this occurred within 6 days of significant rainfall.

Maximum concentrations of mecoprop in the drainage ditch water, from 77 ha of treated fields, corresponded to maximum water flow indicating rapid transport from the field soils to the drainage ditch. The relationship observed in this study between the drainage water volume and the mecoprop concentrations accentuates the dependence of the herbicide mass fluxes upon major rainfall events.

After spray application, mecoprop contamination occurred initially within vegetated and non-vegetated marsh sediments. Over a period of 3 months, mecoprop concentrations exhibited a more rapid decline in sediments from non-vegetated (mudflat) areas then in areas of vegetated salt marsh.

References

Albanis, T.A., 1992. Herbicide losses in runoff from the agricultural area of Thessalonike in Thermaikos Gulf, N. Greece. Sci. Total Environ. 114, 59–71.

Babicka, L., Hubacek, J., 1988. The effect of chlorophenoxyalkanoic herbicides in the environment and their determination. In: Abbou, R. (Ed.), Proc. World Conf. Hazard Waste Detection, Control and Treatment. Elsevier, Amsterdam, pp. 829–838.

Barra, R., Vighi, M., Maffioli, G., Di Guardo, A., Ferrario, P., 2000. Coupling SoilFug model and GIS for predicting pesticide pollution of surface water at watershed level. Environ. Sci. Technol. 34, 4425–4433.

Brampton, A.H., 1992. Engineering significance of British salt marshes. In: Allen, J.R.L., Pye, K. (Eds.), Salt Marshes: Morphodynamics Conservation and Engineering Significance. Cambridge University Press, pp. 115–122.

Buisson, R.S.K., Kirk, P.W.W., Lester, J.N., 1989. Fate of selected chlorinated organics during semicontinuous anaerobic sludge digestion. Arch. Environ. Contam. Toxicol. 23, 285–291.

Chevreuil, T.M., Garmouma, M., 1993. Occurrence of triazines in the atmosphere fallout of the catchment basin of the River Marne, France. Chemosphere 27, 1605–1608.

Colin, F., Puech, C., de Marsily, G., 2000. Relations between triazine flux, catchment topography and distance between maize fields and the drainage network. J. Hydrol. 236, 139–152.

Dixon, A.M., Leggett, D.J., Weight, R.C., 1998. Habitat creation opportunities for landward coastal re-alignment: Essex case studies. J. Inst. Water Environ. Manage. 12, 107–112.

Fletcher, C.A., Bubb, J.M., Lester, J.N., 1994a. Magnitude and distribution of contaminants in salt marsh sediments of the Essex coast, UK I. Topographical, physical and chemical characteristics. Sci. Total Environ. 155, 31–45.

Fletcher, C.A., Meakins, N.C., Bubb, J.M., Lester, J.N., 1994b. Magnitude and distribution of contaminants in salt marsh sediments of the Essex coast, UK. III. Chlorophenoxy acid and *s*-triazine herbicides. Sci. Total Environ. 155, 61–72.

Fletcher, C.A., Bubb, J.M., Lester, J.N., 1995. Agricultural inputs of mecoprop to a salt marsh system: its fate and distribution within the sediment profile. Mar. Pollut. Bull. 3012, 803–811.

Frank, R., Sirons, G.J., 1980. Chlorophenoxy and chlorobenzoic acid herbicides; their use in 11 agricultural watersheds and their loss to stream waters in Southern Ontario, Canada 1975–1977. Sci. Total Environ. 15, 149–167.

Frank, R., Braun, H.E., Van Houe Holdrinet, M., Sirons, G.J., Ripley, B.D., 1982. Agriculture and water quality in the Canadian Great Lakes Basin; pesticide use in 11 agricultural watersheds and presence in stream water 1975–1977. J. Environ. Qual. 113, 497–505.

Gintautas, P.A., Daniel, S.R., Macalady, D.I., 1992. Phenoxyalkanoic acid herbicides in municipal landfill leachates. Environ. Sci. Technol. 263, 517–521.

Gomme, J.W., Shurvell, S., Hennings, S.M., Clark, L., 1991. Hydrology of pesticides in a chalk catchment; surface waters. J. Inst. Water Environ. Manage. 5, 546–552.

Harris, G.L., Bailey, S.W., Mason, D.J., 1991. The determination of pesticide losses to water courses in an agricultural clay catchment with variable drainage and land management. In: Brighton Crop Protection Conference—Weeds 1991, BCPC Publications 3, pp. 1271–1278.

Harris, G.L, Turnbull, A.B., Gilbert, A.J., Christian, D.G., Mason, D.J., 1992. Pesticide application and deposition—their importance to pesticide leaching to surface water. In: Brighton Crop Protection Conference—Pests and Diseases 1992, BCPC Publications 2, pp. 477–486.

Helweg, A., 1993. Degradation and adsorption of 14C-mecoprop MCPP in surface soils and in subsoil. Influence of temperature, moisture content, sterilization and concentration on degradation. Sci. Total Environ. 132, 229–242.

Hill, N.P., McIntyre, A.E., Perry, R., Lester, J.N., 1983. Development of a method for the analysis of chlorophenoxy herbicides in waste waters and waste water sludges. Int. J. Environ. Anal. Chem. 15, 107–130.

Karrickhoff, S.W., Brown, D.S., Scott, T.A., 1979. Sorption of hydrophobic pollutants on natural sediments. Water Res. 13, 241–248.

Klaine, S.J., Hinman, M.L., Winkelmann, D.A., Saucer, K.R., Martin, J.R., Moore, L.W., 1988. Characterisation of agricultural nonpoint pollution: pesticide migration in a West Tennessee watershed. Environ. Toxicol. Chem. 7, 609–614.

Leggett, D., Bubb, J.M., Lester, J.N., 1995. The role of pollutants and sedimentary processes in flood defence. A case study:salt marshes of the Essex coast, UK. Environ. Technol. 16, 457–466.

MAFF, 1994. Evaluation of mecoprop-p and its salts. Disclosure Document No. 96, Advisory Committee on Pesticides. MAFF Pesticides Safety Directorate, London, 151pp.

Standing Committee of Analysts, 1980. Methods for the determination of waters: suspended, settleable and total dissolved solids in waters and effluents. Her Majesty's Stationary Office, London, UK. 54pp.

Wauchope, R.D., 1978. The pesticide content of surface water draining from agricultural fields; a review. J. Environ. Qual. 74, 459–472.

Wauchope, R.D., Buttler, T.M., Hornsby, A.G., Augustijn-Beckers, P.W.M., Burt, J.P., 1992. The SCS/ARS/CES pesticide properties database for environmental decision-making. Rev. Environ. Contam. Toxicol. 123, 1–164.

Williams, R.J., Brooke, D.N., Matthiessen, P., Mills, M., Turnbull, A., Harrison, R.M., 1995. Pesticide transport to surface waters within an agricultural catchment. J. IWEM 9, 72–81.

Willis, G.H., McDowwel, L.L., 1982. The pesticide content of surface water draining from agricultural fields; a review. Environ. Toxicol. Chem. 1, 267–279.