Reviewer comments are in italics, author reply is in plain text

Reviewer 2

The authors have provided a substantially revised manuscript for review. The revised manuscript, after removal of extraneous narrative and figures, is now somewhat more targeted on tractable research questions and the presentation of a nice high-frequency data set. With the revisions, the authors have made good progress towards addressing the multiple reviewers who were fairly consistent in their critiques of the manuscript. The narrative now more narrowly focuses on how and why in-stream flow and solute patterns vary in three different settings under very different wetness conditions. Despite these improvements, I still feel that major revisions are needed to continue to refine the manuscript for publication.

We are glad the reviewer found the manuscript to be substantially improved.

In my previous assessment, I had three primary areas of concern. The inclusion of sensor QA information has addressed my first concern and has given me the opportunity to more comprehensively assess the results and discussions. My other two other concerns (i.e., unsubstantiated inference based upon hysteresis analysis and manuscript organization) were partially addressed, but I have lingering concerns regarding both. In addition, I highly encourage further revisions to address writing style, vagueness, redundancies, and transferability of findings. I have made many comments and suggest many edits in the accompanying marked manuscript. Beyond those detailed edits and comments, I offer these general suggestions:

1) Hysteresis analysis: I still struggle with this portion of the manuscript. It is long, rambling, and mired in details. Much of this analysis could be removed without diminishing the study. The results and discussions on this topic need to be properly separated into the appropriate sections of the manuscript. For example, none of the figures are presented in the results.

The presentation of the hysteresis is now in the Results section as recommended.

Moreover, I feel that the hysteresis analysis is overemphasized and the interpretation of hysteresis is excruciatingly long. It is still my contention that hysteresis analysis, while moderately useful for general assessments of mixing of waters in streams from various potential end-members, is not a robust approach for the assessment of specific processes without explicit measures that have not been undertaken within this research (or at least presented in this draft of the manuscript). The authors focus on minutiae of complex loops, which have limited intepretability within the context of the overarching goals of the research. Additionally, the hysteresis index is hardly used and could be removed. I think that the nearly four pages of discussion/results on hysteresis loops could be reduced to several discussion points and be presented in one or two short, well-crafted paragraphs about source or transport limitations for the various analytes.

We thank the reviewer for this comment and have now substantially reduced the hysteresis discussion by around 1000 words. We have focused the hysteresis discussion on source/transport limitation and removed any repetition with the discussion section 4.1 which discusses more general nutrient dynamics throughout the hydro-year. We have kept the Hysteresis Index in the manuscript, which is now referred to in the hysteresis description in the results, section 3.5. We believe this information helps the reader to see particularly how hysteresis changed between events at the same site and so should remain in the paper.

I did find inclusion of some nitrate concentration data from soils in the Blackwater catchment to be

substantive and informative. And, their discussion of groundwater influence on the chemistry of Wylye basin is nicely supported by the hysteresis analysis. Nonetheless, the entire discussion of hysteresis could be vastly reduced to several key, substantiated findings that directly address their research goals, while avoiding rambling supposition and tangential topics.

We are pleased that the reviewer found the inclusion of this information informative and supportive of the hysteresis interpretation.

2) Organization: While the authors have done a fine job of moving most information to the appropriate sections, the presentation of hysteresis results in the discussion is one area where reorganization is necessary.

The presentation of the hysteresis is now in the Results section as recommended.

3) Writing style and grammar: The authors need to:
** Use the active voice whenever possible. Use direct wording, which along with the use of active voice, will more clearly communicate ideas.*
** Scrutinize the entire document for verb tense. Past tense must be used whenever presenting and discussing results.*
** Avoid subjective or vague statements and provide specifics whenever possible. See the marked manuscript for many examples. As one specific example, it would be more helpful to have "pressures" (p. 5, line 18) specified. In practical terms, it matters what the pressures are, not that they exist. Or, instead of writing that the data provide insight, describe what that insight is.*
** Assure that singular and plural forms are appropriately used such that there is always agreement between subjects and verbs or direct objects or correspondence to the item described.*
** Rewrite runon sentences (several examples noted in the marked manuscript).*
** Remove personification.*
** Remove dangling modifiers.*
** Use appropriate verb tenses, the active voice, and more direct language to clearly distinguish between findings and supposition. That is, make it absolutely clear when findings are supported by presented data, that inference is being made from data, or that inference is supported by findings of others.*

