

1 Assessing the effectiveness of a three-stage on-farm biobed in 2 treating pesticide contaminated wastewater

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11 Abstract

12 Agricultural point source pesticide pollution arising from contaminated machinery washings and
13 accidental spillages pose a significant threat to river water and groundwater quality. In this study, we
14 assess the effectiveness of a three-stage on-farm biobed for treating pesticide contaminated waste
15 water from a large (20 km²) commercial arable estate. The facility consisted of an enclosed
16 machinery wash-down unit (stage 1), a 49 m² lined compost-straw-topsoil biobed (stage 2), and a
17 200 m² drainage field with a trickle irrigation system (stage 3). Pesticide concentrations were
18 analysed in water samples collected fortnightly between November 2013 and November 2015 from
19 the biobed input and output sumps and from 20 porous pots buried at 45 cm and 90 cm depth
20 within the drainage field. The results revealed that the biobed removed 68–98% of individual
21 pesticides within the contaminated washings, with mean total pesticide concentrations reducing by
22 91.6% between the biobed input and output sumps. Drainage field irrigation removed a further 68–
23 99% of individual pesticides, with total mean pesticide concentrations reducing by 98.4% and 97.2%
24 in the 45 cm and 90 cm depth porous pots, respectively. The average total pesticide concentration at
25 45 cm depth in the drainage field (57 µg L⁻¹) was 760 times lower than the mean concentration
26 recorded in the input sump (43,334 µg L⁻¹). There was no evidence of seasonality in the efficiency of
27 biobed pesticide removal, nor was there evidence of a decline in removal efficiency over the two-
28 year monitoring period. However, higher mean total pesticide concentrations at 90 cm (102 µg L⁻¹)
29 relative to 45 cm (57 µg L⁻¹) depth indicated an accumulation of pesticide residues deeper within the
30 soil profile. Overall, the results presented here demonstrate that a three-stage biobed can
31 successfully reduce pesticide pollution risk from contaminated machinery washings on a commercial
32 farm.

33 **Keywords:** Biobed; pesticide; herbicide; biodegradation; water quality; arable

34 **1. Introduction**

35 The widespread use of pesticides in agriculture to kill plant and insect pests which would otherwise
36 reduce crop yields has been instrumental in enhancing global agricultural productivity since the mid-
37 20th century (Oerke and Dehne, 2004; Oerke, 2005; Clarke et al., 2011; Popp et al., 2013). However,
38 the harmful environmental impacts of applying toxic chemicals across large areas of the planet's
39 surface, particularly on the aquatic environment, are coming under increasing scrutiny (Skinner et
40 al., 1997; DeLorenzo et al., 2001; Schwarzenbach et al., 2010). High profile cases, such as the effect
41 of the insecticide DDT on the hatching success of raptors in the 1960s and 1970s, brought into focus
42 the potential for pesticides to bio-accumulate through the food chain and negatively impact upon
43 non-target species (Ames, 1966; Connell, 1988; Arnot and Gobas, 2006). Similarly, recent research
44 has linked the use of neonicotinoid insecticides to the decline of bee populations in Europe and
45 North America (Blacquiere et al., 2012; Whitehorn et al., 2012). Studies have also highlighted the
46 significant economic costs associated with removing pesticides from drinking water. Between 1991
47 and 2000, water companies in the United Kingdom spent £2 billion treating pesticide contaminated
48 water supplies (Jess et al., 2014), whilst in the United States the deleterious impacts of pesticide use
49 were estimated to cost \$9.6 billion in 2005 alone (Pimentel, 2005).

50 In order to tackle pesticide pollution, a range of national and international legislation is currently in
51 force. Under the EU Water Framework Directive (2000/60/EC), specifically the Drinking Water
52 (98/83/EC) and Groundwater (2006/118/EC) Directives, European Union member states must ensure
53 that no individual pesticide concentration in drinking water at the tap exceeds $0.1 \mu\text{g L}^{-1}$ and total
54 pesticide concentrations should not exceed $0.5 \mu\text{g L}^{-1}$. Additionally, the Pesticides Framework
55 Directive (2009/128/EC) aims to reduce the damage caused by pesticides through the adoption of
56 sustainable usage practices. In the United States, similar legislation exists under the Safe Drinking
57 Water Act (1974) which places individual concentration limits on specific pesticides.

58 Pesticide pollution can either arise from diffuse sources, such as spray drift, leaching and overland
59 flow, or from point sources, such as accidental spillages, leakages from equipment or from
60 contaminated machinery washings (Carter, 2000; De Wilde et al., 2007). Whilst diffuse sources can in
61 part be reduced by behavioural changes, such as timing of spraying to avoid periods of wet and
62 windy weather to limit pesticide mobility, biobeds have emerged as a potentially important

63 mitigation strategy for dealing with point source pollution (Fogg et al., 2003a; Reichenberger et al.,
64 2007; Karanasios et al., 2010; Omirou et al., 2012).

