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1 Conservation practice modify soil phosphorus sorption properties
2 and the composition of dissolved phosphorus losses during runoff

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16 **Abstract**

17 It is now well established that shallow tillage (ST) and no-tillage (NT) of cultivated
18 soils can increase the risk of dissolved phosphorus (P) loss during runoff compared to
19 conventional tillage (CT). However, there is still lack of quantitative descriptors of
20 how variations in tillage practices influence soil P sorption properties and how these
21 properties in turn control the dissolved P release. Moreover, the control of tillage

22 practices on the form of dissolved P, particularly molybdate unreactive dissolved
23 phosphorus (MUDP), is still poorly assessed. Thus, we conducted the runoff
24 simulation with the pilot soils benefited from different tillage practices for 20 years.
25 The results showed that shifting from CT to ST and NT changed the capacity of soils
26 to bind P, with Olsen-P, Dyer-P, equilibrium phosphorus concentration (EPC_0), water
27 soluble phosphorus (WSP), and degree of phosphorus saturation (DPS_{OX}) increased
28 obviously. The runoff simulation experiment confirmed that the switching from CT to
29 ST and NT could increase the P loss in the event of runoff. The increase was evident
30 for the two fertilizations (pig manure and poultry manure) with an amplified effect for
31 the poultry manure modality. Switching from CT to ST and NT favored the loss of
32 MUDP, again with an exacerbated effect in the case of the poultry manure modality,
33 for which the proportion of MUDP went up to represent more than 40% of the TDP.
34 Regarding on indication of the risk of dissolved P loss with P sorption property,
35 positive linear correlations were indeed obtained in the five cases, but the results of
36 DPS_{OX} was opposite to that of WSP, EPC_0 and available P, suggesting DPS_{OX} was not
37 suitable as the risk predictor of dissolved P loss. Our results from this study propose
38 that the MUDP should be taken into account in assessing the risk of dissolved P
39 release during change to ST and NT, considering that MUDP are bioavailable and
40 therefore also capable of contributing to the eutrophication of surface waters.

41 *Keywords:* Tillage practices; Soil phosphorus sorption capacity; Dissolved
42 phosphorus loss; Organic and colloidal phosphorus; Risk assessment indicators.

43 **Highlights**

- 44 ● No-tillage increase the three forms of dissolved P loss risk.
- 45 ● No-tillage exacerbates the proportion of molybdate unreactive dissolved P.
- 46 ● High organic matter is the key to exacerbate molybdate unreactive dissolved P
- 47 loss.

48 **1. Introduction**

49 Long-term addition of phosphorus (P) can significantly increase the content of P
50 in cultivated soils, resulting in P loss risks through soil erosion, leaching, or runoff
51 (Jarvie et al., 2017). Phosphorus losses from cultivated fields to aquatic ecosystems
52 can lead to cyanobacterial blooms, which can make water toxic to wildlife and human
53 (Ma et al., 2009; Messiga et al., 2010; Perrotet et al., 2014; Bol et al., 2018). Among
54 various P forms, dissolved inorganic and organic P are major concerns as they are
55 highly reactive and readily bioavailable for algae growth (Li and Brett, 2013; Van
56 Moorlehem et al., 2013; Jarvie et al., 2017). Thus, reducing the loss of dissolved P
57 from cultivated soils is an essential challenge in the fight against eutrophication of
58 surface water (Pizzeghello et al., 2016; Jarvie et al., 2017; Bol et al., 2018; Ulén et al.,
59 2019; Messiga et al., 2021).

60 In the past decades, mitigation of agricultural P loss is mainly addressed by
61 agricultural conservation practices, notably shallow tillage (ST) or no-tillage (NT),
62 which have been shown to reduce soil erosion and soil P losses (Schoumans et al.,
63 2014; Sharpley et al., 2015; Dodd and Sharpley, 2016; Zhang et al., 2017; Ni et al.,

64 2020). However, although efficient in reducing particulate P loss, ST and NT may
65 increase the loads of dissolved P in surface soil and thus increase the loss of dissolved
66 P in case of runoff development (Ulén et al., 2010; Pizzeghello et al., 2016; Daryanto
67 et al., 2017). Two mechanisms are commonly invoked to account for this increase: i)
68 enhancing organic P mineralization from crop residues left at the soil surface leading
69 to higher concentration of dissolved P in the topmost soil layers (Deubel et al., 2011);
70 ii) general enrichment in P (and thus also in potentially soluble P) of the soil topmost
71 layers due to the lack of downward transfer and mixing of the P brought to the soil for
72 crop growth (Baker et al., 2017; Daryanto et al., 2017; Jarvie et al., 2017; Duncan et
73 al., 2019). Two emblematic examples, the first one in Erie Lake Basin, the other in
74 Chesapeake Bay basin, reveal an increase in dissolved P loss associated with areas of
75 increased implementation of conservation tillage practices (Ulén et al., 2010; Dodd
76 and Sharpley, 2016; Jarvie et al., 2017; Ni et al., 2020). These results demonstrate that
77 the implementation of ST and NT practices could lead to a shift from particulate P to
78 dissolved P diffusion in rural catchments, with a potentially highly negative impact on
79 freshwater ecosystems, given that dissolved P is more bioavailable than particulate P
80 (Duncan et al., 2019).

81 Given the growing success of ST and NT practices among farmers and soil
82 health advocates, it is important to develop knowledge to predict the loss risk of
83 dissolved P that adoption of these practices could engage. One option is to determine
84 whether the P tests conventionally used to evaluate the capacity of soils to release
85 dissolved P have the capacity to be good predictors of the potential increase of

86 dissolved P diffusion in case of adoption ST and NT practices. Some existing soil P
87 tests originally developed for agronomic purposes work reasonably well to assess soil
88 dissolved P loss risk, such as total phosphorus (TP), Olsen-P, and Dyer-P (Sims, 2000)
89 and could be suitable for that purpose. Recently, other indicators more directly related
90 to the soil properties engaged in P sorption such as the degree of phosphorus
91 saturation (DPS_{OX}) have received interest as a possible indicator of dissolved P
92 release risk in soils (Pöthig et al., 2010; Wang et al., 2016; Fischer et al., 2017). The
93 determination of adsorption isotherms makes it possible to determine the value of
94 equilibrium phosphorus concentration (EPC_0) in soils, which has been shown to
95 predict well dissolved P concentrations in runoff (Hughes et al., 2000; Munira et al.,
96 2018), and to be also well correlated with DPS_{OX} values (Sims, 2000; Pöthig et al.,
97 2010; Fischer et al., 2017). Another parameter which can be determined from
98 adsorption isotherms is the phosphorus sorption maximum (Q_{max}), which quantify the
99 maximum amount of P that a soil can bind. In addition, the amount of water-soluble
100 phosphorus (WSP) is also viewed has a potential efficient indicator for the dissolved P
101 release risk by soils (Pöthig et al., 2010; Xue et al., 2014; Fischer et al., 2017).

102 Although all these four indicators (DPS_{OX} , EPC_0 , Q_{max} , and WSP) are generally
103 accepted for the assessment of dissolved P release risk of soils, their use has not yet
104 been tested as a mean of predicting the increase in dissolved P release risk in case of
105 shifting a plot from conventional tillage (CT) to ST and NT. A key question in this
106 context, with possible important implication for the deployment of ST and NT
107 practices, is to know which of these four indicators, if any, provides the best

108 prediction of this increase. At the same time, there is the question of the amplitude of
109 the variations that the change from CT to ST and NT is able to induce on soil P
110 sorption properties, all cultivation practices (type and dose of fertilizer applied, in
111 particular) being moreover kept constant, and of the consequences of these changes
112 on the use of these indicators.

113 Of particular interest in this context is the case of soil surface horizons (0-5 cm),
114 given that these horizons are the layers that ST and NT practices are likely to modify
115 the most with respect to their P sorption properties and P concentrations. The
116 enrichment in organic matter (OM) caused by the cessation of plowing is, in particular,
117 expected to increase the capacity of P sorption of surface soils, by creating ternary
118 complexes between OM, metal cations such as Fe, Al, Ca and phosphate oxyanions
119 (Gerke and Hermann, 1992; Messiga et al., 2011). How does this increase, if it exists,
120 affect the release of dissolved P by ST and NT soils compared to CT soils? Likewise,
121 the enrichment in OM of the soil surface horizon induced by ST and NT practices
122 (Daryanto et al., 2017; Duncan et al., 2019) raises the question of the proportion of
123 organic form of dissolved P likely to be released in the event of runoff. How does this
124 proportion evolve according to the evolution of tillage practices? This question has
125 not yet been addressed to our knowledge.

126 The objectives of this study are to i) evaluate how the different tested ploughing
127 modalities affect the P sorption properties of soils and how this affect in turn the
128 potential dissolved P loss risk of soils; ii) screen out which soil indicator(s) can be
129 used to predict the best P loss magnitude in case of runoff development, and how the

130 organic/inorganic dissolved P ratio vary depending on whether the soil is
131 conventionally tilled or benefit from conservation tillage practices.

