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Letter

Significance of Hyporheic Exchange for Predicting Microplastic Fate in Rivers

Jennifer D. Drummond,* Holly A. Nel, Aaron I. Packman, and Stefan Krause

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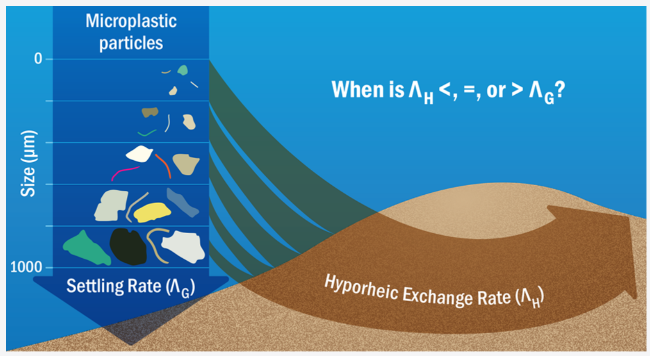
ABSTRACT: Microplastics are abundantly found in streambed sediments, including both small and low-density particles of neutral and positive buoyancy. Although the flow of water into streambed sediments (hyporheic exchange) has previously been shown to increase the rate of delivery of fine particles to the streambed, the influence of hyporheic exchange on microplastic fate in aquatic environments has not yet been assessed in detail. Here we evaluate the effects of hyporheic exchange on microplastics by calculating and comparing the rates of delivery of microplastics to streambed sediments by hyporheic exchange and gravitational settling for combinations of particle size and density most commonly found in streams. In a field stream study, we found that 23% of all microplastic combinations have a hyporheic exchange rate that is higher than their settling rate. This fraction was as high as 42% for microplastics composed of low-density polymers, such as polyethylene. We then expand these findings to consider a wide range of hydrodynamic conditions in rivers and demonstrate that hyporheic exchange is important for the transport and fate of particles that are <100 μm in diameter, irrespective of polymer type. Models that do not include hyporheic exchange are therefore likely to substantially underestimate the deposition, retention, and long-term accumulation of microplastics in streambed sediments.



■ INTRODUCTION

Recent research on plastics in lotic ecosystems shows that rivers and streams are not simply conduits for these materials to propagate to the marine environment. In fact, microplastics are ubiquitous in streambed sediments in all field studies to date.[1,2] In addition, it is now evident that microplastic fate and transport within streambed sediments can vary considerably in space and time.[3,4] A recent study by Frei et al.[5] showed that even the finest size class of microplastics (range of 20−50 μm) was abundantly found in streambed sediments with concentrations of ~30000 particles/kg of dry weight. The hyporheic zone, the region of the streambed porewater that exchanges with the open water column of rivers, is an important ecotone that facilitates nutrient turnover, provides a refuge for aquatic organisms, and retains fine particulate matter.[6−8] However, despite frequent findings that even low-density buoyant plastics accumulate in streambed sediments, hyporheic exchange processes have hardly been considered as mechanisms for microplastic accumulation in streambed environments to date.[5] Hyporheic exchange, the two-way movement between the overlying water and the streambed sediments, is driven by both turbulence in the near bed region and pressure variations at the streambed surface that force water and suspended particles (e.g., microplastics, fine sediments, and microbes) into and out of the sediment porewater.[7,9] In fact, the diverse channel features and in-stream processes that influence hyporheic exchange flow lead to a wide range of characteristic spatial scales and solute residence times (Figure S1), more fully described in the Supporting Information. Therefore, hyporheic exchange is occurring to some extent in most river systems, which leads to the accumulation of even very small particles with very low settling rates within the hyporheic region (Figure S2). However, the relative importance of hyporheic exchange compared to other mechanisms that lead to streambed retention of microplastics has not yet been assessed.

