Preferential flow of surface-applied solutes: Effect of lysimeter design and initial soil water content

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Abstract

Undisturbed lysimeters are widely used to study water and solute transport, where both natural and unnatural preferential flow can greatly influence leaching rates. The objective of this study was to use chemical and isotopic tracers to quantify the effect of initial soil water content and lysimeter design on preferential flows. Ten undisturbed lysimeters (900 cm²) were collected from an agricultural field, with the gap between the soil and lysimeter casing sealed with petroleum jelly for five lysimeters. Lysimeters were subjected to two rainfall simulations (3.3 cm h⁻¹) under contrasting initial soil water contents, and leachate near the soil-casing interface was collected separately from leachate through the bulk soil. Three-component hydrograph separation revealed that event water comprised 21–59% of total leachate irrespective of initial soil water content and lysimeter design. Sealing the edges of the lysimeters with petroleum jelly greatly reduced but did not eliminate edge flow during rainfall simulations. Although water and solute transport were similar in both sealed and unsealed lysimeters under dry antecedent conditions due to the formation of shrinkage cracks on the soil surface, edge flow was substantially greater for the unsealed lysimeters under wet antecedent conditions. Unsealed edges under wet antecedent conditions facilitated the preferential transport of both event and pre-event water, resulting in greater solute leaching. Using multiple tracers to contrast lysimeter designs under different initial soil water contents not only allowed for rigorous testing of a commonly used edge-flow suppression technique but also provided new insights into preferential flow processes and patterns.

1 INTRODUCTION

Preferential flow in soil is a widespread phenomenon, as continuous networks of fast flow pathways have been observed across diverse soil types and landscapes (Jarvis, Koestel, & Larsbo, 2016). Preferential flows are often associated with soil macro pores such as cylindrical biopores (i.e., burrowing soil fauna, plant roots) or planar fissures (i.e., desiccation cracks). Explaining how nutrients and pesticides were being transported to subsurface tile drains and groundwater was one of the primary drivers for initially studying preferential flows (Beven & Germann, 1982); interest in these processes has only increased over
the past 30 yr (Beven & Germann, 2013; Gerke, Germann, & Nieber, 2010). Recent attention on P transport via soil macropores underscores this trend (King et al., 2015; Radcliffe et al., 2015), especially given increasing surface water quality degradation globally as a result of nonpoint-source pollution (Ho, Michalak, & Pahlevan, 2019).

Soil macropores and preferential flows have been studied at the soil pore to watershed scales (see reviews by Beven & Germann, 2013; Jarvis et al., 2016; Singh, Kaur, Williard, Schoonover, & Kang, 2018). Using tracers, dye staining, and tomographic techniques to elucidate water and solute transport, soil column or drainage lysimeter experiments have been widely conducted to measure and model preferential flows because they offer the advantage of working with a known soil volume and providing greater control over experimental initial and boundary conditions. The initial soil water content (Shipitalo & Edwards, 1996), rainfall intensity and duration (Jarvis, 2007), and saturated hydraulic conductivity of the soil matrix, which is strongly controlled by soil texture (Lin, McInnes, Wilding, & Hallmark, 1999), determine when preferential flow occurs. Although the effects of initial and boundary conditions on macropore flow are complex, especially for soils where the structure is a dynamic function of water content (i.e., shrink–swell clay soils), laboratory lysimeter studies that allow for greater control over these variables can provide detailed insights into processes.

Numerous variations and modifications to lysimeter design have been proposed and described in the literature (Singh et al., 2018). Many of these modifications have been aimed at better managing edge-flow effects, one of the largest recognized limitations for lysimeter studies (Titus & Mahendrappa, 1996). Edge flow between the soil and the lysimeter casing can be problematic because water transported as unnatural preferential flow along this interface can have a profound influence on results such as nutrient leaching rates (Cameron, Harrison, Smith, & McLay, 1990). As a result, many lysimeter studies employ methods to minimize edge-flow effects, including (a) using sealants such as petrolatum, plaster of paris, or liquid polyurethane foam to seal the gap between the soil and the casing (Feyerisen & Folmar, 2009); (b) constructing annular rings to move rainfall water away from the casing wall (Corwin & Le Mert, 1994); (c) roughening the lysimeter casing to improve contact with the soil (Powelson & Gerba, 1994); (d) collecting lysimeters with a large surface area and under appropriate antecedent field conditions (Bergström, 1990); or (e) collecting leachate from only the central core of the lysimeter (Till & McCabe, 1976). Modifying boundary conditions to reduce preferential flows along the lysimeter casing are often described as effective; however, few studies have directly compared lysimeters with and without boundary condition modifications (Williams, McAfee, & Kent, 2019).

In this study, 10 undisturbed soil blocks were collected from an agricultural field with fine-textured soils to quantify the effect of initial soil water content (wet vs. dry) and lysimeter design (sealed vs. unsealed edges) on preferential flows. Petroleum jelly was chosen to seal the gap between the soil and the lysimeter casing for five of the soil blocks given its widespread use in the literature (Williams et al., 2019). Leachate during two rainfall simulations was collected such that flow along the casing–soil interface could be separated from flow through the center of the soil block in order to separate edge flow from flow through bulk soil. Specific objectives were (a) to quantify preferential flow processes in sealed and unsealed lysimeters under contrasting initial antecedent conditions, and (b) to examine how differences in preferential flows influenced the transport of surface-applied solutes. Although previous studies have reported the effectiveness of petroleum jelly at reducing edge flow under ponded conditions (Cameron et al., 1990) and after long periods of soil drying (Williams et al., 2019), a secondary objective was to build upon this previous work and determine the effectiveness of petroleum jelly at reducing edge flow under varying antecedent conditions and related effects on solute transport. Comparison of lysimeter design and initial soil water content has the potential to provide new insights into preferential flow processes and patterns.