132 **2. Materials and methods**

133 *2.1. Site description*

134 The study site was established in 2000 at the Kerguéhennec Agricultural
135 Experimental Station in Brittany, western France (48° N, 3° W; Fig. S1). Brittany is a
136 major agricultural region in France, with agriculture occupying approximately 66% of
137 the region, which also is a hot spot region of eutrophication of surface waters at the
138 scale of France and Europe (Bol et al., 2018). The highest soil P budget was observed
139 in Brittany where the P budget was 17 kg P ha⁻¹ in 2006 (national average was 4.4 kg
140 P ha⁻¹) (Senthilkumar et al., 2012). In Brittany, almost 76,000 t of nitrogen was
141 discharged into the sea in 2008 (Perrotet al., 2014). Climate is temperate oceanic, with
142 20-years average of annual precipitation and temperature of 1000 mm and 10.6 °C at
143 the station, respectively. Soils at the Kerguéhennec station are well-drained silty
144 loamy Luvisols.

145 The studied soils were sampled from two distinct series of plots separated from
146 each other by ca. 50 m (Fig. S1). The A plots comprise nine contiguous plots (6×50
147 m), with three plots being continuously maintained in CT (tillage depth 0-25 cm),
148 three maintained in ST (tillage depth 0-15 cm), and three maintained in NT (tillage
149 depth 0 cm). This “tillage comparison” experiment is a long-term trial which started
150 in 2000, and still continue today. Crop rotations on these plots consist of

151 corn-wheat-rapeseed succession. The A plots are fertilized annually with pig manure.
152 The average annual P input is 122 kg ha⁻¹ (pig manure = 4.8 g of P per kg).

153 The B plots comprise three plots (12×25 m, Fig. S1). The fertilization
154 management is receiving poultry manure with three tillage practices (CT, ST, and NT).
155 Cropping system on B plots consists of a corn-wheat rotation. The average annual P
156 input is 156 kg ha⁻¹ (chicken manure = 17.2 g of P per kg). The amount of fertilizer
157 supplied to soils is adjusted annually to ensure the balance between the input and
158 output of nitrogen, P and carbon, and to provide ideal nutrient amounts for the crop.

159 Crop residues are left on the soil surface of A and B plots managed with NT,
160 while being incorporated in the surface soils by chisel plough for soils of the other
161 two tillage modalities. The average annual coverage rates of crop residues returned to
162 the soil CT, ST and NT is 0.5%, 8.5%, and 26.5%, respectively.

163 *2.2. Soil sampling and preparation*

164 Sampling 100 different surface soils (0-5cm) from 5 different sampling sites in
165 each plot. The composite soil samples were air-dried, thoroughly mixed, and passed
166 through a sieve (2 mm) and analyzed for physicochemical properties, isothermal
167 adsorption experiment and WSP determination.

168 Soils from six plots were selected for the rainfall simulation experiments. Three
169 plots (A (Pig)-CT, A-ST, and A-NT) were from the A trial, and three plots from B trial
170 (B (Poultry)-CT, B-ST, and B-NT). The surface (0-5cm) soil samples from each
171 excavation were combined to form a composite sample of each studied plot, each
172 composite sample being then air-dried, thoroughly mixed, and sieved (20 mm). Note

193 2.4. *Laboratory analyses*

194 All soil samples used in rainfall experiments were analyzed at the soil analysis
195 laboratory of INRAE in Arras, France, on soil aliquots of about 2 kg sieved at < 2 mm.
196 The soil particle size was classified into three categories: clay (<0.002 mm), silt
197 (0.002-0.05 mm), and sand (0.05-2 mm) (Day, 1965). The organic matter (OM) was
198 measured as loss on ignition (Jensen et al., 1992). The dissolved organic carbon (DOC)
199 concentrations in runoff samples were analyzed with a total organic analyzer
200 (Shimadzu TOC-5050A) (using potassium hydrogen phthalate as the standard solution)
201 (Gu et al., 2018). Soil pH was determined with a pH electrode in a soil to water ratio
202 of 1:5 (NF ISO 10390). The Si-ox/Al-ox/Fe-ox concentrations were determined with
203 ICP-AES after extraction with ammonium oxalate and oxalic acid (Ross and Wang,
204 1993). The TP content was also measured (ICP-AES) after total solubilization with
205 hydrofluoric and perchloric acid (NF X 31-147). Dyer-P was extracted with 20 g L⁻¹
206 monohydrate citric acid solution (NF X 31-160) and Olsen-P was extracted with 42 g
207 L⁻¹ NaHCO₃ (pH = 8.5) (Sims, 2000).

208 The WSP concentrations was determined following the standard method
209 developed by Murphy and Riley (1962). The Q_{max} and EPC₀ were determined based
210 on Langmuir P sorption isotherms obtained using the method proposed by Pöthig et
211 al., (2010). Briefly, the air-dried and sieved soil (< 2 mm) of 1.0 g was suspended in
212 50 mL of 0.01 M CaCl₂ solution containing various initial P concentrations: 0, 0.5, 1,
213 5, 10, 25, 50, 75 mg L⁻¹, and centrifuged at 1500 rpm for 10 min, suspensions were
214 filtered (<0.45 μm), and soluble reactive P concentration of the supernatant was

215 measured colorimetrically. The sorbed P (SP) was calculated from the difference
216 between the measured P concentration of the filtrate (EPC) and the initial P
217 concentration of the solution before equilibrium with the soil. The P sorption
218 isotherms are obtained by plotting SP against EPC (Pöthig et al., 2010). The Q_{max} was
219 determined by fitting a Langmuir equation to the last three points (25, 50, 75 mg P
220 L^{-1}), as follows:

$$221 \quad Q = (c * K * Q_{max}) / (1 + K * c) \quad (1)$$

222 where Q is the total amount of P sorbed ($mg\ mg^{-1}$), c is the concentration of P in
223 the equilibrium solution, which is also EPC ($mg\ L^{-1}$) and K is a constant parameter (L
224 mg^{-1}). The EPC_0 represents the solution P concentration at which no net sorption or
225 desorption of P would occur between soil and solution. To calculate EPC_0 values, the
226 data points corresponding to the first three concentrations (0, 0.5, 1 $mg\ P\ L^{-1}$) of the
227 isotherms were plotted as a straight line, and the EPC_0 value calculated as the
228 intersection of this straight line with the X axis ($SP = 0$) (Pöthig et al., 2010).

229 The DPS_{OX} was calculated as the molar ratio of acid ammonium oxalate extracts
230 as follows (van der Zee et al., 1987):

$$231 \quad DPS_{OX} = P_{OX} / \alpha (Al + Fe)_{OX} \quad (2)$$

232 where P_{OX} , Al_{OX} , and Fe_{OX} are quantified in acid ammonium oxalate extracts ($mmol$
233 kg^{-1}). The sum of Al_{OX} and Fe_{OX} is termed P sorption capacity (PSC, $mmol\ kg^{-1}$). We
234 used an average α value of 0.5 to calculate DPS_{OX} (van der Zee et al., 1987; Messiga
235 et al., 2021).

236 Molybdate reactive dissolved phosphorus (MRDP) runoff filtrates were
237 determined colorimetrically by direct reaction with ammonium molybdate (Murphy

238 and Riley, 1962). The same method was used for total dissolved phosphorus (TDP),
239 but after digestion of filtrates in acidic potassium persulfate. The precision of MRDP
240 and TDP measurements was ± 4 and $\pm 13 \mu\text{g L}^{-1}$, respectively. The difference
241 between TDP and MRDP was defined as the molybdate unreactive dissolved
242 phosphorus (MUDP), which includes truly dissolved organic P as well as both organic
243 and inorganic P associated with colloidal particles (Haygarth and Sharpley, 2000).

244 The DOC concentrations were analyzed with a total organic analyzer (Shimadzu
245 TOC-5050A), with a precision of $\pm 5\%$ (using potassium hydrogen phthalate as the
246 standard solution) (Gu et al., 2018).

247 *2.5. Statistical analyses*

248 Pearson correlation analysis was carried out to identify relationships among the
249 various soil parameters (P content, DPS_{OX} , WSP, EPC_0 , Q_{max}) and the amount of
250 dissolved P in runoff waters. All statistical analyses were conducted using the SPSS
251 22.0 (IBM, USA) software package. Figures were created using the Origin 8.0
252 (OriginLab, USA) software package. Measurements in triplicate of each sample were
253 conducted, and their arithmetic mean values were reported.

254 **3. Results**

255 *3.1. Soil physicochemical properties*

256 Comparison of soil physicochemical properties showed that tillage management
257 had no regular effect on soil particle size, metal oxide contents, and pH values (Table
258 1). Conversely, ST and NT obviously increased TOC, OM, TP, Dyer-P, Olsen-P, and

259 P_{OX} contents of soils compared to CT. For TOC, the increase was 23-42% for plot A
260 soils (pig manure), and 34-50% for plot B soils (poultry manure). The TP increases
261 were also obvious, ranging from 6-9% for plot A soils and 18-20% for plot B soils,
262 respectively. The highest increases were found for Dyer-P and Olsen-P, with increases
263 in between 36-64% and 28-79%, respectively, for plot A soils, and in between 57%
264 and 25-59%, respectively, for plot B soils. Thus, the switch from conventional tillage
265 to conservation tillage led to a strong enrichment of soils in OM, TP, Olsen-P and
266 Dyer-P in the topmost layers (0-5cm). It was noteworthy that the addition of poultry
267 manure yielded to the highest increases of OM, TP, Dyer-P and Olsen-P at comparable
268 tillage modality.