65 The biobed concept originated in Sweden in the 1990s as a way of using microbial activity to degrade
66 waste pesticide residues (Torstensson, 2000). A biobed is essentially a moderately sized pit (typically
67 tens of cubic metres in volume) which can be lined or unlined and is filled with a 1:2:1 matrix of
68 compost, straw and topsoil. The surface is covered with grass and onto this the waste pesticide
69 residues are deposited. In principle, microorganisms (e.g. bacteria and fungi) within the biobed
70 matrix chemically and physically interact with the pesticides leading to structural changes and/or
71 complete degradation (Pinto et al., 2016). To work effectively, the biobed mixture needs to have
72 high pesticide absorption capacity and be able to facilitate high rates of microbial activity (Castillo et
73 al., 2008). For this reason, straw is included to enhance microbial activity, particularly that of lignin-
74 degrading fungi (e.g. white rot fungi) which produce phenoloxidase enzymes that have a broad
75 specificity and are thereby able to degrade a wide range of pesticide residues (Bending et al., 2002).
76 Soil is included to increase the sorption capacity of the matrix material so that it holds onto the
77 pesticides and also provides a source of microorganisms for biodegradation. Lastly, compost is
78 added to increase sorption capacity, improve moisture content and decrease the pH to make
79 conditions favourable for fungi growth. The surface grass layer aids water regulation and prevents
80 surface crusting, thus limiting the formation of cracks that would open up preferential pathways for
81 pesticides to escape the biobed prior to degradation (Fogg et al., 2004; Castillo and Torstensson,
82 2007; Castillo et al., 2008). In lined biobed systems, common in the United Kingdom (UK), the
83 leachate is typically collected from the bottom of the biobed and re-used for either irrigation,
84 sprayer washing or as a carrier for further herbicide applications. Irrigation can be on infield crops or
85 a designated drainage area. In order to minimise pollution risk and comply with UK environmental
86 protection legislation, the drainage area must be vegetated, be neither frozen or water logged,
87 be >10 m away from any surface waterbody, be >50 m from any spring, well or borehole not used
88 for domestic supply or food production, and be >250 m away from any borehole that is used for
89 domestic supply or food production (Environment Agency, 2007).

90 Established in 2010, the River Wensum Demonstration Test Catchment (DTC) project is a part of a UK
91 government funded initiative to evaluate the extent to which on-farm mitigation measures can be
92 employed to cost effectively reduce the impacts of agricultural pollution on river ecology whilst
93 maintaining food production capacity (Outram et al., 2014). Draining a catchment area of 660 km² in
94 Norfolk, UK, of which ~63% is arable land, the River Wensum supplies drinking water for the city of
95 Norwich and is affected by agricultural pesticide pollution. A small unpublished water quality

96 monitoring study carried out at 20 locations on the River Wensum over a 16-week period in autumn
97 2012, revealed that 23% of samples contained individual pesticide concentrations greater than the
98 0.1 $\mu\text{g L}^{-1}$ drinking water limit. Five key pesticides (metaldehyde, metazachlor, dimethenamid,
99 flufenacet and propyzamide) accounted for 90% of all detected compounds, with 21% of samples
100 containing metaldehyde concentrations $>1 \mu\text{g L}^{-1}$ (further details of this study can be found in the
101 electronic supplementary material). Partly in response to this pesticide pollution pressure, an on-
102 farm biobed unit capable of treating contaminated machinery washings was installed at Manor
103 Farm, Salle, in the Blackwater sub-catchment of the River Wensum. This was part of a trial package
104 of on-farm mitigation measures, co-funded under the Catchment Sensitive Farming (CSF) initiative
105 (Natural England, 2014), aimed at reducing agricultural pollution.

106 The primary objectives of this paper are as follows:

- 107 (i) To assess the efficiency of the Manor Farm biobed at reducing pesticide concentrations in
108 agricultural machinery washings;
- 109 (ii) To assess the effectiveness of drainage field irrigation at further reducing pesticide
110 concentrations in biobed leachate;
- 111 (iii) To determine if biobed pesticide removal is more efficient for certain types of pesticide;
- 112 (iv) To assess temporal variability in the effectiveness of the biobed.

113

114 **2. Methods**

115 **2.1 Study Location**

116 This study focuses upon a biobed unit installed in 2013 at Manor Farm, Salle Park Estate, Norfolk, UK
117 ($52^{\circ}46'57''\text{N}$, $01^{\circ}08'07''\text{E}$). The large, commercial Salle Park Estate covers 20 km^2 of which 79% is
118 intensive arable land managed with a seven-year crop rotation of winter wheat, winter and spring
119 barley, winter oilseed rape, spring beans and sugar beet. The estate also comprises 15% improved
120 grassland, 5% mixed woodland and 1% rural settlements. Across the estate, 16,387 litres of
121 concentrated liquid pesticide and 1,230 kg of solid pesticide granules were applied in 2014, the
122 majority of which was applied during spring (March – May). Prior to the installation of the biobed,
123 the risk of pesticide pollution occurring was relatively high. Farm machinery was washed down in the
124 farmyard on concrete hard standing and the wastewater was collected in a drain with an isolation

125 valve from where it was subsequently transported to a designated disposal area 0.8 km from the
126 farm. However, the drain isolation valve was manually operated and human error could result in the
127 contaminated washings discharging directly into a nearby pond.

128

129 **2.2 Biobed Facility**

130 The Manor Farm biobed facility consists of three main components (Figures 1 and 2):

131 **(i) Wash-down unit:** a 20 m x 9 m enclosed concrete wash-down unit is used to both
132 remove pesticides residues from farm machinery and to contain any pesticides spilt
133 during the filling of the pesticide sprayer. A drain running down the centre of the unit
134 channels contaminated washings into a concrete storage tank (the input sump);

135 **(ii) Biobed:** the biobed itself is an uncovered, indirect, lined (impermeable geomembrane)
136 design covering an area of 49 m² (7 m x 7 m) to a depth of 1.2 m, thus providing a large
137 surface area for biological and photo-degradation. The organic bio-mix matrix material is
138 composed of a 1:2:1 mix of peat-free compost, chopped wheat/barley straw and local
139 topsoil. The surface is seeded with grass. Contaminated water from the input sump is
140 pumped onto the biobed surface via a trickle irrigation system, with the leachate
141 collected at the base of the biobed in a concrete output sump;

142 **(iii) Drainage field:** the leachate from the output sump is pumped onto a 200 m² (20 m x 10
143 m) drainage field via a second trickle irrigation system buried just below the surface to
144 promote further removal of residual pesticide residues. This drainage field is covered
145 with grass and is surrounded by seven mature trees. A network of 20 porous pots were
146 installed (30° angle) across the drainage area at 45 cm and 90 cm depth (ten pots for
147 each) to monitor soil water pesticide concentrations at depth for signs of further
148 removal or accumulation. As far as the authors are aware, this is the first time that
149 pesticide removal in a drainage field on a commercial farm has been routinely
150 monitored.