The delivery of microplastics to the streambed is normally calculated in terms of particle settling rates in the water column, which are dependent on plastic density, size, and shape.[10,11] Deposition leads to the retention of some proportion of microplastic inputs within rivers, instead of direct transport to the oceans.[12,13] Only a few studies have assessed the retention of smaller microplastics in rivers, with Nizzetto et al.[14] estimating that plastic particles that are <200

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μm in diameter and specific gravities (i.e., particle density/density of water) of <1 will be transported directly to the oceans with minimal interaction with riverbed sediments. However, particle settling varies in space and time based on factors such as microbial colonization,[15] homoaggregation,[16] and heteroaggregation with organic matter, suspended sediments, and phytoplankton aggregates.[17−19] Besseling et al.[12] showed that although biofouling and homoaggregation had a minimal effect on particle settling rates in streams, heteroaggregation increased the retention of microplastics with primary particle diameters down to 100 nm, ranging from 60% to 100% of plastic inputs. These estimates were used to predict large-scale transmission of microplastic loads from land to sea[20] but did not include hyporheic exchange.

Within previous models, microplastics are assumed to stay immobilized in the sediments under baseflow conditions, until the next stormflow event mobilizes streambed sediments and resuspends microplastics back into the water column. However, flow in the hyporheic zone transports particles through porewater even during baseflow conditions. Porewater flow causes both deeper penetration of particles, especially if aggregation occurs, into the streambed and reemergence of microplastics via hyporheic flowpaths that return to the water column.[21−24] Therefore, we hypothesize that current models[12,14,20] predicting microplastics in rivers that do not consider hyporheic exchange and porewater flow underestimate the rate of delivery of small and light (i.e., specific gravities of ∼1) particles into the streambed and overestimate particle retention time scales during baseflow.

The aim of this paper is to determine when hyporheic exchange influences microplastic fate in streams, which will help to improve future sampling and modeling strategies used to measure the delivery of microplastics from the continents to the oceans. Here we present a field study evaluating the effects of particle size, particle density, and river characteristics on microplastic deposition, retention, and remobilization. We identify combinations of plastic properties and hydrological parameters that make hyporheic exchange important to microplastic fate and transport and discuss the implications of short- and long-term retention within the hyporheic region on microplastic characteristics (e.g., size, density, and aggregation state) and time scales of delivery to the oceans.

■ MATERIALS AND METHODS

Microplastic Characteristics. Plastic polymers such as polypropylene (PP), polyethylene (PE), polyvinyl chloride (PVC), polyurethane (PU), polyethylene terephthalate (PET), and polystyrene (PS) are major global commodities,[25,26] and their unique polymer signature is detected throughout a wide range of freshwater environments.[27−29] Although still a current topic of discussion among researchers, on the basis of the recent study by Hartmann et al.,[25] microplastics are defined as particles ranging in size from 1 to 1000 μm, and the most common polymer types found in the environment have a density between 830 and 1580 kg/m^3.[1,30] While microplastics occur in a wide range of shapes (i.e., fragments, fibers, spheres, and films) in the environment, this study focuses on fragments and fibers because these are the most prominent types found in the natural environment.

For the purpose of the analysis presented here, we group polymers into five categories based on their densities:[16,30,31] (1) 830−980 kg/m^3 (PP and PE), (2) 1000−1100 kg/m^3 (PS and PU), (3) 1100−1300 kg/m^3 [polyamide (PA) also known as nylon, polycarbonate (PC), and acrylic], (4) 1300−1400 kg/m^3 [PVC, PET, and polybutylene terephthalate (PBT)], and (5) >1500 kg/m^3 [polyvinylidene fluoride (PVDF), polytetrafluoroethylene (PTFE), and plastics with a high percentage of fillers].

