Climate and contaminant transport: the role of within-storm variability on contaminant transport by surface runoff

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Abstract The impact of climate drivers on contaminant transport has been largely neglected. Rainfall variability within a storm event can have a significant impact on the amount of contaminant transport by surface runoff. We seek to better understand how rainfall patterns impact contaminant transport depending upon pesticides characteristics. A bounded random cascade approach is used to generate an ensemble of rain events with specific Intensity–Duration–Frequency characteristics. We explore the effects of the partitioning of rainfall and chemical between fast surface runoff and slow flow in the soil matrix. The hydraulic properties, i.e. effective porosity, suction head and saturated hydraulic conductivity, of a vineyard clay loam soil are investigated. Ten years of 6-min resolution rainfall data from the Alsatian vineyard in France are used to derive the cascade and Intensity–Duration–Frequency relationships. Pesticide transport, both by runoff and infiltration, are modelled by a near-surface mixing model and Green-Ampt infiltration. Much smaller pesticide loading occurred for more weakly and more strongly adsorbing pesticides. The patterns of rainfall generating large surface runoff did not necessarily associate with large pesticide loadings depending on pesticide adsorption. We show that potential shifts in rainfall patterns within storms dramatically impact the frequency of contamination events, even without changes in storm duration and mean intensity return intervals. The framework developed allows a better understanding of risk of pesticide transport by rapid flow processes under changing climate conditions.

Key words pesticides; rainfall; runoff; modelling

INTRODUCTION

The timing and nature of rainfall since the last pesticide application are often described as significant controls on pesticide transport via rapid flow processes as surface runoff (Leonard, 1990; Gregoire *et al*., 2010). Rainfall variability within an intense, high magnitude event can preferentially increase pesticide transport into surface runoff (Zhang *et al*., 1997) compared with a steady rainfall. McGrath *et al*. (2008) showed that rainfall variability during an event strongly affects the variability of pesticide transport by preferential flow, a process that is triggered by soil moisture thresholds similar to surface runoff.

As highlighted by recent studies (Rocha *et al*., 2008; Hancock, 2009; Floris *et al*., 2010; Mailhot *et al*., 2010), impacts of global change on rainfall would result in greater variability in the intensity of rain events. Therefore, shift in rainfall patterns and rainfall intermittency within storms may dramatically impact the frequency of contamination events, even without changes to mean storm duration and mean intensity. This is especially true for surface applied chemicals, such as pesticides, for which rapid flow processes such as surface runoff trigger off-site movement.

The focus of this paper is to investigate how solute transport by runoff occurs in response to natural rainfall variability within events. An existing method initially developed to analyse pesticide transport by preferential flow is adapted to assess the impact of rainfall variability by runoff (McGrath *et al*., 2008). The approach will focus on: (1) the rainfall signal controlling the triggering of pesticide transport; and (2) the impact of rain event duration and intensity on the subsequent transport of pesticides. The rainfall and soil characteristics applied in this analysis were derived from measurements made in the Hohrain catchment (Rouffach, Haut-Rhin, France) (Gregoire *et al*., 2010).

The paper is organised as follows: We first present the minimalist model used to simulate runoff and pesticide transfer from the soil matrix to surface runoff. We then describe the method for the simulation of the within-storm variability of rainfall for a series of rain events with a fixed return interval but variable intensity and duration. The results of this modelling exercise is then presented and discussed in the context of changing rainfall conditions.

MODELLING APPROACH

Surface runoff and solute transport modelling

Rapid flow processes such as preferential flow and runoff have repeatedly been shown to be initiated as a near surface process (Leonard, 1990; Lecompte *et al.*, 2001; Heppel *et al.*, 2004). Therefore, simple near surface mixing models tend to reproduce the transfer of pesticide from soil to runoff (Leonard, 1990; Zhang *et al.*, 1997).