2 | MATERIALS AND METHODS

2.1 | Soil block collection and edge flow suppression

Ten blocks of undisturbed soil were collected from an agricultural field located in northeastern Indiana, USA (48° 28′ 20.19″ N, 84° 59′ 28.50″ W). The field was farmed with a 4-yr rotation of corn (Zea mays L.), soybean (Glycine
max [L.] Merr., oat (Avena sativa L.), and wheat (Triticum aestivum L.). Soil block collection occurred in the spring prior to tillage and oat planting. Soils at the site were Glynwood loam (fine, illitic, mesic Aquic Hapludalfs), which are characterized as moderately well drained. Saturated hydraulic conductivities decrease from 30 mm h$^{-1}$ in silt loam surface horizons (0–20 cm) to <3 mm h$^{-1}$ in silty clay to clay subsurface horizons (15–60 cm).

Soil blocks were collected with square steel casings (30 × 30 × 30 cm, 0.5 cm thick) using the methods of Cameron et al. (1992). Briefly, casings were lined with 1.0-cm polyvinyl chloride (PVC) spacers, and a removable cutting head was mounted on the bottom (Figure 1a). The casing with cutting head was placed on the soil surface and a trench was dug around the casing, exposing a soil block approximately 40 × 40 × 10 cm. The casing with cutting head was pushed down over the soil block using the bucket of a front-end loader, and excess soil was sheared off. Digging around the casing helped minimize soil compaction during lysimeter collection. The process of digging a trench and pushing the casing into the soil was repeated several times until it was filled to within 2.5 cm of the top (final soil depth = 27.5 cm). The soil block was then separated from the soil below, the cutting head and extra soil were removed, and a perforated PVC plate (1.3 cm thick) with geotextile fabric was attached to the bottom of the casing (Figure 1b). All soil blocks were collected near one another along a single crop row. Volumetric soil moisture was 0.33 and 0.37 cm$^3$ cm$^{-3}$ at depths of 5 and 20 cm, respectively, when soil blocks were collected.

Upon return to the laboratory, PVC spacers lining the casing were removed from five randomly selected soil blocks by pulling them out from the top. Petroleum jelly was heated until liquefied and poured into the gap created after spacer removal to within 1.0–2.0 cm of the soil surface. Petroleum jelly was selected as a sealant to prevent edge flow due to its inherent properties (e.g., low melting temperature, ability to be poured or injected as a low-viscosity liquid) and widespread use in lysimeter studies (Williams et al., 2019). After petroleum jelly cooled and returned to a semisolid state, paraffin wax was heated and poured on the petroleum jelly to the level of the soil surface (see Feyereisen & Folmar, 2009). Soil blocks with sealed edges are hereafter referred to as the “jelly” treatment, whereas the five remaining soil blocks with PVC spacers in place were considered the unsealed or “spacer” treatment.

2.2 Instrumentation and leachate collection

Each soil block was equipped with a ceramic suction-cup lysimeter installed to a depth of 15 cm from the soil surface. Suction cups were located in one corner of each soil block ~5 cm from the casing. A soil probe (2.5-cm diam.) was used to create a void in the soil, the suction-cup lysimeter was inserted, the remaining void was backfilled with soil, and bentonite clay slurry was used to seal the soil surface around the suction-cup lysimeter. To collect leachate from the perforated PVC bottom, a collector pan was

![Cross-sectional view of the soil block with (a) cutting head and plastic spacers in place for insertion into the soil, (b) soil block extracted, perforated polyvinyl chloride (PVC) bottom attached, and plastic spacer or petroleum jelly, and (c) bottom view of the perforated PVC bottom showing the inner and outer flow collectors. Note: drawing not to scale.](image-url)
fabricated and mounted to each soil block such that leachate near the casing–jelly or casing–spacer interface could be separated from leachate through the middle of the soil block (Figure 1c). The outer flow collector extended from the steel casing inward 3.5 cm (0.5 cm casing, 1.0 cm of the PVC spacer or jelly, and outermost 2.0 cm of soil), which equated to 26.5% of the drainable surface area (208 of 784 cm$^2$) of the soil block. The inner flow collector captured leachate from the middle of the soil block (576 of 784 cm$^2$).

### 2.3 Rainfall simulations

Soil blocks were wetted to field capacity by saturating them from the bottom up by placing them in a pool of water (4 d) and subsequently removing them from the pool and allowing them to drain (3 d). A vacuum (60 kPa) was placed on the ceramic suction-cup lysimeter in each soil block 24 h before the rainfall simulation, with samples collected prior to the simulation. Potassium bromide solution (160 ml 10 g KBr L$^{-1}$) was uniformly surface applied to each soil block immediately prior (30–60 min) to a 60-min rainfall simulation (33 mm h$^{-1}$). The soil surface was positioned 2.0 m below the nozzles (VEEJET 80-100) of the simulator, and deionized water was used as the source of rainfall. Leachate from both the inner and outer flow collectors was sampled every 5 min during the rainfall simulation, with samples collected periodically up to 24 h.

After the first rainfall simulation, soil blocks were stored in the laboratory (average air temperature 22–25°C) for 28 d. No additional water was added to the soil blocks during this time. Due to the dry antecedent conditions, no samples were able to be collected from the ceramic suction-cup lysimeters prior to the second rainfall simulation. Calcium chloride (CaCl$_2$) solution (160 mL; 10 g CaCl$_2$ L$^{-1}$) was uniformly surface-applied to each soil block immediately prior to a 90-min rainfall simulation (33 mm h$^{-1}$). The duration of the second rainfall simulation was increased compared to the first rainfall simulation, as it was expected it would require more rainfall to generate leachate after the soil blocks had been drying for several weeks. Leachate from both the inner and outer flow collectors was sampled every 5 min during the second rainfall simulation, with samples collected periodically up to 24 h.

### 2.4 Water sample analysis

All water samples, including those collected from the pool used to saturate the soil blocks, suction-cup lysimeters, rainfall during the simulation, and leachate from soil blocks, were analyzed for concentrations of K$^+$, Ca$^{2+}$, Mg$^{2+}$, Br$^-$, Cl$^-$, SO$_4^{2-}$, and NO$_3^-$ by ion chromatography (Thermo Fisher Scientific, Dionex ICS-2100). Water samples were also analyzed for stable water isotope ratios using a liquid water isotope analyzer (Los Gatos Research, LWIA-45-EP). Samples were analyzed against reference values calibrated to Vienna Standard Mean Ocean Water. Instrument precision for δ$^{18}$O and δ$^2$H was ±0.11 and ±0.5‰, respectively.

