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5

6 **SPATIAL AND TEMPORAL VARIATIONS OF NUTRIENT LOADS IN**
7 **OVERLAND FLOW AND SUBSURFACE DRAINAGE FROM A MARGINAL LAND**
8 **SITE IN SOUTH EAST IRELAND**

9

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18 **Keywords:** Grasslands, Nutrient losses, Food Harvest 2020, Sustainable Intensification

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20

21 **ABSTRACT**

22 In Ireland, Food Harvest 2020 focuses on increasing productivity whilst enhancing
23 environmental sustainability of agricultural land. On under-utilised or marginal land, drainage
24 systems may be installed to expand agricultural enterprises. Mixed nutrient losses are
25 inevitable from any drained system, but assessing processes leading to differences in nutrient
26 speciation, fractionation and losses in grasslands between locations or flowpaths is important
27 to achieve sustainability. This study investigates these processes in overland flow and
28 subsurface drains over three rainfall events from four non-grazed plots recently converted
29 from marginal land in the southeast of Ireland. A shallower water table and smaller plot size
30 resulted in greater water and nutrient losses in overland flow per unit of land area. Nutrient
31 losses were less in subsurface drains. Dissolved organic nitrogen dominated, but dissolved
32 inorganic nitrogen was more abundant in the drains. Particulate phosphorus generally
33 dominated in drains, except in plots with a shallow water table where dissolved unreactive
34 phosphorus (DUP) was more abundant. In overland flow, a shallower water table resulted in a
35 switch from dissolved reactive phosphorus (DRP) to DUP. Fertilization strongly increased
36 phosphorus losses in overland flow, with DRP dominating. These results highlight the
37 importance of an integrated assessment of the controls on flow and nutrient losses to design
38 drainage systems in marginal lands.

39 INTRODUCTION

40 In Ireland, Food Harvest 2020 (Anon 2010) outlines the future vision for Irish agriculture. It
41 focuses on increasing productivity while enhancing the environmental sustainability of
42 agricultural land. Within this framework, abolition of European Union (EU) dairy milk quota
43 in 2015 (CEC 2008) will allow for the expansion of the dairy sector. Approximately 29% of
44 Irish grasslands have poorly-drained soils (EEA 2009; Fealy *et al.* 2009). On such marginal
45 land, the water table in floodplains or in areas of a perched aquifer is often shallow (Misstear
46 *et al.* 2009). These conditions, as well as high rainfall, result in excess water in the soil,
47 which can strongly reduce grass yields, as well as limit access to the fields by grazing
48 animals and machinery during wet periods (Mulqueen 1985; Brereton and Hope-Cawdery
49 1988; Armstrong and Garwood 1991; Lalor and Schulte 2008). Often in these areas, artificial
50 drainage systems need to be installed to allow sustainable grass production (Galvin 1983).
51 Thus, implementation of designed drainage systems (artificial pipes with drain spacing and
52 depth estimated using soil physical parameters) and renovation or maintenance of existing
53 systems (tile drains and mole drains) on under utilised or marginal land, is likely to increase
54 in the coming years in order to develop new grassland areas for use by the dairy sector. In
55 addition, previously drained land will need modernization.

56 In agricultural landscapes, nutrients in water can originate directly through fertilizer
57 application or animal excreta, or indirectly due to the effect of poaching of land by cattle on
58 nutrient infiltration and cycling in soils (Richards *et al.* 2009; Watson and Foy 2001).
59 Enhancing the drainage capacity of poorly-drained soils can increase or decrease losses of
60 these nutrients to the environment (Skaggs *et al.* 1994). Artificially lowering the water table
61 can also result in bypassing areas of high natural attenuation and further increase nutrient
62 losses (Gold *et al.* 2001). In turn, increasing nutrient loading to surface water and

63 groundwater bodies above the natural recycling or retention capacity of these ecosystems can
64 lead to: 1) exceeding drinking water quality standards, as defined by the EU Water
65 Framework directive (EU-WFD; CEC 2000); 2) increasing primary production favouring
66 eutrophication in a surface waterbody (Smith *et al.* 1999; Watson and Foy 2001; Khan and
67 Ansari 2005; Rivett *et al.* 2008); and 3) enhancing greenhouse gas emissions from
68 unsaturated and saturated zones (Watson and Foy 2001; Reay *et al.* 2003; Stark and Richards
69 2008b).

70 At present in Ireland, land drainage in agriculture areas is regulated through the European
71 Communities (Environmental Impact Assessment) (Agriculture) Regulations 2011 (CEC
72 2011). Under this legislation, an Environmental Impact Assessment (EIA) has to be
73 undertaken for works involving more than 15 ha of land or where the farmer feels that
74 drainage may have a significant effect on the environment (DAFF 2011). Nevertheless, this
75 legislative framework does not provide guidelines for the design of subsurface drainage
76 systems in Ireland. Failure in adapting a drainage design to local soil, hydrogeological or
77 climatic conditions can result in enhanced nutrient losses from grassland fields (Skaggs *et al.*
78 1994; Gilliam *et al.* 1999). There is a need to propose guidelines for the design of drainage
79 systems in contrasting natural Irish settings for variable soil, subsoil and aquifer types in
80 different rainfall regimes.

81 Before such guidelines are developed, a sound understanding of the controlling factors on
82 nutrient losses in surface and subsurface hydrological pathways is needed. Briefly, spatial and
83 temporal variations in surface and subsurface nutrient losses relate to: 1) anthropogenic
84 factors, such as design of drainage systems (Skaggs *et al.* 1994; Kladvko *et al.* 2004), timing
85 of fertilizer application (Olness *et al.* 1980; Hart *et al.* 2004), excessive fertilization (Jordan
86 *et al.* 2000; Watson and Foy 2001), or modification of soil properties through the use of

87 machinery or animal grazing (Watson and Foy 2001); and 2) natural factors, such as
88 variations in event and antecedent hydrometeorological conditions (Heathwaite and Dils
89 2000; Hart *et al.* 2004; Haygarth *et al.* 2004; Kurz *et al.* 2005; Doody *et al.* 2006), soil types
90 and microbial activity (Daly *et al.* 2001; Watson *et al.* 2007; Stark and Richards 2008a;
91 Ghani *et al.* 2010), plot size or slope, or depth of the water table. Furthermore, spatial and
92 temporal patterns of total nutrient losses and nutrient bioavailability from surface runoff and
93 subsurface drainage systems can differ strongly (Gilliam *et al.* 1999). Overall, these
94 differences relate to the total amount of flow generated by these systems for a given rainfall
95 input, and to the contribution of water from different surface and subsurface flow-paths
96 (Haygarth *et al.* 2005; Ghani *et al.* 2010), which will differ in terms of intensity of soil-water
97 interactions and/or groundwater inputs.

98 The objectives of the current study were to investigate the division of nutrient speciation and
99 loads between surface and subsurface drainage over three rainfall events. More specifically,
100 the research assesses to what extent spatial variations of plot size and slope within a similar
101 landscape position, soil characteristics and groundwater patterns impact flow generation, as
102 well as losses and bioavailability of nutrients in overland and subsurface drain flow systems.

103 In order to address these issues, this paper formulates the following hypotheses:

104 1-Spatial variation of flow and solutes between plots are smaller than temporal variations for
105 each plot between rainfall events;

106 2-Nutrient load variability reflects overland and subsurface flow patterns and anthropogenic
107 inputs;

108 3-The occurrence and proportions of different species and fractions of P and N are controlled
109 by the contribution of different proportions of water originating from a rainfall event (event
110 water) or stored in the subsurface (pre-event water).

111

112 **MATERIALS AND METHODS**

113 **FIELD SITE**

114 This study was conducted between January and April 2009 on a 4.2-ha study area, divided
115 into six un-grazed grassland plots within the same landscape position, located on a beef farm
116 at the Teagasc, Johnstown Castle, Environmental Research Centre, Co. Wexford, SE Ireland
117 (52° 17' 36" N, 6° 31' 6" W). The soil and subsoil onsite originates from heterogeneous
118 glacial parent material, which is underlain by Pre-Cambrian greywacke, schist and massive
119 schistose quartzites that have been subjected to low grade metamorphism (Fenton *et al.*
120 2009). Four plots (1, 2, 3 and 4) were used in the study (Fig. 1). Plot areas and slope, and soil
121 texture and nutrient concentrations are presented in Tables 1 and 2. The design of the site
122 ensures no runoff from adjacent (1.5-m-deep drains) or up-gradient (3-m-deep drains) sites
123 can enter the isolated plots. Overland flow was collected in a surface drain at a low point
124 within each plot and transported off site to a v-notch weir setup (Fig. 1). The site had a
125 subsurface herring bone drainage system at 1 m bgl (tapping into higher permeability subsoil)
126 made of corrugated pipes (15 cm, inner diameter) fitted with a gravel pack (washed and less
127 than 20 mm in size), with drain spacing of 10 m and connected to another set of v-notch
128 weirs (outflow of the drainage network). This allowed for the monitoring and sampling of
129 subsurface drain flow. Flow through the v-notch weirs (for both runoff and drain flow) was
130 measured using a calibrated pressure transducer (Sigma, Hach Company, USA). Three

131 shallow piezometers (4.5-m-depth, screen interval 1 m at end of casing) were drilled to below
132 lowest water strike within each plot (Fig. 1), each fitted with a mini-diver (Schlumberger
133 Water Services, Delft, Netherlands) to record variations of water level at 15-min-intervals.

