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Hydrology and Earth System Sciences Discussions

Interactive comment on "The chemical signature of a livestock farming catchment: synthesis from a high-frequency multi-element long term monitoring" *by* A. H. Aubert et al.

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General comments

Firstly, we thank the two referees for their interest in the paper and their encouragement towards publication, and above all for the constructive comments they made to improve the paper. We agree to most of their comments and we propose to modify the text accordingly. As most of the two referee comments are about the same parts of the paper, we propose a common answer to their two interactive comments. When a paragraph raised several comments on which we agreed, we propose below a corrected version of the full paragraph.

The answer to the general comment on the functioning of the groundwater table, and particularly the wetland, made by Referee2, was included below in the section 3.2.1. (comments on the annual pattern part §P9727, l.16-20). We decided to include additional data to be more accurate. In the previous version of the paper, we focused on deep down-slope groundwater. Data collected during one hydrological year in the wetland compartment have been added. Therefore, the paper now presents three groundwater compartments: deep upland domain, deep down-slope domain and superficial wetland domain. The comment relative to sulphate reduction is addressed in the section 3.1 (comment on static signature part, §P9724, l.3-22.

We agree with all the comments on English language and typographic errors and we corrected the manuscript following the reviewers suggestions (comments number 3, 4, 5, 5bis, 6, 8bis, 13, 14, 16, 18, 29, 30, 33, 34, 38, 48, 49, 60, 61, 66, 69, 73, 74, 77, 78, 81 and 82 from referee1 and p9716, l.8; p9716, l.10; p9717, l.6; p9718, l.3; p9718, l.6; p9718, l.14; p9718, l.25; p9720, l.11; p9721, l.21; p9722, l.8; p9722, l.25; p9723, l.23; p9723, l.25; p9727, l.1; p9728, l.7; p9728, l.21; p9729, l.10 and p9729, l.20 from referee2).

In all the text, the word "element" is replaced by the word "solute" (comment 2 r1). When the word "pattern" was wrongly used for explanations of observed patterns, it is changed into "conceptualisation" (56, 64 r1). We simplify "concentration levels" by "concentrations" P9721, l.21. Rather than using "process-controlled" that is too general, we use "supply-limited". Considering the comment on p9721, l.14-15, we realised that what we called in the manuscript "storm events" can be more accurately called "floods".

Title (1,2 r1+ p9716, l.15-16 r2)
We maintain that Kervidy-Naizin is an intensive livestock farming catchment. It is not

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so visible on the land use data given because pig and poultry production is off-land. Therefore, it is part of the 30% of the total land surface not defined in the manuscript. These 30% of the land include minor agricultural crops, woods and hedges and all infrastructures (roads, pig and hen houses, other buildings). We add the number of livestock equivalent units (about 13 LSU ha^{-1}) to show how intensive the livestock breeding activity is. We also specify that the cereals and maize are mostly produced for animal feeding within the catchment.

We propose an improved title as follow:
Solute transport dynamics in small, shallow groundwater-dominated agricultural catchments: insights from a high-frequency, multi-solute 10-year-long monitoring.

Comments on the structure
The paragraphs p9721, l.21-25 and p9724, l.25-27 will go to Method section and p9721, l.24-p9722, l.4 will go to conclusion. Title of paragraph 2.3. will be changed to "Data treatment".

Comments on the abstract
P9716, l.12-14. Replace with: Nitrate and chloride exhibit rather smooth variations.

The abstract was re-written as follow:
High frequency, long-term and multi-solute measurements are required to assess the impact of human pressures on water quality due to (i) the high temporal and spatial variability of climate and human activity and (ii) the fact that chemical solutes combine short and long-term dynamics. Such data series are scarce. This study, based on an original and unpublished time-series from the Kervidy-Naizin headwater catchment, aims to determine solute transfer processes and dynamics that characterise this strongly human-impacted catchment.
The Kervidy-Naizin catchment is a temperate, intensive agricultural catchment,