### 2.5 Hydrograph separation

Hydrograph separation was used to assess flow pathways through the soil blocks for both inner and outer flow collectors. Three components (rainfall [event water, $Q_e$], soil water [water from suction-cup lysimeter, $Q_s$], and uncharacterized soil water [$Q_u$]) and two tracers (δ$^{18}$O and Mg$^{2+}$) were used in a mass balance approach to solve for the leachate mixing fractions (i.e., $Q_u/Q$, $Q_s/Q$, and $Q_u/Q$; see equations for three-component hydrograph separations in Genereux, 1998). The third component, $Q_u$, was included as part of the hydrograph separation because δ$^{18}$O values for some leachate samples were lower than both rainfall (−6.72 ± 0.48‰) and soil water (−8.45 ± 0.73‰) and approached the δ$^{18}$O value of the water used to saturate the soil blocks (−13.68‰); that is, saturating the soil blocks did not produce a uniform δ$^{18}$O depth profile. The δ$^{18}$O signature of $Q_u$ was set equal to the measured δ$^{18}$O of the water used to saturate the soil blocks, whereas the Mg$^{2+}$ concentration was assumed to be equal to that measured in the suction-cup lysimeters. To separate leachate from the second simulation when dry antecedent conditions precluded collection of a suction-cup lysimeter sample, the Mg$^{2+}$ concentration of $Q_u$ and $Q_u$ was assumed to be the same as samples collected from the suction-cup lysimeter prior to the first simulation. Values of δ$^{18}$O for both $Q_u$ and $Q_u$, however, were increased by 1.33‰ to account for evaporative effects during the period between rainfall simulations based on previous laboratory results under similar conditions (Williams, Larrey, & Sanders, 2018).

### 2.6 Data analysis

Time to flow initiation, time to surface ponding, peak flow rate, discharge to precipitation ratio ($Q/P$), and leachate mixing fractions were determined for each of the 10 soil blocks. Mixing fractions were also multiplied by $Q/P$ to determine $Q_u/P$, $Q_s/P$, and $Q_u/P$ as described by von Freyberg, Studer, Rinderer, and Kirchner (2018). Although $Q_u/Q$ quantifies the fraction of total storm discharge that comes from rainfall, $Q_u/P$ quantifies the fraction of total rainfall that will be discharged during the same event and
has been previously used as a surrogate for the fraction of drainage area that generates surficial runoff (Buttle & Peters, 1997). Similarly, $Q_u/Q$ (or $Q_o/Q$) quantifies the fraction of total discharge that comes from storage, whereas the ratios $Q_s/P$ and $Q_o/P$ quantify how much pre-event soil water and uncharacterized soil water, respectively, is mobilized by, not contained in, a unit volume of precipitation (i.e., $Q_u/P$ and $Q_o/P$ do not represent a physical fraction of a whole). These metrics have been shown useful in elucidating major sources and controls on streamflow generation, especially when storm characteristics and antecedent conditions vary among events (von Freyberg et al., 2018).

To determine the effect of treatment (i.e., spacer vs. jelly), antecedent condition (i.e., wet vs. dry), and flow collector (i.e., inner flow collector vs. outer flow collector) on hydrograph characteristics (e.g., time to flow initiation, peak flow rate, $Q/P$) and hydrograph components ($Q_u/Q$, $Q_s/Q$, $Q_o/Q$, $Q_u/P$, $Q_s/P$, and $Q_o/P$) a three-way ANOVA was completed. Main effects and interaction effects were considered significant at $\alpha = 0.05$. Temporal patterns in hydrograph components were also visually assessed to infer dominant processes. The relationship between hydrograph components and surface-applied solutes ($\text{Br}^-$ and $\text{Cl}^-$) were analyzed using analysis of covariance (ANCOVA) as outlined by Clausen and Spooner (1993). Initial evaluation showed no differences between the slope or intercept of the inner and outer flow collectors; thus, data were pooled and the slope and intercept of the linear relationship between spacer and jelly treatments were compared. Significantly different slopes (or intercepts when slopes were not significantly different) indicated a treatment effect. All data were analyzed using R statistical software (R Development Core Team, 2011).

3 | RESULTS

3.1 | Flow characteristics

Leachate commenced quicker during the first simulation compared with the second simulation, with a significant interaction effect found between antecedent condition and treatment ($p = .028$) (Table 1). Leachate was observed for the spacer treatment ($15.2 \pm 2.4$ min; mean $\pm$ SD) prior to the jelly treatment ($22.4 \pm 10.1$ min) under wet antecedent conditions. In contrast, during dry antecedent conditions, time to leachate initiation was $37.9 \pm 17.1$ min and $28.1 \pm 12.1$ min for the spacer and jelly treatments, respectively. Ponding of rainfall on the soil surface occurred during both rainfall simulations (Table 1). For the wet antecedent conditions, ponding on the soil surface started to occur at $13.9 \pm 6.9$ min and $47.3 \pm 6.5$ min for the jelly and spacer treatments, respectively. Ponding on the soil surface began at $30.3 \pm 17.8$ min for the jelly treatment and $37.0 \pm 7.9$ min for the spacer treatment under dry antecedent conditions.

Peak flow rate was significantly different between inner ($0.35 \pm 0.14$ mm min$^{-1}$) and outer ($0.68 \pm 0.41$ mm min$^{-1}$) flow collectors ($p < .001$) (Table 1). A significant interaction effect of antecedent condition and treatment was also found for peak flow rate ($p = .013$) (Table 1). For the spacer treatment, peak flow rate decreased from $0.75 \pm 0.50$ to $0.53 \pm 0.24$ mm min$^{-1}$ between wet and dry antecedent conditions, whereas for the jelly treatment, peak flow rate increased from $0.31 \pm 0.24$ to $0.48 \pm 0.20$ mm min$^{-1}$. Since rainfall duration varied between simulations, peak flow rate during the first 60 min ($\text{Peak}_{60}$) was also compared, assuming that if rainfall ceased at 60 min during the second simulation, then flow rate would have subsequently decreased. A significant interaction effect between the antecedent condition and treatment was found for $\text{Peak}_{60}$ flow rate ($p = .006$) (Table 1). $\text{Peak}_{60}$ flow rate was greater for the spacer treatment and wet antecedent condition ($0.75 \pm 0.50$ mm min$^{-1}$) compared with the other combinations of treatment and antecedent condition ($0.31 \pm 0.23$, $0.29 \pm 0.19$, $0.29 \pm 0.05$ mm min$^{-1}$).