We would like to thank the reviewer for the time they spent making recommendations to the manuscript text in such great detail. We have tried to take these recommendations on board, using the active voice wherever possible, which has been an effective way of cutting the word count down and making the manuscript more succinct. Examples where we have changed the wording include:

“The flashy nature of Newby Beck is attributed to the operation of rapid runoff response pathways (surface runoff and preferential flow in drains) and the lower baseflow index”, originally p. 14 lines 16-18

Which has been changed to:

The operation of rapid runoff response pathways (surface runoff and preferential flow in drains) and the lower baseflow index (Table 1) explain the flashy nature of Newby Beck.”, page 15 lines 3-4.

“The frequent and rapid response in TP concentration at the sampling point is attributed to rapid surface runoff generation exacerbated by extensive soil compaction from livestock trampling and silage production fields and transport of particulate P with eroded soil, with farm machinery tramlines also promoting high connectivity between sources and receiving waters.” Page 14 lines 28-

32.

Which has been changed to:

“Rapid surface runoff generation exacerbated by extensive soil compaction from livestock trampling and silage production is thought to be responsible for soil erosion, resulting in the rapid response in TP concentration to increasing flow at the sampling point. Tramlines created by farm machinery also promote high connectivity between sources and receiving waters by channelling flow downslope” page 15 lines 14-18

4) Redundancies and irrelevant information: The manuscript is long. Some topics reappear numerous times and some topics are not well integrated into the manuscript or are simply irrelevant. I have made some suggestions for deletions. The authors need to consider what information is really most relevant to the telling of a concise story and remove topics that are repeated or tangential.

Again, we thank the reviewer for their extensive edit recommendations and have removed much extraneous information, for example removing all of the suggested sentences from section 4.4. The manuscript text has been reduced from ~9400 words to ~8300 words which we think is a substantial improvement and it is now a more concise script.

5) This manuscript, with a discussion focused on the DTCs and little comparison to other catchment studies, comes across as a case study with little transferability to other locations or situations. I still find the introduction of the broader DTC study to be excessive and something that could be trimmed. While an important case study, I urge the authors to find ways to explicitly link their points about management to other locations and situations. If they generalize portions of their discussions and conclusions, the work could contribute to a broader science. For example, they devote considerable text to setting up a hydrogeologic framework for the study design, but have not really capitalized upon how that framework may offer particular insights to other locations. I do point out one example in the marked manuscript wherein theirs results are portrayed in a broader context – I highly urge the authors to follow that example when revising. Overall, I encourage the authors to focus on their specific findings that are of general relevance to other researchers, resource managers, and geographic areas. Otherwise, the work is likely to resound with only a small group having an interest in the particular DTC catchments.

We thank the reviewer for this comment. We have now added in much more information about the transferability of our results, particularly page 15 line 26 – page 16 line N regarding measures required in permeable catchments to reduce nitrogen losses to groundwater, and page 19 line 26 – page 20 line 6 regarding the significance of acute, storm-dependent nutrient transfers which were identified in the three catchments here but likely to be operating in catchments throughout Europe, posing a challenge for meeting WFD targets. We think that the results from this manuscript are now put into a broader framework making the information much more applicable to a wider scientific audience.

6) My previous comment on a more specific title still stands. It would be more informative to potential readers to know about the focus on N and P. The manuscript only describes 2 of the well over 20 nutrient elements.

We have now changed the title to reflect the focus on N and P.