134 **AGRONOMIC MANAGEMENT**

135 Plots received fertilizer inputs of nitrogen (N) fertilizer (applied twice a year) and phosphorus
136 (P) and potassium (K) (applied once a year). For the period of study, P and K were applied on
137 the 21st and 22nd March 2009 at a rate of 37 Kg ha⁻¹ P and 74 Kg ha⁻¹ K, respectively, while N
138 fertilizer application in the form of urea was applied to all plots at a rate of 118 Kg ha⁻¹ N on
139 the 23rd March 2009. The plots were re-seeded at establishment in 2001 and left ungrazed
140 until the experiment started. All plots were sown with mid-season yielding variety of *Lolium*
141 *perenne*. Subject to soil conditions, plots were cut three times a year for silage (last week in
142 May, last week in July and last week in September/first week in October). After the study
143 period, a second application of N, in the form of calcium ammonium nitrate (101.8 kg ha⁻¹
144 N), was applied after the first cut. Plots received no other inputs.

145 **CLIMATE AND SOIL MOISTURE DEFICIT**

146 The study area has a cool maritime climate, with mean annual precipitation 1002 mm,
147 effective rainfall ranging 400 to 500 mm, and mean annual air temperature of 9.6°C (Ryan
148 and Fanning 1996). Daily weather data were recorded at the Johnstown Castle Weather
149 Station and were used to estimate soil moisture deficit (SMD) for moderately-drained soil
150 using the Hybrid model (Schulte *et al.* 2005).

151 **WATER SAMPLING AND ANALYSIS**

152 Water samples from v-notch weirs were taken on a flow-weighed basis using a 900 Max
153 Portable auto-sampler (Sigma, Hach, USA). Filtered (0.45 µm membrane) and non-filtered

154 water samples (20 ml) were analysed using a Thermo Konelab 20 analyser (Technical Lab
155 Services, Ontario, Canada). Total dissolved nitrogen (TDN) consists of dissolved inorganic
156 nitrogen (DIN, sum of nitrate (NO₃-N), nitrite (NO₂-N), ammonia (NH₃-N) and ammonium
157 (NH₄-N)) and dissolved organic nitrogen (DON = TDN-DIN). Total P consists of particulate
158 P (PP) and total dissolved P (TDP), with TDP being the sum of dissolved reactive P (DRP)
159 and dissolved unreactive P (DUP).

160 **GEOPHYSICAL SURVEY AND SOIL ANALYSIS**

161 Several 2D resistivity profiles (to a depth of 50 m) and electromagnetic surveys (to a depth of
162 5 m) were used to develop a conceptual model of the site, to ascertain soil/subsoil material
163 and bedrock type and quantify depth to bedrock on site (Fig. 1). Soil samples were taken
164 from the top, middle, bottom of each plot, and soil texture was obtained using particle size
165 distribution techniques using the sieving and pipette method (BS 1796; BSI 1989) (Table 1).
166 Plot soil nutrient status (Table 2) was determined on soil samples (0-0.3m) at the top, middle
167 and bottom segments of each plot and were analysed using Morgan's extractant (Morgan
168 1941) for soil test P, K⁺ and magnesium Mg²⁺ (Table 2). Total Nitrogen was analysed after
169 KCl extraction on a TN analyser (TNM-1, Shimadzu Corporation, Kyoto, Japan). According
170 to previous soil analysis (unpublished Johnstown Castle soil and land use summary report),
171 Carbon (C) /N ratios at the sites are between 8.3 and 11.4.

172 **RAINFALL EVENT DELINEATION AND CHARACTERIZATION**

173 Three events, the two first in Winter (Event 1 and 2, starting on the 11th January and 24th
174 January 2009, respectively) and the second in Spring (Event 3, starting on the 24th April
175 2009) were used in this study. These events were chosen to represent contrasting pre-event
176 and event rainfall and SMD patterns (Figs 2 and 3) and to discuss incidental losses of

177 nutrients following a fertilizer application. Event 1 was the first significant event occurring
178 after a dry period (peak of 12.9 mm of SMD immediately before the event), while Event 2
179 occurred during a wet period (peak of -7.0 mm of SMD immediately before the event). Event
180 3 occurred in intermediate weather conditions (SMD ranging from -4.7 to 10.6 mm before the
181 event), at 36 days after fertilizer application. Absence of rainfall for more than 12 h was used
182 to separate one rainfall event from the other (Kurz *et al.* 2005). The slopes of cumulative
183 overland and drain flow were computed at hourly intervals. According to Vidon and Cuadra
184 (2010), the start of a flow event can be delineated when a “perceptible rise in discharge”
185 occurs. Accordingly, in the present study, the start of an overland or subsurface drain flow
186 event occurred when the slope of the cumulative flow exceeded the maximum slope observed
187 for the 12 h preceding the rainfall event. Similarly, the end of the flow event occurred when
188 the slope of the cumulative flow reached smaller value than the maximum observed before
189 the event.

190 Runoff coefficients in overland and subsurface drain flow were defined as the ratio of total
191 runoff and rainfall depth (both in mm) for the event (Macrae *et al.* 2010). Loads of dissolved
192 and particulate N and P species and fractions for a time interval were calculated by
193 multiplying the mean concentration of the species and fractions by the corresponding flow
194 occurring during the interval (Kurz *et al.* 2005). Total loads for single flow events were
195 computed by adding the loads from each sampling interval occurring during the event.

196

197 **RESULTS**

198 **PRE-EVENT AND EVENT HYDROMETEOROLOGICAL CONDITIONS**

199 Total precipitation was the greatest for Event 3 and the smallest for Event 2 (29.3 and 12.3
200 mm, respectively, Fig. 2a), while rainfall intensity was the greatest for Event 3 and the
201 smallest for Event 1 (1.5 and 0.5 mm hr⁻¹, respectively, Fig. 2a). Maximum rainfall intensities
202 were more comparable between events, with a minimum value observed for Event 2 and a
203 maximum for Event 1 (3.6 and 6.8 mm hr⁻¹, respectively). Pre-event cumulative rainfall (Fig.
204 2b) was the lowest for Event 1 (0.1 mm in the pre-event 14 days, and 30.4 mm in the 30 pre-
205 event days) and the highest for Event 2 (up to 92.5 mm in the 30 pre-event days), while Event
206 3 had intermediate values. This resulted in generally 1) shallower groundwater depths before
207 Event 2 (Fig. 2c) and larger depths before Events 1 and 2), a sharp decrease from positive to
208 negative SMD at the start of Event 1 and 3, and fluctuating negative SMD between Event 1
209 and 2 (Fig. 3).

210 **OVERLAND AND SUBSURFACE DRAIN FLOW PATTERNS**

211 *Flow variations across plots*

212 Overland total flow values (Fig. 4a) and runoff coefficients (Fig. 4c) for the three events
213 increased from Plot 2 to Plot 4 (Plot 2 < Plot 1 < Plot 3 < Plot 4, up to 2.5 times more flow
214 for Plot 4 than for Plot 2). Subsurface drain total flow patterns (Fig. 4b) were less consistent
215 (e.g. flow in Plot 4 was higher than in Plot 1 and 2 for Events 2 and 3, but smaller than for
216 Plot 1 for Event 1). Nevertheless, runoff coefficients for subsurface drain flow (Fig. 4d) were
217 generally ordered in a similar way than total flow values.

218 For all three events, there was between 2.1 to 6.9 more total flow measured in overland flow
219 than in the subsurface drains. For Event 1 and 2, Plot 1 had the smallest ratios of overland
220 and subsurface drain flow (down to 2.1), while Plot 4 had the biggest (up to 4.9); in contrast,
221 Plot 1 had a greater ratio than Plot 4 for Event 3 (6.9 and 6.3, respectively).

222 ***Flow variations across events***

223 In general, total overland flow (Fig. 4a) was slightly smaller for Event 2 than for Event 1 and
224 increased greatly for Event 3 (up to 2.8 times more flow for Plot 1 for Event 3 than for Event
225 1). Runoff coefficients in overland flow (Fig. 4c) had different patterns. They were minimal
226 for Event 1 and maximal for Event 2, except for Plot 1, where a small increase was observed
227 between Events 2 and 3; differences in runoff coefficients between Plots 1 and 2, and Plots 3
228 and 4 were also more variable, with maximum differences observed for Event 2 (coefficient
229 2.5 times higher for Plot 4 than for Plot 2).

230 As in overland flow (Fig. 4a), subsurface drain total flows (Fig. 4b) decreased from Event 1
231 to Event 2 for Plots 1 and 2 (up to 2.1 times less for Plot 2). For these two plots, even if the
232 flows increased for Event 3, they remained lower than for Event 1. A reverse pattern was
233 observed for Plot 4: an increase was observed between Events 1 and 2 (1.5 times more flow),
234 and a subsequent decrease between Events 2 and 3 (1.4 times less flow). Runoff coefficients
235 in subsurface drain flow (Fig. 4d) had similar patterns than total flow for Plot 4 (Fig. 4b), but
236 they were reversed for Plots 1 and 2.

237 For Plots 1 and 2, ratios of overland and subsurface drain total flow increased from Event 1 to
238 Event 3 (up to 3.3 times greater for Plot 1). In contrast, for Plot 4, they decreased from Event
239 1 to Event 2 (1.6 times less), and increased from Event 2 to Event 3 (2.1 times more).

240 **GROUNDWATER CHEMISTRY**

241 The concentrations of PP, TDP, NH₄-N and NO₃-N for selected piezometers sampled on the
242 5th March 2009 are presented in Table 3. Particulate phosphorus concentrations were all
243 below detection limits, while TDP concentrations were similar at Plots 2, 3 and 4 (overall
244 range of 0.008 to 0.020 mg L⁻¹) and below detection limits at Plot 1. There were no spatial

245 concentration trends within the plots. Ammonium concentrations were similar across plots
246 (overall range of 0.054 to 0.071 mg L⁻¹). Average NO₃-N concentrations were lower and
247 displayed more variations in Plots 1 and 2 than in Plots 3 and 4 (average of 4.7 and 8.7 mg L⁻¹,
248 respectively).