151 The biobed is designed to treat >15,000 L of contaminated wastewater from the wash-down unit
152 per year. The trickle irrigation pumps are controlled by float-switches within the input and
153 output sumps so that irrigation commences automatically once the water depth within the
154 sumps has reached a predefined level. During the winter, the irrigation systems are switched off
155 to prevent ice damage.

156

157 **2.3 Sample Collection**

158 Water samples were collected from the input and output sumps and the 45 cm and 90 cm porous
159 pots at approximately two week intervals between November 2013 and November 2015. No
160 sampling took place between June 2014 and November 2014 due to a hiatus in funding. On each
161 sampling occasion eight water samples were collected to enable a range of analyses – three from
162 each of the input and output sumps and one each from the 45 cm and 90 cm porous pots. Water
163 from the sumps was collected using a stainless steel bucket lowered into the chambers on a chain
164 and was decanted into a 1 L glass bottle (sample code = PESTP) and two 250 mL polyethylene
165 terephthalate (PET) bottles for each sump. To preserve the samples, one PET bottle had 2 mL of 3
166 molar formic acid added (HERBP), whilst the other contained 2 mL of 2.65 molar formic acid and 5
167 molar ammonium acetate (URON). For the drainage field, each 45 cm and 90 cm porous pot was put
168 under vacuum for 20 minutes to extract soil water. Recovered soil water was bulked together to
169 produce a single sample for each depth and was decanted into a 250 mL PET bottle containing 2 mL
170 of 3 molar formic acid preservative (HERBP). The volume of soil water collected varied seasonally
171 depending on soil moisture conditions, with up to 200 mL collect during the winter and <50 mL
172 collected during the summer. Throughout summer and autumn 2015, dry soil conditions meant no
173 samples could be collected from the 45 cm porous pots. Note that in any given week, samples
174 collected from the input sump, output sump and the drainage field did not correspond to the same
175 body of contaminated water. Instead, samples collected from the drainage field corresponded to
176 water that was in the output sump several days/weeks prior to sampling.

177

178 **2.4 Sample Analysis**

179 All samples were analysed by the Environment Agency's National Laboratory Service. Three different
180 analytical techniques were employed to determine a wide variety of pesticide compounds:

- 181 (i) Phenoxy acidic herbicides (HERBP): a 1000 μ L aliquot was transferred into a silanised vial
182 and an internal standard was added. 400 μ L of the sample was then injected into a high
183 performance liquid chromatograph (HPLC) interfaced to a triple quadrupole mass
184 spectrometer (TQMS) operated in positive and negative atmospheric pressure electrospray

185 mode. Tandem mass spectroscopy data (MS/MS) were acquired in multiple reaction
186 monitoring mode;

187 (ii) Phenyl urea herbicides, *n*-methyl carbamates, fungicides and asulam (URON): a 1000 μL
188 aliquot was transferred into a silanised vial and ethylenediaminetetraacetic acid (EDTA) and
189 an internal standard were added. A 100 μL sample was then injected into the HPLC and
190 analysed as for HERBP;

191 (iii) Triazines, organophosphorus and miscellaneous pesticides (PESTP): pesticides were
192 extracted into dichloromethane using liquid-liquid extraction. The extract was then
193 concentrated and injected into a gas chromatograph interfaced with a mass spectrometer
194 (GC-MS) operating in electron ionisation mode. The collected results were then compared
195 with data obtained from a series of similarly treated standard solutions in data handling
196 software;

197 In total, 86 pesticides were detected and here we primarily focus on 15 compounds which were
198 regularly used, had high input concentrations ($>100 \mu\text{g L}^{-1}$) and/or are CSF key indicator pesticides.
199 The physico-chemical properties of these pesticides, which are all herbicides and which accounted
200 for $\sim 98.6\%$ of all compounds measured in the input sump, are presented in Table 1. Insufficient
201 water was collected from the drainage field to enable the full suite of analyses to be carried out and
202 therefore the porous pot analysis was restricted to a smaller number of compounds (HERBP only).

203

204 3. Results

205 3.1 Total Pesticide Concentration

206 The total concentrations for all 86 pesticides measured at the four monitoring points between
207 November 2013 and November 2015 are shown in Figure 3. Mean pesticide concentrations over this
208 period were: $43,334 \mu\text{g L}^{-1}$ (range = $1037\text{--}508,873 \mu\text{g L}^{-1}$) in the input sump; $3647 \mu\text{g L}^{-1}$ ($47\text{--}42,260$
209 $\mu\text{g L}^{-1}$) in the output sump; $57 \mu\text{g L}^{-1}$ ($0.5\text{--}192 \mu\text{g L}^{-1}$) in the 45 cm depth porous pots; and $102 \mu\text{g L}^{-1}$
210 ($2\text{--}396 \mu\text{g L}^{-1}$) in the 90 cm depth porous pots. Overall, this corresponds to a 91.6% reduction in
211 pesticide concentration between the biobed input and output sumps, with a further 98.4% and
212 97.2% reduction between the output sump and the 45 cm and 90 cm drainage field porous pots,
213 respectively. Substantial temporal variability in the input sump concentrations reflect both variations
214 in the amount of pesticide being applied across the farm at any one time and in the amount of water

215 used during the washing of farm machinery (i.e. lower pesticide concentrations result when more
216 water is used). Similarly, fluctuations in the output sump and porous pot concentrations will also
217 reflect variability in precipitation which has the potential to both dilute and flush out pesticide
218 residues within the biobed and drainage field.