hydrologically controlled by shallow groundwater. Over 10 years, five solutes (nitrate, sulphate, chloride, and dissolved organic and inorganic carbon) were monitored daily at the catchment outlet and roughly every four months in the shallow groundwater. The concentrations of all five solutes showed seasonal variations but the patterns of the variations differed from one solute to another. Nitrate and chloride exhibit rather smooth variations. In contrast, sulphate, organic and inorganic carbon are dominated by flood flushes. The observed nitrate and chloride patterns are typical of an intensive agricultural catchment hydrologically controlled by shallow groundwater. There, nitrate and chloride mainly from organic fertilisers accumulated over several years in the shallow groundwater. They are seasonally exported when upland groundwater connects with the stream during the wet season. Conversely, sulphate, organic and inorganic carbon patterns are not specific to agricultural catchments. These solutes do not come from fertilisers accumulated in soil or shallow groundwater; instead, they are biogeochemically produced in the catchment. These results allowed development of a generic classification system based on the specific temporal patterns and source location of each solute. It also considers the stocking period and the dominant process that limits transport to the stream, i.e. the connectivity of the stocking compartment. This mechanistic classification can be applied to any chemical solute to help assess its origin, storage or production location and transfer mechanism in similar catchments.

Comments on the introduction (1.)

P9717, l.7-20. As Referee 2 found it useful, this part is kept.

P9717, l.26-27. We deleted the sentence, as it was not really necessary in this context and because clear cutting is only one of the various modalities of local forest policies (and therefore, it is implicitly mentioned in the previous sentence).

P9718, l.5. Reynolds's statement was wrongly reformulated, mixing vary and variability. The original statement is as such "long-term water quality data show considerable variability over a variety of time scales" and that various processes explain this variability.

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Replace with: Long-term water quality data exhibit high variability patterns depending on the timescales at which they are analysed (Reynolds, 1997).

P9718, l.6. Replace with: The conclusions that can be drawn about the impact of human activities on water quality and solute transfer mechanisms are influenced by time-series length and sampling frequency as they create a temporal filter that emphasizes some timescales and processes and hides others (. . .).

P9718, l.13. Replace with: (Gascuel-Odoux et al., 2010; Howden et al., 2010; Hrachowitz et al., 2012; Monteith et al., 2000)

P9718, l.14. Replace with: intermediate scale studies focus on seasonal variations (Dawson et al., 2008, 2011; Martin et al., 2004; Mulholland and Hill, 1997). This intermediate timescale (. . .).

Comments on the study site part (2.1.)

P9719, l.8-12. Shorten to: As of January 2012, 21 journal articles based on the catchment observations are referenced in the Web of Knowledge.

P9719, l.15-18. Equivalences in the WRB 2006 are corrected: Soils on the hill-slopes are well-drained and consist of Cambisols Dystric and Luvisols. "Soils on the hill-slopes are well-drained and consist of Cambisols Dystric and Luvisols. Down-slope soils, in the wetland domain, consist of Epistagnic Luvisol and Epistagnic Abvuvisols, in which Mn and Fe-oxyhydroxides are depleted due to seasonal waterlogging by the rising groundwater and reduction of these oxides by heterotrophic bacteria."

P9719, l.21. Replace with: The climate is temperate with oceanic influence.

P9720, l.4-5. Replace with: N efficiency, defined as N output (sold animal and vegetal products, exported manure. . .) divided by N input (mineral fertilizers, bought animal feed, bought animals, N fixation, imported manure), varied greatly (19 - 79 %) depending on the farm types and on the farms within each farm type.

Comments on the data collection part (2.2.)
P9720, l.16-18. Replace with: Stream-water samples used to determine solutes concentration are collected manually at approximately the same hour (5 p.m.), without specific sampling during floods. These instantaneous grab samples are immediately filtered (0.2 μm) on site and stored in the dark at 4°C in propylene bottles. The bottles are top-full. Analyses are performed within a maximum of a fortnight.