Discharge to precipitation ratio was not significantly influenced by treatment ($p = .279$); however, $Q/P$ varied significantly according to main effects of both flow collector ($p = .004$) and antecedent condition ($p = .002$) (Table 1). Discharge to precipitation ratio was greater for the outer flow collector ($0.90 \pm 0.64$) compared with the inner flow collector ($0.50 \pm 0.21$). For wet and dry antecedent conditions, $Q/P$ was $0.92 \pm 0.61$ and $0.48 \pm 0.25$, respectively.

3.2 | Three-component hydrograph separation

The $\delta^{18}\text{O}$ signature of rainfall during the first simulation averaged $-6.72 \pm 0.48\%$, whereas the $\delta^{18}\text{O}$ signature of soil water collected from suction-cup lysimeters averaged $-8.45 \pm 0.73\%$. Leachate during the first simulation had $\delta^{18}\text{O}$ values between $-11.45$ and $-6.33\%$, with minimum and maximum $\delta^{18}\text{O}$ values approaching the signature of water used to saturate the soil blocks prior to the simulation ($-13.68\%$) and rainfall, respectively. The $\delta^{18}\text{O}$ signature of rainfall during the second simulation was $-6.33 \pm 0.26\%$, and leachate $\delta^{18}\text{O}$ values ranged from $-8.24$ to $-5.58\%$. Magnesium concentration in rainfall was near the analytical detection limit ($0.1 \pm 0.1$ mg L$^{-1}$) for both simulations. Suction-cup lysimeter soil water $\text{Mg}^{2+}$ concentration averaged $16.0 \pm 3.5$ mg L$^{-1}$, whereas leachate had $\text{Mg}^{2+}$ concentrations ranging from $0.4$ to $25.5$ mg L$^{-1}$ and $0.1$ to $22.3$ mg L$^{-1}$ during the first and second simulations, respectively. Mixing diagrams ($\delta^{18}\text{O}$ vs. $\text{Mg}^{2+}$) for each of the soil blocks are shown in Figure 2.
### TABLE 1 Flow characteristics including time to flow initiation, time to ponding, peak flow rate, and discharge/precipitation ratio for each of the soil blocks during the first and second rainfall simulations

| Treatment Block | Flow initiation | Peak flow rate | Peak 60 flow rate | Q/P | Time to ponding |
|------------------|-----------------|----------------|-------------------|-----|-----------------|
|                  | Inner | Outer | Inner | Outer | Inner | Outer | Inner | Outer | Inner | Outer |
| **Rainfall Simulation 1 (wet)** |       |       |       |       |       |       |       |       |       |       |
| Spacer 1         | 16.0  | 16.8  | 50.5  | 0.57  | 1.03  | 0.57  | 1.03  | 0.87  | 1.20  |       |
| 3                | 15.8  | 15.8  | 51.8  | 0.54  | 0.78  | 0.54  | 0.78  | 0.80  | 1.09  |       |
| 5                | 13.0  | 18.0  | 39.7  | 0.57  | 0.38  | 0.57  | 0.38  | 0.84  | 0.52  |       |
| 8                | 15.3  | 15.0  | n/a   | 0.34  | 1.53  | 0.34  | 1.53  | 0.49  | 2.30  |       |
| 9                | 16.8  | 9.6   | n/a   | 0.14  | 1.64  | 0.14  | 1.64  | 0.19  | 2.57  |       |
| Jelly 2          | 21.3  | 31.5  | 9.2   | 0.12  | 0.06  | 0.12  | 0.06  | 0.67  | 0.13  |       |
| 4                | 12.1  | 21.2  | 23.7  | 0.39  | 0.76  | 0.39  | 0.76  | 0.63  | 1.18  |       |
| 6                | 15.5  | 20.0  | 16.5  | 0.46  | 0.59  | 0.46  | 0.59  | 0.75  | 0.97  |       |
| 7                | 36.5  | 32.6  | 14.3  | 0.07  | 0.32  | 0.07  | 0.32  | 0.29  | 1.39  |       |
| 10               | 21.1  | 21.2  | 6.0   | 0.13  | 0.22  | 0.13  | 0.22  | 0.70  | 0.80  |       |
| **Rainfall Simulation 2 (dry)** |       |       |       |       |       |       |       |       |       |       |
| Spacer 1         | 37.8  | 25.3  | 55.0  | 0.34  | 0.68  | 0.14  | 0.16  | 0.31  | 0.54  |       |
| 3                | 30.6  | 35.0  | 14.0  | 0.35  | 0.93  | 0.21  | 0.50  | 0.31  | 0.75  |       |
| 5                | 35.5  | 76.0  | 32.5  | 0.41  | 0.25  | 0.32  | 0.00  | 0.42  | 0.07  |       |
| 8                | 41.5  | 25.8  | n/a   | 0.41  | 0.83  | 0.28  | 0.69  | 0.39  | 0.84  |       |
| 9                | 29.3  | 42.0  | 46.4  | 0.40  | 0.72  | 0.29  | 0.36  | 0.38  | 0.56  |       |
| Jelly 2          | 48.0  | 17.0  | 43.0  | 0.44  | 0.31  | 0.24  | 0.24  | 0.43  | 0.31  |       |
| 4                | 31.5  | 56.7  | 16.7  | 0.37  | 0.59  | 0.27  | 0.13  | 0.41  | 0.36  |       |
| 6                | 34.0  | 22.0  | 26.0  | 0.40  | 0.65  | 0.26  | 0.50  | 0.38  | 0.89  |       |
| 7                | 22.5  | 22.3  | 11.6  | 0.34  | 0.45  | 0.17  | 0.27  | 0.44  | 0.40  |       |
| 10               | 13.3  | 13.3  | 54.0  | 0.28  | 0.96  | 0.16  | 0.64  | 0.28  | 1.17  |       |

*Peak flow rate observed during the first 60 min of the rainfall simulation.
Discharge/precipitation ratio.
Inner and outer flow collector.
Not applicable.