A minimalist model previously developed to simulate preferential flow and solute transport modelling (McGrath *et al.*, 2008, 2010) was used to assess the impact of rainfall intensity on pesticide transport by runoff. This model includes both the partitioning of rainfall between infiltration and runoff, and a simple mixing layer model to simulate the retention and release of pesticides at the soil surface.

The partitioning of rainfall between the soil matrix and runoff pathway was performed by the Green-Ampt method (Green & Ampt, 1911; Mein & Larson, 1973). By using this method we assume an infiltration excess mechanism for runoff triggering which is supported by the field observation in the Rouffach catchment (Gregoire *et al.*, 2010). The method uses two effective parameters: $\Delta\theta\varphi$ [L] the product of the volumetric water content difference across the wetting front and the suction head at the wetting front; and K_s [L T^{-1}] the saturated hydraulic conductivity. The Green-Ampt method allows us to estimate the time until ponding t_p [T]. The cumulative depth of runoff will be denoted Q_r. The two parameters were estimated from field data (Tournebize, 2001). The retained values correspond to soil properties in an untilled row of the vineyard which correspond to a clay loam (Table 1).

The simple mass balance for a solute in the near surface fully mixed zone uses three parameters: z_l [L] the depth of the mixing layer, θ [L^3 L^{-3}] the volumetric water content and R [-] the retardation factor describing the role of sorption (McGrath *et al.*, 2008).

The mass of solute released to preferential flow from the surface layer m_r [M] is assumed to be proportional to the amount of rainfall converted to runoff in each time step. Table 1 summarizes the model parameters for the Rouffach catchment.

Table 1 Runoff and pesticide transport model parameters for the Rouffach catchment.

Parameter	Units	Values
K_s	mm h^{-1}	10.32
$\Delta\theta\varphi$	mm	51
z_l	mm	10
θ	cm^3 cm^{-3}	0.36
R	-	1; 5; 25

Three contrasting values of the retardation factor R were retained to investigate the different behaviour of pesticides in solution during rainfall events according their sorption. The first case analysed, R = 1, corresponds to a tracer, i.e. without sorption to the soil. With R = 5 and 25, progressively more strongly sorbed pesticide behaviour is investigated.

Within-storm variability of rainfall modelling

To investigate how the rainfall variability within a storm is reflected in the variability of pesticide transport by surface runoff, random within-storm rainfall patterns were generated. Among the existing method of rainfall disaggregation (Gaume *et al.*, 2007; Rupp *et al.*, 2009), a micro-canonical, also called Bounded Multiplicative Random Cascades approach (BMRC) was adopted (Menabde & Sivapalan, 2000). As discussed in McGrath *et al.* (2008), the BMRC approach was retained both to ensure mass balance in rainfall simulations and to preserve the statistical properties of both low and high intensity rainfall.

We analysed six-minute resolution rainfall data recorded at the Rouffach station (Meteo France, station no. 68287003; 47°57′9 N, 07°17′3 E; 284 m a.m.s.l.), from January 1998 to June 2010. The data were recorded to a precision of 0.2 mm. A power law relationship between the temporal resolution of interest *t* [T] (in relation to the rainfall resolution t_0 [T]) and *r* the shape parameter of the beta function [-] describing the variability of the ration of rainfall amounts between two temporal resolutions was obtained as in Menabde & Sivapalan, (2000) and Rupp *et al.*, (2009):

$$r_t = r_0\ (t/t_0)^{-H} \qquad (1)$$

where r_0 [-] and *H* were empirically derived (Menabde & Sivapalan, 2000). The values of r_0 and *H* were found to be 12.8 and 0.53, respectively.

Based on a Duration–Intensity–Frequency analysis of the 12 years of rainfall measured at Rouffach, four events were retained corresponding to a 10-year return period with durations of 24, 48, 96 and 192 minutes. The temporal resolution of rain simulation was fixed at 6 minutes, in accordance with the resolution of the rainfall database. Therefore, between 4 and 32 constant intensity rainfall periods were modelled corresponding to events from 24 minutes to 192 minutes length, respectively. A total of 5×10^4 realisations of the distribution of rainfall periods were simulated by the BMRC disaggregation procedure.