269 **Table 1** Basic surface soil (0-5 cm) characteristics of each field experiment site. The data
270 represent the mean ± standard deviation.

271 3.2. Soil sorption properties

272 **Fig. 2.** Plot A (pig manure applied) (a) and plot B (poultry manure applied) (b) soil adsorption
273 isotherm fitting curves. CT = conventional tillage; ST = shallow tillage; NT = no-tillage.

274 **Table 2** Phosphorus sorption parameters of the studied soils. Each value is the mean of three
275 separate measurements.

276 The NT and ST changed the parameters of soils to bind P (Table 2). Compared
277 with CT, the dissolved P indexes (EPC_0 and WSP) that equilibrated with the solid
278 phase P were increased due to the long-term implementation of NT and ST. The
279 average EPC_0 and WSP of soils in CT were 0.60 and 3.10 mg L⁻¹, but they were
280 increased to 1.73 and 4.85 mg L⁻¹ after converted to NT and ST, respectively. The
281 potential of soils to bind P exhibited similar characteristics with Q_{max} and DPS_{OX}
282 increased by different extents after converted to NT and ST. The average magnitudes
283 of Q_{max} and DPS_{OX} were increased by 10.3% and 19.7%, respectively.

284 The change of soils in sorption property highly depended on the type of
285 conservation tillage practices. Soils in NT had a high Q_{max} and DPS_{OX} , also a large
286 EPC_0 and WSP, compared with those in ST. For example, the average values of EPC_0 ,
287 Q_{max} , WPS and DPS_{OX} of soils in ST were merely 1.60 mg L⁻¹, 1249 mg kg⁻¹, 4.33 mg
288 L⁻¹ and 15.13% respectively, but the corresponding values of soils in NT were as high
289 as 1.87 mg L⁻¹, 1326 mg kg⁻¹, 5.36 mg L⁻¹ and 16.54%, respectively.

290 The change of soils in sorption property was also related to the type of manures.
291 The application of poultry manure had a greater impact on soil sorption properties

292 than pig manure. Except for Q_{\max} , all the indicators of soils P sorption in plot B
293 (poultry manure applied) were higher than those in plot A (pig manure). For example,
294 EPC_0 of soils in B-NT increased by 517% compared with A-NT.

295 *3.3. Runoff P concentration and flux*

296 **Fig. 3.** Hydrographs of rainfall simulation experiments for plot A (pig manure applied) (a) and
297 plot B (poultry manure applied) (b) soils. CT = conventional tillage; ST = shallow tillage; NT =
298 no-tillage.

299 Runoff development in the rainfall simulation experiments could be divided into
300 four steps (Fig. 3). The first step, which lasted between 20 and 50 minutes, started
301 from the onset of rainfall and went on until the first drops of runoff water could be
302 collected in the pipe at the downstream edge of the experimental plate. During this
303 step, water-saturated areas were observed at the soil surface but the lack of connection
304 between these areas prevented the collection and downward transfer of the surface
305 accumulated water. The second step corresponded to the onset of the connection of
306 water saturated surface areas each other and with the runoff collecting pipe device,
307 but runoff flux was low and constant. The third step corresponded to the sudden
308 connection of all water saturated surface area and was marked by a sudden increase of
309 the runoff flux. Finally, the fourth step corresponded to the installation of a
310 steady-state regime during which runoff volumes were high and temporally constant.

311 **Table 3** Average concentrations and total fluxes of different P forms and DOC in rainfall water

312 during simulated rainfall.

313 Table 3 showed the total amount of collected runoff water, as well as average
314 MRDP, TDP, MUDP, and DOC concentrations and total fluxes for each of the six
315 rainfall simulation experiments. The volumes of collected runoff water were quite
316 similar for all experiments (between 9400 and 11950 mL), except that soil B-NT,
317 which yielded to a much lower runoff volume (6150 mL). The average TDP, MRDP,
318 MUDP and DOC in runoff waters were in an order $NT > ST > CT$. Thus, ST and NT
319 markedly increased dissolved P in runoff waters, compared with CT. However, a
320 notable exception was the B-NT experiment, whose highest concentrations were not
321 passed on to the level of total P losses/fluxes due to the delay in the onset of step 4 of
322 the runoff episode recorded and the much lower collected volume of runoff water.

323 In MRDP concentrations increased by 143% and 104% for plot A and B soils,
324 respectively (Table 3). The increase was even greater for MUDP, which was increased
325 by 165 and 434%, respectively. The impact of tillage methods on DOC in runoff was
326 similar to that of dissolved P, with NT yielding higher DOC than other modalities.
327 However, the amplitude of the variations was much lower (increase $<10\%$) than that
328 observed for dissolved P. Clearly, the application of poultry manure increased the
329 concentration of all dissolved P forms in runoff as compared to pig manure, especially
330 MUDP.

331 **Fig. 4.** Influences of tillage (a, b, c, d) and fertilization (e, f, g, h) practices on concentrations of
332 TDP, MRDP, MUDP and DOC in runoff waters during simulated rainfall experiments. CT =

333 conventional tillage; ST = shallow tillage; NT = no-tillage; MRDP = molybdate reactive dissolved
334 phosphorus; MUDP = molybdate unreactive dissolved phosphorus; TDP = total dissolved
335 phosphorus; DOC = dissolved organic carbon.

336 The change of MRDP in runoff showed a tendency to decrease during the
337 experiment (Fig. 4). The magnitude of the decreases clearly increased with the level
338 of initial MRDP in runoff, with decreases in the range 20-30% for the high initial
339 concentrations, as against <5% for the lowest initial concentrations. As the MRDP
340 concentrations increased in the experiments in the CT < ST < NT direction, the
341 amplitudes of decrease of concentrations are also CT < ST < NT.

342 Temporal decreases were also observed for TDP and MUDP (Fig. 4), with the
343 same positive correlations between the relative decrease amplitude and the initial
344 levels as observed for MRDP. A major difference, however, was that the decrease
345 rates of TDP and MUDP were not constant over time. In fact, the decay amplitudes
346 were very strong during the stage 2 of runoff development, and declined then
347 stabilized at very low levels. This dynamic was particularly well expressed by the
348 treatments of A-NT, B-NT, and B-ST, which had highest initial concentrations of TDP
349 and MUDP.

350 To conclude, compared to CT, the conversion of soil management to ST and NT
351 led to more export of MUDP (along with more export of reactive P), a large part of
352 these MUDP being solubilized and exported quickly during the early stages of runoff.

353 **4. Discussion**

354 *4.1. Impact of soil management on soil P sorption property and dissolved P loss*

355 Our results show that NT and ST significantly increase total P content of the
356 topmost soil layers (0-5 cm), indeed, consistent with those reported (Ulén et al., 2010;
357 Baker et al., 2017; Daryanto et al., 2017; Jarvie et al., 2017; Duncan et al., 2019). It is
358 also true that the enrichment of TP in the topmost soil layers in NT and ST increases P
359 agronomical availability (indicated by Olsen-P and Dyer-P) (Table 1), which have
360 been observed in loamy soils (Sokolowski et al., 2020). Compared with soils under
361 CT, soils under ST and NT treatment usually contained high crop residues, which
362 upon degradation by soil organisms are expected to release significant proportions of
363 P (Sokolowski et al., 2020; Ni et al., 2020; Daryanto et al., 2017), which will tend to
364 stay on the soil surface due to lack of mixing imposed by the ST and NT management
365 practices, thus leading to an accumulation of the released P into the topmost soil
366 layers (Zhang et al., 2017).

367 Many previous studies on P sorption characteristics of agricultural soils
368 emphasizes the dominant role of mineral (Al, Fe, Ca), OM content and other soil
369 properties, all of which can increase the capacity of soils to fix newly applied P and
370 thus reduce P loss from soil through (Messiga et al., 2021; Pradhan et al., 2021;
371 Messiga et al., 2011). In this study, compared with conventional tillage, the DPS_{OX} of
372 conservative tillage always increased, regardless of the content of (Fe and Al)_{OX}
373 increased or decreased, thus (Fe and Al)_{OX} may not be the key factor affecting soil
374 DPS_{OX} .

375 Increasing soil OM can improve the crop availability of soil P, but it can also
376 increase the risk of P loss. The main mechanism is that OM can reduce the sorption
377 strength of soils to bind P. The substances in soils that directly bind P through
378 chemical bonds mainly include Fe, Al and Mn oxides, secondary clay minerals, and
379 calcium carbonate. The OM will compete with P for binding sites on the surface of
380 these substances. Moreover, OM may bind to the sorption sites thereby increasing the
381 negative charges of the mineral surface, which results in a reduction in the
382 electrostatic attraction between phosphate and soil surface. The reduction of bonding
383 energy with conservative tillage and poultry manure application, could increase
384 dissolved P loss through runoff as suggested by Pradhan et al. 2021 and Abboud et al.
385 (2018) as well as this study (Table 3 and Fig. 4).

