

## ***Interactive comment on “Contrasting physical controls on phosphorus transport to shallow groundwater at the hillslope scale” by Maelle Fresne et al.***

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General comments:

The authors submitted results of a study investigating controls on phosphorus (P) transport from soils to groundwater by application of the one-dimensional transport model Hydrus 1D. The manuscript is well-written and comprehensible in applied methods. Processes influencing P transport in soils and export to ground-/head water were investigated before in many studies for different ecosystems, but still is highly relevant in research because understanding of P mobilization, accumulation, and translocation within soils is incomplete. Therefore, the transfer from observations into adequate

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modeling approaches too is still a challenge. In this regard, the basic approach of the presented study to combine water and P fluxes in a model to simulate P transport under different boundary conditions is very interesting. However, I have serious concerns about the lack of consideration of biogeochemical understanding of P cycling, especially in the modelling approach presented and in the interpretation and discussion of the study results. My main concerns are: (1) to treat P as conservative tracer in the model and therefore the lack of consideration of any biological or chemical controls on P transport. It is well-known in literature that P behaves not conservative in soils. (2) to ignore preferential flow P transport, whose importance has been shown in recent studies. (3) the lack of validation of model results both by means of own observations and literature data. (4) to discuss spatial and temporal variability based on results of only 1-year observation with 1 value per month and on results of a 1D-model with exactly 1 vertical flow process (matrix flow) at two single points on a hillslope. Nevertheless, I appreciate the approach of investigating soil physical controls on water movement and this should be, in my opinion, the central theme of the paper. This is the crucial requirement to simulate solute fluxes in a next step, by including solute-specific information on processes in soils. For the named reasons, I recommend rejection of the manuscript in the present form, but recommend resubmission with a change focus (water fluxes).

Reply: We thank the reviewer for his/her comments and acknowledge the limitations of this work regarding the consideration of chemical/biological P attenuation processes, the lack of validation and the difficulty to precisely discuss temporal variability in GW P using monthly data. On reflection and following the reviewer's recommendations, we have made the changes suggested and re-focused the models on water flow and shortly discussed the implication for P attenuation processes and P transport to GW that would need to be further investigated.

Detailed comments:

Introduction:

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Line 35: Introduction: As the physical controls on P transport are derived from modelling results in this study, I miss a (short) description of available tools and why the chosen Hydrus 1D might be suitable for this purpose. There are some interesting reviews available regarding models for P transport, such as Lewis & McGechan (2002), Vadas et al. (2013), Radcliffe et al. (2015), Qi & Qi (2016), Pferdmenges et al. (2020) – just to name some of them.

Reply: Thank you for providing some good references, we have added a description of models available for water and P transport modelling and the strengths of Hydrus for this purpose on pages 4-5 lines 75-86, along with presenting recent work on P transport to GW using Hydrus.

Line 86: not clear what is meant with ‘: : with pressure assumed to be from GW P pathways.’ Please rephrase.

Reply: We have modified the sentence on page 6 lines 119-120.

Materials and methods:

Line 107: Sources for information shown in Table 1 should be named (when not gained within the presented study). Include information on classification system for soil type (FAO, USDA, etc.) and drainage class (what is ‘well drained’?)

Reply: Information on soil types and soil drainage classes come from the Agricultural Catchments Programme (carrying the present study) and geology information comes from Geological Survey Ireland. We have included this source in Table 1 on page 7. Soil types are classified according to the Irish soil classification system and have been converted into soil World Reference Base classes (without soil profile examination). We have also included these specifications. Drainage classes are assigned according to the Irish classification system based on the presence or absence of features visible in the profile. Well-drained soils show no obvious sign of impeded drainage (mottling) throughout the solum. Exception where under pasture, sparse mottling may occur in

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topsoil.

Line 111: ‘transect of multi-level piezometers’ How many piezometers per slope position were installed (one each in DS, MS, and US)?

Reply: There are 3 piezometers per well/slope position; we only used the shallower one for this study as we focused on shallow groundwater. We have modified the sentence on page 8 line 155 to remove references to other piezometers which are not part of the present study.