249 **NUTRIENT PATTERNS**

250 *Total P and TDN variations*

251 Flow-weighted mean concentrations of TP and TDN for all plots and events are presented in
252 Table 4, and the corresponding total loads (in g ha⁻¹) in Figs 5a to 5f. For the events before
253 fertilization (Events 1 and 2), flow-weighted mean concentrations of TP and TDN in overland
254 flow increased from Plot 1 to Plot 3 and 4 (up to 1.6 times more TP and TDN). Plot 2 had the
255 highest concentrations for Event 1, and intermediary concentrations between those of Plot 1
256 and 3 for Event 2. Similarly, total loads of TP and TDN in overland flow increased from Plot
257 1 to Plot 4, but to a greater extent than flow-weighted mean concentrations (up to 3.3 times
258 more TP and 2.9 times more TDN for Plot 4 than for Plot 1, Fig. 5a to 5d). For the event after
259 fertilization (Event 3), Plot 2 had the highest flow-weighted mean concentrations of TP and
260 TDN. Otherwise, there were no clear spatial trends in flow-weighted mean concentrations
261 across plots for this event. Both TP and TDN loads were slightly smaller for Plot 2 than for
262 Plot 1 (Fig. 5e to 5f), but still greater for Plots 3 and 4 (1.5 more TP and 1.4 more TDN for
263 Plot 4 than for Plot 2).

264 In overland flow, Event 3 had the highest flow-weighted mean concentrations of TP and the
265 highest loads of TP and TDN (up to 183.2 and 144.4 g ha⁻¹, respectively, Fig. 5e to 5f).
266 Similarly, Event 2 had the lowest flow-weighted mean concentrations of TP and the lowest
267 loads of TP and TDN (down to 2 and 31.2 g ha⁻¹, respectively, Fig. 5c to 5d). Increases in

268 total loads of TP in overland flow for Event 3 (16.1 to 30.4 times more loads for Event 3 than
269 for Event 1) were much higher than for TDN (1.3 to 3.1 times more loads for Event 3 than for
270 Event 1).

271 In subsurface drains, flow-weighted mean concentrations of TP and TDN were more variable
272 between plots than in overland flow. Total dissolved nitrogen loads increased from Plot 2 to
273 Plot 4 for Event 1 (Fig. 5b), and from Plot 1 to Plot 4 for Event 2 (Fig. 5d). Total phosphorus
274 loads behaved similarly for Event 2 between Plots 1 and 4 (Fig. 5c), but they decreased from
275 Plot 1 to Plot 3 for Event 1 (Fig. 5a).

276 In contrast to overland flow patterns, ranges of flow-weighted mean concentrations and total
277 loads of TP and TDN in subsurface drains between events were very comparable (total loads
278 of 0.5 to 4.7 g ha⁻¹ and 6.2 to 61.9 g ha⁻¹ for TP and TDN, respectively, across all plots and
279 events). For the same plots, flow-weighted mean concentrations of TP and TDN were
280 generally smaller for Event 2, while they were more similar for Events 1 and 3. There were
281 few variations in loads of TP and TDN between events at the same plots, except for Plot 1,
282 where they were maximum for Event 2 (6.4 and 9.7 times more P and N at Event 2 than
283 Event 1, respectively) and minimum for Event 1.

284 ***Phosphorus and N speciation and fractionation patterns***

285 For the events before fertilization, dissolved reactive phosphorus was the most abundant P
286 fraction in overland flow for Plots 1 and 2 for Event 1 (49.9 and 53.1% of TP loads,
287 respectively, Fig. 5a) and for Plot 2 for Event 2 (39.2% of the TP loads, Fig. 5c), and in much
288 greater proportions in all plots for Event 3 (from 78.8 to 83.3% of TP loads, Fig. 5e). In
289 contrast, Plots 3 and 4 had DUP as the dominant P fraction in overland flow for Events 1 and
290 2 (up to 41.8 and 55.9% of TP loads for Events 1 and 2, respectively, Fig. 5a and 5c). In

291 contrast, PP was the most abundant fraction in subsurface drain flow for Plots 1 and 2 (up to
292 80.6% of TP loads) for Events 1 and 2. This was also the case for Plot 4-Event 1 (Fig. 5a);
293 otherwise Plots 3 and 4 had DUP as the dominant P fraction in subsurface drains (up to
294 89.2% of total P).

295 Dissolved organic nitrogen was the most abundant fraction of N in both overland and
296 subsurface drain flow (up to 99.6% of total N for Plot 1-Event 2, Fig. 5d), except for Plot 6-
297 Event 2. The relative abundance of DIN was nevertheless generally greater in subsurface
298 drain flow. For Events 1 and 2, this trend was more marked for Plots 3 and 4 (overall range of
299 25.8 to 58.2% of total N as DIN in subsurface drain flow, and only 1.3 to 3.5% in overland
300 flow, Fig. 5b and 5d) than for Plots 1 and 2 (overall range of 6.6 to 26.2% of total N as DIN
301 in subsurface drain flow, and 0.4 to 3.3% in overland flow). For Event 3, subsurface drain
302 flow at Plots 1 and 4 had a relative abundance of DIN of 38.5 and 32.5% of total N as DIN,
303 respectively (Fig. 5f).

304 The relative proportions of NH_4^+ and NO_3^- in DIN are presented in Figures 6a to 6c. In
305 overland flow, NH_4^+ was the only N species detected in samples for Event 1 for all plots, for
306 Event 2 for Plots 2 and 3, and for Event 3 for Plot 2. Nitrate was also the less abundant
307 fraction of DIN in overland flow for Plots 1 and 3-Event 3, while it was more abundant than
308 NH_4^+ for only two occasions (Plot 1-Event 2 and Plot 4-Event 3). In contrast, NO_3^- appeared
309 to be the dominant fraction in all samples of subsurface drain flow, except for Plot 2-Event 1
310 and Plot 3-Event 3 (43.3% and 31.7% of total DIN as NO_3^- , Fig. 6a and 6c). In general, the
311 relative abundance of NO_3^- was greater for Plots 3 and 4 than for Plots 1 and 2.

312

313 **DISCUSSION**

314 SPATIAL AND TEMPORAL VARIATIONS OF FLOW PATTERNS

315 This section evaluates the hypothesis that spatial variations of flow patterns between plots for
316 the same event are smaller than temporal variations for each plot between events.

317 *Overland Flow*

318 Total flow and runoff coefficients values followed a consistent ordering in overland flow
319 across plots for all events (Plot 2 < Plot 1 < Plot 3 < Plot 4, Fig. 4a and 4c). This pattern links
320 to spatial differences in physical controls on runoff generation. In particular, the overall
321 increase in runoff coefficients from Plot 2 to Plot 4 (up to 2.5 times for Event 2) can be in
322 part linked to a decrease in surface area of up to 47% from Plots 1 and 2 to Plots 3 and 4
323 (Table 1). This scale dependency of runoff coefficients has been observed in numerous
324 studies, as reviewed by Wainwright and Parsons (2002), Cerdan *et al.* (2004) or Norbiato *et*
325 *al.* (2009). Joel *et al.* (2002) found that over eight rainfall events, experimental plots with a
326 surface area of 50 m² had surface runoff coefficients of 40% less than plots with a surface
327 area of 0.25 m². When looking at a wider range of scales over a 5-year period in
328 predominantly arable land in Normandy (France), Cerdan *et al.* (2004) found that 450 m²
329 plots had mean runoff coefficients 10 and 30 times greater than larger catchments of 90 and
330 1100 ha, respectively. In the present study, differences in size – and slope length – of the
331 different plots were smaller than in the above studies. This suggests that the effect of plot size
332 can only explain a part of the increase in runoff coefficients from Plots 1 and 2 to Plots 3 and
333 4.

334 Runoff coefficients also tend to increase with increasing slope (Scherrer and Naef 2003;
335 Alaoui *et al.* 2011). Even if Plots 3 and 4 were of similar size (4080 and 4070 m²,
336 respectively, Table 1), the slightly greater slope in Plot 4 than in Plot 3 (5.4 and 4.2%,

337 respectively) may be responsible for the increase in overland flow runoff coefficient. Runoff
338 coefficients also tend to increase with decreasing soil permeability. The influence of
339 permeability was shown at the catchment scale in northern Italy by Norbiato *et al.* (2009) and
340 in a study of preferential flow in forest and grassland sites in Switzerland by Alaoui *et al.*
341 (2011). In the present study, soil texture based on soil sampling to depths of 0.1 and 0.3 m
342 was very similar between plots (Table 1), but geophysical surveys, which include subsoils,
343 showed some differences (Fig. 1). Plots 3 and 4 had sandy, gravely clay and less silt clay
344 horizons than Plots 1 and 2. These patterns may have influenced the difference in runoff
345 coefficients between these two groups of plots, but they were probably not sufficient to
346 counterbalance the effect of other parameters, such as plot size. Nevertheless, when
347 comparing Plots 1 and 2, the greater proportion of silt-clay horizons in Plot 2 may explain the
348 higher overland runoff coefficients observed in Plot 2 than in Plot 1.

349 In addition to these differences in plot size and sediment characteristics, a shallower water
350 table in Plots 3 and 4 than in Plots 1 and 2 (Fig. 2c) also possibly implies that a greater
351 amount of rainfall was required in these latter plots to reach saturation of the soils, further
352 enhancing the effect of plot size on runoff generation. As pointed out by Doody *et al.* (2010),
353 infiltration excess overland flow can dominate over saturation excess overland flow in
354 poorly-drained soils in Irish grasslands, in particular in areas of high soil water repellency. In
355 the present study, the respective importance of both processes was difficult to assess, as no
356 field measurements were available to compare the volumetric soil moisture to the soil field
357 capacity. As all plots had a very similar soil texture, it is therefore likely that they had very
358 similar infiltration capacity. Nevertheless, saturation excess overland flow would probably
359 occur more often where the water table is shallower i.e. in Plots 3 and 4 (water table depth

360 often shallower than 0.5 m for the period of study), as well as towards the bottom of the
361 slopes.