386 It should be noted that the soil would have a different structure in situ (the
387 porosity of the soil and the straw coverage, etc.), which will affect water infiltration,
388 runoff, as well as P losses (Daryanto et al., 2017; Baker et al., 2018). In this study, we
389 are only concerned with the impact of tillage and fertilization on soil physicochemical
390 properties and thus on dissolved P loss. Therefore, the soil samples were screened
391 before simulating rainfall, soil structures in situ are not considered.

392 *4.2. Significance and origin of the MUDP compartment*

393 **Fig. 5.** Proportion of MUDP flux to TDP flux for plot A (pig manure applied) (a) and plot B
394 (poultry manure applied) (b) soils during simulated rainfall experiments. CT = conventional tillage;
395 ST = shallow tillage; NT = no-tillage; MUDP = molybdate unreactive dissolved phosphorus.

396 The tillage practice change from CT to ST and NT stimulates the loss of MUDP.
397 The P loss in MUDP accounts for 11-27% of TDP flux with the highest loss occurring
398 at the onset of runoff development, reaching to 50% of total TDP flux in ST and NT
399 (Fig. 5). This result is very important insofar as i) this compartment is rarely
400 quantified in the role of conservation agriculture practices on the risks of dissolved P
401 transfer (Daryanto et al., 2017), and ii) like MRDP, MUDP compartments (organic P
402 and colloidal P) also contains some bioavailable P forms to aquatic organisms (Li and
403 Brett, 2013; Van Moorlehem et al., 2013; Li et al., 2015). Thus, the loss of MUDP is
404 of great concern as regards eutrophication risks in cultivated landscapes, and should
405 be taken into account during implementation of conservation agriculture.

406 As shown in Fig. 5, the formation and rapid exhaustion of a MUDP peak
407 resemble very closely to the flush of MUDP observed by Gu et al. (2018)
408 demonstrated that MUDP consists in P bound to a mixture of OM and
409 Fe(Al)oxide-rich colloids, which is a well-established feature (Liu et al., 2014). It is
410 also well established that drying of the soil matrix generates soil colloids, which can
411 be mobilized and flushed out of the soil matrix during rewetting (Mohanty et al.,
412 2015). Runoff development as simulated here is similar to the rewetting of dried soils,
413 so we can reasonably speculate that it is the rewetting processes that results in the loss
414 of P bound to organic-rich colloids as observed by Gu et al. (2018). Based on this
415 origin of MUDP, it is not difficult to explain the observed transient peaks of MUDP at
416 the onset of runoff development.

417 The hypothesis of a colloidal origin of MUDP compartment does not rule out the

418 possibility that it also contains a part of truly dissolved organic P. In the soil solution,
419 indeed, there exist a part of dissolved organic P molecules from microbial cell lysis
420 due to osmotic shock upon soil rewetting (Blackwell et al., 2013). This part of organic
421 P is not directly reaction with ammonium molybdate, but truly available to aquatic
422 organism.

423 *4.3. Prediction of P loss from soil indicators*

424 **Fig. 6.** Relationships between the concentration of TDP (a), MRDP (b) and MUDP (c) in runoff
425 and the different indicators tested here to predict the risk of dissolved P release. EPC_0 =
426 equilibrium phosphorus concentrations; Q_{max} = phosphorus sorption maximum; WSP =
427 water-soluble phosphorus; MRDP = molybdate reactive dissolved phosphorus; MUDP =
428 molybdate unreactive dissolved phosphorus; TDP = total dissolved phosphorus; DOC = dissolved
429 organic carbon.

430 Both the results of soil sorption properties and simulation experiments showed that
431 NT and ST aggravated the loss of dissolved P in the topmost soil layers. To explore
432 the possibility using soil sorption indicators to predict the dissolved P loss, the
433 relationship between EPC_0 , WSP, Q_{max} and DPS_{OX} indicators and the concentration
434 and fluxes of TDP, MRDP, MUDP are analyzed. The results shows that EPC_0 , WSP,
435 and DPS_{OX} are positively and significantly correlated with the concentration and
436 fluxes of TDP, MRDP, MUDP in runoff, suggesting that these three indicators could
437 be used to assess P loss risks (Fig. 6; Table S1 in Supplementary Data). The Q_{max} has
438 no correlation with P concentrations and fluxes in runoff (Fig. 6; Table S1 in

439 Supplementary Data). This indicates that Q_{\max} is not a suitable indicator for assessing
440 the risk of P release. Because Q_{\max} is related to the number P sorbing sites in the soil,
441 and not to the strength of these sites, which is the key parameter to consider when
442 dealing with the dissolved P loss risk (Ma 2014; Schoumans and Chardon, 2015).

443 **Fig. 7.** Relationships between concentrations of available P (Dyer-P (a, b) and Olsen-P (c, d))
444 in soils and soil EPC_0 and WSP values. EPC_0 = equilibrium phosphorus concentrations; WSP =
445 water-soluble phosphorus.

446 However, the soil DPS_{OX} was always lower than 25%, indicating that the risk of
447 soil P release was not large (van der Zee et al., 1987), which is contrast to the results
448 of WSP, EPC_0 , available P as well as simulated rainfall experiments. Furthermore,
449 DPS_{OX} is also not a good indicator to evaluate the marked increases in dissolved P
450 loss caused by the change in tillage, because this switch not being accompanied by a
451 significant change in DPS, whether in the poultry or pig fertilization modality.
452 Compared with DPS_{OX} , the calculation of EPC_0 and WSP are convenient and simpler
453 soil P release risk assessment indicators. Therefore, EPC_0 and WSP are more suitable
454 for assessing changes in P release risk when shifting soil management from CT to ST
455 and NT.

456 The EPC_0 values show the most significantly positive correlation with MRDP,
457 TDP, and MUDP concentrations and fluxes in runoff ($R^2 = 0.910-0.997$, $P < 0.01$).
458 The EPC_0 values $> 0.03 \text{ mg L}^{-1}$ such as observed in all investigated plots, especially
459 those fertilized with poultry manure, are indicative of a high potential risk of
460 eutrophication due to the capacity of these soils to easily exchange large quantities of

461 dissolved P with runoff waters (US EPA, 1988). Indiatl and Sharpley (1998) and
462 Munira et al. (2018) have found that there is generally a threshold of EPC_0 in soils, the
463 position of which depends on the physicochemical properties of soils (Indiatl and
464 Sharpley, 1998; Hughes et al., 2000; Munira et al., 2018). Given that soil EPC_0 is
465 above the threshold, it strongly increases as a function of Olsen-P concentrations. We
466 also find that there are significantly linear correlations between EPC_0 values and
467 bioavailable P contents (Dyer-P and Olsen-P) (Fig. 7a, b). The slopes of the
468 correlations are quite high, indicating that the bioavailable P contents in these soils
469 may have exceeded the threshold.

470 Positive correlations are also obtained between WPS values and dissolved P and
471 fluxes in runoff (Fig. 6; Table S1 in Supplementary Data). This feature has been
472 already observed in several previous studies (Pöthig et al., 2010; Xue et al., 2014;
473 Fischer et al., 2017). Furthermore, the very significant positive correlation between
474 WSP and DPS_{OX} with r^2 of 0.9995 is found. van der Zee et al. (1987) report that the
475 risk of soil P loss can be predicted with the single quadratic regression and a critical
476 DPS_{OX} . When soil DPS_{OX} is more than 25%, the loss of dissolved P by runoff water is
477 high (Messiga et al., 2021). However, the linear positive correlation between WSP
478 and DPS_{OX} in this study illustrated that the critical value of DPS_{OX} is smaller than 25%
479 or insufficient amount of data. In a recent study, Messiga et al. (2021) have shown
480 that the concentration of WSP was 3.7 mg L^{-1} with critical DPS_{OX} value.
481 Comparatively, the same DPS_{OX} has a higher WSP value, indicating that the P in the
482 this studied soil is easier to lost.

483 Finally, soil Olsen-P and Dyer-P contents are also significantly positively
484 correlated with TDP, MRDP and MUDP concentrations and fluxes in runoff (Fig. 6;
485 Table S1 in Supplementary Data). Many published studies have reported such positive
486 correlations between Olsen-P and Dyer-P concentrations in soils and P loss from
487 runoff (Mcdowell et al., 2003; Hopkins and Ellsworth, 2005; Pizzeghello et al., 2016).
488 This means that agronomic soil tests for P can be also used as indicators to assess
489 dissolved P loss risk in soils, since these tests are widespread and not very expensive
490 (Pizzeghello et al., 2016). Some authors have reported a threshold of ca. 60 mg
491 Olsen-P kg⁻¹, above which the dissolved P loss risk increases sharply (Heckrath et al.,
492 1995; Johnston and Poulton, 1997). In this study, only for the soils amended with
493 poultry manure had Olsen-P concentrations > 60 mg kg⁻¹, suggesting the application
494 of poultry manure increases the loss risk of dissolved P in the topmost soil layers.