RESULTS

Simulations with constant rain intensities were run as a reference to assess the impact of within storm variability on the runoff coefficient. Of the constant intensity events only the two shortest duration (greater mean intensity) events, A and B, generated surface runoff (Fig. 1). Runoff occurred for all event types when within storm variability was considered. These results imply that using high resolution data is a necessity to identify critical source areas for runoff-related pesticides. The percentage of the 5×10^4 realizations generating runoff is 100%, 96%, 77% and 52% for events A to D, respectively. For a fixed return period, the shortest events, with the highest intensity, are associated to the highest risk of runoff. The mean runoff coefficients were found to be significantly different from each other ($p < 0.013$ between C and D, and $p < 2.2\ 10^{-6}$ for other event combinations using the paired nonparametric Signed Rank test).

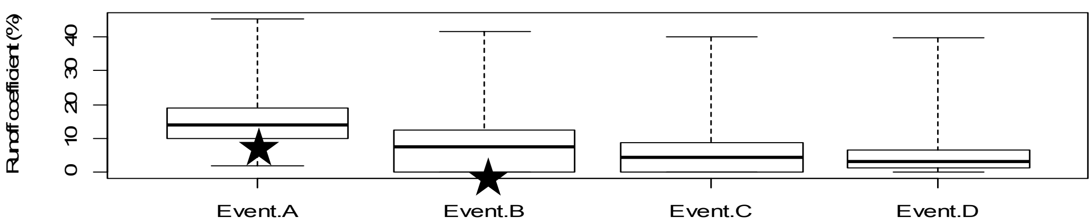

Fig. 1 Runoff coefficient associated to the four events with constant intensity *vs* bounded Multiplicative Random Cascades realizations. Box whisker plots show maximum, minimum, mode, 25% and 75% quartiles of the runoff coefficients for those realizations which generated runoff. For events A and B the star (★) indicates the runoff coefficient obtained with constant rainfall intensity.

Event-based runoff coefficients vary widely with event B showing greatest variation. This apparently stems from the balance between the mean intensity required to generate runoff, higher for the event B than for C and D, and the number constant intensity rainfall periods, i.e. 8 periods of 6 minutes for event B.

From ensembles of rainfall distributions within each event type it can be seen that large runoff generating events tend to begin with low intensities and build up in intensity throughout the event. This pattern was similar for the four events from A to D. Figure 2(a) shows this in terms of the ensemble average rainfall distributions which contributed to the largest 25% of runoff events. These events begin with below average intensities building throughout the event and peak towards the end. Figure 2 (b) and (c) shows the temporal rainfall distributions dominating solute loading corresponding to tracer (R = 1) and strongly sorbed pesticide (R = 25), respectively. The largest 25% of tracer-loading storms describe earlier peaking intensities in the rainfall (Figure 2(b)) than the 25% of sorbed pesticide-loading storms (Fig. 2(c)). The largest 25% sorbed pesticide-loading events appears to be strongly associated with the equivalent quartiles of runoff, i.e. the largest 25% of runoff events. The largest 25% of loading patterns emerge from the balance between the volume of water required to initiate and sustain runoff and the amount of chemical that is available for transport whenever runoff is triggered. Because conservative tracers are so mobile, early runoff initiation has a greater impact on the volume of runoff.