Temporal patterns of $Q_e/Q$ during the first simulation were consistent among replications and between flow collectors for the spacer treatment (Figure 3; Soil Blocks 3 and 9 shown as examples). Event water comprised a small fraction of discharge at the onset of flow (12.6 ± 13.2%). For both inner and outer flow collectors, $Q_e/Q$ increased with flow rate, with maxima (65.8 ± 7.8%) observed near peak flow. On the falling limb of the hydrograph, $Q_e/Q$ declined but remained between 40 and 60%. Similar to the spacer treatment, $Q_e/Q$ increased with flow rate for the inner flow collector of the jelly treatment, with $Q_e/Q$ maxima observed near peak flow (Figure 3; Soil Blocks 2 and 6 shown as examples). Maximum $Q_e/Q$ for the inner flow collector of the jelly treatment (15.4–50.9%), however, was substantially less than the spacer treatment. Event water fractions of discharge tended to remain elevated on the falling limb of the hydrograph. In contrast with the inner flow collector, maximum $Q_e/Q$ for the outer flow collector of the jelly treatment was observed at the onset of flow (70.0 ± 23.0%). Event water fraction of discharge continuously decreased throughout the simulation, with values on the falling limb of the hydrograph averaging 31.6 ± 8.0%.

During the second simulation, maximum $Q_e/Q$ for the inner flow collector of the spacer treatment (61.3 ± 12.8%) was observed early on the rising limb of the hydrograph, with values decreasing substantially after the cessation of rainfall (22.4 ± 9.2%) (Figure 3). The outer flow collector of Soil Blocks 1, 8, and 9 exhibited a similar temporal pattern of continuously decreasing $Q_e/Q$; however, $Q_e/Q$ for the outer flow collector of Soil Blocks 3 and 5 increased with flow rate, with maximum values observed near peak flow (Figure 3). Substantial variability in patterns of $Q_e/Q$ were also observed among replicates for the inner flow collector of the jelly treatment. Soil Blocks 2, 4, and 10 exhibited maximum $Q_e/Q$ near the onset of flow (53.6 ± 12.4%), with values declining throughout the remainder of the rainfall simulation. The $Q_e/Q$ for Soil Blocks 6 and 7 initially decreased during the first 60 min of...
FIGURE 2  The $\delta^{18}$O and Mg$^{2+}$ for the inner and outer flow collectors during both first and second rainfall simulations. Black symbols with solid and dashed lines represent the mixing space defined by the three components used in hydrograph separation for the first and second rainfall simulation, respectively. Soil blocks in the spacer and jelly treatments are shown in the top and bottom row, respectively.

FIGURE 3  Flow rate and event water fraction of discharge ($Q_e/Q$) for inner and outer flow collectors during both the first (RF-1) and second (RF-2) simulations.
rainfall but subsequently increased prior to the end of rainfall (Figure 3). Once rainfall ceased, $Q_e/Q$ decreased again. Event water fractions of discharge for the outer collector of all soil blocks in the jelly treatment were initially elevated at the onset of flow (79.9 ± 22.6%) and decreased throughout the simulation (Figure 3).

Event water fraction of discharge varied significantly according to main effects of treatment ($p = .009$), flow collector ($p < .001$), and antecedent condition ($p = .031$) (Figure 4). The $Q_e/Q$ ratio was significantly greater for the spacer treatment (0.49 ± 0.11) compared with the jelly treatment (0.38 ± 0.20). The outer and inner flow collectors had $Q_e/Q$ values of 0.53 ± 0.16 and 0.34 ± 0.11, respectively, whereas dry antecedent conditions resulted in greater $Q_e/Q$ (0.48 ± 0.18) compared with wet antecedent conditions (0.39 ± 0.14). The $Q_e/Q$ ratio was also significantly different between inner (0.59 ± 0.14) and outer (0.40 ± 0.16) flow collectors ($p < .001$, Figure 4). Both treatment ($p = .031$) and antecedent condition ($p = .022$) significantly influenced the ratio of $Q_e/Q$ (Figure 4). The jelly and spacer treatments had $Q_u/Q$ of 0.10 ± 0.08 and 0.04 ± 0.09, respectively, whereas wet and dry antecedent conditions were 0.10 ± 0.10 and 0.04 ± 0.05.

The $Q_e/P$ ratio was significantly different between inner (0.17 ± 0.08) and outer (0.45 ± 0.34) flow collectors ($p < .001$) (Table 2). A significant interaction effect of treatment and antecedent condition and was also found for $Q_e/P$ ($p = .026$) (Table 2). The $Q_e/P$ ratio was greater for the spacer treatment and wet antecedent condition (0.52 ± 0.42) compared with the other combinations of treatment and antecedent condition (0.25 ± 0.17, 0.21 ± 0.13, and 0.25 ± 0.24). Both $Q_s/P$ and $Q_u/P$ were significantly different between wet and dry antecedent condition ($p = .002$ and $p = .016$, respectively) (Table 2). Wet antecedent conditions resulted in greater $Q_s/P$ and $Q_u/P$ (0.47 ± 0.33 and 0.08 ± 0.10, respectively) than dry antecedent conditions (0.21 ± 0.10 and 0.02 ± 0.02).