362 Overland flow runoff coefficients (Fig. 4c) were at a maximum for Event 2 for all plots
363 except Plot 1, and were at a minimum for Event 1. Event and pre-event - or antecedent -
364 hydrologic conditions need to be accounted for in the discussion of these patterns. An
365 increase in total rainfall or rainfall intensity, as well as wetter antecedent conditions, have
366 been shown to increase surface runoff coefficients (Norbiato *et al.* 2009; Macrae *et al.* 2010;
367 Vidon and Cuadra 2010; Vidon and Cuadra 2011). Accordingly, an increase in rainfall
368 intensity and total rainfall between Events 1 and 3, as well as wetter pre-event conditions, as
369 indicated by higher pre-event cumulative precipitation and a shallower water table in the
370 majority of wells, can explain the increase in runoff coefficients between Events 1 and 3. In
371 contrast, the high increase in runoff coefficients from Event 1 to Event 2 is not related to an
372 increase in event precipitation, but rather to much wetter antecedent conditions for Event 2
373 than for other events.

374 ***Subsurface drain flow***

375 Subsurface drains generated less flow than surface flow systems (Fig. 4b). They also lacked a
376 consistent ordering of total flow and runoff coefficients observed across plots in overland
377 flow. There were nevertheless clear differences in flow behaviour between plots for each
378 event. Vidon and Cuadra (2010) also observed large variations (often > 50%) of flow
379 generation between two nearby tile drains of the same design installed within a similar soil
380 type. In the present study, the design of the drainage system was identical for all plots.
381 Therefore, differences in drain flow patterns could not be related to factors related to drainage
382 design criterion (Kladviko *et al.* 2004), but rather to the inherent soil and subsoil

383 heterogeneity of the plots, which impact the hydrological connectivity between the surface
384 and the subsurface drains.

385 Instead, variations in drain flow patterns across plots for different events highlight some
386 similarities with overland flow behaviour. For Event 2, wetter pre-event hydrological
387 conditions, in particular, appeared to result in the greatest runoff coefficients (Fig. 4d), with
388 the greatest increase for Plot 4 and the lowest for Plot 2. This pattern was probably related to
389 spatial variations in water table depth across plots, with groundwater inputs to the subsurface
390 drains being more important in wetter periods in areas of shallower water table (Plot 4) than
391 in areas of deeper water table (Plots 1 and 2). Similarly, the small increase in subsurface
392 runoff coefficient between Events 1 and 3 for Plot 4 may be related to increased inputs in
393 both pre-event and event water, as indicated by both wetter pre-event conditions and higher
394 precipitation for the event (total precipitation and rainfall intensity, Fig. 2a). In contrast to
395 what the hypothesis suggested, spatial differences in overland and subsurface drain flow
396 patterns can be greater between plots than between events for the same plots. Overland flow
397 patterns were nevertheless clearly different between Plots 1 and 2, and Plots 3 and 4; indeed,
398 if these two pairs of plots are considered separately, their flow patterns confirm our
399 hypothesis.

400 **CONTROLS ON SPATIAL AND TEMPORAL VARIATIONS IN TDN AND TP** 401 **LOADS**

402 This section evaluates the hypothesis that TDN and TP load variability is inherited from
403 overland and subsurface flow patterns and anthropogenic inputs.

404 ***Overland flow***

405 In overland flow, variations of loads of TP and TDN across plots appeared to follow the same
406 ordering than that of total flow and runoff coefficients for Event 3 (Plot 2<Plot 1<Plot 3<Plot
407 4, Fig. 4a, 5e and 5f), while for the other events, values were smaller for Plot 1 than for Plot
408 2. Furthermore, increases in loads of TP and TDN between plots were similar to the increase
409 in total flow generation. For example, in Event 2, 2.1 times more flow was generated for Plot
410 3 than for Plot 1, resulting in a 3.3 and 2.3 times increase in loads of TP and TDN,
411 respectively. For the same plots during Event 3, 1.2 times more flow resulted in a similar
412 increase in loads of TP and TDN. Kurz *et al.* (2005) showed in a similar setting that P losses
413 would increase in overland flow with increasing soil test P concentration in soils. This
414 confirmed the findings of other studies (Sharpley 1995; Smith *et al.* 1995; Heckrath *et al.*
415 1995; Hesketh and Brookes 2000; Hart *et al.* 2004; Watson *et al.* 2007). Similarly, high N
416 losses in surface runoff often occur as incidental losses under standard or excessive fertilizer
417 applications (Cuttle and Scholefield 1995; Scholefield and Stone 1995). The present study
418 considered soils with very similar Morgan's P concentrations (arithmetic mean of 2.8 to 3.9
419 mg L⁻¹, low P-index, Table 2) compared to a range of 4 to 17 mg L⁻¹ in Kurz *et al.* (2005), as
420 well as identical fertilizer applications across plots. The lack of strong variability in P and N
421 availability in soils, or through fertilizer application across plots, explains the general absence
422 of large differences in TP and TDN flow-weighted mean concentrations in overland flow.
423 The increase in flow-weighted mean TP concentrations from Plot 1 to Plot 4, possibly related
424 to the contribution of water with higher TP concentrations at Plots 3 and 4, contributes to the
425 overall increase in TP loads across plots. Nevertheless, the absence of such patterns for TDN,
426 and the good correlations between flow and total load variations suggest that spatial
427 variations in nutrient losses in overland flow are primarily controlled by differences in flow
428 generation across plots.

429 Variations in TP and TDN loads between Event 1 and 2 in Plot 3 were proportional to those
430 of flow generation i.e. 1.2 times more flow for this plot between these two events resulted in
431 1.4 greater P but similar N losses (Fig. 4a, and Fig. 6a to 6d). Nevertheless, the higher TP and
432 TDN flow-weighted mean concentrations for Event 1 than for Event 2 for Plot 2 (Table 4)
433 resulted in 2.5 times more TP loads for Event 1 than for Event 2, and up to 1.7 times more
434 TDN loads, for similar volumes of water (Fig. 4a, and Fig. 5a to 5d). The strong increase in
435 TP losses for Event 3 relates to the application of fertilizer on the 20th and 21st March 2009
436 (i.e. 36 days before the start of the event). This pattern relates to a change from small critical
437 P losses in a low P-index context to high incidental losses resulting from the occurrence of
438 rainfall events after the spreading of fertilizers (Haygarth *et al.* 1999). Increases in TP of 16.1
439 to 30.4 times greater than for Event 1 are within the range shown by other studies (see Hart *et*
440 *al.* (2004) for a review). For example, TP losses for native grassland watersheds in Oklahoma
441 (USA) increased 10 to 25 times after application rates of 75 kg P ha⁻¹ of ammonium
442 phosphate fertilizer (Olness *et al.* 1980; Hart *et al.* 2004). In contrast to this, Kurz *et al.*
443 (2005) reported no obvious increase in DRP levels in overland flow 44 days after the
444 application of P fertilizer in mid-March; they attributed this result to a “time lag” effect, as
445 rainfall only occurred 44 days after the fertilizer application. Ideally, advice on the timing of
446 fertilizer spreading should account for variations in rainfall patterns occurring over similar
447 periods; nevertheless, present weather forecast capabilities do not allow to do so. In the
448 present study, N fertilizers were applied two to three days after P fertilizer. The small
449 increase in TDN losses in overland flow at Event 3 reflects an increase in flow generation for
450 this event (up to 2.8 times greater flow, for up to 3.1 times greater TDN losses). This pattern
451 suggests that the time lag between fertilizer application and Event 3 was enough to mobilize
452 excess N, either by biological processing or through losses along surface and subsurface
453 flowpaths.

454 ***Subsurface drain flow***

455 As in overland flow, differences in loads of TP and TDN between plots in subsurface drain
456 flow often followed the flow generation sequence for the same event, whereas differences
457 between events were more variable (Fig. 4b and 5). Furthermore, Event 3 was not
458 characterised by a large increase in TP loads as in overland flow (Fig. 5e and 5f). Indeed, for
459 this event, loads of TP and TDN in subsurface drain flow were similar to those observed for
460 Event 1. This pattern suggests that soil-water interactions, as well as losses of nutrients in
461 overland flow, were sufficient to significantly buffer the effect of P fertilisation on incidental
462 losses, or that transit times of P towards the subsurface drains are longer than the period
463 between the application of fertilizers and Event 3.

464 ***Overland flow versus subsurface drain flow***

465 Loads of TP and TDN in subsurface drain flow were, in general, significantly smaller than
466 those in overland flow (2.3 to 123.1 times higher TP loads, and 2.6 to 5.0 times higher TDN
467 loads in overland flow), except for Plot 1-Event 1 (1.1 and 1.6 times less TP and TDN loads,
468 respectively, in overland flow than in subsurface drain flow, Fig. 5). Haygarth *et al.* (1998)
469 related higher mean concentrations of TP in the shallowest horizons of the soil to a greater
470 soil test P. Indeed, P is often considered to be retained in the shallow subsurface. In the
471 present study, the smaller loads of TP and TDN in the subsurface drainage system could be
472 attributed to a combination of such processes, and to the fact that smaller amounts of water
473 were generated by the subsurface drainage system. For example, in Event 1, ratios of loads of
474 TP between overland and subsurface drainage flow were larger than ratios of overland versus
475 subsurface drainage flow for all Plots except Plot 1 (up to 17.5 times less P in subsurface
476 drainage flow for 6.5 times less flow for Plot 3), while there was an overall decrease in TP
477 flow-weighted mean concentrations between overland and subsurface drainage flow. This

478 suggests that the reduction in TP loads in the subsurface drainage system comparatively to
479 the surface system is potentially a result of: 1) P retention in the shallow soil, 2) dilution with
480 water from subsurface flowpaths less concentrated in TP and 3) smaller amount of flow in the
481 subsurface drains. In contrast, for the same event, ratios of loads of TDN between overland
482 and subsurface drainage flow were slightly smaller than ratios of overland and subsurface
483 drainage flow, except for Plot 2, while TDN flow-weighted mean concentrations were often
484 higher in the subsurface drainage system. This specific pattern suggests losses of N from the
485 subsurface flowpaths to the subsurface drains.