P9720, l.27-28 and P9721, l.4. To account for the added data, replace the whole paragraph on groundwater data with: “Shallow groundwater (GW) data were collected from two four-meters-deep piezometers set along a topographic transect (Gueriniec transect; see Fig. 1) and from eight shallow (20 to 40 cm) zero-tension, lysimeters set in the wetland domain (Mercy wetland; see Fig. 1) were also used in this study. Water-table depth on this catchment is measured every 15 minutes by pressure probes (Orpheus OTT) since 2000. Water-table chemistry is measured at different frequency depending on water-table depth. In the four-meters-deep piezometers, measurements are performed roughly every three months, providing 24 analyses for each piezometer since 2000. This is justified by the relative chemical inertness of the GW at that depth. In the 20- to 40-cm-deep lysimeters, a much higher sampling frequency was used due to water-table movements and changes in redox conditions. Several high-frequency sampling campaigns were performed during hydrological years 2000-2001, 2001-2002, 2007-2008 and 2010-2011, providing ca. 100 analyses for each lysimeter since 2000. Throughout the rest of this paper, the three GW compartments will be referred to as upland GW, deep down-slope GW and wetland GW.”

Comments on the statistical analyses part (2.3.)
P9721, l.6. Title changed to “data treatment”. The §P9721, l.21-23 will be inserted at the beginning of this part, immediately followed by the §P9724, l.25-27.

P9721, l.8-9. Sentence deleted. We meant that cutting the dataset by month, lead to about 300 data point in each month, which strengthen our monthly averages. But, it seems unclear and is not very useful, so it was deleted.

P9721, l.16. Kervidy-Naizin is quickly reactive to rainfall events when it is saturated. Therefore, even 1 mm of rainfall can lead to significant increase in discharge. Replace with: As no specific sampling strategy was implemented to account for floods, we used the following decision rule (adapted from Morel (2009) and Molenat (2008)) to distinguish base-flow from flood-flow periods (...).

Comments on static signature part (3.1.)

P9722, l.22-25. Replace with: (...)(Kirchner et al., 2000) and about 13 mg l^{-1} in Tillingbourne (Hill et al., 2002). High chloride concentrations may come from the combined effect of dry deposition of sea salt aerosol and concentration increases due to evapotranspiration. However, even if Kervidy-Naizin is relatively close to the coast line (50 km to the South, 60 km to the North), stream chloride concentrations seem too high to originate only from the rain. Similarly high chloride concentrations were obtained in a catchment with the same soil, climate and land-use context. They were ascribed to an agricultural origin (...)

P9723, l.3. From the 43 rainfalls analysed between February 2000 and April 2001, the precipitation weighted mean value of chloride concentration is 4.75 mg l^{-1}.

P9723, l.23. Replace with: as that in Wytham (England) and Scottish catchments (...).

P9723, l.26-27. Replace with: Contrarily to nitrate and chloride, concentrations in sulphate, DOC and DIC are not so different from those found in natural catchments.

P9724, l.1-22. Replace with: The concentrations of the four water compartments (stream, upland GW, deep down-slope GW and wetland GW) define the static signature. This static signature enabled to develop two points: one on solute origin and one on spatial source patterns. First, we distinguished two origins of solutes, anthropogenic and natural (Ouyang et al., 2006; Vega et al., 1998). In the first group, we set nitrate and chloride whose high concentrations clearly originate from excessive crop fertilisation. These two solutes, reacting little with the soil mineral phase, were leached and more or less stored in the GW. Sulphate, DOC and DIC constitute the second group of solutes. They have low concentrations in the upland GW and the most concentrated compartment is located down-slope. Solutes from this group depend for a significant part on biogeochemical production processes taking place down-slope (in the wetland for DOC (Morel et al., 2009) and in the deep down-slope GW for sulphate (Pauwels et al., 2010). Second, from this static signature (Tables 1 and 2, Fig. 2), we identified three spatial sources. Mean stream nitrate concentrations were 20% lower than those in upland GW but still much higher than in wetland and down-slope GW. Mean stream chloride concentrations were about the same as those in upland and wetland GW and higher than in the deep down-slope GW. The dominant spatial source of nitrate and chloride is the upland GW. Nitrate spatial distribution showed that the hill-slopes were not directly connected to the wetland but rather to the stream. The low wetland nitrate concentrations were also due to the absence of fertilisation and to denitrification (Molenat et al., 2008). Mean sulphate concentrations were similar in the stream and upland and wetland GW, but high in deep down-slope GW. In the wetland GW, sulphate reduction to sulphur did not occur as the reduction chain stops with iron reduction. In deep down-slope GW, sulphate is produced by the pyrite layer. DIC stream concentration would be in the same case as sulphate. For sulphate and DIC, a deep down-slope GW contribution is not necessary to explain the level and variability in stream-water concentrations: wetland GW concentrations and variability are sufficient. Last, mean stream DOC concentration had high variability and was much higher than in both upland GW and deep down-slope GW, but not as high as in wetland GW. It indicated that DOC originated from the wetland domain. The static signature led to solute differentiations based on origins and dominant spatial sources.