495 It can also be seen that the four tested indicators (EPC₀, WSP, Olsen-P, Dyer-P)
496 define linear correlations of very good quality when they are plotted against each
497 other, with no evidence of a break in the correlations. It can be concluded that in the
498 range of the P content values encountered in the soils of this study, EPC₀, WSP,
499 Olsen-P and Dyer-P values can be used indifferently as soil indicators to assess the
500 surplus of dissolved P loss risk caused by changing soil management from CT to ST
501 and NT. Obviously, one will favor the use of Olsen-P and Dyer-P concentrations for
502 reasons of ease of measurement and lower cost.

503 *4.4. Implication for management*

504 Results from this study confirm that if beneficial for limiting soil erosion and

505 export of particulate P, conservation practice can have the perverse effect of
506 exacerbated dissolved P losses in case of intense runoff development. Here, the
507 potential rise in dissolved P losses induced by switching soil management from CT to
508 NT is of a factor of 2 to 3 times (Table 3), and appears independent of the type of
509 fertilizer used. As indicated in the introduction, Brittany, the region in which the
510 Kerguehenenc station is located, is a region of intensive agriculture suffering from a
511 recurring problem of eutrophication of these surface waters, with a clear role of P as
512 the limiting nutrient causing these blooms (Senthilkumar et al., 2012; Perrotet al.,
513 2014; Bol et al., 2018). In a recent study, Guillemot et al. (2021) revealed that ca. 50%
514 of the catchments have median dissolved P concentrations already higher than the
515 eutrophication threshold of 0.03 mg L^{-1} (Yang et al., 2019), the other 50% catchments
516 having median concentration in the range $0.08\text{-}0.026 \text{ mg L}^{-1}$. In Brittany, the relative
517 surface area of soils conducted using NT techniques is only 25% of the total
518 cultivated area, and there is therefore room for progress to develop these techniques.
519 Taking into account the results of our study and those obtained by Guillemot et al.
520 (2021) we anticipate that the risk is great and a generalization of these techniques will
521 amplify the eutrophication problem that suffers Brittany given the already high P
522 contents of most of fresh water.

523 In light of these findings, we may recommend to combine NT with other land
524 management techniques (e.g., injection instead of broadcast application of P fertilizer,
525 inclusion of cover crops to scavenge residual P) to limit the risk of dissolved P losses.
526 Occasional soil harrowing as suggested by Daryanto et al. (2017) may be also a

527 solution to overcome the surface P accumulation problem. Above all, it seems that
528 Brittany farmers should be primarily recommended to limit soil fertilization in the
529 case of guaranteed crop yield crop yield, so as to reduce the overall P content of
530 Brittany soils.

531 **5. Conclusion**

532 Our experimental results confirm that conservation agriculture may increase the
533 risk of all dissolved P loss during runoff. The prime effect causing this risk is clearly
534 the P enrichment of the topmost soil layers caused by ST and NT. Moreover,
535 conservation agriculture contributes to release MUDP into the runoff, thereby
536 increasing the total flux of dissolved P emitted out of the plots. The main species is
537 most probably colloidal P, followed in lesser proportion by dissolved organic P. These
538 species are also bioavailable for aquatic organisms, and in particular algae.

539 The concentration and flux of TDP, MRDP, and MUDP in runoff were B (poultry
540 manure)-NT > B-ST > B-CT > A (pig manure)-NT > A-ST > A-CT. Clearly, the
541 application of poultry manure increased the concentration and flux of all dissolved P
542 in runoff waters as compared to pig manure, especially as regards MUDP. Special
543 attention is required to MUDP that accounts for 11-27% of TDP fluxes, the highest
544 value can reach 50%, which suggests that MUDP should not be ignored, especially in
545 the early stages of rainfall. The EPC₀, WSP, agronomically Olsen-P and Dyer-P
546 content can be used as indicators of the risk of P loss, since they had a significantly
547 positive correlation with dissolved P concentration and fluxes in runoff. The Olsen-P

548 and Dyer-P are also commonly measured by farmers to control crops and are
549 technically easy.

550 Considering the fairly high dissolved P concentrations of most fresh water in
551 Brittany, local authorities in charge of water quality protection and farmers must be
552 aware of the risk involved in systematizing the development of conservation
553 agriculture. It is recommendation that decrease in the amounts of P brought to soil and
554 development of complementary practices specifically dedicated at maintaining low
555 runoff such as permanent cover crops.

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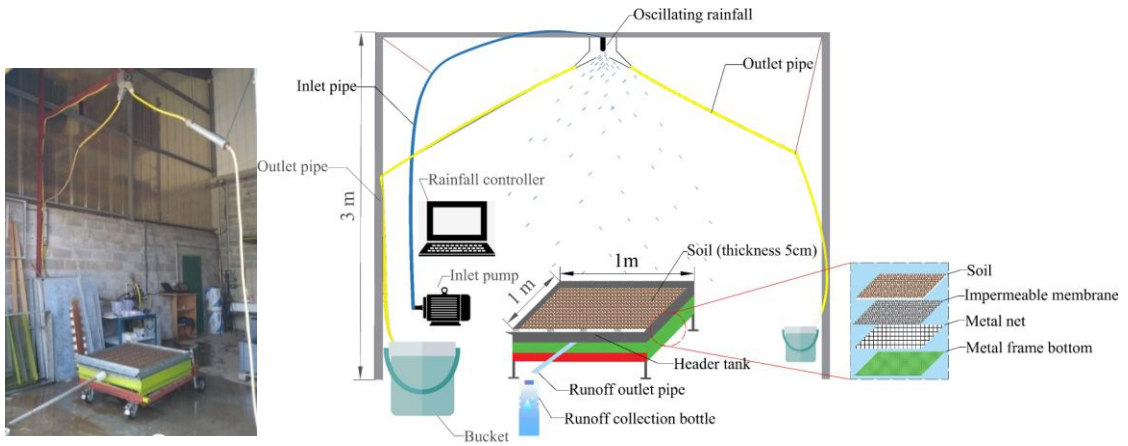
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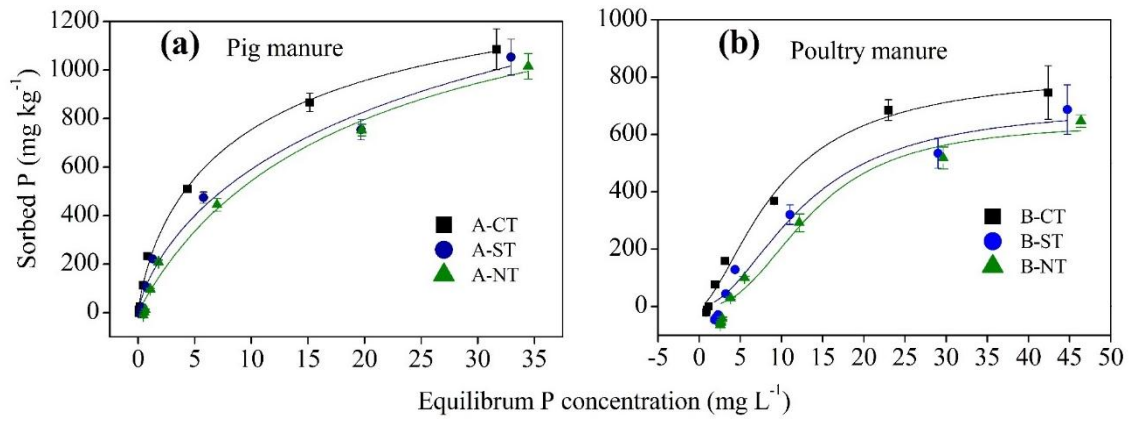
730 **Figure**



731

732

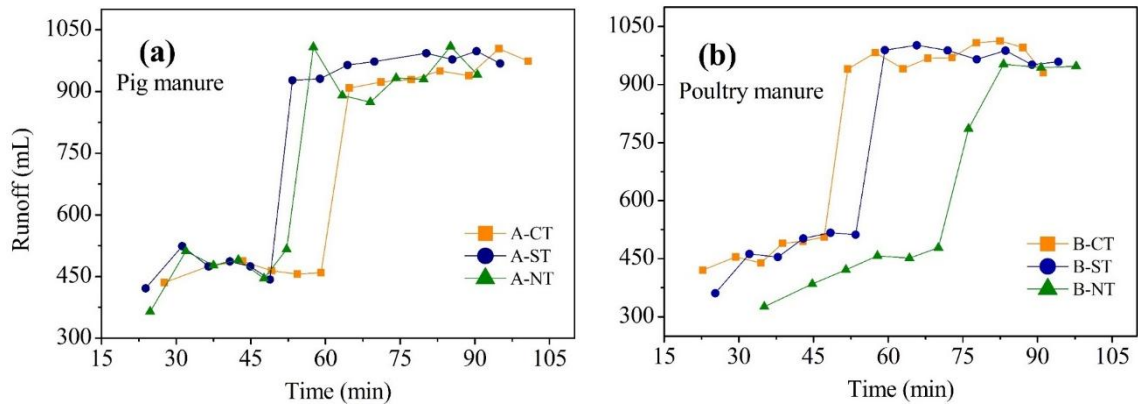
Fig. 1. Rainfall simulation device.