219

220 **3.2 Individual Pesticide Concentrations**

221 Individual pesticide concentration data for the 15 key pesticides are presented in Table 2. The
222 highest mean pesticide concentration recorded in the input sump ($26,935 \mu\text{g L}^{-1}$) was for
223 ethofumesate, a widely applied herbicide to kill grass and broadleaf weeds in sugar beet crops.
224 The lowest mean concentration ($15.3 \mu\text{g L}^{-1}$) recorded was for carbetamide, a grass/broadleaf
225 herbicide applied to oilseed rape. The efficiency of individual pesticide reduction between the
226 input and output sumps ranged from 97.6% for propyzamide to 68.4% for metazachlor, with
227 seven out of 15 pesticides achieving >90% reduction in mean concentration. Mean concentrations
228 in the 45 cm depth drainage field porous pots varied between $1.1 \mu\text{g L}^{-1}$ for bromoxynil and
229 MCPA, to $9.3 \mu\text{g L}^{-1}$ for fluroxypyr. Similarly, in the 90 cm porous pots, bromoxynil and MCPA had
230 the lowest mean concentrations ($1.6 \mu\text{g L}^{-1}$), whilst clopyralid had the highest concentration (16.2
231 $\mu\text{g L}^{-1}$). The efficiency of pesticide removal between the output sump and the 45 cm porous pots
232 ranged from 99.0% for 2,4-D to 77.1% for MCPA, whilst in the 90 cm porous pots efficiencies
233 ranged from 97.0% for 2,4-D to 68.3% for dicamba.

234

235 **4. Discussion**

236 **4.1 Biobed Efficiency**

237 The biobed proved to be highly effective in reducing the concentrations of pesticide within the
238 contaminated machinery washings, lowering total pesticide concentrations by an average of 91.6%.
239 This compares with pesticide removal efficiencies of 52–100% recorded for a wide range of
240 chemicals in other biobed studies conducted across Europe (De Wilde et al., 2007). Nevertheless, the
241 mean total pesticide concentration ($3647 \mu\text{g L}^{-1}$) and the mean concentrations of individual
242 pesticides ($3\text{--}1755 \mu\text{g L}^{-1}$) within the output sump remained sufficiently large to pose an
243 environmental risk. These output concentrations are consistent with the results of similar studies
244 assessing biobed removal efficiencies (e.g. Spliid et al., 2006). Irrigation of the biobed leachate in the

245 drainage field was therefore necessary for promoting further pesticide removal. In the top 45 cm of
246 the soil, total pesticide concentrations were reduced by 98.4% to $57 \mu\text{g L}^{-1}$, whilst individual pesticide
247 concentrations were reduced by 77.1–99.0% to $1.1\text{--}9.3 \mu\text{g L}^{-1}$. These results clearly demonstrate that
248 collecting the leachate from the biobed output sump and applying it onto a drainage field to allow
249 further pesticide removal within the soil profile is essential to reduce concentrations down to more
250 environmentally acceptable levels and represents a significant reduction in risk over the previous
251 farm practice described in Section 2.1.

252

253 **4.2 Individual Pesticide Removal**

254 With the mean pesticide removal efficiency varying by 29.2% between the best (propyzamide) and
255 worst (metazachlor) performing herbicide, it is apparent that the degree of removal achieved is
256 dependent upon the chemical structure of the pesticides used. The environmental mobility and
257 persistence of any given pesticide is primarily controlled by its soil sorption characteristics, water
258 solubility and half-life (Arias-Estévez et al., 2008). Highly soluble pesticides with low sorption
259 capacity will tend to move more quickly through the biobed matrix than pesticides with high
260 sorption capacity, and this reduced residence time will diminish the opportunities for
261 microorganisms to degrade these chemicals (i.e. bioavailability will be reduced) (Spliid et al., 2006;
262 De Wilde et al., 2007). Furthermore, most pesticides are degraded by co-metabolic processes. By
263 metabolising constituents within the biobed (e.g. straw), bacteria and fungi produce enzymes which
264 are able to break down toxic chemicals that they otherwise would not be able to degrade (Castillo
265 and Torstensson, 2007). However, different pesticide chemical structures have different
266 susceptibility to the oxidative enzymes produced by bacteria and fungi (Ferris and Lichtenstein,
267 1980), and therefore even pesticides with a high sorption capacity that are retained within the
268 biobed may experience low degradation rates.

269 Evidence of these processes can be seen in Figure 4, which shows the relationships between biobed
270 removal efficiency and the typical soil sorption (K_{oc}), water solubility and half-life (DT_{50}) values of the
271 15 pesticides monitored here (data from Lewis et al. (2016)). Despite considerable scatter, there is a
272 positive linear relationship ($R^2 = 0.19$, $p = 0.10$) between soil sorption and removal efficiency, with
273 five out of six pesticides with the highest sorption coefficients ($K_{oc} >100$) having high removal
274 efficiencies (>93%). Similarly, there is a significant negative relationship ($R^2 = 0.28$, $p = 0.04$)
275 between pesticide solubility and removal efficiency, with the six least soluble ($<440 \text{ mg L}^{-1}$)
276 pesticides exhibiting the highest levels of removal (>93%). A significant positive relationship ($R^2 =$

277 0.34, $p = 0.02$) is also apparent between removal efficiency and pesticide half-life, indicating that
278 more persistent pesticides were removed from the leachate more readily than less persistent
279 compounds. However, pesticide sorption coefficients are strongly and significantly correlated with
280 both solubility ($r = -0.79$, $p < 0.01$) and DT_{50} ($r = 0.50$, $p < 0.05$) and this in part helps to explain the
281 positive and negative relationships observed between removal efficiency and DT_{50} and solubility,
282 respectively. In general, pesticides with higher soil sorption coefficients, lower solubility and longer
283 half-lives experienced the greatest removal rates within the Manor Farm biobed.