Quantification of Microplastic Gravitational Settling Velocity. The Stokes settling velocity (V_S) for the settling of particles is calculated as follows:[10,11]

$$V_S = \left(\frac{\rho_{tot} - \rho_w}{\rho_w} g w_* \nu \right)^{1/3} \tag{1}$$

where ρ_{tot} is the density of the plastic particle (kilograms per cubic meter), ρ_w is the density of water (1000 kg m^{-3}), g is the gravitational acceleration (9.81 m s^{-2}), and ν is the kinematic viscosity of water (1 × 10^{-6} m^2 s^{-1}). The dimensionless settling velocity w_* is calculated from the dimensionless particle diameter (D_*^2) as

$$w_* = 1.74 \times 10^{-4} D_*^2 D_* < 0.05 \tag{2}$$

where the dimensionless particle diameter is calculated from the equivalent spherical diameter D_n (meters):

$$D_* = \left(\frac{\rho_{tot} - \rho_w}{\rho_w} \right) g D_n^3 / \nu^2 \tag{3}$$

Although the Stokes settling velocity has been previously shown to underestimate fine particle and microplastic deposition in streams,[24,32] especially under turbulent conditions,[33] this value is still used within available freshwater microplastic models. We calculate V_S for 4000 combinations of microplastic size and density: 1000 sizes (D_n = 1, 2, 3, ..., 1000 μm) for four different densities, ρ_{tot}, of 1050, 1200, 1350, and 1580 kg/m^3, corresponding to categories 2−5 above, respectively. Because PP and PE have specific gravities of <1, these particles are buoyant, yielding a negative settling velocity. Therefore, V_S was not calculated for these plastics.

Quantification of Hyporheic Exchange. Solute tracer injections are commonly used to estimate stream transport parameters. In these experiments, a solute is injected upstream and then its passage is measured over time at a downstream location.[34] The tracer concentration versus time profile (breakthrough curve) is fit to a transport model that accounts for advection, dispersion, and short-term retention (storage) of the solute in the stream and hyporheic zone.[35,36] A full description of the models used in this study is presented in the Supporting Information. The rate of hyporheic exchange (Λ_H, inverse seconds) is a key transport parameter describing the exchange of the solute from the water column into slower moving regions, including porewater, estimated from model fitting to the breakthrough curve. The particle settling rate in the stream (Λ_G, inverse seconds) is calculated from the settling velocity (V_S) and stream depth (h):

$$\Lambda_G = V_S / h \tag{4}$$

In eq 4, the stream depth is used for normalization because the deposition process occurs over the streambed surface and h is the ratio between the volume of streamwater and the streambed surface area. We compare here the ratio of Λ_H to Λ_G (Λ_H/Λ_G), where larger Λ_H/Λ_G ratios indicate that hyporheic exchange dominates microplastic transfer from the water column to the riverbed.

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Hydrologic Conditions. We first assess the significance of hyporheic exchange with a field study, previously published to characterize fine particle and microbial transport in streams, that used a tracer injection of solute (Rhodamine WT) and fluorescent polymer microplastic particles (Dayglo Fluorescent AX Pigments, fragments with effective diameters ranging from 1 to 10 μm, average of ~4 μm, ρ = 1360 kg/m^3) in Toenepi Stream, Hamilton, Waikato, New Zealand.[37] A site description and additional experimental methods of this study are further described in the Supporting Information. Approximately 1.6 × 10^{13} particles were injected in this experiment. The microplastics were recently manufactured and were not subject to any biofouling, and the mixture was continuously agitated during the tracer addition to prevent aggregation. The average stream discharge was 35 L/s; the stream depth was ~21 cm, and the hyporheic exchange rate (Λ_H) was estimated to be 2.29×10^{-2} s^{-1} using a model fitting for solute and fine particle transport in streams.[38] To assess the relative effects of hyporheic exchange and particle settling in delivering microplastics to the streambed, we calculated Λ_H/Λ_G for each combination of microplastic density and size.

