

***Interactive comment on “Spatial variability of herbicide mobilisation and transport at catchment scale: insights from a field experiment” by T. Doppler et al.***

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The authors observed pesticide transport behavior in an agricultural catchment over a period of approx. two months and several storm events with different precipitation characteristics. They controlled the pesticide application in the catchment, sampled the pesticide transport along the flow paths (soil, surface runoff, surface depressions, tile drains, stream) and observed hydrological processes and hydrological connectivity during the study period. They linked pesticide transport with catchment behavior, underlined the importance of field-stream connectivity, and found that pesticide mobilization was independent of the chemical properties. The study and the results are definitely of

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interest to the reader of HESS and contribute to process understanding in agriculture hydrology. The scientific approach is appropriate and the data set is very valuable and comprises a big and rare effort. Nevertheless the manuscript needs several improvements before it can be published in HESS. The necessary improvements are related to clear objectives/hypotheses, needed citations, structure, and language/style. I highly recommend that a native speaker improves the language throughout the manuscript. A serious problem in the manuscript is the structure of the different sections. E.g. methods are reoccurring in the result section while discussion is occurring in both, the results and the conclusion. A more consistent structure with a more precise story line linked to clear expressed objectives will significantly improve the readability and impact of the work. Additionally the discussion section is mainly based on work of the EAWAG group itself. I imagine that, at least for pesticide transport in infiltration excess overland flow, more work exists. I think these improvements are achievable. If they are included I see no obstacle for publication. In the following I present some suggestions for improvements, and (if necessary) outline scientific concerns.

#### Detailed comments Abstract

The abstract is clear, well-structured, and summarizes the paper well. But it is clearly too long (ca. 400 words) and should be shortened by 100-150 words.

#### Introduction

p.2359,L.21: “. . .of little importance for most pesticides.” Citation needed.

p.2359.L22/23: “large differences of pesticide losses” In what: amount? What is large (too subjective)?

p.2359,L25: add: “of a catchment can cause the”

p2360L5 “or contributin areas” (Citation needed)

p2361/2362 L25-L2: PrefFlow is linked to surface runoff, but it can also occur and various other boundary conditions. E.g. Jarvis (2007) showed that PF can start with

precipitation intensities as low as 1mm/h.

p2363/L1-L19: in the objectives you missed that you also linked the chemical properties of the herbicides to behavior. A large part of results/discussion consist of that. Also you writing the objective in the past: “..procedure allowed us. . .”. You take the results ahead of the story (in parts). I think the last part of the introduction need to be reworked. The objectives/hypothesis must be defined more clear at the end of the section. Right now you give a vague formulation of the objective (L5-7), then you present another problem (L8ff) and give there another objective (transferability), that is not even handed in the following. The objectives have to be the end of the introduction, clearly formulated, and completely addressed in the manuscript. Methods and material Please describe/mention the calculation of the runoff ratio

P2364, please add the percentage of paved area. Also are all roads paved?

P2364, L24, rewrite: “Discharge was measured at five locations” (Try to bring verbs in front of your sentences)

P2364, L26, change “Isco” to “ISCO”

P2365, L25/26: I haven’t found that you used the temperature data in this paper, so don’t describe that they were measured in the method section.

P2366/2367 L22-L2, please rewrite point two.

P2367L3: exchange position of “16 overland flow detectors” and “eleven runoff sensors”, point 1 before point 2. I can not see the locations that were equped with both sensors in figure 1. Do they have different marker?

P2367L7. “Figure 4” Here you haven’t introduced figure 2 and 3, yet. The introduction of the figures has to be in order.

P2367L7-10. You could formulate that section more positive.

P2367L12, delete the comma

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P2368L14. Change “In the 13 rain...” to “during the 13 rain...”

P2368L17: “PP”=Polypropylene? Full word. Issues with sorption in those bottles?

P2369L14: delete “which was also...” Citation is not necessary here.

P2372L21: Based on what assumption the 15 m drainage width was chosen? Some knowledge about catchment processes, data from other studies? Work about drainage e.g.: Shipitalo&Gibbs (2000, Soil Science Society Am. Jour.), Klaus&Zehe (2010, Hydrol.Proc.)