### 3.3 Surface-applied solutes

Leachate Br– concentration during the first simulation ranged from 108.9 to 775.1 mg L$^{-1}$ for the spacer treatment and from 12.4 to 403.9 mg L$^{-1}$ for the jelly treatment. Bromide concentration was negatively related to $Q_e/Q$ for both treatments and positively related to $Q_s/Q$. The $Q_e/Q$ ratio was significantly different between inner (0.17 ± 0.08) and outer (0.45 ± 0.34) flow collectors ($p < .001$) (Table 2). A significant interaction effect of treatment and antecedent condition and was also found for $Q_e/P$ ($p = .026$) (Table 2). The $Q_e/P$ ratio was greater for the spacer treatment and wet antecedent condition (0.52 ± 0.42) compared with the other combinations of treatment and antecedent condition (0.25 ± 0.17, 0.21 ± 0.13, and 0.25 ± 0.24). Both $Q_s/P$ and $Q_u/P$ were significantly different between wet and dry antecedent condition ($p = .002$ and $p = .016$, respectively) (Table 2). Wet antecedent conditions resulted in greater $Q_s/P$ and $Q_u/P$ (0.47 ± 0.33 and 0.08 ± 0.10, respectively) than dry antecedent conditions (0.21 ± 0.10 and 0.02 ± 0.02).
### Table 2

| Treatment | Block | Rainfall Simulation 1 (wet) | Rainfall Simulation 2 (dry) |
|-----------|-------|-----------------------------|-----------------------------|
|           |       | $Q_e/P$         | $Q_s/P$         | $Q_u/P$         | $Q_e/P$         | $Q_s/P$         | $Q_u/P$         |
|           |       | Inner  | Outer | Inner  | Outer | Inner  | Outer | Inner  | Outer | Inner  | Outer | Inner  | Outer | Inner  | Outer |
| Spacer    | 1     | 0.39   | 0.72  | 0.17   | 0.29  | 0.31   | 0.19  | 0.13   | 0.23  | 0.19   | 0.31  | 0.00   | 0.00  |
|           | 3     | 0.26   | 0.54  | 0.51   | 0.54  | 0.02   | 0.00  | 0.16   | 0.51  | 0.15   | 0.22  | 0.00   | 0.02  |
|           | 5     | 0.30   | 0.32  | 0.52   | 0.19  | 0.02   | 0.01  | 0.19   | 0.04  | 0.21   | 0.03  | 0.03   | 0.00  |
|           | 8     | 0.18   | 0.99  | 0.31   | 1.31  | 0.00   | 0.00  | 0.13   | 0.52  | 0.26   | 0.30  | 0.00   | 0.02  |
|           | 9     | 0.10   | 1.44  | 0.06   | 1.11  | 0.03   | 0.03  | 0.16   | 0.40  | 0.22   | 0.16  | 0.00   | 0.00  |
| Jelly     | 2     | 0.12   | 0.07  | 0.39   | 0.03  | 0.16   | 0.03  | 0.14   | 0.26  | 0.24   | 0.03  | 0.05   | 0.01  |
|           | 4     | 0.20   | 0.31  | 0.41   | 0.81  | 0.02   | 0.06  | 0.14   | 0.22  | 0.21   | 0.08  | 0.06   | 0.06  |
|           | 6     | 0.24   | 0.45  | 0.51   | 0.35  | 0.00   | 0.17  | 0.08   | 0.38  | 0.25   | 0.46  | 0.05   | 0.06  |
|           | 7     | 0.05   | 0.35  | 0.18   | 0.75  | 0.06   | 0.29  | 0.11   | 0.21  | 0.33   | 0.18  | 0.00   | 0.01  |
|           | 10    | 0.13   | 0.23  | 0.54   | 0.47  | 0.04   | 0.11  | 0.06   | 0.88  | 0.22   | 0.27  | 0.00   | 0.02  |

*Fraction of total precipitation that was leached during the same event.*

*How much pre-event soil water was mobilized by a unit volume of precipitation.*

*How much uncharacterized soil water was mobilized by a unit volume of precipitation.*

### Figure 5

Flow rate and bromide concentration for inner and outer flow collectors during both the first (RF-1) and second (RF-2) simulations. Chloride concentration also shown for RF-2.

(Figures 3, 5, and 6). As such, Br$^-$ concentration decreased with increasing flow rate for the inner and outer flow collectors of the spacer treatment and inner flow collector of the jelly treatment, with Br$^-$ concentration minima observed near peak flow (Figure 5). In contrast, Br$^-$ concentration tended to increase within increasing flow rate for the outer collector of the jelly treatment (Figure 5). The slope of the regression line was not significantly different between treatments; however, the difference in $y$ intercept was significant ($p < .001$; spacer: $y = -428.5x + 478.3$; jelly: $y = -357.8x + 307.1$), as Br$^-$ concentration observed for the spacer treatment was greater than Br$^-$ concentration for the jelly treatment (Figure 6).
FIGURE 6 Relationship between the event water fraction of discharge \( (Q_e/Q) \) and Br\(^{-} \) concentration for the first (RF-1) and second (RF-2) rainfall simulation (top and middle panels). Relationship between \( Q_e/Q \) and Cl\(^{-} \) concentration for the second rainfall simulation (bottom panel).

Bromide concentration during the second simulation tended to be greater than the first simulation for both spacer (14.6–1,032 mg L\(^{-1} \)) and jelly (8.0–662.6 mg L\(^{-1} \)) treatments. Bromide concentration was again negatively related to \( Q_e/Q \) for both treatments and positively related to \( Q_s/Q \) (Figures 3, 5, and 6). The slope of the regression line was significantly greater for the spacer treatment compared with the jelly treatment \( (p < .001; \text{spacer: } y = -982.2x + 939.7; \text{jelly: } y = -479.6x + 440.0) \) (Figure 5). Chloride, which was surface applied prior to the second simulation, also followed in similar temporal pattern in leachate, with Cl\(^{-} \) concentration negatively related to \( Q_e/Q \) for both treatments (Figures 3, 5, and 6). Leachate Cl\(^{-} \) concentration ranged from 4.0 to 302.3 mg L\(^{-1} \) and 1.6 to 255.4 mg L\(^{-1} \) for spacer and jelly treatments, respectively. Neither the slope nor y intercept was significantly different between treatments (spacer: \( y = -263.3x + 238.8 \); jelly: \( y = -196.7x + 202.9 \)) (Figure 5).