We extended the analysis of hyporheic particle transport by considering the range of streamflow conditions, water column depths, and hyporheic exchange rates found in 38 rivers worldwide.[39] We calculate Λ_H/Λ_G for each combination of microplastic density and size and six hydrologic scenarios: h = 1 and 10 m, and $\Lambda_H = 5 \times 10^{-6}$, 5×10^{-4}, and 5×10^{-3} s^{-1}. These results are first presented for all 24000 parameter combinations. Then, for the cases in which hyporheic exchange dominates microplastic deposition ($\Lambda_H/\Lambda_G > 1$), density and size classes are presented separately.

■ RESULTS AND DISCUSSION

Field Study Comparing Microplastic Gravitational Settling to Hyporheic Exchange Rates. For the Toenepi field study, the V_s of the injected particles is 3.15×10^{-6} m/s, corresponding to a gravitational settling rate Λ_G of 1.50×10^{-5} s^{-1} and a Λ_H/Λ_G of 1.5×10^3. Therefore, hyporheic exchange dominated transport of these microplastics to the streambed. Once delivered to the hyporheic zone, particles can deposit within the sediments or propagate through porewater to deeper depths or back to the water column. Excluding hyporheic exchange, and considering only gravitational settling, would have wrongly assumed that particles are transported directly downstream and do not interact with the streambed.

For the full range of microplastic size and density combinations with the hydrodynamic conditions in the Toenepi stream, we find that 23% have $\Lambda_H > \Lambda_G$ (Figure 1A, values above black dashed line indicate $\Lambda_H = \Lambda_G$). By density class (i.e., polymer type), the fraction with $\Lambda_H > \Lambda_G$ is 12% for PVDF, PTFE, and plastics with a high percentage of fillers (ρ = 1580 kg/m^3), 16% for PVC, PET, and PBT (ρ = 1350 kg/m^3), 21% for nylon, PC, and acrylic (ρ = 1200 kg/m^3), and 42% for PS and PU (ρ = 1050 kg/m^3) (Figure 1A, values above the black line $\Lambda_H = \Lambda_G$). In addition, for buoyant microplastics (density of <1000 kg/m^3) such as PP and PE, hyporheic exchange is the primary mechanism of particle transport to the riverbed as these particles float instead of settle.

Microplastic size is classified by D_n = 1−100, 100−300, 300−500, and 500−1000 μm. Within each particle size class, Λ_H/Λ_G is grouped by values of <1, 1−10, 10−100, or >100. All microplastics in the smallest particle size class with $D_n \leq 100$ μm have $\Lambda_H/\Lambda_G > 1$, indicating that hyporheic exchange is the dominant process delivering these microplastics to the riverbed (Figure 1B). In fact, for $D_n \leq 100$ μm, the fractions of combinations in this particle class with $\Lambda_H/\Lambda_G > 100$ and Λ_H/Λ_G = 10−100 are 21% and 42%, respectively (Figure 1B). For the larger particle size class with D_n = 100−300 μm, 49% of the combinations still have $\Lambda_H > \Lambda_G$ (Figure 1C).