Line 113: ‘shallow piezometers’ implies that there is more than one per slope position. What means ‘shallow’ - the piezometer screening depth (line 114) are quite deep from the pedological point of view. Is ‘screening depth’ also sampling depth for monthly taken samples? How this screening depth of 4-7 m for DS fits to the average GW levels which are surface-near (line 129). Please clarify.

Reply: Shallow means shallow bedrock (either weathered rock at DS or bedrock at MS). We have added this clarification on page 8 line 156. The screening depth is indeed the depth where monthly samples are taken. We have added this clarification on pages 8 line 160. The piezometer measures water potential. The water enters the screen interval at 4-7 m and rises to a height equal to that of the unconfined water table (i.e. around 0.3 m).

Line 205: Figure 3: Why the ‘unequal’-sign is needed between DS and MS column? The information left to the column make clear that they are related to specific physical soil parameters. Thus, it is clear that they are not equal. Why upper slope (US) was not modelled? Why 10% dispersivity of solutes? Why not 30 % or 50 % - so, how you determined that value? As far as I understood, this has nothing do to with the solute itself and its chemical properties but is more a theoretical approach. As different elements and compounds which are translocation through soils behave quite different considering interactions within seepage water and adjacent soil, neglecting element specific processes is a rather rough approach. Or asking the other way around: Does

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it make any different if I replace in your results the phosphorus by nitrate (or any other solute). The model is a nice approach to simulated vertical water movement, but as it contains no biogeochemical information, it cannot be applied for any solute in my opinion. It may be able to simulate rough estimate for really conservative tracer, such as chloride and to a certain extent also nitrate, but phosphorus (and dissolved reactive P too) is not a conservative tracer and this is well known in literature (also line 226 ff). P is highly sorptive in soil matrix – and the model simulates matrix transport only. The role of preferential flow for P transport was mentioned in the introduction. Was this somehow included in the modelling approach or would it be possible with Hydrus 1D?

Reply: We agree that the unequal sign was not needed and we have removed it from Figure 3 on page 14. Upper slope (US) was not modelled because we focused on locations where the unsaturated zone was thinner and where soil structure was assumed to be different because of the distance from stream, topography location and land management. We chose the “average” value of longitudinal dispersivity in soils which is dependent on the scale and is on average equal to 1/10th of the soil profile depth. However, we agree that because variations have also been observed regarding flow conditions (saturated or unsaturated) or soil texture for example, this value may not be the optimal one. We did not investigate the effect of this value on model output. We agree that not considering chemical information and attenuation processes to model P transport is critical and we have modified the models to only integrate water transport. Changes have been made throughout the manuscript. However, we integrated preferential flow in the Hydrus model as we chose the bimodal/dual-porosity model of Durner which fits better to structured soils with bimodal porosity. We observed that this model fitted better to our data, compared to the unimodal model, especially for the grassland soils of this present study.

Line 211: The model was parameterized for soil depths down to 55 cm, right? So this is also the depth for which P breakthrough was modelled (results in Fig. 6 and Table

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3)? Please clarify. You should also explain how you will conclude from modelled solute transport in 55 cm to GW solute concentrations in several meters' depth. There is a gap where a lot can happen depending on deeper soil properties, geology, etc.

Reply: Yes, the models and results described are the one observed at the bottom of the soil profile. We have clarified this point on page 14 line 266, and on page 15 line 294. We agree on the difficulty to conclude on P or water transport to GW when working only on the first soil 55 cm. We added this concern for water flow in the discussion section on page 32 lines 599-602.

Line 230: Based on which information you defined the initial concentration of 10 mmol cm<sup>-3</sup>, which is around 310 g L<sup>-1</sup>. How sensitive is this value for model results?

Reply: We chose this initial concentration arbitrarily, based on previous studies on tracer transport. We did not investigate the effect of this value on models outputs and acknowledge this problem as it is surely controlling model outputs.

Line 288 Table 2: The soil samples were taken in 5-10 and 30-35 cm depth and thus are within the first two horizons you listed in Tab. 2. For the third horizon in DS you assumed same values as in the second. Did you found (in the field survey) that these horizons actually were very similar in soil properties or did you observed considerable changes with depth (what I would expect in the soils you described earlier)?