486 **PHOSPHORUS FRACTIONATION AND N SPECIATION**

487 This section tests the hypothesis that the occurrence and proportions of different species and
488 fractions of P and N are controlled by the contribution of different proportions of water
489 originating from a rainfall event (event water) or stored in the subsurface (pre-event water).

490 ***Phosphorus fractions***

491 The predominance of dissolved forms of P in overland flow possibly reflects particle
492 retention by the vegetation and limited erosion in the absence of livestock (Haygarth *et al.*
493 1997; Hart *et al.* 2004; Heathwaite and Dils 2000). McDowell and Sharpley (2002) observed
494 through rainfall experiments that PP increased with increasing distance along the slope in un-
495 manured grass plots. In the present study, differences in plot size – and slope length – did not
496 seem to impact on the relative proportion of PP in overland flow. The predominance of DRP
497 in overland flow for Plots 2 and 1-Event 1 and for Plot 2-Event 2 (Fig. 5a and c) reflects the
498 result of previous studies (Haygarth *et al.* 1997; Heathwaite and Dils 2000; Hart *et al.* 2004).
499 The higher contribution of pre-event water for Plots 3 and 4 is probably responsible for the
500 fact that DUP was the major fraction of P released at these locations for the first two events.

501 Indeed, processing of P by plants and soil biota, as well as higher adsorption of DRP in soils,
502 cause increases in organic forms of P in pre-event water (Haygarth *et al.* 1998; Reynolds and
503 Davies 2001; Gburek *et al.* 2005). Dissolved reactive phosphorus was nevertheless the most
504 abundant P fraction in overland flow for all plots for Event 3 (Fig. 5e). This pattern probably
505 relates to the application of mineral P fertilizers 36 days before the event (Hart *et al.* 2004),
506 which effectively overrides differences in the respective contribution of pre-event and event
507 water across plots or events. As in overland flow, the predominance of DUP in subsurface
508 drainage flow at Plots 3 and 4 is probably linked to the predominance of pre-event water. In
509 contrast, in Plots 1 and 2, where the water table is deeper, the predominance of PP can be
510 indicative of the development of preferential flow along macropores, in areas of finer soil
511 texture than those found in Plots 3 and 4 (Heathwaite and Dils 2000; Kramers *et al.* 2009,
512 2012).

513 ***Nitrogen species***

514 As in few previous studies, DON was generally the predominant form of N in overland and
515 subsurface drain flow for all events. Streeter *et al.* (2003) also found that DON amounted to
516 over 90% of TN in soil water and surface water (lake water, as well as lake inflow and
517 outflow) in upland semi-managed grassland in Cambria (UK). Similarly, Willett *et al.* (2004)
518 found that DON represented from 40% to over 85% of the total nitrogen pool in streams and
519 lakes in Wales (UK), in areas predominantly covered by grazed grasslands and forests. A
520 similar predominance of DON over inorganic forms of N was observed in soil water under
521 pasture (Ghani *et al.* 2010) or un-grazed grassland (Dijkstra *et al.* 2007; van Kessel *et al.*
522 2009). In the present study, this pattern probably relates to the use of Urea as N fertilizer in
523 Spring, to the uptake of DIN inputs through fertilization by grass species as well as to high
524 rates of organic matter decomposition in the soil (Stark and Richards 2008a; van Kessel *et al.*

525 2009). Several studies and reviews show the need to better assess the importance of DON
526 losses to groundwater and surface water systems (Seitzinger and Sanders 1997; Pellerin *et al.*
527 2006; Willett *et al.* 2004; van Kessel *et al.* 2009). These works especially highlight that the
528 bioavailability of DON in aquatic ecosystems - and its contribution to eutrophication - and
529 the overall function of DON in the N cycle should be more accounted for.

530 The shift from NH_4^+ dominated water in overland flow to NO_3^- dominated water in
531 subsurface drain flow (Fig. 6), and the greater loads of DIN in the subsurface drain flow
532 further indicate the impact of soil microbial activity (i.e. nitrification; Stark and Richards
533 2008a) on the chemical transformation of N in the subsurface. Here again, a shallower water
534 table for Plots 3 and 4 probably relates to higher inputs of NO_3^- in the subsurface drainage
535 system than for Plots 1 and 2. Higher concentrations of $\text{NO}_3\text{-N}$ than $\text{NH}_4\text{-N}$ in groundwater,
536 as well as higher $\text{NO}_3\text{-N}$ concentrations at Plots 3 and 4 than at Plots 1 and 2, confirm this
537 process. In contrast, higher NH_4^+ concentrations in Plots 1 and 2 possibly relate to quick
538 transfer of NH_4^+ in macropores (Kramers *et al.* 2009); the absence of an increase in P in the
539 subsurface drains for Event 3 nevertheless suggest that this pathway is of small importance.
540 Alternatively, limited nitrification in areas of higher than optimal soil water content
541 (Grundmann *et al.* 1995) could explain this pattern.

542 **MANAGEMENT IMPLICATIONS**

543 This study highlights the necessity to adopt a common holistic and interdisciplinary
544 framework, which seeks to identify the controlling factors for water and solute losses in both
545 surface and subsurface environments in Irish marginal lands. Due to the mixed nature of
546 nutrient losses (i.e. both P and N losses) in surface and subsurface drainage, any end-of-pipe
547 solution to ameliorate discharges must consider both nutrient types. In addition, buffer strip
548 areas positioned at the bottom of slopes can decrease such losses, providing the subsurface

549 drainage system does not extend into these areas and the outflow still exists further down-
550 gradient.

551 ***Plot size***

552 In this study, the size of the plots – and the correlated slope length – appears to relate to the
553 volume of water generated by unit of land area (i.e. in mm) and total loads of P and N (in g
554 ha⁻¹) in overland flow. As pointed out above, it was difficult to quantify the importance of
555 these controls with respect to others such as watertable depth. It is nevertheless likely that in
556 this kind of setting, significantly increasing field size will result in a decrease of nutrient
557 losses in overland flow. In contrast, the decrease in flow volume and nutrient loads with
558 increasing field size were not compensated by correlated increases in subsurface drain flow.
559 This suggests that other pathways such as evapotranspiration, losses to groundwater, or
560 retention in the soil, are enough to compensate for the difference. In Ireland, the mean size of
561 permanent pastures not classified as commonage is 5.5 ha (standard deviation of 12.2 ha). A
562 median size of grassland fields of 3 ha was determined by excluding extreme outliers (CEC
563 2001). In areas where the natural attenuation capacity of nutrients in subsurface flowpaths is
564 enough to accommodate increasing nutrient loads, increasing field size could be considered
565 as a measure to limit nutrient losses to a connected waterbody.

566 ***Water table depth***

567 Generally, fields with similar soil characteristics will generate more overland flow where the
568 water table is shallow when compared with areas where more rainfall is needed to reach full
569 saturation (Doody *et al.* 2010). In the present study, this mechanism, in conjunction with the
570 increase in plot size, was probably responsible for the increase in the volume of water and in
571 the total loads of P and N generated in overland flow from Plots 1 and 2 (deeper water table)

572 to Plots 3 and 4 (shallower water table). Subsurface drain flow also increased more at Plots 3
573 and 4, further enhancing nutrient losses. In addition, for events preceding the application of
574 fertilizer, a shallower water table depth across plots related to: 1) an increase in the
575 proportions of DUP over DRP in overland flow, and those of DUP over PP in subsurface
576 drain flow; 2) an increase in the proportions of DIN relatively to DON in subsurface drain
577 flow; and 3) a decrease in the proportions of NH_4^+ relatively to NO_3^- in subsurface drain flow.
578 Dissolved inorganic forms of P and N (i.e. DRP and DIN) are said to be more bioavailable
579 than organic (i.e. DON and the majority of DUP) or particulate fractions (i.e. PP) (Seitzinger
580 and Sanders 1997; Reynolds and Davies 2001; Willett *et al.* 2004). Similarly, NH_3 is highly
581 toxic to aquatic ecosystems (Camargo and Alonso 2006), whereas in the current water
582 legislation in Ireland, NO_3^- is mostly considered to be a limiting nutrient in coastal waters and
583 a contaminant in drinking water. Implementing subsurface drainage may enhance crop
584 growth and nutrient uptake and thereby decrease nutrient loads. Nevertheless, in many
585 circumstances, artificially lowering the water table could result in decreasing total loads of N
586 and P lost from surface pathways, but increasing the nutrient bioavailability in quick flow
587 (e.g. increasing the proportions of inorganic forms of nitrogen) through the subsurface
588 drainage system. In order to address this issue, technologies such as permeable reactive
589 barriers or constructed wetlands (Brix *et al.* 2012; Healy *et al.* 2012), which aim at
590 remediating mixed contaminant sources could be implemented.