Comments on the annual pattern part (3.2.1.)

P9725, l.5. Replace with: All rainfall events led to a dilution of nitrate concentrations (Fig. 2).

P9725, l.9-22. Replace with: In other words, nitrate concentration remains high in the stream as long as the latter was hydrologically connected to the upland GW which behaves as an infinite nitrate reservoir (Molenat et al., 2008). Then, from June to August, nitrate stream concentration decreases slightly as (i) the upland groundwater progressively contributes less because the catchment is drying and thus reduced nitrate transport capacity is available (Molenat et al., 2008) and as (ii) other processes, such as denitrification and plant uptake also contribute (Betton et al., 1991). The combination of these processes explains the low concentrations observed in October. Mulholland and Hill (1997) also observed a sharp decrease of stream nitrate concentration in autumn in their forested catchment: leaf input to the stream increased, as well as photosynthetically active radiation, leading to more active in-stream decomposition. The Kervidy-Naizin stream is bordered by riparian hedges, which could have the same effect as forest trees on stream nitrate concentration. In autumn, there is a transition period when the upland water-table progressively rises (Molenat et al., 2008), thus increasing nitrate transport capacity.

P9725, l.23-P9726 l.2. Replace with: We propose a new seasonal conceptualisation for chloride. The stream chloride pattern was characterised by a flush of high concentrations at the beginning of the hydrological year (Fig. 3b), as the catchment is wetting up fast, thus providing increased transport capacity. Concentrations then decreased and remained relatively stable until the end of the hydrological year when a slight increase occurred. A seasonal change concerning the influence of floods on concentrations (called hereafter seasonal flood effect) was noticed (Fig. 2), in which only floods occurring at the beginning of the hydrological year led to an increase in concentration, whereas later floods mostly had a dilution effect. Therefore, floods did not influence annual chloride concentration means (Table 1): chloride concentration was 33.4 and 34.4 mg l^{-1} during and between floods, respectively. This seasonality of flood effects may have two explanations. First, at the beginning of the hydrological year, the rise of the water-table allows the transport of chloride that was concentrated by summer evapotranspiration, particularly in the wetland domain. Second explanation could be that the chloride signal in rain is itself seasonal (Hrachowitz et al., 2009; Neal et al., 1988; Shaw et al.; 2008). However, we believe that this second explanation has a minor effect in Kervidy-Naizin where (i) the mean concentration in GW is much higher than that of rainfall and (ii) rainfall water contributes to less than 10% of the flood flow, the major part coming from soil water.

P9726, l.7-12. Replace with: However, some authors (Bastviken et al., 2007; Chen et al., 2002; Viers et al., 2001) have warned that chloride is not systematically a conservative element, i.e. the yearly balance is not null. They reported that adsorption-like processes in soil organic matter, hydrology and microbial soil activity influence stream chloride concentration. Other studies reported local storage of chloride in soils, for instance under hedges due to higher evapotranspiration (Grimaldi et al., 2009), and in groundwater (Rouxel et al., 2011). Imbalances of the chloride budgets are also reported in catchments where land-uses changes occurred (Guan et al., 2010; Oda et al., 2009).

P9726, l.18-20. Replace with: At this period, the chloride pool is more superficial and closer to the stream, i.e. wetland GW, than the nitrate pool. After connection of the concentrated upland GW with the stream and according to the seasonality of the chloride signal in rainfall (albeit negligible here), chloride base-flow concentrations remain high but are diluted by floods, as nitrate concentrations.