Result section: The results section is a mixture between results and discussion. Usually the results section should only include a description of the results without any discussion. In my opinion the discussion part in the result section should be moved to the discussion section. That would increase readability of the section. Of course it is also possible to join section 3 and 4 together and rename them as “results and discussion”. Further section 3.1 should also include the runoff ratio that are presented in Table 2, but remain unmentioned throughout the manuscript. Is there a correlation between runoff coefficient and processes? A link to pesticide transport in mass and timing? I guess they will also allow a better interpretation if used in the discussion.

P2373L14: “The period... was very dry”. How many mm? What was the water deficit evap. versus precip.?

P2373L18-20. Rewrite: “Four of the five largest events...”

P2378L22. “The human...” to “Human modification...”

P2378L26: Change “Very peaky” suggestions: “sharp”, “pronounced” etc.

P2374L4-L22: This paragraph has to be restructured and rewritten. There is no clear story line, it is too much of a mixture between results, a rudimental discussion, and methods (the description of mixing). Line 17ff is similar to hydrograph separation or a mixing of two end-member with a tracer. The mixing proportions can be quantified by calculations resulting in 52%-84% if rain water in surface runoff (when assuming that

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EC is conservative and that rain and groundwater are the only endmember). Might help to give a more quantitative description, although problems with nutrients at the soil surface will exist.

P2374L15; “regional” is too coarse description. Regional groundwater usually describes a way larger scale than in this study.

P2374L17 change “as measured in the” to “of”

P2374L18 “main effect”, calculations would help here

P2375L1, Repetition: it is already stated that infiltration excess overland flow is the main process

P2375: Does saturation excess overland flow has a different chemical/isotopic composition than infiltration excess overland flow so that EC can distinguish them? Please give citation for that. It could be that mixing during sat. over. Flow is limited and thus rain is also the main source of overland flow.

P2376L4: Explain the process via catch basins better, I have difficulties with the understanding of how that works.

P2376L6/7: “The connectivity analysis” better: “GIS analysis”

P2376L8-10: “This connectivity analysis. . .worst-case assumption” That is method, no point to describe it in the result section

P2376L10-18: Discussion, not results

P2377L5-25: Please shorten. Not so much method detail. Maybe present data in table?

P2378L2: “. . .decreased with time(.)”

P2378L3: “while the absolute. . .” to “The absolute. . .”, delete: “at the stations”

P2378L4-5: “depended on the proportions of the sample water. . .” Can you quantify

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that, do you have numbers? Otherwise it is discussion not results.

P2378L11-P2379L5: This section belongs to the method section. Considering the citation of Neitsch et al. (2005). Is there also a journal article that you could cite?

P2379L14: Retardation is used in the title while retention is used in the text. I think Retardation is already defined by the difference in travel times/travel distances of 2 solutes (e.g. Jury&Horton, 2004 Soil Physics).

P2379L15-P2380L6: It is very difficult to follow this section. It is again mixing of methods and results, this hides the important findings within this section. Separating them will lead to a better and clearer message. Can you give a process explanation how this retention process works in detailed? (or discuss possibilities in the discussion section, would be very interesting)

P2380L7ff. Can you give an example of the correlation factors between the different pesticides, to have a value of similarity? Also correlation coefficient for similarity between chemograph and hydrograph.

P2380L19. Is that a singular pattern for one solute and one event, or is that more frequent?

P2380L28: "observations" What observations do you mean?

P2382L2: "On the other hand", Language problem, you have to use "On the hand" before. A singular use of "... other hand" is not possible. Occurs several time in the manuscript.

P2382L5-7: What do you exactly mean with higher? Please give number. Can this higher EC derive from solutes that are dissolved from the surface?

P2382L8/9: change "where the situation ... only one experiment" to "with less complexity"

P2382L20 "dominated by road runoff". On what measurement is this conclusion

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based?

P2382L24: “the two (add: possible) flow paths”, delte: “that the water from the fields could take”. How does the concentration dynamic reflect the possible flow paths. Explain.