591 Similar studies should be undertaken to evaluate the impact on nutrient losses of installing
592 mole or gravel mole drains to enhance the infiltration capacity of soils (Galvin 1983). In the
593 1970s and 1980s, the priority was to drain land without an environmental framework. As
594 drainage implementation is now being re-visited to provide environmentally sustainable

595 solutions for Irish farming, these patterns of nutrient losses and bioavailability need to be
596 accounted for.

597

598 **CONCLUSION**

599 This study investigated the controls on flow generation and P and N losses and
600 fractionation/speciation in surface and subsurface drainage for three rainfall events in four
601 un-grazed grassland plots in Ireland. Spatial differences in runoff coefficients and in the total
602 amount of water by unit area (in mm) generated by the overland flow systems mainly linked
603 to 1) the size of the plots (an increase in size linked with a decrease in flow) and 2) water
604 table depth (shallower water table linked with an increase in flow). Temporally, both event
605 and pre-event hydrometeorological conditions were the main controls. In contrast, controls on
606 spatial differences in flow generation in the subsurface drainage systems were more difficult
607 to assess. Overall, these controls imposed greater spatial differences in overland and
608 subsurface drainage patterns between all plots than temporal variations between events for
609 the same plot. In turn, an increase in overland and subsurface drain flow induced higher TP
610 and TDN total losses. Subsurface drain flow generated smaller loads of TDN and TP than
611 overland flow. Before the application of fertilizer, the proportions of different P fractions and
612 N species reflected the influence of pre-event water to overland and subsurface drain flow.
613 Even if DON was generally the dominant form in both systems, the proportion of DIN was
614 higher in the subsurface drains, especially in areas of shallower water table. Nitrate also
615 dominated over NH_4^+ in the subsurface drain flow, but not in overland flow. Similarly, DUP
616 was the dominant P fraction in subsurface drain flow where the water table was shallow,
617 while PP was the most important fraction elsewhere. In overland flow, a shallower water
618 table implied a switch from DRP to DUP-dominated water. The application of fertilizer

619 resulted in a strong increase in TP concentrations in overland flow, and a dominance of DRP
620 in all plots. Both P and N total losses and speciation/fractionation will affect the nutrient
621 bioavailability in aquatic ecosystems. This study highlights the importance of an integrated
622 assessment of the controls of flow and solute patterns in both surface and subsurface flow
623 systems when aiming at identifying the impact of grassland management on nutrient losses in
624 water. Further research is needed to test whether the implementation of design criteria
625 specific to local soil and groundwater conditions can further reduce P and N losses from
626 grasslands.

627

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862 **TABLES**

863 Table 1-Plot area, slope and soil texture. Clay Loam (CL), Sandy Silt Loam (SSL), Sandy
 864 Clay Loam (SCL), Sandy Loam(SL).

Plot Number	Area (m ²)	Slope (%)	Soil Texture (0-10 cm) (Top/Middle/Bottom of slope)	Soil Texture (0-30cm) Top/Middle/Bottom of slope
1	7780	3.9	CL/CL/CL	CL/CL/SSL
2	7470	4.1	CL/SSL/CL	CL/SL/CL
3	4080	4.2	CL/SSL/SL	CL/SSL/SCL
4	4070	5.4	CL/CL/CL	CL/SSL/SSL

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866 Table 2-Mean soil nutrient concentrations (mg L⁻¹) and standard deviation (mg L⁻¹, in
867 brackets) of TN, P, K and Mg at all plots from 0-30 cm depth soil samples

Plot	TN	P*	K*	Mg*
1	4.3 (0.9)	2.8 (0.8)	107.3 (69.7)	183.3 (36.4)
2	2.5 (0.6)	3.7 (0.5)	77.4 (30.3)	235.2 (18.1)
3	2.5 (0.7)	3.9 (1.7)	66.6 (12.5)	167.7 (32.1)
4	1.9 (0.4)	3.4 (0.9)	56.0 (15.8)	158.9 (45.4)

868 *Morgan's soil nutrient concentration

869 Table 3-Groundwater nutrient concentrations (mg L⁻¹) of PP, TDP, NH₄-N and NO₃-N for
870 selected piezometers sampled on the 5th March 2009.

Piezometer	PP	TDP	NH ₄ -N	NO ₃ -N
A	bdl	bdl	0.065	10.417
B	bdl	bdl	0.068	0.863
D	bdl	0.020	0.064	0.840
E	bdl	0.012	0.060	6.424
F	bdl	0.010	0.054	9.848
G	bdl	0.012	0.051	9.420
H	bdl	0.008	0.059	8.123
I	bdl	0.009	0.071	5.680
J	bdl	0.009	0.060	8.670
K	bdl	0.008	0.065	10.270

871 bdl refers to concentrations below detection limits

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881 Table 4-Flow-weighted mean nutrient concentrations (mg L⁻¹) for all plots (1-, 2-, 3- and 4-)
882 in overland flow (OF) and subsurface drain flow (DF) for the three events (Event 1/Event
883 2/Event 3). bdl refers to concentrations below detection limits and n.a. to samples not
884 analyzed.

Plot	TP	PP	DRP	DUP	TDN	DON	DIN	NH ₄ -N	NO ₃ -N
1- OF	0.096/0.061 /1.381	0.024/0.02 8/0.227	0.048/0.01 5/1.088	0.025/0.01 9/0.066	0.861/0.88 5/1.268	0.833/0.88 2/1.183	0.027/0.00 4/0.084	0.027/0.00 1/0.050	bdl/0.003/0 .035
2- OF	0.218/0.077 /1.586	0.060/0.01 9/0.127	0.096/0.03 0/1.289	0.062/0.02 7/0.170	1.492/0.94 4/1.351	1.443/0.90 9/1.310	0.049/0.03 5/0.042	0.049/0.03 5/0.042	bdl/bdl/ bdl
3- OF	0.114/0.095 /1.237	0.022/0.03 1/0.105	0.045/0.01 1/1.002	0.048/0.05 3/0.129	0.876/1.00 0/1.145	0.845/0.98 7/1.088	0.031/0.01 3/0.057	0.031/0.01 5/0.039	bdl/bdl/ 0.018
4- OF	0.123/bdl/ 1.380	0.035/n.a./ 0.096	0.042/n.a./ 1.128	0.046/n.a./ 0.156	1.365/n.a./ 1.067	1.332/n.a./ 1.060	0.033/n.a./ 0.007	0.033/n.a./ 0.001	bdl/n.a./ 0.006
1- DF	0.233/0.065 /0.217	0.188/0.05 2/0.150	0.008/0.00 1/0.008	0.037/0.01 2/0.059	2.956/0.55 1/2.138	2.555/0.50 6/1.315	0.401/0.04 5/0.822	0.085/0.00 5/0.773	0.315/0.04 0/0.049
2- DF	0.098/0.082 /n.a.	0.070/0.05 8/n.a.	0.010/0.00 6/n.a.	0.018/0.01 8/n.a.	0.786/0.86 8/n.a.	0.734/0.63 6/n.a.	0.052/0.23 1/n.a.	0.029/0.04 8/n.a.	0.022/0.18 4/n.a.
3- DF	0.042/n.a./ n.a.	0.004/n.a./ n.a.	0.002/n.a./ n.a.	0.036/n.a./ n.a.	2.227/n.a./ n.a.	1.654/n.a./ n.a.	0.574/n.a./ n.a.	0.025/n.a./ n.a.	0.549/n.a./ n.a.
4- DF	0.106/0.076 /0.076	0.072/0.02 1/0.004	0.005/0.00 2/0.005	0.029/0.05 3/0.067	2.918/2.92 1/1.524	1.559/1.22 0/1.028	1.359/1.70 1/0.496	0.015/0.01 4/bdl	1.343/1.68 7/0.496

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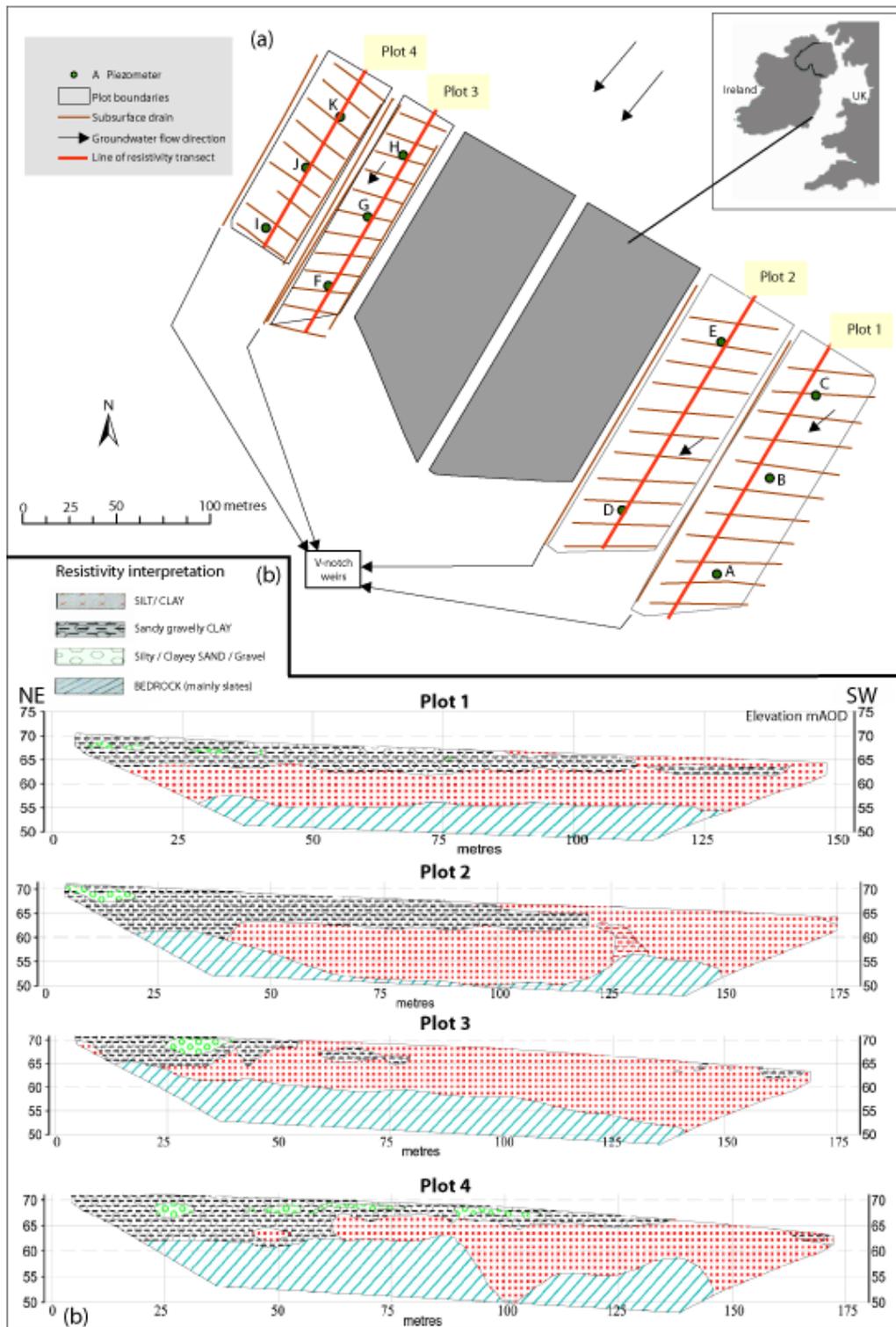
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890 **LIST OF FIGURES**