284

285 **4.3 Pesticide Accumulation**

286 Although total pesticide concentrations were reduced by 98.4% between the output sump and the
287 45 cm porous pots, the mean total pesticide concentration in the 90 cm drainage field porous pots
288 ($102 \mu\text{g L}^{-1}$) was nearly double that recorded at 45 cm depth ($57 \mu\text{g L}^{-1}$) (Figure 3). Similarly, all
289 individual pesticide concentrations were higher at 90 cm depth compared with 45 cm (Table 2),
290 indicating an accumulation of pesticides residues at depth within the drainage field. A potential
291 explanation for this observation comes from examining 1 m depth soil cores taken from the drainage
292 field during porous pot installation which revealed that a silty clay layer dominates the upper 0.5 m
293 whereas sandier material dominates at 0.5–1.0 m depth (Lewis, 2011; Figure SM2 in supplementary
294 material). The clay-rich surface layer would be expected to favour greater pesticide attenuation via
295 sorption onto soil, thus lowering pesticide concentrations in the pore water extracted for analysis.
296 Conversely, the sandier layer at depth would be expected to have lower sorption capacity, thus
297 leaving higher pesticide concentrations in the pore water collected in the porous pots. Additionally,
298 desiccation and fissuring of the surface clay-rich layer could form preferential flow paths deeper into
299 the soil profile, potentially allowing the pesticide leachate to bypass the aerobic surface layers
300 where most biological degradation occurs. Ultimately, these processes could result in the drainage
301 field itself acting as a point source of pesticide pollution, particularly if interactions with
302 groundwater increase the lateral mobility of the pesticide residues. These findings emphasise the
303 importance of drainage field design and siting in maximising the removal of pesticides and
304 minimising potential off-site transport.

305

306 **4.4 Temporal Trends**

307 Successful removal of pesticides within a biobed is dependent upon the biobed matrix supporting a
308 high level of microbial activity and, as such, temperature and moisture content are important factors
309 in determining biobed efficiency. A study by Castillo and Torstensson (2007) demonstrated higher
310 rates of pesticide dissipation when the biobed temperature was at 20°C (compared to 5°C and 10°C)
311 and moisture levels were at 60% (compared to 30% or 90%) of the water holding capacity.
312 Therefore, it might be expected that greater pesticide removal will occur during the summer when
313 temperatures are higher, provided the biobed matrix maintains high moisture content. However,
314 there was no clear evidence of such a trend with the Manor Farm biobed (Figure 3), suggesting that
315 temperature and moisture content may be secondary factors in determining the performance of
316 operational biobeds when compared with laboratory studies. Mean pesticide removal efficiencies
317 between the input and output sumps were 94.5% during the winter (DJF), 97.5% during the spring
318 (MAM) and 92.5% during the summer (JJA). Only autumn (SON), with an efficiency of 75.1%, had
319 significantly lower pesticide removal. This was predominantly due to the very high concentrations of
320 metazachlor recorded in the input (up to 73,900 $\mu\text{g L}^{-1}$) and output (up to 27,900 $\mu\text{g L}^{-1}$) sumps
321 during September – October 2015 after spraying of the autumn sown oilseed rape crop. Prior to
322 autumn 2015, concentrations of metazachlor in the input sump were relatively low (mean = 192 μg
323 L^{-1}) and the efficiency of biobed removal was high (mean = 94.9%). However, the removal efficiency
324 declined sharply in autumn 2015 (mean = 63.4%), indicating that the biobed was unable to cope with
325 very high metazachlor loading. Although none of the other 14 pesticides analysed here
326 demonstrated this behaviour, similar declines in removal efficiency due to high pesticide loadings
327 have previously been reported in other biobed studies (Fogg et al., 2003b; Vischetti et al., 2008). The
328 effect of poor metazachlor removal in autumn 2015 reduced the overall biobed total pesticide
329 removal efficiency by 2.8%, from 94.4% to 91.6%.

330 In the UK, it is suggested that the entire biobed matrix is replaced every five years since
331 decomposition of organic matter gradually reduces the efficiency of pesticide removal (Castillo et al.,
332 2008). Over the two-year monitoring period of this study, there was no evidence of a reduction in
333 the biobed performance, with mean biobed removal efficiencies of 91.1% prior to July 2014 and
334 91.6% after December 2014.

335

336 **4.5 Biobed Maintenance**

337 The biobed facility required limited maintenance following its construction in 2013. The biobed
338 matrix was topped up with fresh material in July 2015 after two years of operation as decomposition

339 of organic material had reduced the depth of the bio-mix. At the same time, some re-profiling of the
340 biobed surface was carried out to address slumping in one corner which was causing minor runoff
341 away from the biobed onto the adjacent grassed area. Previous research by Fogg et al. (2004) found
342 that uncovered lined biobeds treating large volumes of machinery washings, such as this one here,
343 can become waterlogged without some form of water management, thus resulting in reduced
344 microbial activity and lower rates of pesticide degradation. Some evidence of water accumulation on
345 the surface of the Manor Farm biobed was observed during very heavy rainfall events, although such
346 incidences were infrequent and of short duration. There was no evidence of reduced biobed
347 performance during the winter when the matrix moisture content would be at its highest level. This
348 confirms that the biobed design was appropriate for handling machinery washings from the Salle
349 Park Estate.

350

351 **4.6 Implications and Economics**

352 The results presented here clearly demonstrate the effectiveness of a straw-compost-topsoil biobed
353 at reducing pesticide residues in substantial volumes of contaminated water generated from
354 machinery washings on a large, arable farm. It is also clear that further treatment of the biobed
355 leachate by irrigating the contaminated water through the soil profile of a substantially sized
356 drainage field is beneficial to further reduce pesticide concentrations down to environmentally
357 acceptable levels. Furthermore, the enclosed sprayer wash-down area provides a secure
358 environment when handling pesticide concentrate during sprayer filling operations, thus minimising
359 the risk of accidental spillage leading to surface water contamination. Wider scale adoption of
360 biobeds as an on-farm mitigation measure could therefore result in a significant reduction in point
361 source pesticide pollution of streams and rivers draining agricultural catchments. Biobeds are
362 effective in reducing the risks associated with farm pesticide spraying operations since they contain
363 and breakdown pesticides in effluent that could otherwise escape the farm via drainage water.
364 Hence, biobeds are an efficient pesticide reduction measure and are an important tool used by
365 catchment level pollution reduction schemes such as Catchment Sensitive Farming (Environment
366 Agency, 2014; Natural England, 2014). The farmers of the Salle Park Estate also reported that the
367 three-stage biobed significantly improved the efficiency of pesticide handling operations, with
368 pesticide dispensing, machinery washing and wastewater disposal now occurring at a single, purpose
369 built facility.