4 | DISCUSSION

4.1 | Sources of soil block leachate

Three-component hydrograph separation revealed that event water comprised between 21 and 59% of total leachate (inner flow collector + outer flow collector) through the soil blocks (Figure 7). Similar proportions of event water have been reported from plot- and field-scale studies with similar soil types across the U.S. Midwest. For instance, Vidon and Cuadra (2010) observed that event water represented between 11 and 50% of subsurface tile drain discharge from fields in Indiana. Leachate collected in pan lysimeters from 12 plots across northwestern Ohio during rainfall simulations was also composed of 6–46% event water (Williams, King, Duncan, Pease, & Penn, 2018). Excluding the potential for edge flow in the current study, preferential flows likely resulted from soil cracking, as annual tillage operations in the field where the soil blocks were collected largely precluded the formation of surface-connected biopores. It is widely recognized that agricultural tillage practices disrupt soil macropores created by earthworms compared with no-tillage (Kladivko, Akhouri, & Weesies, 1997). Indeed, in a subsequent dye tracer and dissection experiment using the same soil blocks as the current study (see Williams et al., 2019), earthworm burrows were observed at depth in several of the soil blocks, but they were often undyed because they were not directly connected to the soil surface.

Saturating the soil blocks from the bottom up prior to the rainfall simulations produced a nonuniform \( \delta^{18}O \) profile evidenced by measured leachate \( \delta^{18}O \) signatures lower than \( \delta^{18}O \) values in the suction-cup lysimeters and approaching the \( \delta^{18}O \) of the water used to saturate the soil blocks. This resulted in two sources of pre-event water in leachate, characterized (suction-cup lysimeter, \( Q_s \)) and uncharacterized (\( Q_u \)). Suction-cup lysimeters in the current study were installed within the silt loam surface soil layer (0–20 cm) overlying a silty clay subsoil (20+ cm). It is hypothesized that the uncharacterized soil water originated from this lower soil depth. Differences in soil texture
4.2 Effect of initial and boundary conditions on flow rates, water sources, and solute transport

4.2.1 Flow rates

Wet antecedent conditions resulted in greater $Q/P$ compared with dry antecedent conditions for both treatments (Figure 7). Precipitation on soils near field capacity mobilized more pre-event soil water (i.e., larger $Q_s/P$ and $Q_u/P$) compared with dry soils yielding larger discharge volumes. von Freyberg et al. (2018) also noted that antecedent conditions were strongly correlated to the amount of pre-event water mobilized at the small catchment scale. As expected, $Q/P$ did not vary between spacer and jelly treatments, as water was confined within the lysimeter casing and not allowed to runoff. Given the faster time to ponding and greater ponding depth for the jelly treatment during the first simulation, it is likely that leachate volume would have been substantially less compared with the spacer treatment if surface runoff would have occurred.

Although differences in ponding between spacer and jelly treatment suggest that the petroleum jelly provided edge-flow suppression, the outer flow collector had greater $Q/P$ and peak flow rate than the inner flow collector irrespective of treatment. This finding indicates that edge effects were present in both treatments. It should be noted, however, that differences in $Q/P$ and peak flow rate between inner and outer flow collectors were substantially greater in magnitude for the spacer treatment relative to the jelly treatment. For example, Peak$_{60}$ for the inner and outer flow collectors of the jelly treatment were, on average, less than or the same as Peak$_{60}$ for the inner...
flow collector of the spacer treatment, whereas Peak\textsubscript{60} for the outer flow collector of the spacer treatment was two to three times greater. Comparing unsealed lysimeters with lysimeters sealed with petroleum jelly, Cameron et al. (1990) also reported differences in soil hydraulic conductivity. The authors concluded that the petroleum jelly provided a water-tight seal and edge flow could be avoided, as they found nearly two times greater hydraulic conductivity rates for unsealed lysimeters compared with sealed lysimeters when a 2-cm constant head of water was applied to the soil surface. Findings from the current study suggest that sealing the gap between the lysimeter casing and the soil with petroleum jelly greatly constrained the magnitude of edge-flow effects but did not entirely prevent their occurrence.

### 4.2.2 Water sources

Event water fractions of discharge and precipitation (Q\textsubscript{e}/Q and Q\textsubscript{e}/P) were greater in the outer flow collector compared with the inner flow collector for both spacer and jelly treatments (Figure 7). This is consistent with greater Q/P and peak flow rates observed for the outer flow collector and provides additional evidence that edge effects were present in both treatments, but to a much lesser degree for the jelly treatment. Overall, event water comprised a greater fraction of discharge for the spacer treatment; however, Q\textsubscript{e}/P was dependent upon the interaction of treatment and antecedent condition. Although Q\textsubscript{e}/P was similar for the jelly treatment under both wet and dry antecedent conditions (0.21 and 0.25, respectively), Q\textsubscript{e}/P for the spacer treatment was substantially greater under wet (0.52) vs. dry (0.25) antecedent conditions. Combined with differences in temporal patterns of Q\textsubscript{e}/Q between wet and dry antecedent conditions, results support the hypothesis that soil cracking was the dominant preferential flow pathway through the soil blocks. During wet antecedent conditions, the unsealed edges for the spacer treatment acted as a preferential conduit of water, as no visible surface soil cracking was present on either treatment. The Q\textsubscript{e}/Q increased for both treatments during the rising limb of the hydrograph as the soil became saturated and ponding started to occur, but maximum Q\textsubscript{e}/Q was substantially greater for the spacer treatment. During the intervening time between simulations, soil cracks formed on the soil surface of all soil blocks and, as a result, Q\textsubscript{e}/P was similar between treatments during the second simulation. Declining Q\textsubscript{e}/Q on the rising limb for both treatments indicate that the soil cracks on the surface that initially transported event water closed as soil moisture increased, which resulted in decreased event water transport throughout the rainfall simulation.