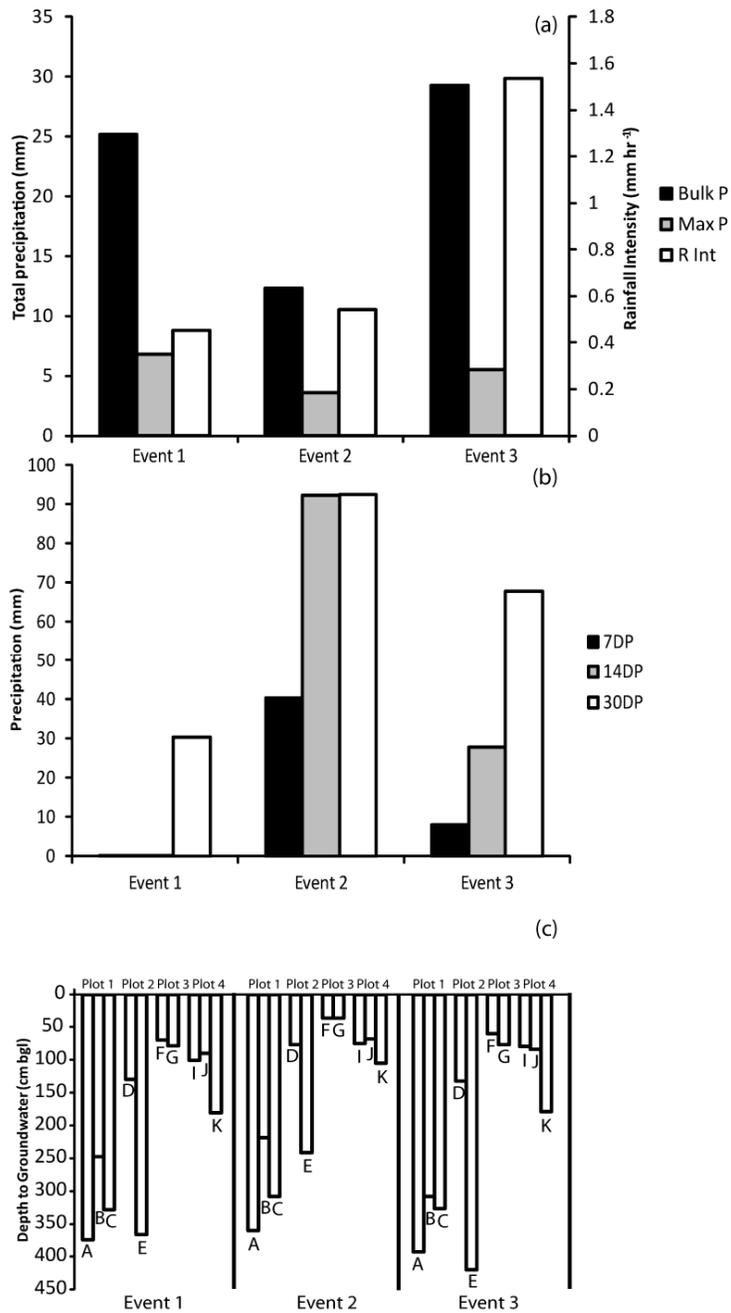
891 Fig. 1-(a) Map of the study area, (b) interpreted resistivity profiles.



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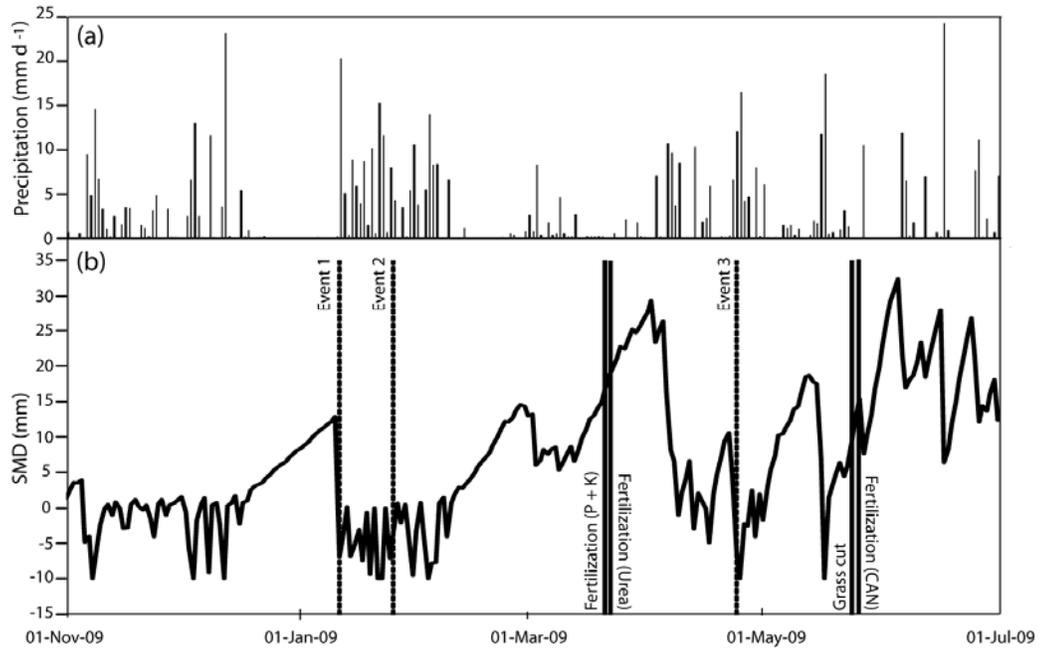
894 Fig. 2-Event precipitation characteristics (a) total precipitation (Bulk P), maximum
 895 precipitation (Max P) and rainfall intensity (R Int), (b) pre-event cumulative precipitation for
 896 7 days (7DP), 14 days (14DP) and 30 days (30DP) preceding the event, (c) water table depth
 897 at the start of the event for selected piezometers (see Fig. 1 for position on the plots).



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900 Fig. 3-(a) Precipitation and (b) soil moisture deficit (SMD) computed from the model by
901 Schulte et al., (2005). Vertical dashed lines indicate start of the three rainfall events and
902 vertical plain lines dates of fertilization and grass cut events.