Reply: We did not conduct the soil profiles examination but we know that the 23-43 cm horizon at DS is an OB horizon rich in organic matter and minerals whereas the 43-55 cm horizon is a C horizon (parent material). Differences in saturated water content (lower in C) or saturated hydraulic conductivity (higher in C?), for example, could be expected between these 2 horizons and are not included here. We have included this point in the discussion section on page 32 lines 597-599.

Results:

Line 334: 'comparable to concentrations at MS on some occasions' – It is known from

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earlier studies that P concentration in different hydrological pathways is highly variable over the year with storm events as one of the main drivers. A monthly sampling strategy can give a rough overview on occurring concentrations but for detailed data analysis values should be handled very carefully.

Reply: We agree with this comment and agree that monthly data can hide strong temporal variability. We have slightly modified the paragraph on page 25 lines 456-460 to describe more carefully the variations observed between January-June (where P concentrations are variable at DS) and July-December (where P concentrations are always higher at DS) without referring to single peaks.

Line 344: Figure 6: Looking on the temporal development of tracer concentrations, it is not clear to me how exactly the model simulated solute transport. When I understood it right, there is an injection point (e.g. starting with the rain event R1) and then the model simulated the solute curves. During this curves are further rain events – how do they influence solute transport (with additional water input or additional water + solute input or not at all). When I see the modelled tracer concentrations, I am concerned, both with regard to value range and to curve development. There are many studies available, which worked on P transport through soils. You should consider them to carefully validate your results (just as examples: Heathwaite & Dils (2000), Haygarth et al. (1998, 2012, and more), Verheyen et al. (2015)). The peak concentrations of 0.6 mmol cm<sup>-3</sup> (or 18.6 g L<sup>-1</sup>) seem to be very low and an often observed curve development would be short and high concentration peak directly after or during every rain events (especially after dry periods). The graphs in Fig. 6 confirm my concern above, that the model approach is not suitable to simulate P transport.

Reply: Yes, we injected the tracer just before a rainfall event (R1, R2 and R3) but then additional rainfall events occurred until the end of the simulation and the complete recovery of the tracer at the bottom of the soil profile. We agree that having several rainfall events occurring in a same simulation make it difficult to clearly see the effect of a single rainfall event on solute transport. Moreover, we also agree that we did not

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strongly validate the models for P transport. For these reasons, we have modified the models to account only for water flow and only considered one single rainfall event for each simulation; we have modified the results on pages 25-28 lines 463-498 and made the corrections needed throughout the manuscript.

Line 348: Same comment as line before: The results are not in any way comparable to results of monitoring studies in literature. The occurrence of tracer (or phosphorus) in the breakthrough depth of 55 cm several days after the rain event is not plausible. Previous observations showed clear ‘first flush’ effects for P during the rain events (within hours not days!) followed by fast decrease of concentrations (also within the first hours) because of the high importance of preferential flow for P transport. The model results of your study must be validated before interpreting and discussing them!

Reply: We recognise the lack of validation of the models and we have modified the models to account only for water transport. We have made the corrections needed on pages 25-28 lines 463-498 and throughout the manuscript.

Discussion:

Line 374: I think you should be careful to discuss ‘spatial variability’, because a 1D model with no variation of hydrological pathways (e.g. differentiation in matrix and preferential flow) was applied at two single positions at a slope with no lateral connections (thus, line 378 ‘along the hillslope’ is also not valid). Conclusion on spatial variability is not possible in my opinion.

Reply: We agree and have modified sentences and the paragraph to clarify this point on page 28 lines 502-503, 507 and 515-516. However, variation in hydrological pathways has been integrated as we used the dual porosity model of Durner and not the single porosity of van Genuchten.

Line 385: ‘favourable soil properties’ – consider rephrasing, because P transport to GW is not favourable but should be prevented

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Reply: We have deleted this sentence on page 28 line 519 as it was later discussed in section 4.1. to shorten the introduction of the discussion section and avoid redundancy.