Differences in Q\textsubscript{e}/Q and Q\textsubscript{e}/P further highlights the importance of antecedent condition on preferential flows. The Q\textsubscript{e}/Q was greater under dry conditions compared with wet conditions, but Q\textsubscript{e}/P was lower under dry conditions compared with wet conditions. Although there were fewer preferential flow paths (i.e., soil cracks) visible under wet antecedent conditions, Q\textsubscript{e}/P was greater than under dry antecedent conditions, but Q\textsubscript{e}/Q was lower. Ford, King, Williams, and Confesor (2017) also noted that although soil cracking was prominent during dry summer months, preferential flow was more likely to occur during wet winter months when these flow pathways were less likely to be visible. Even after soil cracks are visibly closed at the soil surface, preferential flow pathways may remain open and transport event water through the soil profile (Greve, Andersen, & Acworth, 2012). Thus, preferential flow generation in cracking soils requires a combination of pathway and adequate supply of source water (e.g., due to antecedent conditions or rainfall intensity).

The fraction of pre-event soil water from the upper soil layer (Q\textsubscript{u}/Q) did not vary based on initial or boundary conditions; however, the fraction of pre-event soil water from the lower soil layer (Q\textsubscript{u}/Q) was greater for the jelly treatment and under wet antecedent conditions (Figure 7). Generally, if event water comprised a larger fraction of discharge during the first simulation, then less pre-event soil water from the lower soil layer was mobilized (Q\textsubscript{u}/P). Similarly, under dry antecedent conditions, Q\textsubscript{u}/P decreased relative to wet antecedent conditions. Previous studies have shown that matrix–macropore interaction was a function of soil properties (Gerke & Köhne, 2002) and soil water content (Weiler & Naef, 2003). Findings from the current study therefore suggest that less pre-event water from the lower soil depth tended to be mobilized when preferential flow paths were present and that preferential flow interaction with the lower soil depth was likely minimal due a combination of high soil water content (especially under wet antecedent conditions) and low matrix permeability. Dye tracing studies in soil profiles with fine-textured soils have also observed low matrix–macropore interaction and bypassing of subsurface soil layers as preferential flow (Grant, Macrae, & Ali, 2019).

### 4.2.3 Solute transport

During both simulations, Br\textsuperscript{−} and Cl\textsuperscript{−} concentrations were negatively related to Q\textsubscript{e}/Q and positively related to Q\textsubscript{u}/Q. There were no observations of high solute concentration and a large fraction of event water. Under wet antecedent conditions, the highest Br\textsuperscript{−} concentration for both treatments was observed at the onset of flow. The coupled timing of Br\textsuperscript{−} delivery and source water indicates that
preferential transport of pre-event soil water from the upper soil layer was occurring. Previous studies have also observed that macropore flow was a mixture of both event and pre-event water (Klaus et al., 2013). The similarity in slope and difference in y intercept for the Br\(^{-}\) vs. \(Q_e/Q\) relationship between spacer and jelly treatments indicate that not only did the unsealed edges of the spacer treatment result in greater fractions of event water, but they also facilitated the preferential transport of pre-event water and associated solutes from the upper soil layer. It is likely that pre-event water from the upper soil layer entered preferential flow paths (including along the soil–casing interface) during the initialization of flow as the soil surface became saturated, with the fraction of pre-event water from the upper soil layer (and solute concentration) decreasing as more event water entered the flow pathways as the simulation progressed.

After soil block drying, there were no differences in Cl\(^{-}\) transport observed between spacer and jelly treatments during the second simulation, suggesting that the flow pathways and sources of water were similar relative to the first simulation (Figure 7). That is, soil cracks either in the middle of the soil block or along the soil–casing interface transported initially large fractions of event water at the beginning of the simulation, with pre-event water (and solute concentration) increasing on the falling limb of the hydrograph. Differences in the Br\(^{-}\) vs. \(Q_e/Q\) relationship for the second simulation between spacer and jelly treatments; however, highlights how differences in flow pathways and sources of water through the soil during the rainfall immediately after application can influence solute transport patterns in subsequent rainfall events. It is hypothesized that differences in flow pathways and sources of water during the first simulation between spacer and jelly treatments resulted in differences in the spatial distribution of Br\(^{-}\) throughout the soil, as patterns of Cl\(^{-}\) transport were similar during the second simulation between treatments. Destructive soil sampling after the first simulation, although not completed in the current study, could yield greater insight into how the surface-applied Br\(^{-}\) was redistributed and additional information regarding sources and flow pathways through the soil during the second simulation (Klaus et al., 2014).

5 CONCLUSIONS

In the current study, multiple tracers were used to contrast lysimeter initial and boundary conditions to provide new insights into preferential flows of water and surface-applied solutes. Unsealed lysimeters with initial soil water content near field capacity prior to the rainfall simulation represented the largest risk for edge flow for the soils studied. Since desiccation cracking was the primary preferential flow path through the lysimeters, water and solute transport did not vary considerably between sealed and unsealed lysimeters under dry antecedent conditions. Sequential rainfall simulations also showed that flow pathways present during the first rainfall after surface application can substantially influence solute transport during that rainfall plus subsequent rainfalls due to presumed differences in solute redistribution in the soil profile. The coupled timing of Br\(^{-}\) delivery and hydrograph separation indicated that preferential transport of pre-event soil water from the upper soil layer was occurring. That is, preferential flow pathways were transporting both event and pre-event water through the soil profile. When preferential flow pathways were present in the soil, the vast majority of pre-event water mobilized during the rainfall simulations was from the upper soil layer (0–15 cm), with little to no mobilization of pre-event water from deeper in the soil profile. This suggests that most water delivered to depth (e.g., subsurface tile drain depth) would be sourced from a combination of rainfall and soil water from near surface soil horizons, which has implications for nutrient and pesticide transport. Additional research examining pre-event water delivery in preferential flow pathways across soil textures is needed to better understand how and when this process occurs.

Unnatural preferential flow along the soil–lysimeter casing interface is one of the largest recognized limitations of lysimeter studies; thus, many methods have been used to suppress edge-flow effects. Results from the current study indicate that using petroleum jelly to seal the gap between the soil and lysimeter casing greatly constrains the magnitude of edge-flow effects but does not entirely prevent its occurrence. Sealing the edges of lysimeters may have differential effects on water and solute leaching rates depending upon the initial soil water content, and these effects on solute transport may persist during subsequent rainfall events. Further, previous work has shown that if soil blocks are allowed to dry for substantial periods of time, then the effectiveness of the petroleum jelly may greatly decrease (Williams et al., 2019).

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