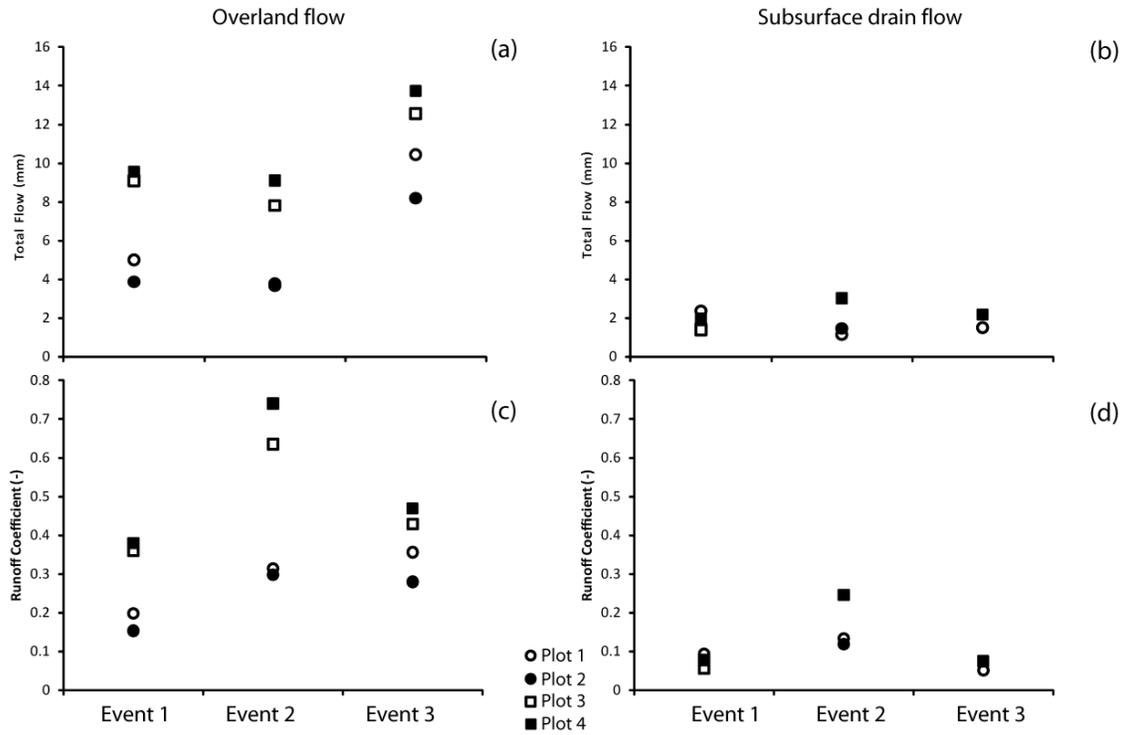


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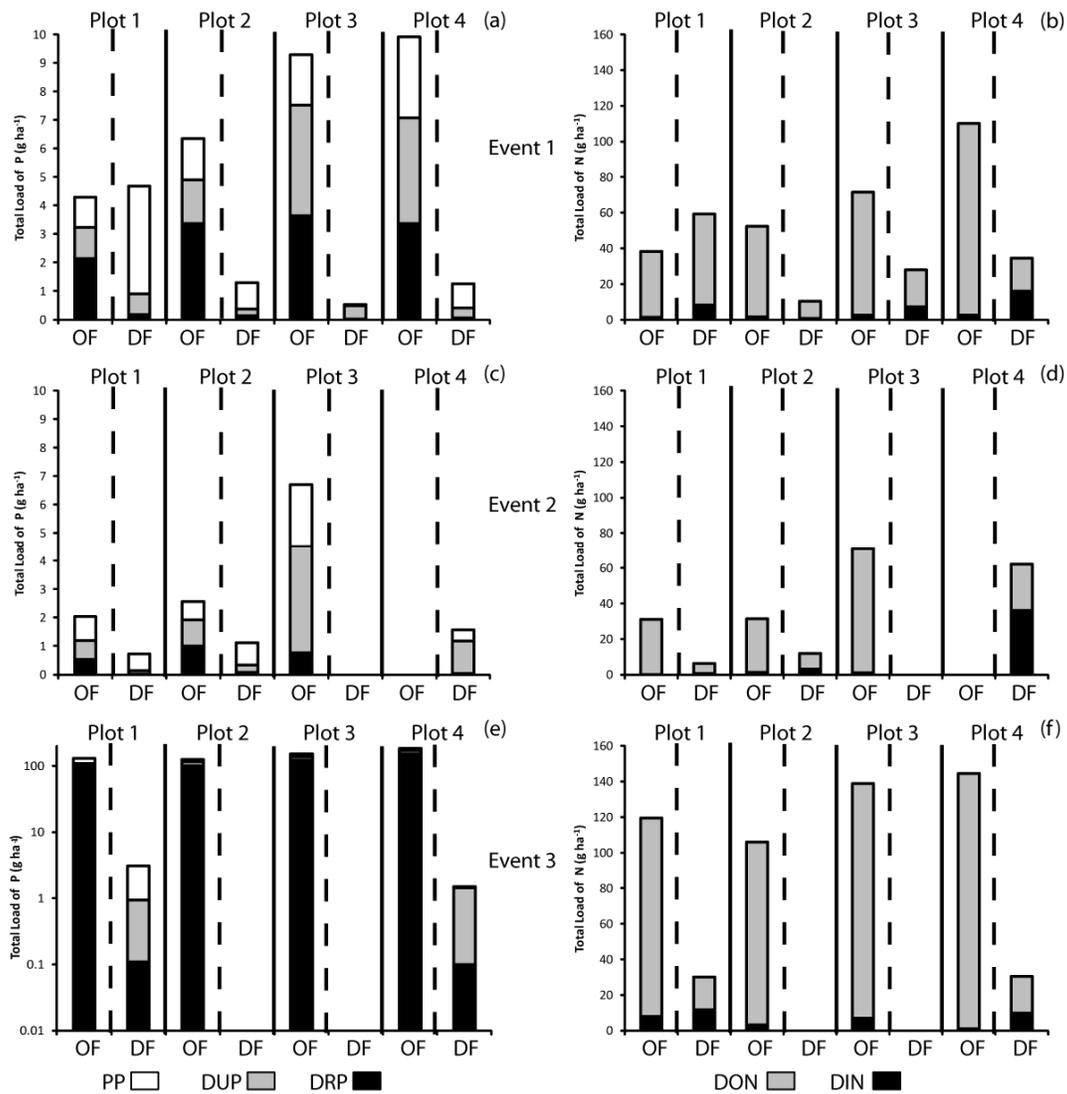
906 Fig.4-Overland and subsurface drain flow patterns per event and plot. (a) ; (b) Total
 907 subsurface drain flow; (c) Runoff coefficient for overland flow (ratio of total overland flow
 908 and total precipitation per event); (d) Runoff coefficient for drain flow (ratio of total
 909 subsurface drain flow and total precipitation per event).



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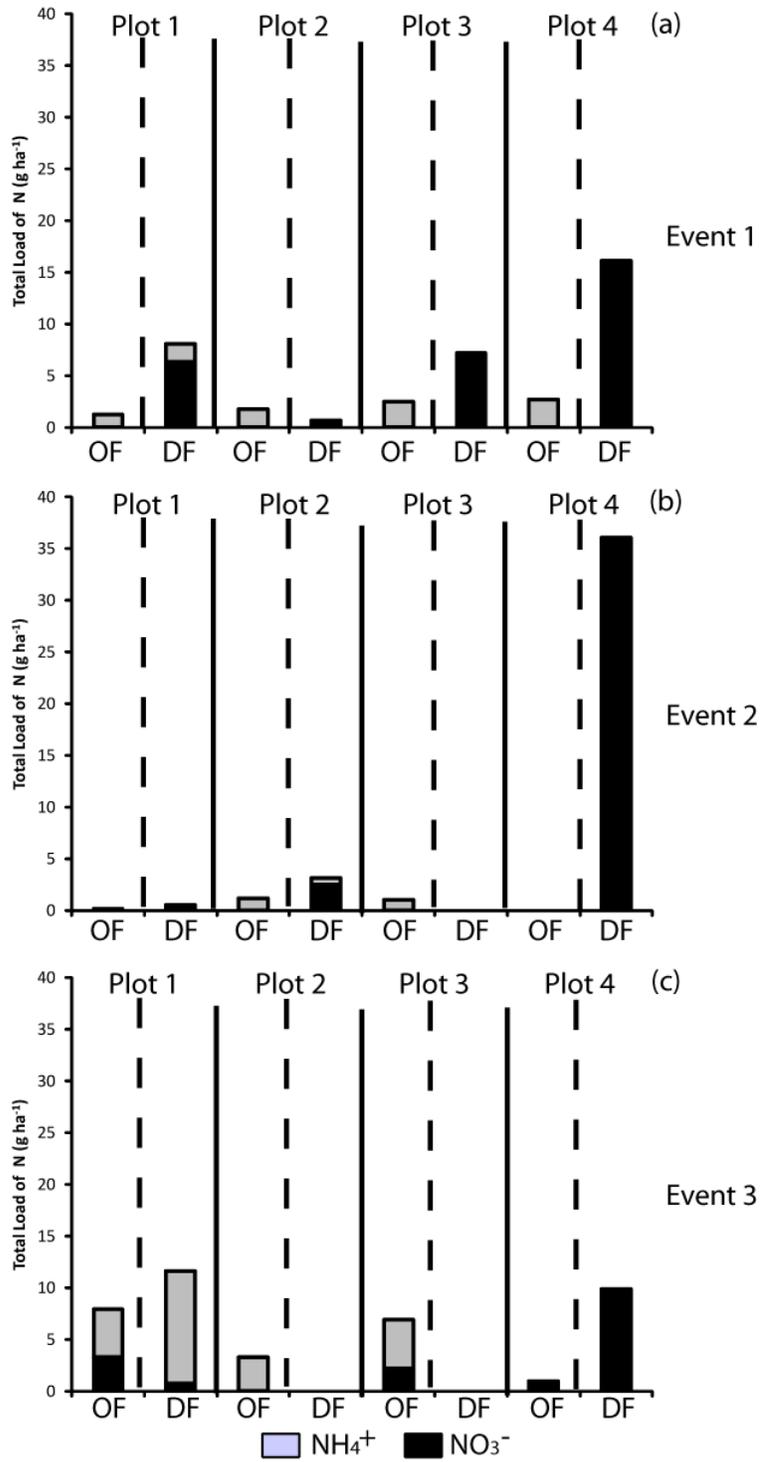
912 Fig. 5-Total loads of P per plot in overland flow (OF) and drain flow (DF) for Event 1 (a),
 913 Event 2 (c) and Event 3 (e) and the relative contribution of particulate phosphorus (PP),
 914 dissolved unreactive phosphorus (DUP) and dissolved reactive phosphorus (DRP). Total
 915 loads of N per plot in overland flow (OF) and drain flow (DF) for Event 1 (b), Event 2 (d)
 916 and Event 3 (f) and the relative contribution of dissolved organic nitrogen (DON) and
 917 dissolved inorganic nitrogen (DIN).



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920 Fig. 6-Total loads of DIN expressed as N per plot in overland flow (OF) and subsurface drain
 921 flow (DF) for Event 1 (a), Event 2 (b) and Event 3 (c) and the relative contribution of
 922 ammonium (NH_4^+) and nitrate (NO_3^-).



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