P9727, l.2-10. Replace with: There is a large stock of organic carbon in shallow soil horizons, particularly in the wetlands where soil organic matter concentration is about 0.1 g g^{-1}). During winter and spring floods, DOC is flushed, and the magnitude of the release depends on the hydrological state of the catchment and the temperature (Dawson et al., 2008, 2011). From our results, based on 10 years, we concluded that in normally humid years, the high concentrations of DOC are depleted when raining. During summer, DOC concentrations increase due to biological soil activity and lack of transport. In-stream production contributes to the seasonal increase in the summer and beginning of autumn.

P9727, l.16-20. Replace with: Shallow groundwater connectivity plays an important role in controlling inter-flood stream concentration, as shown in the annual signature. The importance of upland GW connectivity is obvious on nitrate and chloride concentration. It confirmed previous conclusion drawn on Kervidy-Naizin or similar catchments stating that, for nitrate, (i) groundwater store is the major control of nitrate concentration in stream-water (Ruiz et al., 2002); (ii) groundwater concentration differs down-slope, where denitrification induces lower nitrate concentrations (Martin et al.,2004); (iii) water-table level dynamics along hill-slope controls annual nitrate concentration variations in the stream, along with the spatial distribution of the solutes in groundwater (Molenat and Gascuel-Odoux, 2002); for DOC, (iv) hill-slope soils are rapidly DOC depleted when the water-table rises uplands whereas wetland soils, from which >80% of the stream DOC come from, behave as an almost infinite DOC reservoir (Morel et al., 2009). Our study questions the connection of wetland GW with upland GW and stream. Upland GW must be contributing in minor proportions to the wetland during inter-flood periods, having direct flow pathways to the stream. The wetland GW contributed more to the stream during floods or high flow periods.

Comments on the inter-annual pattern part (3.2.2.)

P9727, l.25-26. Replace with: This result emphasised the role of alternating dry-and-wet periods and theirs consequences on hydrological connectivity as well as production processes.

P9728, l.2-6. Replace with: Therefore, variation of the recharge period (duration, quantity and intensity of rain) and temperature between years partly explains the high variability in the autumn-winter period.

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P9728, l.8. We confirm that Figure 4 presents variograms, i.e. the variance of the differences between measured values at two times across realisations. This method, usually applied for spatial variation analysis (2D), is applied here for time variation (1D). Periodograms are the estimates of spectral density of a signal (spectral analyses).

P9728, l.13-18. Replace with: This pointed out the hydrological control on the export of chloride to the stream. The nitrate variogram seemed a mixture of the two previous groups. These observations confirm the grouping of solutes according to their origins. At the 10-years scale, DOC, sulphate and DIC production relies mainly on surface biological processes influenced by temperature. Their temporal dynamic is linked with yearly processes occurring in the soil wetland domain. Nitrate and chloride are less influenced by the own catchment production, because they are in excess in the GW, and what is emphasized at the 10 years scale are transport conditions they depend on.

Comments on the generic classification part (3.3.)

P9728, l.20 - P9729 l.7. This §is meant to be generic, introducing the concepts of transport-limited versus supply-limited processes. Therefore, it is intentional that it is not more detailed. However, the following §is modified.

P9729, l.8-26. Replace with: Applying these concepts developed from single solutes (either nitrate or DOC) in several catchments to five solutes in one catchment, and keeping in mind the solute distinctions presented in this paper allows further classification (Fig. 5). First, nitrate and chloride depend on upland-groundwater-connection-limited processes: the seasonal pattern of shallow groundwater connectivity to the stream determines whether they can be exported. The timing, rate and duration of the connectivity are controlling factors of the export. Nitrate differs from chloride as nitrate production exists, even if it is not the main source, whereas chloride is not produced in the catchment. In contrast, sulphate and DOC depend on wetland-groundwater-connection-limited processes: the exported solutes are produced in the catchment within the period preceding the export but exports occur only when the wetland is hydrologically connected to the stream, i.e. floods and when the quickly reactive groundwater fluctuates in the upper soil organic layer. There is little time lag between production and export, even though production can be seasonal (sulphate) or continuous (DOC). Unlike nitrate which export is continuous during the rainy period, DOC and sulphate exports are highly discontinuous: every flood leads to a peak in concentration. For DOC and sulphate, the wetland domain is the main productive compartment close to the stream and contributes mostly during floods. It would be interesting to analyse data from other agricultural catchments to corroborate our classification. A longer dataset would provide longer temporal variograms, which may verify the periodicity identified in this catchment.