733

734 **Fig. 2.** Plot A (pig manure applied) (a) and plot B (poultry manure applied) (b) soil adsorption

735 isotherm fitting curve. CT = conventional tillage; ST = shallow tillage; NT = no-tillage.

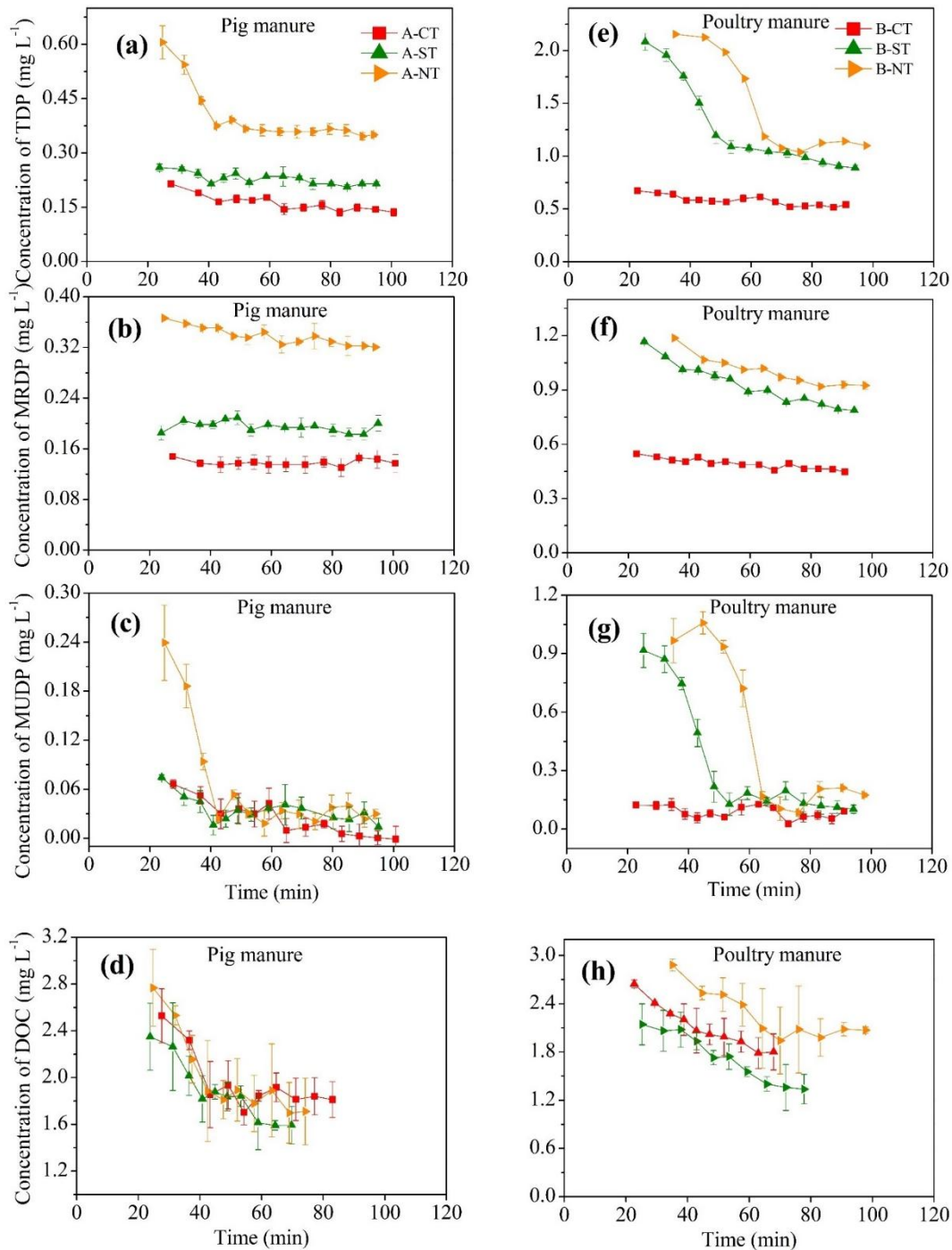


736

737 **Fig. 3.** Hydrographs of rainfall simulation experiments for plot A (pig manure applied) (a) and

738 plot B (poultry manure applied) (b) soils. CT = conventional tillage; ST = shallow tillage; NT =

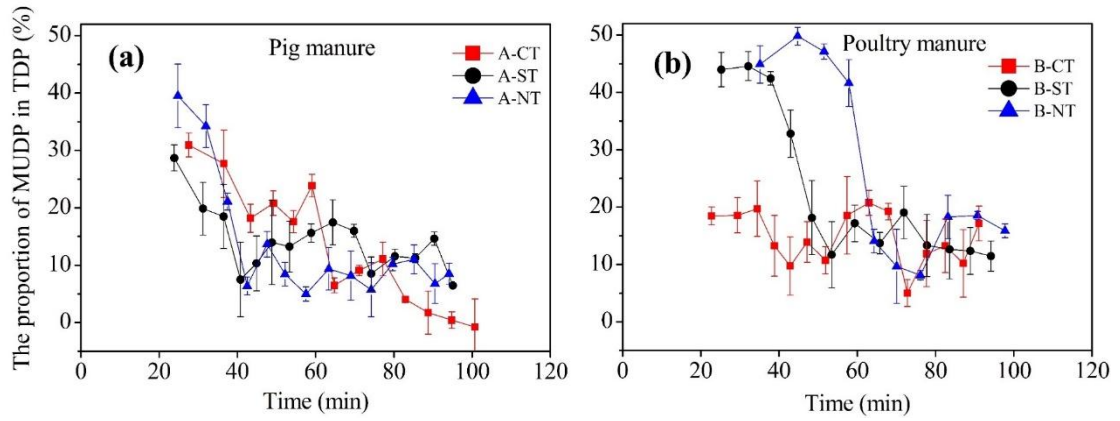
739 no-tillage.



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741

742 **Fig. 4.** Influences of tillage (a, b, c, d) and fertilization (e, f, g, h) practices on concentrations of
 743 TDP, MRDP, MUDP and DOC in runoff waters during simulated rainfall experiments. CT =
 744 conventional tillage; ST = shallow tillage; NT = no-tillage; MRDP = molybdate reactive dissolved
 745 phosphorus; MUDP = molybdate unreactive dissolved phosphorus; TDP = total dissolved
 746 phosphorus; DOC = dissolved organic carbon.

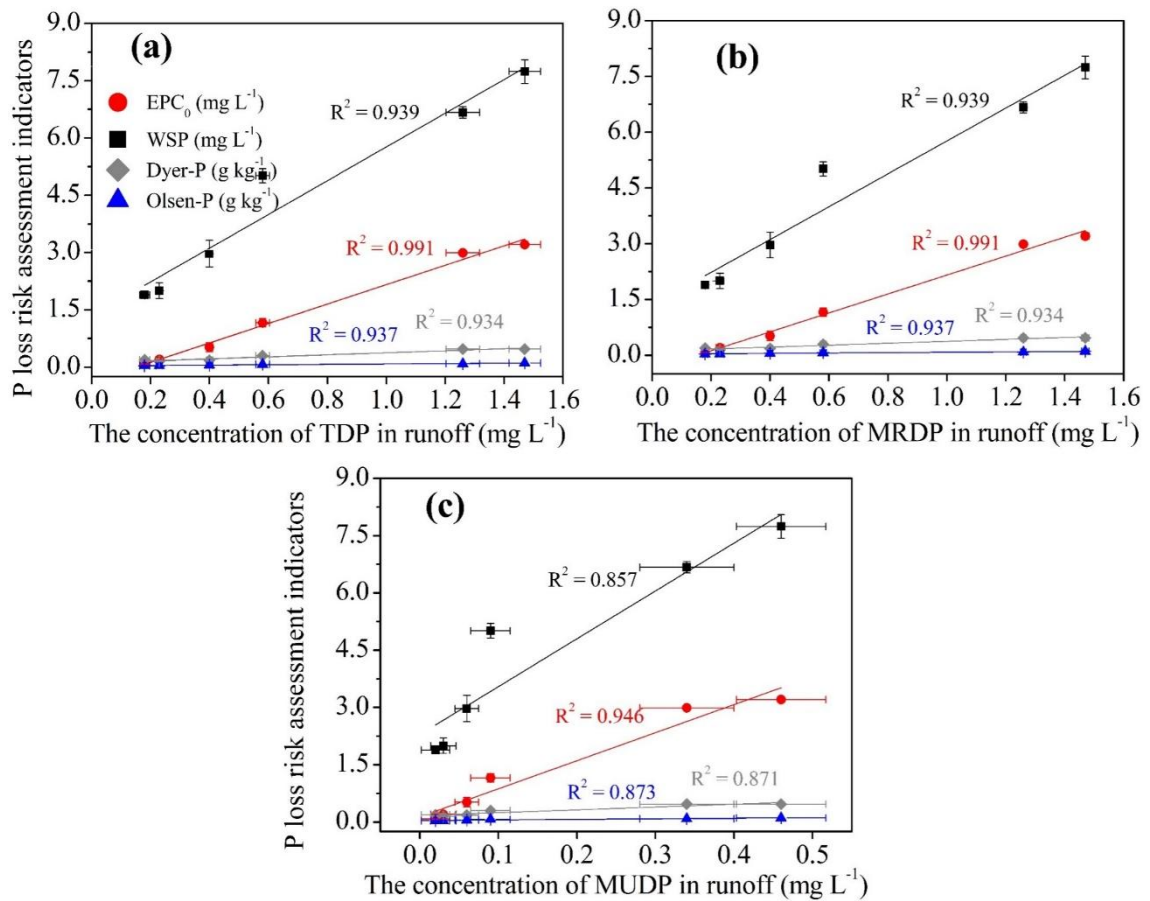


747

748 **Fig. 5.** Proportion of MUDP flux to TDP flux for plot A (pig manure applied) (a) and plot B