Line 417: The section discussion hydraulic properties, but completely neglects some influencing factors which might change these properties. For example, land management affects bulk density, infiltration, and runoff (vegetation cover) as well as soil chemistry (fertilization). Therefore, both water and solute transport through soils depend on many factors which are not included in your study, what should at least be part of a critical discussion of the results.

Reply: We agree that we did not discuss the effect of land management; we have included this point on pages 31-32 lines 588-595.

Line 446: An interpretation of inter-annual variability based on monthly-taken sample for only one year should be handled very carefully. Besides a high variation of seasonal variability from year to year, the within-month variability with rain events as main driver is very high, what was highlighted by some previous studies. However, the influence of anoxic conditions on P release for the downslope position is an interesting aspect.

Reply: We agree on this aspect and we have modified and shortened the section 4.2. on pages 33-35 lines 630-677 to discuss more carefully the variations observed; we discussed the difference observed between January-June where P concentrations can be low (similar to MS) and high at DS and July-December where P concentrations are always high at DS.

Line 463: 'higher soil labile inorganic P' – no data show, what is 'higher'? Can you include some values?

Reply: We have added values of soil labile inorganic P and DPS on page 34 lines 650-652 with a reference.

Line 478-479: You conclude on soil moisture effect based on one single sample in May, that's critical.

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Reply: We agree that it was critical and we have removed it from the discussion.

Line 481: You cannot compare explain GW P peaks with particle-bound P because your samples were filtrated. Nevertheless, the processes you name here are important and therefore should be also considered in models (biological controls, preferential flow, all P fractions).

Reply: Some particles are smaller than 450 nm (colloids, nanoparticles) and can contribute to facilitated-P transport. However, we have deleted this sentence as it was based on one or two observation points.

Line 489: ': : : suggesting that P reaction time : : : ' the model can simulate water flux (and P or any other solute is attached to that), thus you could conclude that water flux behaves different depending on type of rainfall event. For P, the model contains no 'reaction' routine within the soil matrix.

Reply: We agree with the fact that the model only considers water flow but it can suggest differences in reaction time with the soil matrix even though we did not integrate the chemical component in the models. We have shortly discussed this point on page 32 lines 602-605 and on page 33 lines 636-638.

Line 520: I wonder, why remediation measures are part of the discussion as no land management-dependent effects were considered in the study. In my opinion, the methodical approach is not suitable to derive management measures.

Reply: We agree that we did not conduct a specific study of the effect of land management factors on water flow. Thus, instead of discussing remediation measures in a separated section, we have integrated some implications for agricultural management while discussing the effect of soil properties (section 4.1.) or rainfall patterns/GWL (section 4.2.) on water flow.

Line 252: Why you assume long-term legacy of P when your soils are well-drained?

Reply: The time needed to reduce soil P content does not depend on soil drainage

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class but more on the clay content. This suggests that depletion of soil P will be a longer process in the upslope (higher clay content) than at DS (lower clay content).

Technical comments:

Line 85: change 'knowedge' to 'knowledge'

Reply: We have rectified this on page 6 line 118.

Line 118: what is MDL? I think this is detection limit, but all abbreviations have to be explained when firstly mentioned.

Reply: This is method detection limit; we have explained this abbreviation on page 8 line 163.

Line 131: Figure 1: Font size should be increased.

Reply: We have increased the front size of Figure 1 on page 9 to improve the readability.

Line 230: I assume the unit mmol cm<sup>-3</sup> is right here, please prove

Reply: Indeed, mmol cm<sup>-3</sup> is the right unit, not mmol cm<sup>-1</sup>, as we considered initial concentration in the liquid phase and not solid phase. However, this has been removed in the re-focus to water flux only.

Line 246: Tables S3 and S4 as well as table 2 include a lot of parameters/symbols. It would be nice for the reader to include a list of abbreviations in the manuscript.

Reply: We agree on the numerous parameters involved in the manuscript and we have included a list of abbreviations as Table 2 on page 13.

Line 322: What is ED? Clarify abbreviation

Reply: This is effective drainage which includes both infiltration and runoff in the SMD model we used. However, throughout the manuscript we assumed that effective drainage was equal to infiltration as soils are well-drained. We have modified ED

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to infiltration on page 24 line 445.

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