370 Table 3 lists the approximate construction costs for the three main components of the Manor Farm
371 biobed. Whilst total costs were £96,827, the majority of this (£90,454) was for building the large,

372 insulated, wash-down unit and equipping it with mains electricity and steam cleaning equipment.
373 Such a high quality design is not essential to achieve good operational performance and much
374 simpler facilities would be more appropriate for wider deployment across multiple farms within a
375 catchment. The cost of the biobed itself, which included the pipework, pumps, liner, matrix material
376 and labour, was relatively inexpensive (£4311). Replenishment of the matrix material two years after
377 construction cost £8 m⁻². The cost of the drainage field infrastructure was approximately £1684, of
378 which the porous pots accounted for £1466. Installing porous pots in other commercial biobeds
379 would not be necessary as their installation here was purely for research purposes. Much simpler
380 designs could likely be constructed for £5000–10,000, increasing the feasibility of uptake by a larger
381 number of farms, particularly if such measures were financially incentivised under government agri-
382 environment schemes.

383

384 **5. Conclusion**

385 Pesticide pollution threatens the sustainable ecosystem functioning of rivers draining agricultural
386 catchments and therefore mitigation measures are required to reduce the amount of pesticides
387 entering freshwater environments. In this study, we have demonstrated how an on-farm biobed is
388 capable of reducing the risk of point source pesticide pollution by substantially decreasing pesticide
389 concentrations in large volumes of contaminated machinery washings from a 20 km² arable estate.
390 The three-stage biobed facility, consisting of an enclosed machinery wash-down unit, a 49 m² lined
391 compost-straw-topsoil biobed and a 200 m² drainage field, provided an efficient and secure
392 environment for pesticide handling and mixing operations, containing contaminated washings and
393 removing waste pesticide residues. Water quality monitoring over a two-year period revealed
394 individual pesticide concentrations reduced by 68–98% between the biobed input and output
395 sumps, with mean total pesticide concentrations reducing by 91.6%. Further treatment of the
396 contaminated washings in the drainage field removed an additional 68–99% of individual residual
397 pesticides, with total mean pesticide concentrations reducing by a further 98.4% and 97.2% in the 45
398 cm and 90 cm depth porous pots, respectively. Mean total pesticide concentrations at 45 cm depth
399 (57 µg L⁻¹) after drainage field irrigation were 760 times lower than that recorded in the untreated
400 machinery washings (43,334 µg L⁻¹). Although the treated effluent still requires careful handling to
401 avoid contaminating freshwater bodies, this nevertheless represents a substantial reduction in
402 groundwater pesticide pollution risk compared with the previous farm practice of disposing of
403 untreated waste washings in a designated disposal area. The biobed has also reduced the risk of
404 point source surface water pollution by removing reliance upon a manually operated isolation valve

405 to prevent contaminated washings discharging directly into a farm pond. No evidence of seasonality
406 in the efficiency of pesticide removal was detected, nor was there any evidence of a decline in
407 biobed performance over the two-year monitoring period. However, elevated pesticide
408 concentrations at 90 cm depth within the drainage field potentially indicate an accumulation of
409 pesticide residues deeper within the soil profile which could pose a risk to groundwater quality.
410 Nevertheless, the results presented here clearly demonstrate the effectiveness of a three-stage on-
411 farm biobed at reducing pesticide residues in substantial volumes of contaminated water generated
412 from machinery washing on a large, commercial arable farm.

413

414 **Acknowledgements**

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423

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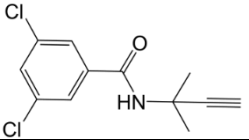
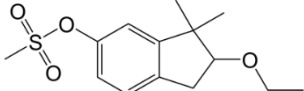
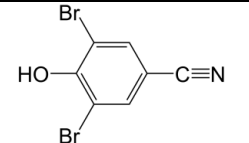
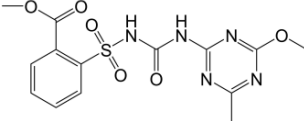
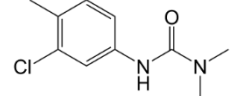
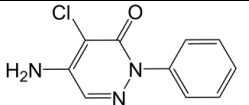
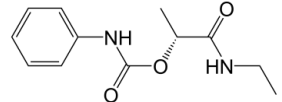
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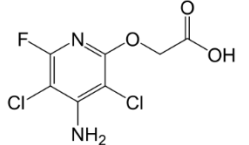
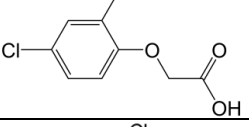
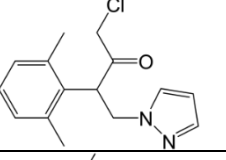
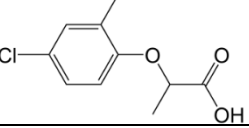
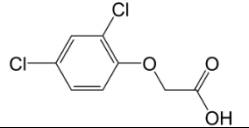
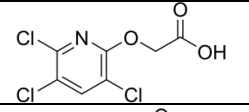
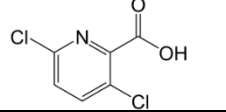
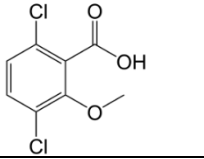
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Table 1: Summary of the 15 pesticides analysed in the Manor Farm biobed, Salle, which were either regularly used, had high input concentrations (>100 µg L⁻¹) or are CSF key indicator pesticides. Typical physico-chemical characteristics derived from Lewis et al. (2016).