749 (poultry manure applied) (b) soils during simulated rainfall experiments. CT = conventional tillage;

750 ST = shallow tillage; NT = no-tillage; MUDP = molybdate unreactive dissolved phosphorus.



751

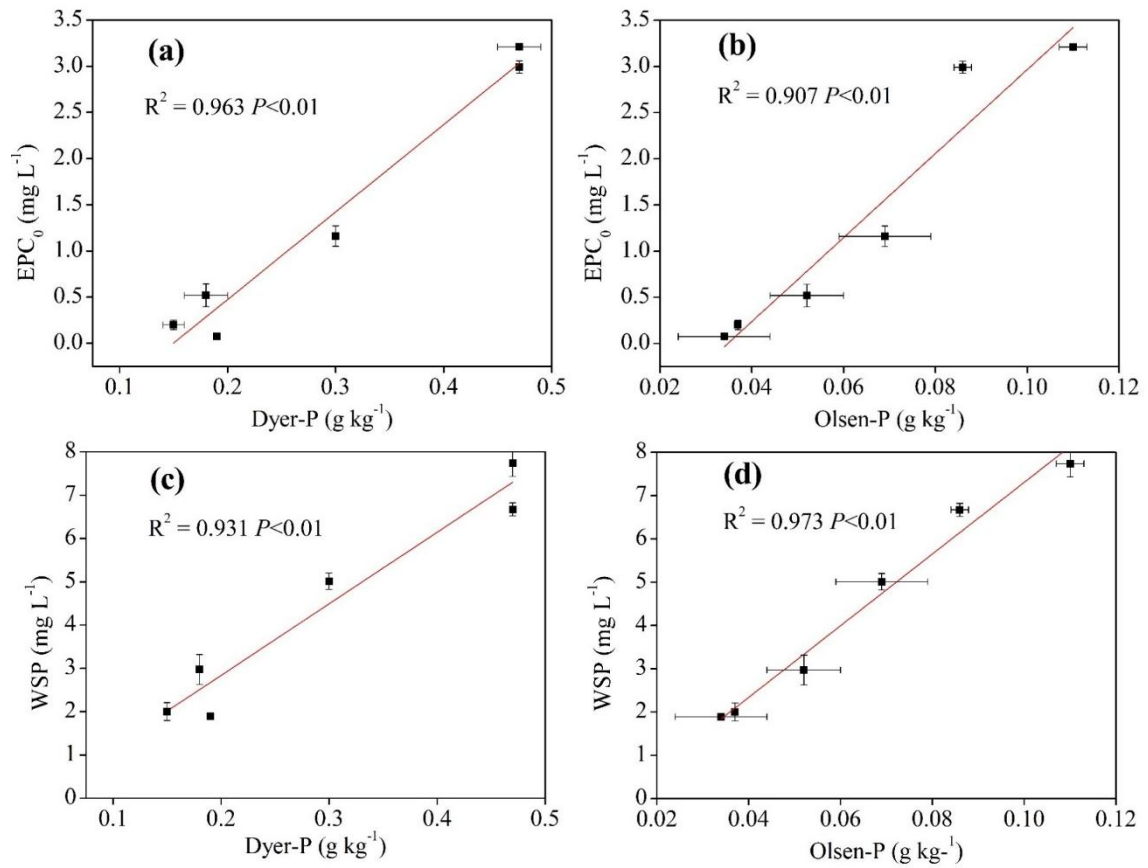
752 **Fig. 6.** Relationships between the concentration of TDP (a), MRDP (b) and MUDP (c) in runoff

753 and the different indicators tested here to predict the risk of dissolved P release. EPC₀ =

754 equilibrium phosphorus concentrations; WSP = water-soluble phosphorus; MRDP = molybdate

755 reactive dissolved phosphorus; MUDP = molybdate unreactive dissolved phosphorus; TDP = total

756 dissolved phosphorus.



757

758 **Fig. 7.** Relationships between concentrations of available P (Dyer-P (a, c) and Olsen-P (b, d) in

759 soils and soil EPC₀ and WSP values. EPC₀ = equilibrium phosphorus concentrations; WSP =

760 water-soluble phosphorus.

761

762 **Table**

763 **Table 1**

764 Basic surface soil (0-5 cm) characteristics of each field experiment site. The data represent the mean \pm standard deviation.

	Clay	Silt	Sand	pH	TOC	OM	Al-ox	Fe-ox	Si-ox	TP	Dyer-P	Olsen-P	P _{OX}
	%				g kg ⁻¹								
A-CT (Pig)	15.5	47.4	37.0	6.29	18.7	32.4	2.26	5.35	0.31	1.16	0.11	0.029	0.29
	± 0.6	± 0.5	± 1.2	± 0.04	± 0.5	± 0.9	± 0.21	± 0.16	± 0.03	± 0.03	± 0.01	± 0.00	± 0.01
A-ST (Pig)	15.7	47.4	36.9	6.35	23.0	39.7	2.23	5.20	0.31	1.23	0.15	0.037	0.32
	± 0.8	± 0.5	± 0.5	± 0.04	± 0.7	± 1.2	± 0.15	± 0.27	± 0.02	± 0.06	± 0.02	± 0.02	± 0.01
A-NT (Pig)	15.3	50.5	34.2	6.45	26.5	45.9	2.25	5.04	0.31	1.26	0.18	0.052	0.35
	± 0.4	± 0.8	± 0.8	± 0.05	± 0.9	± 1.5	± 0.05	± 0.24	± 0.01	± 0.06	± 0.00	± 0.01	± 0.02
B-CT (Poultry)	15.2	43.0	41.8	6.10	21.8	37.6	2.05	6.14	0.31	1.74	0.30	0.069	0.46
	± 0.0	± 0.7	± 0.1	± 0.09	± 0.2	± 0.6	± 0.03	± 0.17	± 0.00	± 0.01	± 0.02	± 0.02	± 0.02
B-ST (Poultry)	15.6	43.2	41.2	6.37	29.3	50.7	2.27	6.35	0.33	2.05	0.47	0.086	0.57
	± 0.4	± 0.3	± 0.8	± 0.09	± 0.2	± 0.5	± 0.06	± 0.07	± 0.00	± 0.01	± 0.00	± 0.01	± 0.01
B-NT (Poultry)	15.8	45.2	39.0	5.90	32.7	56.7	2.37	6.28	0.31	2.09	0.47	0.11	0.63
	± 0.3	± 0.5	± 0.8	± 0.02	± 0.2	± 0.6	± 0.00	± 0.11	± 0.01	± 0.00	± 0.02	± 0.02	± 0.01

765 TOC = total organic carbon; OM = organic matter; Oxalate-Al = Al-ox; Oxalate-Fe = Fe-ox; Oxalate-Si = Si-ox; TP = total phosphorus; CT = conventional tillage;

766 ST = shallow tillage; NT = no-tillage.

767 **Table 2**

768 Phosphorus sorption parameters of the studied soils. Each value is the mean of three separate
 769 measurements.

	A-CT (Pig)	A-ST (Pig)	A-NT (Pig)	B-CT (Poultry)	B-ST (Poultry)	B-NT (Poultry)
EPC ₀ (mg L ⁻¹)	0.039	0.20	0.52	1.16	2.99	3.21
Q _{max} (mg kg ⁻¹)	1315	1404	1515	1020	1094	1137
WSP (mg L ⁻¹)	1.19	2.00	2.97	5.01	6.67	7.74
DPS _{OX} (%)	10.27	11.64	12.83	16.12	18.62	20.25

770 CT = conventional tillage; ST = shallow tillage; NT = no-tillage; EPC₀ = equilibrium phosphorus
 771 concentrations; Q_{max} = phosphorus sorption maximum; WSP = water soluble phosphorus; DPS_{OX}
 772 = degree of phosphorus saturation.