Comments on the conclusion (4.)

P9730, l.2; P9730, l.9-11; P9730, l.12-16. To account for the reviewers comments and the added data, the conclusion is rewritten as follows:

From this original dataset, the hydro-chemical signature of a livestock farming catchment dominated by shallow groundwater flow has been defined. This signature was identified by analysing (i) mean concentrations in four water compartments in the catchment (stream-water, upland, wetland and deep down-slope groundwater), considered as a static signature, and (ii) annual patterns of stream-water concentrations and their inter-annual variability, considered as a temporal signature. Our results broadened some previously proposed conceptual models, based on short-term studies, for nitrate and DOC. The data enabled us to explain annual patterns for chloride and sulphate. Both aspects of the signature, static and temporal, lead to a generic conceptual model of stream-water quality.

The static signature is defined by average long-term concentrations in the stream and the contributing compartments. Nitrate and chloride concentrations were high in both the shallow groundwater and the stream due to fertilisation, while sulphate, DOC and DIC were present at similar concentrations as in other catchments. The

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temporal signature was characterised at annual and inter-annual scales. By analysing the annual patterns observed, we proposed a classification of solutes. In-stream nitrate and chloride concentrations depended on the connection of upland shallow groundwater with the stream, while in-stream DOC, DIC and sulphate concentrations were influenced by the connection of wetlands with the stream. Inter-annual patterns emphasised that nitrate and chloride in the stream were influenced mostly by discharge, thus transport, while sulphate, DOC and DIC showed clear sinusoidal signals, similar to those of temperature and ET_0. It also showed that solute concentration variability was much higher during the re-wetting stage than during other periods.
Stream nitrate and chloride concentrations depended on the connection of their storage compartment (upland groundwater), while sulphate, DOC and DIC concentrations depended on catchment production and the connection of the producing compartment (wetland soil) during rainfall events. These hydrological connections control solute transport. From these hydrological connections of the stocking compartment, the concept of "transport-limited" processes was further developed. This classification could be applied to any chemical solute and help assess its origin, storage or production location and transfer mechanism in similar catchments.

Figures and Tables

The location of the meteorological station is added to Figure 1.
In Figure 2, discharge is shown in another panel; more contrasting symbols are chosen to make the figure readable.
As correctly deduced by referee 2, the blue lines in Figure 5 symbolize typical flood event. Figure 5 is updated according to the text of the present comment and the added data. Therefore the new legend is the following: "Generic classification of the temporal patterns of elements and their determinants. Thick lines represent stream concentrations between floods throughout a year. Blue lines represent the reaction during a flood. Horizontal (red) dotted lines represent mean upland groundwater (GW) concentrations, while horizontal dot-dashed (purple) lines represent mean deep

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down-slope GW concentrations and dashed (purple) lines represent mean wetland GW concentration. Criterion (1) is deduced from the bibliography. Criterion (2) is deduced from the comparison of stream concentrations to GW concentrations. Criterion (3) is deduced from the comparison of GW concentrations. Criterion (4) is deduced from the temporal patterns. DOC: dissolved organic carbon; DIC: dissolved inorganic carbon. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)".
Table 2 is updated with the added data.

Added references

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Oda, T., Asano, Y., Suzuki, M.: Transit time evaluation using a chloride concentration input step shift after forest cutting in a Japanese headwater catchment, Hydrol Process, 23, 2705-2713, 2009.

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Suppressed references

Carluer et al. (2004); Cognard et al. (1995) ; Dia et al. (2000) ; Payraudeau et al. (2007) ; Pourret et al. (2007)

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