Reviewer 3

High-frequency monitoring of catchment nutrient response in the Demonstration Test Catchments to the end of the 2011-12 drought in England.
By Outram FN, Lloyd C, Jonczyk J, Benskin CMc.WH, Grant F, Perks MT, Deasy C, Burke SP, Collins AL, Freer J, Haygarth PM, Hiscock KM, Johnes PJ and Lovett AA.
General points
The revised version of the paper is much improved. The changes made are in line with the reviewers' comments. The paper now has a much better balance in terms of content. The focus of the work remains the analysis of the 25-29 April 2012 storm event rather than an analysis of the responses of all storms on record (Referee#1 General Comment, Referee #3 General Comment 4, Referee #4 General Comment). The rationale for the use of the hysteresis analysis is much clearer, there is now a useful description of the dynamics during the 25-29 April storm event, and the work has been placed into the context of the broader literature much better. The addition of the detailed description of the changes in flow and streamwater nitrate and TP concentrations is very useful (sections 3.1 and 3.2, Figure 2).

We thank the reviewers for this comment and are glad they found the revised manuscript to be clearer and better contextualised in the literature.

Interestingly, the 25-29 April 2012 storm event does not look exceptional when compared with other events with rapid transitions (in API) from dry to wet conditions evident in the Blackwater Tributary in March 2012, and in the Newby Beck Tributary in December 2011 (Figure 2). The API transition in the Wylye does look exceptional in the period October 2011 to September 2012. Whilst there may have been exceptional meteorological conditions in the first five months of 2012 it is notable to see the response in the Blackwater Tributary and Newby Beck Tributary catchments, which does not seem very different from other storm event responses through the October 2011-September 2012 period. That is a really interesting finding, though it is not really commented upon in this way.

The effect of the storm isn't just observed in the API but the nutrient responses during a transition from drought conditions to a wet summer in all three catchments. The drought commenced in April 2011 and we therefore propose that during this period the accumulation of nutrients occurred in the catchments (page 19 line 18-27).

There was only really one large hydrological event in the Blackwater before the one studied in depth here which was in March, but as stated on page 9 line 5-6, the discharge receded rapidly until the larger hydrological event examined in this manuscript at the end of April marked a transition into a wet summer (page 9 line 29-31 and page 19 line 20-21). The point is made that despite the large event in March, the nitrate response to the late April event showed no signs of exhaustion, and doesn't until late summer (page 16 line 29 – page 17 line 2).

With regards the Newby Beck, the API transition is not that dramatic, which we state is due to the rainfall total in this storm (page 10, line 9-11). However, the TP response is disproportionate to the size of the flow generated – peak TP in the late April storm was higher than in the next large storm which has a higher maximum API. We attribute this to lack of source exhaustion from previous storms and that the mobility of particulate P increases as soil conditions become drier (page 18, line 19-24 and page19 line21-26).

Because of these aspects which were already included in the discussion we feel that we have in fact already explored this idea sufficiently.

As requested by all referees, the authors now include a section on data quality assurance (section 2.3). Based on the evidence of Table 2, the quality of the streamwater data in general look good though streamwater nitrate data for Newby Beck appear questionable. The correlation co-efficient is 0.45 suggesting some dispersion between the sub-daily and grab sample measurements of the streamwater nitrate concentrations. The standard deviation of the residuals is 0.60 mg N l-1 against a
mean concentration of approximately 2 mg N l-1. How large is the range of the residuals? It would be useful to see a time-series or one-to-plot of the sub-daily and grab samples concentrations to see how the errors vary with flow and concentration (for all sites, for all determinands).

In the original submission, the Eden DTC (Newby Beck) nitrate probe data had a poor relationship with monthly grab samples and storm samples collected with automatic samplers which were analysed in the laboratory. The nature of this relationship has now been fully investigated, resulting in the removal of some laboratory analysed data from the validation of the nitrate probe.

The removal of some laboratory analysed data was based on the relationship of nitrate to total nitrogen concentrations and the probable transformation of nitrate into other nitrogen species during transportation. All grab samples are collected, transported and ready for analysis within 24hrs and so it is assumed that these samples will have limited deterioration. By contrast, the storm samples are collected by automatic water samplers which are programmed to take a 1L sample every hour for 24hrs which means that there is at least a 24hr delay from when the first sample is taken until it is collected and transported for analysis at the laboratory. At least 98% of total nitrogen in the grab samples throughout the hydro-year was in the form of nitrate. Data was therefore, discarded from the storm sample dataset when nitrate was less than 98% of total nitrogen.