P2383L1-6: Repetition, not necessary

P2383L11-12. Is it surprising that infiltration excess overland flow is an important transport process. I think that previous studies just didn't observed rain with the intensities of this study.

P2383/84L25-5 I think citations are needed in this section

P2384L7: This equation does not consider preferential flow that is generated without surface runoff. I think that is ok so far, but has to be mentioned in the discussion.

P2384L12-15 Maybe a own section on management implications, or in conclusion?

P2384L15-22: Please shorten this section, especially on precip variability, maybe just state that precip variability causes variability in  $A_{inf\_ex}$

P2385L2-5. It is nice that this work confirmed previous work, but where does it go beyond? Here is the change to make an important statement that can show why this study should be cited.

P2385L6/7 delete: “-which is. . . areas-“

P2385L21 “. . .risk for herbicide transport (add: to streams) can . . .”

P2385L24 “On the other hand. . .”, missing “on the one hand”, see above

P2386L1 change “fields. . .network” to: “fields showed no surface connectivity”

P2386L4-5. “In this context. . . “ clarify sentence

P2386L7 rewrite and clarify “miss the inlet to the shortcut”

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P2386L9-11: rewrite “In addition. . . water.”

P2386L13-14. “It has already. . . van Beinum, 2009)” Why? Give explanation?

P2386L19-21 Suggestion, change: “had shown” to “showed”, “that the loss rates” to “the loss rates”, “depended to depend”, “substances” to “chemicals”.

P2386L21. Citation needed for Previous observations, for both the loss rates and the sorption effect

P2386L24-25. What about the strong effect of the macropore coating on sorption?

P2386L25 Delete “interestingly”

P2386L26: here you mentioned what you expected in the work. It would be great using this in the introduction to formulate objectives/hypotheses

P2386L27: When you write about mobilization you are referring to the timing of the mobilization? Because to overall recovery might me reduce compared to non reactive chemicals. Please clarify what you mean with “mobilization” in the manuscript. Is there an effect of sorption on pesticide recovery? High sorptivity leads to lower recovery during events with same chemograph dynamics?

P2387L6: Retardation, I think you mean your defined retention? Retardation is measured by the travel time of the center of mass (Jury&Horton, 2004)

P2387/88L15-L2: In my opinion that does is a little bit contradicting the finding that herbicides applied at the same field have the same temporal dynamics in concentration. If some solutes will enter the aggregate and some stay at the surface of the aggregate the temporal dynamics of pesticides applied in the same field should be different (different time to get in the mobile phase. But it is not.

Conclusion: Overall the conclusion need to be clearly shortened. You have to link it directly to the objectives and hypothesis and present the novelty of this study. In this manuscript the conclusion is an extension of the discussion. P2388L14-19 p2389L1-5

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are discussion and should not be a part of the conclusions.

P2388L20-21. Is that really a new finding? No other study found pesticide transport in inf.ex. surface runoff? I doubt that this was unknown that this is new, since we know that surface runoff will transport pesticides, and we also know that infil.excess surface runoff can occur in humid (agricultural) catchments.

References: After the year of the publications there are often a bunch of numbers. That seems to be strange to me. E.g. P2390L24 “J. Hydrol., 365, 23-36, 2009. 2362, 2386”; P2390L32/33, etc.

Figures: The font size might be too small in the final version of the paper, in both the maps and the figures. Please have a look at that in the final version before publication Using solid lines for the pesticide concentration (Fig.8, 9, 10) is not accurate, since there is no information between the sampling points. Using data points without lines is more appropriate to the sampling conditions. With the (low) sampling frequency your sampled peak concentrations cannot be assumed to be the real concentration peak.

Figures 1, 2, 4, 9 add coordinates. Also in the text figure 4 follows figure 1. Need to be corrected

Figure 3. y-captions, Unit ( $L s^{-1}$ ), I think HESS uses small l symbol for liters (but check again), if so please change, letters might be too small in the publication

Figure 6 and 7, add unit to  $K_d$ , also needed throughout the manuscript

Figure 8. l instead of L

Figure10. Maybe avoid overlapping of the discharge and precipitation peak. Use l for liter instead of L.

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