773 **Table 3**

774 Average concentrations of runoff waters and total P and DOC losses measured during simulated
775 rainfall.

	Runoff volume (mL)	Average P concentration (mg L ⁻¹)				Total flux (mg)			
		MRDP	TDP	MUDP	DOC	MRDP	TDP	MUDP	DOC
A-CT(Pig)	9405	0.14	0.16	0.023	1.96	1.30	1.46	0.16	12.5
A-ST(Pig)	11945	0.20	0.23	0.033	1.88	2.32	2.68	0.37	12.0
A-NT(Pig)	9506	0.34	0.40	0.061	2.01	3.19	3.66	0.47	12.6
B-CT(Poultry)	11554	0.49	0.58	0.086	2.11	5.60	6.55	0.96	13.5
B-ST(Poultry)	9652	0.93	1.26	0.335	1.73	8.64	11.10	2.46	11.0
B-NT(Poultry)	6149	1.00	1.47	0.460	2.26	6.00	8.20	2.20	13.4

776 The average P concentration in the runoff is the weighted average value of the P concentration in

777 the runoff in each time period; total flux is the sum of P flux in each time period. CT =

778 conventional tillage; ST = shallow tillage; NT = no-tillage, MRDP = molybdate reactive dissolved

779 phosphorus; MUDP = molybdate unreactive dissolved phosphorus; TDP = total dissolved

780 phosphorus; DOC = dissolved organic carbon.

781

782 **Supplementary materials**

783 **Table S1**

784 Pearson's correlation analysis between P loss flux and concentration in runoff and P release risk
785 assessment parameters (n = 6).

	Concentration			Flux		
	TDP	MRDP	MUDP	TDP	MRDP	MUDP
EPC ₀	0.991**	0.991**	0.976**	0.932**	0.892*	0.990**
Q _{max}	-0.625	-0.653	-0.560	-0.732	-0.740	-0.672
WSP	0.939**	0.959**	0.857**	0.928**	0.903**	0.948**
DPS _{OX}	0.969**	0.980*	0.923**	0.928**	0.903*	0.949**
Dyer-P	0.934**	0.934**	0.871**	0.943**	0.910**	0.981**
Olsen-P	0.937**	0.937**	0.873**	0.881*	0.847*	0.928**

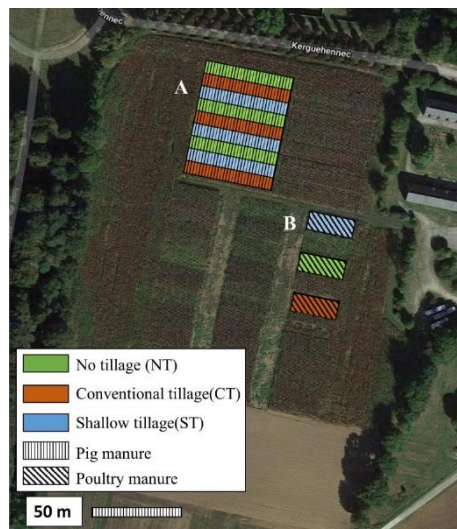
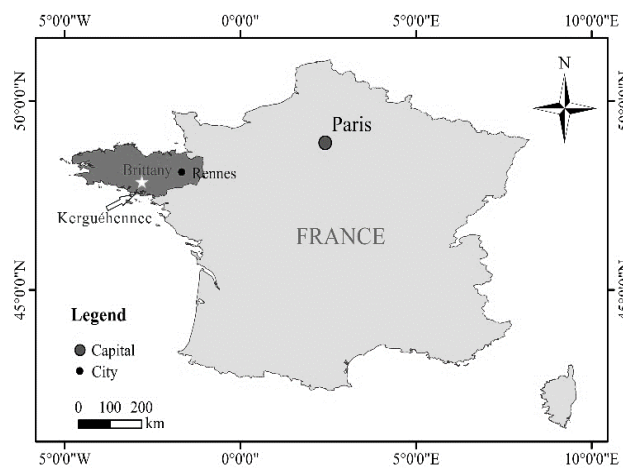
786 * significant at $P < 0.05$, ** significant at $P < 0.01$; TDP = total dissolved phosphorus; MRDP =

787 molybdate reactive soluble phosphorus; MUDP = molybdate unreactive dissolved phosphorus;

788 EPC₀ = equilibrium phosphorus concentrations; Q_{max} = phosphorus sorption maximum; WSP =

789 water-soluble phosphorus; DPS_{OX} = degree of phosphorus saturation.

790



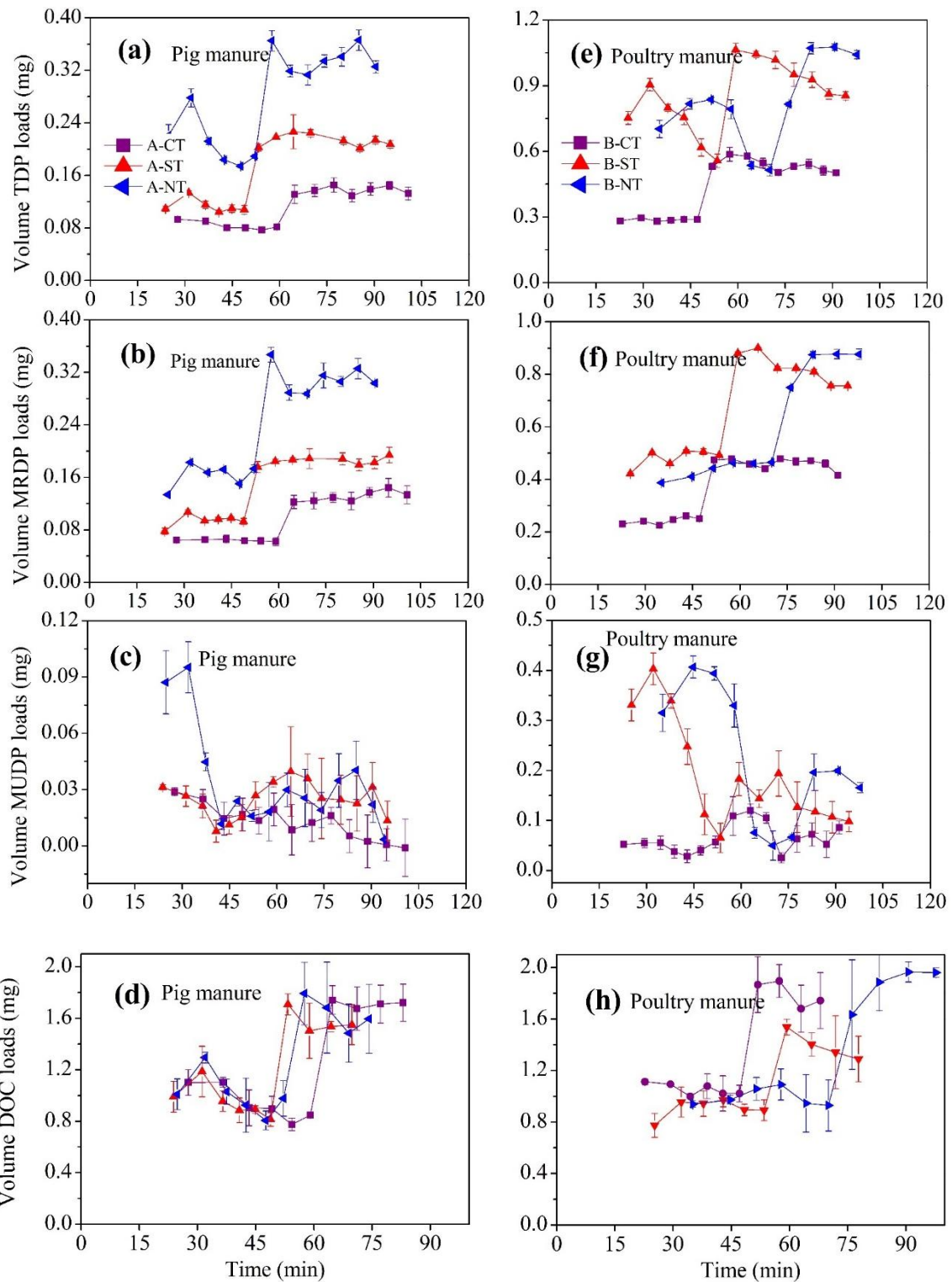
791

792 **Fig. S1.** Location map (left) and aerial photograph (right) of the Kerguéhennec experimental

793 station. Plot A represents the fertilization management with pig manure. Plot B represents the

794 fertilization management with poultry manure.

795



796

797

798 **Fig. S2.** Influences of tillage (a, c, e, g) and fertilization (b, d, f, h) on flux of TDP, MRDP
 799 and DOC in runoff during simulated rainfall. TDP = total dissolved phosphorus; MRDP =
 800 molybdate reactive dissolved phosphorus; MUDP = molybdate unreactive dissolved phosphorus;
 801 DOC = dissolved organic carbon; CT = conventional tillage; ST = shallow tillage; NT = no-tillage.