Pesticide	Formula	2D Structure	Type	Primary Crop Use	Soil Sorption Coefficient (K_{oc} mL g ⁻¹)	Solubility in Water (mg L ⁻¹)	Half Life in Field DT ₅₀ (days)
Propyzamide	C ₁₂ H ₁₁ Cl ₂ NO		Grass/broadleaf herbicide	Oilseed rape/field beans	840 (Slightly mobile)	9 (Low)	56 (Moderately persistent)
Ethofumesate	C ₁₃ H ₁₈ O ₅ S		Grass/broadleaf herbicide	Sugar beet	55-500 (Moderately mobile)	50 (Moderate)	37.8 (Moderately persistent)
Bromoxynil	C ₇ H ₃ Br ₂ NO		Broadleaf herbicide	Cereals	302 (Moderately mobile)	90 (Moderate)	8 (Non-persistent)
Metsulfuron-methyl	C ₁₄ H ₁₅ N ₂ O ₆ S		Grass/broadleaf herbicide	Cereals	120-320 (Moderately mobile)	2,790 (High)	13.3 (Non-persistent)
Chlorotoluron	C ₁₀ H ₁₃ ClN ₂ O		Grass/broadleaf herbicide	Cereals	196 (Moderately mobile)	74 (Moderate)	34 (Moderately persistent)
Chloridazon	C ₁₀ H ₈ ClN ₃ O		Broadleaf herbicide	Sugar beet	120 (Moderately mobile)	422 (Moderate)	34.7 (Moderately persistent)
Carbetamide	C ₁₂ H ₁₆ N ₂ O ₃		Grass/broadleaf herbicide	Oilseed rape	89 (Moderately mobile)	3,270 (High)	8 (Non-persistent)

Fluroxypyr	$C_7H_5Cl_2FN_2O_3$		Broadleaf herbicide	Cereals	74 (Mobile)	6,500 (High)	51 (Moderately persistent)
MCPA	$C_9H_9ClO_3$		Broadleaf herbicide	Cereals	74 (Mobile)	29,390 (High)	25 (Non-persistent)
Metazachlor	$C_{14}H_{16}ClN_3O$		Grass/broadleaf herbicide	Oilseed rape	54 (Mobile)	450 (Moderate)	6.8 (Non-persistent)
Mecoprop	$C_{10}H_{11}ClO_3$		Broadleaf herbicide	Cereals	47 (Mobile)	250,000 (High)	8.2 (Non-persistent)
2,4-D	$C_8H_6Cl_2O_3$		Broadleaf herbicide	Cereals	39.3 (Mobile)	24,300 (High)	28.8 (Non-persistent)
Triclopyr	$C_7H_4Cl_3NO_3$		Broadleaf herbicide	Cereals	27 (Mobile)	440 (Moderate)	30 (Moderately persistent)
Clopyralid	$C_6H_3Cl_2NO_2$		Broadleaf herbicide	Cereals/oilseed rape	5 (Very mobile)	143,000 (High)	11 (Non-persistent)
Dicamba	$C_8H_6Cl_2O_3$		Broadleaf herbicide	Cereals	2 (Very mobile)	250,000 (High)	3.9 (Non-persistent)

1 **Table 2:** Mean concentration data for 15 pesticides which were either regularly used, had high
 2 input concentrations (>100 µg L⁻¹) or are CSF key indicator pesticides. Data are for the period
 3 November 2013 to November 2015. The efficiency of the biobed sumps refers to the reduction
 4 in pesticide concentration between the input and output sumps. The efficiencies of the porous
 5 pots reflect the reductions in pesticide concentration between the output sump and the 45 cm
 6 and 90 cm porous pots. Missing values relate to non-detected pesticides.

Pesticide	Biobed Sump			Porous Pot			
	Mean Concentration (µg L ⁻¹)			Mean Concentration (µg L ⁻¹)			
	Input	Output	Efficiency (%)	45 cm	Efficiency (%)	90 cm	Efficiency (%)
Propyzamide	2551.3	60.0	97.6	-	-	-	-
Chloridazon	2547.7	81.9	96.8	-	-	-	-
Triclopyr	958.5	32.8	96.6	1.2	96.3	2.5	92.4
Ethofumesate	26935.1	980.9	96.4	-	-	-	-
Chlorotoluron	150.4	6.9	95.4	-	-	-	-
Bromoxynil	167.3	11.3	93.2	1.1	90.3	1.6	85.8
2,4-D	2944.9	213.7	92.7	2.2	99.0	6.5	97.0
Mecoprop	803.7	112.7	86.0	3.0	97.3	6.6	94.1
MCPA	30.4	4.8	84.2	1.1	77.1	1.6	66.7
Fluroxypyr	1162.0	224.6	80.7	9.3	95.9	16.0	92.9
Dicamba	223.5	43.8	80.4	9.1	79.2	13.9	68.3
Carbetamide	15.3	3.0	80.4	-	-	-	-
Clopyralid	1025.5	238.1	76.8	5.5	97.7	16.2	93.2
Metsulfuron-methyl	32.9	8.1	75.4	-	-	-	-
Metazachlor	5561.0	1754.9	68.4	-	-	-	-

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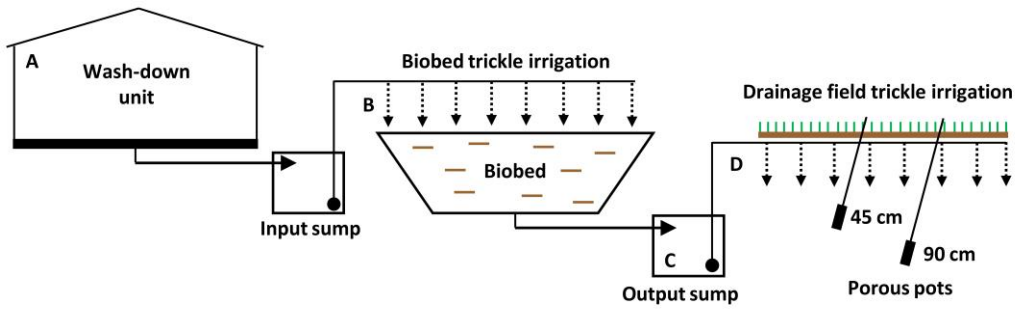
9 **Table 3:** Approximate construction costs (including labour) for the Manor Farm biobed installed in
 10 2013.