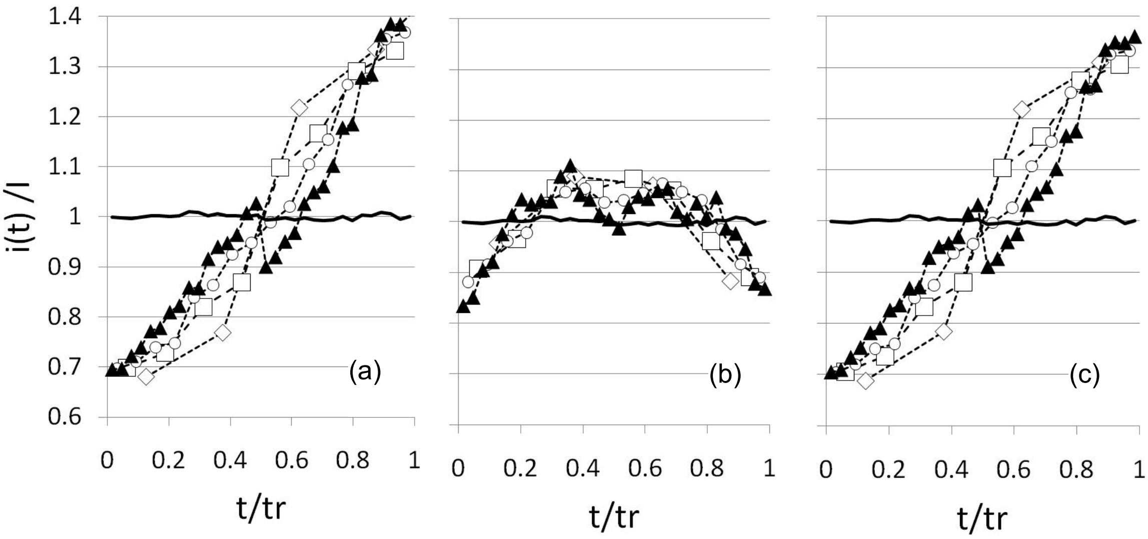

Fig. 2 Temporal rainfall distributions dominating the largest 25% (a) of runoff events, (b) of solute loading of tracer (R = 1) events, and (c) of solute loading of sorbed pesticide (R = 25) events. The y axis denotes the ensemble averaged rainfall intensity (normalised by the mean event intensity $< i(t) / I >$ occurring at time *t* (normalised by rainfall event duration *tr*). The symbols correspond to the events A (◇), B (□), C (○) and D (▲). The solid line shows the ensemble averaged rainfall intensity throughout the event.

The range of pesticide loading to runoff as a function of the retardation factor R for the four events from A to D are shown in Fig. 3. Greater R values indicate stronger sorption to soil. The mean loads of each of the 12 combinations, i.e. four events and three retardation factors are significantly different from one another ($p < 2.2 \times 10^{-6}$ using the paired nonparametric Wilcoxon Signed Rank test). As sorption increases the mean mass transported by runoff increases with increasing sorption before decreasing at higher sorption capacities. Because weakly sorbing solutes (R = 1) are more likely to be transported into the soil matrix and away from the soil surface before the initiation of runoff, stronger sorbing solutes are more likely to be transported by surface runoff. Stronger sorption (R = 5) means more pesticide is retained near the surface of the soil when runoff occurs irrespective of when it occurs during the rainfall event. However, very strong sorption (R = 25) leads to smaller losses in runoff because it is held so tightly to the soil and hence it is unavailable for transport through either runoff or matrix flow. As discussed in McGrath *et al.*, (2008), this behaviour is consistent with observations performed to link amounts transported and pesticide sorption. This study, further demonstrates that the variability of pesticide loadings also increases as the event duration increases in conjunction with decreasing intensity.