In total, 50 samples in 2011-12 hydrological year (monthly grab and storms samples) were used to determine the relationship of the nitrate sonde with those analysed in the laboratory to give an improved correlation value of 0.885. The manuscript has been corrected accordingly.

Grab samples have only been taken at monthly intervals for the Blackwater and it is unclear if high flow events have been captured to verify the sub-daily data across the flow range; this is important given the focus of the paper is storm event hydrochemcial analysis.

In fact, it is stated in the manuscript that grab samples were taken weekly in the Blackwater, not monthly (see page 7 line 12), which gave a good range of flow conditions for performing the correlation analysis between grab sample and in situ nitrate and phosphate data.

There is also an issue in that the authors refer to the concentration-duration plots as being for one hydrological year for the Wylye and Newby Beck (see specific comments) but, from Figure 2, it appears that there only six months of data for the Wylye for streamwater nitrate and TP concentrations, and perhaps five-six months of streamwater nitrate data for Newby Beck.

In the manuscript we had already made reference to the fact that the concentration-duration plots were made with a limited data set in the Wylye to inform the reader, see page 10 line 16-18. The reviewer is mistaken regarding the nitrate data in the Newby Beck. There is almost a complete dataset for the hydro-year (see Fig 2), the only section missing is around the time of the late April storm as mentioned in the manuscript.

My recommendation is that this paper can not be accepted at this time. This is based on two main reasons. Firstly, further verification of the sub-hourly streamwater nitrate data needs to be done for the Newby Beck nitrate concentrations to provide robustness if these data are to be included.

This has now been corrected for, see comment above.

Secondly, the paper focuses on an analysis of the storm event dynamics, but the hysteresis curves for only one storm are considered and therefore the question still remains of how general is this behaviour (as in the original review)? The authors have the data, or are gathering the data, to do this analysis with much greater depth. There is much in the literature already which is cited in the paper about the range of catchment responses during storms, what is needed to push the science on are detailed analyses of what is happening. The authors have the opportunity to do this, but this paper is not there yet with the sufficient depth needed to make use of this opportunity.

As we said in our reply to the first set of comments, the inclusion of the time-series data from the hydrological year of nitrate, TP, discharge, rainfall and API provides the reader with a good deal of information about the general hydrological regimes of the three catchments and how the transition storms at the end of April fit in to the wider context, which is discussed in section 3.1 and 4.1. We felt that it was unnecessary to include hysteresis analysis for other periods as we were using the hysteresis as a tool to explore the different catchment responses to this particular storm period. An in depth analysis of the range of catchment responses would indeed be interesting but would be the topic of a different paper. The main thrust of this paper is that these unusual meteorological conditions allowed for an analysis of the three catchment responses acting under relatively similar conditions, from drought to flood risk. We chose to focus on the transition from dry to wet by examining the variety of responses to the late April storm, which revealed large peaks of N and/or P in all three catchments, attributed to nutrient build up in soil reserves during the drought period. Such acute, storm-dependant transfers of N and P indicate the scale of the challenge to environmental managers in tackling nitrogen and phosphorus pollution in rural catchments under variable antecedent conditions. This is discussed in section 4.3, particularly page 19 line 19 – page 20 line 13.

The title of the paper suggests an analysis of hydrochemical conditions leading up to the end of the 2011-12 drought but this is not really done given the lack of data in the Wylye for the period October 2011 to February 2012.

It is unfortunate that we only have the N and P data for the Wylye form February 2012 onwards but as can be seen from Figure 2 the complete record of rainfall, discharge and API show that the winter was very dry; it is clear that we have captured the wetting up of the catchment in response to this late April event and associated N and P response.

If the intention of this paper is to provide an introduction to the Demonstration Test Catchments research programme then this is done already in Owen et al. (2012).

This is incorrect. The Owen et al paper specifically refers to the DTC setup in the Eden catchment only. The manuscript presented here is the only paper comparing the results from the three DTC catchments.

Specific points

P2. Line 4. Can you identify 'trends' in one year of data? I would suggest changing this to 'patterns'.

This has been done

P2. Abstract. The abstract does not list the three sites nor the key results.