Component	Area (m ²)	Cost	
		(£)	(£ m ⁻²)
Sprayer wash-down area	270	90,454	335
Biobed	49	4311	88
Drainage field	200	1684	8
Matrix replenishment after 2 years	49	378	8
Total cost		96,827	

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13 **Figures**



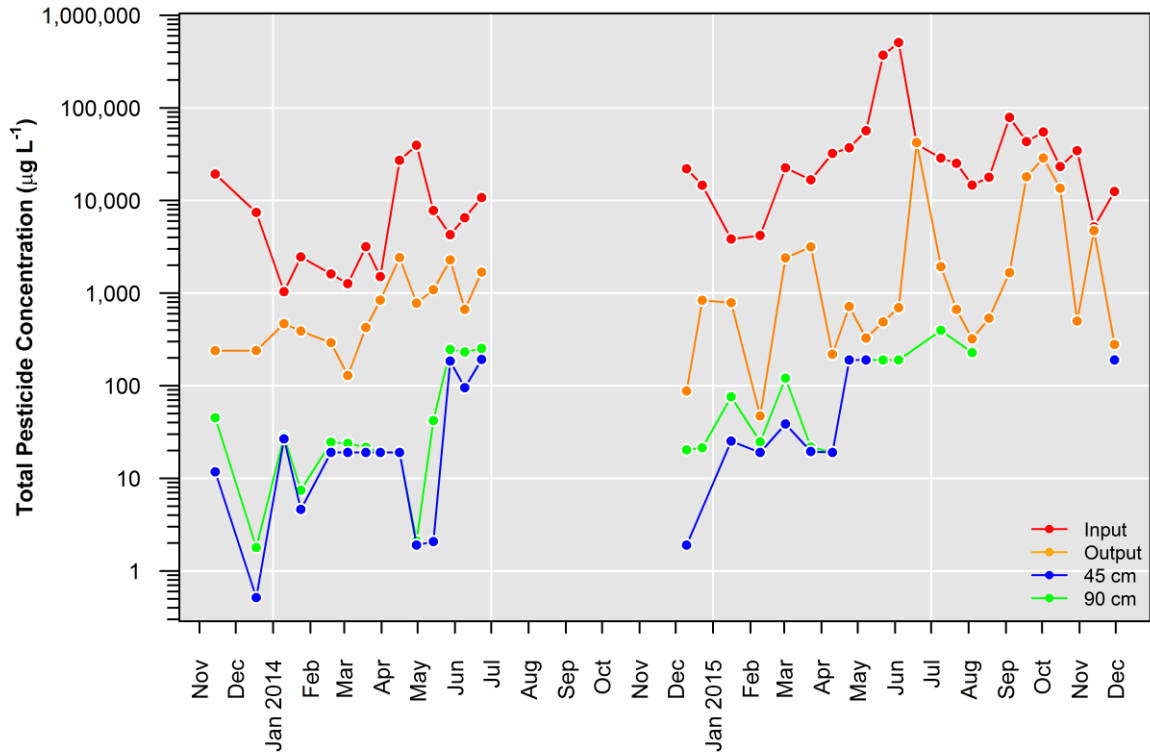
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Figure 1: Schematic of the biobed unit installed at Manor Farm, Salle. Letters refer to the photographs in Figure 2.



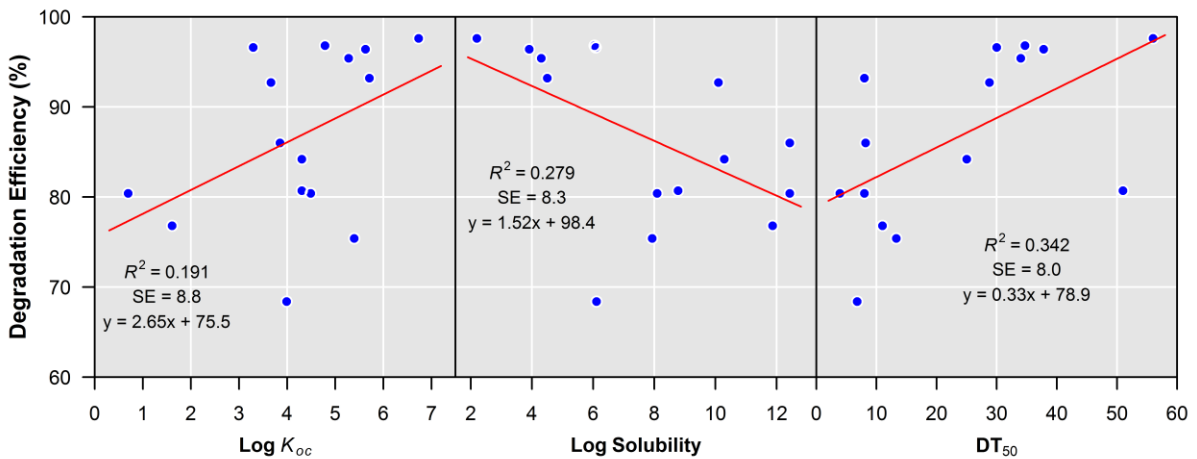
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Figure 2: Images of the biobed facility installed at Manor Farm, Salle. (A) Pesticide sprayer inside the machinery wash-down unit during construction; (B) biobed operational area (7 m x 7 m) with the completed enclosed wash-down unit in the background; (C) biobed output sump and trickle irrigation system during construction; (D) drainage field trickle irrigation area, with porous pot outlets located underneath terracotta pots.



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 28 Figure 3: Total pesticide concentrations recorded in the input and output sumps and in the drainage
 29 field porous pots (45 cm and 90 cm depth) between November 2013 and November 2015.

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 33 Figure 4: Linear regression relationships between biobed removal efficiency at the output sump and
 34 the typical physico-chemical properties of the 15 key pesticides monitored. Physico-chemical
 35 properties derived from Lewis et al. (2016).