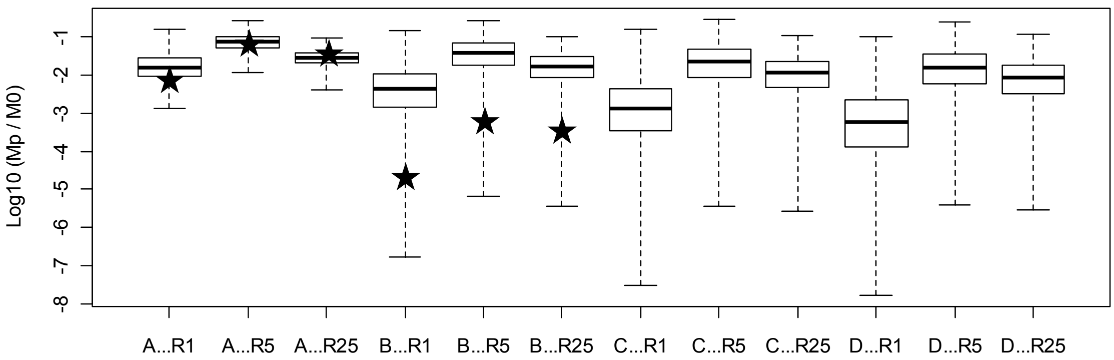

Fig. 3 Effect of sorption on event pesticide loading of runoff dependence for events A to D and tested retardation factor values R equal to 1, 5, 25. Box whisker plots show maximum, minimum, mode, 25% and 75% quartiles of the log of total mass loaded of runoff log(M_p/M_0) for those realizations which generated runoff. For the event A and B, the star (★) indicates the pesticide loading obtained with constant rainfall intensity.

In a global change context, it is relevant to analyse the potential impact of an increase of intensity variability within changes in storm duration and mean intensity return intervals. According to the Bounded Multiplicative Random Cascades approach, the increase of rainfall intensity variability is simulated by an increase of H as defined in the equation (1). As a first step of sensitivity analysis we analysed the impacts of an increase of 10% of the H for the four events. The results show an increase both of runoff and number of contamination events, i.e. with pesticide transports, after the increase of H (Table 2). The impact of H increase on runoff is higher for long duration (Event D) than for short duration (Event A) with an increase of 26% and 5%, respectively. The increase of the number of contamination events is also higher for event D than for event C and B with an increase of 21%, 7% and 1%, respectively.

Table 2 Effect of increase of rainfall intensity intermittency on runoff coefficient (RC) and pesticide loading expressed by the percentage of the simulations that produce off-site pesticide transport.

			$H = 0.53$		$H = 0.583$	
Event	Event duration (mm)	Rainfall intensity (mm h^{-1})	RC %	Contamination events %	RC %	Contamination events %
A	24	54.2	13.8	100	14.5	100
B	48	31.7	8.7	96.4	9.8	97.3
C	96	18.1	4.4	76.7	5.5	82.2
D	192	9.8	3.1	51.5	3.9	62.5

CONCLUSION

The results suggest the following recommendations to assess the risk of pesticide transport by surface runoff under changing climate conditions:

– Using rainfall intensity data at low temporal resolutions of hours to days is not adequate to assess surface runoff and the risk of pesticide off-site movement associated with it. Indeed the results show that variations in rainfall intensity within events have a significant effect on the triggering of point-scale rapid flow and transport processes such as surface runoff.

– Monte Carlo simulations investigating the role of within-storm rainfall variability during the four events suggested that the average amount of solute released to runoff pathways increases rapidly for weakly sorbing solutes, then decreases for more strongly sorbed solutes with increases in sorption. This result casts doubt on the utility of sorption capacity as a measure of the mobility of solute by runoff.
– The riskiest events in terms of runoff were end-of-event peaking while they are middle-of-event or end-of-event for pesticide loading depending on pesticide sorption characteristics.
– A preliminary sensitivity analysis shows that potential shifts in rainfall patterns within storms dramatically impact the frequency of contamination events, even without changes to storm duration and mean intensity return intervals.
– A better understanding of how or whether climate change will alter scaling relationships of rainfall variability, particularly at fine temporal scales, is required for water quality risk assessment.
– A key way forward is the extension of this framework from point-scale to hillslopes to investigate the threshold nature of surface runoff connectivity at the landscape scale.

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