This has now been done (Page 2 line 27 – page 3 line 2 and page 3 line 7-11)

P4 Lines 1-3. Will the contributions from all point sources, such as diary shed retention ponds or septic tanks, arrive together on the falling limb? I would have expected a distribution in travel times reflecting the spread of these sources around a catchment, and this could complicate the hysteresis patterns.

This has been removed in the editing process.

P5. Lines 17 and 30. Which nutrient and sediment pressures do the Wylye and Blackwater experience?

This has been changed to read "Diffuse sources of N and P dominate stream loads in the headwaters of the Wylye (Yates and Johnes, 2013) compromising its ecological status under the WFD" (page 5 lne 13-14) and "The ecological status of the Blackwater tributary is compromised by high rural N, P and sediment inputs" (page 5 line 25 -26) respectively, to show that the pollutants have led to bad ecological status in both sub-catchments.

P6. Line 12. What is meant by 'harsher' – 'wetter, colder'?

This has been changed to wetter and colder (page 6 line 6-7).

P9. Line 18. Please can you clarify 'early part of the winter' – 01 October 2011 to 01 December 2011?

This has been done (page 9 line 8-9).

P10. Line 13. The antecedent conditions for the 25-29 April event were not as dry as those in December 2011.

This paper is about the transition from a drought period to a wet period. We have not said that we captured storms at the time of lowest API but that we captured the transition from a dry winter to a wet summer.

P10. Lines 19-21. Only six months of data have been measured on Wylye and for streamwater nitrate on the Blackwater Tributary.

A few sentences later, this is mentioned to inform the reader of this (page 10 line 16 – 18).

P11. Lines 5-6. Only six months of data have been measured on Wylye.

This has been changed to "for the period monitored in the year" (page 10 line 31).

P11. Lines 16-19. Please also add a reference to Figure 2 and/or Figure 4 as these illustrate the time series referred too.

These Figures are discussed in subsequent sections.

P11. Line 25-29. Looking at Figure 2, is this event really exceptional? Perhaps it is in the Wylye, but it doesn't look that way in the Newby Beck Tributary (e.g. early December 2011) and the Blackwater Tributary (March 2012, when the flow appears to rise to approximately 1.5 m3s-1).

See comments above.

P12. Line 29. *Why did you decide not to estimate uncertainty in the loads?*

Because that is an entire topic worthy of a great deal of discussion, which would make this paper over-long. The loads were not the central topic of discussion.

P13. Line 15. Only six months of data have been measured on Wylye.

The word 'complete' has been removed, the data available in the Wylye were from the same water year.

P13. Line 19. Could the peak in the streamwater TP concentrations be due to instream remobilisation of phosphorus, or nearby point sources?

Our interpretation is backed up by a paper by Yates and Johnes (2013) which looked in detail at processes in the Wylye.

P14. Line 24-28. Has mineralisation taken place in the soils following re-wetting of the soils? Which figure are you referring to here?

The re-wetting of soils and mineralisation is discussed in section 4.3 (page 19 line 5-7) in which we discuss general mechanisms for nitrate build up in soil in dry periods. The figure number has been added.

P14. Line 27-33. Do you evidence to support these statements?

We use the phrase "thought to be responsible.." (page 15 line 15) to show that this is conjecture based on our conceptual knowledge of the catchment.

P15. Line 6. Only six months of data have been measured on Wylye.

This sentence was removed in the editing process.

P18. Line 5. 'Top soil remobilised bed-sediment' – I am confused by this description, please could you clarify what you mean?

A comma has been added: "topsoil (Cooper et al., 2013), remobilised bed-sediment (Ballantine et al., 2009)" (page 17 line 28)

P20. Lines 1-2. Whilst the statement is true, the effect of these unusual hydro-meterological conditions doesn't seem to be manifest in all three catchments according to Figure 2.

See comment above.

P21. Line 25. Only six months of data have been measured on Wylye and for streamwater nitrate on the Blackwater Tributary.

This sentence is referring to the general benefit of collecting high-frequency monitoring data and does not relate directly to the data presented here (page 20 line 17 – 18). Incidentally, the statement about there only being 6 months of nitrate data for the Blackwater is incorrect.

P23. Lines 14-23. These does not seem relevant. I would suggest that it is deleted.

These sentences were removed in the editing process.