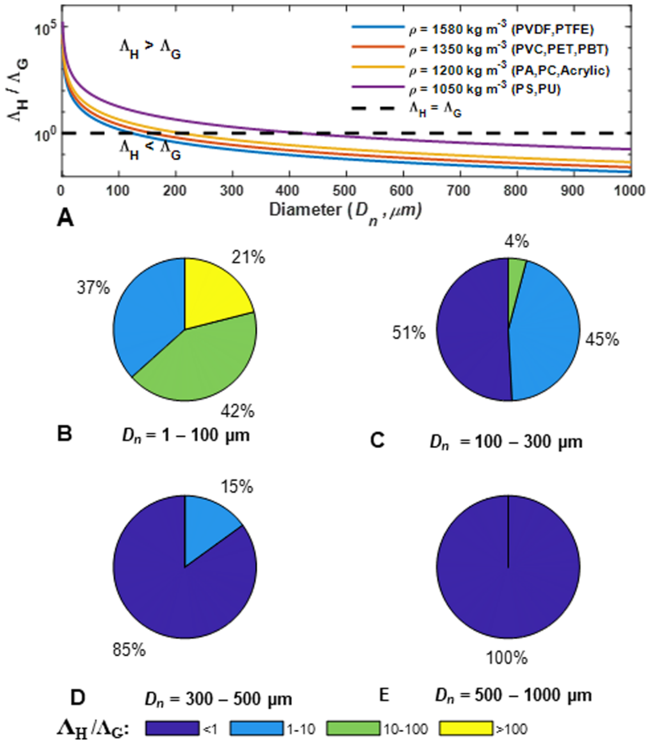

Figure 1. (A) Ratio of hyporheic exchange rate to settling rate (Λ_H/Λ_G) vs particle diameter (D_n = 1−1000 μm) for all microplastic combinations classified by density (i.e., polymer type). The black dashed line indicates $\Lambda_H = \Lambda_G$. (B−E) Microplastic combinations classified by particle size. Within each particle size class, Λ_H/Λ_G is grouped by ranges of values to show the relative importance of hyporheic exchange for each size group.

Relative Importance of Hyporheic Exchange for Different Microplastics with Varying Properties and Hydrologic Conditions. Particle settling velocity depends on only basic particle and fluid properties, while the relative importance of hyporheic exchange to microplastic deposition depends primarily on streamflow conditions (Figure 2). Hyporheic exchange is more important for particle accumulation in streams with greater hyporheic exchange rates (Λ_H increases from left to right in Figure 2A) and larger stream depths (h increases from the first to second row in Figure 2A). The gravitational settling rate decreases with an increase in stream depth, as Λ_G is calculated on the basis of the ratio between the volume of streamwater and the streambed surface area, or h. Depending on the stream specific channel features and in-stream processes, the extent of spatial and temporal scaling of hyporheic exchange will greatly vary (Figure S1) and therefore will influence most river systems. It is important to note that the Stokes law is not always accurate,[24,32] especially under turbulent conditions,[33] and therefore, even when $\Lambda_H \leq \Lambda_G$, hyporheic exchange may dominate the exchange and be a more accurate predictor of microplastic delivery to the streambed sediments.

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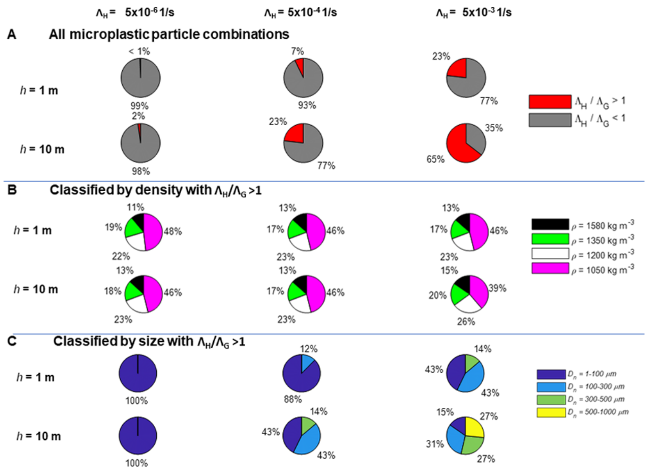

Figure 2. For each hydrologic scenario, 3 × Λ_H (columns) and 2 × h (rows) microplastic combinations of size and density are shown (A) with Λ_H/Λ_G either greater than or less than 1, (B) classified by density (i.e., polymer type) for only $\Lambda_H/\Lambda_G > 1$ (red in panel A), and (C) classified by particle size for only $\Lambda_H/\Lambda_G > 1$ (red in panel A).

Microplastic combinations with $\Lambda_H/\Lambda_G > 1$ are classified by density (i.e., polymer type) (Figure 2B) and by particle size (Figure 2C). When classified by density, the fractions are similar in all scenarios (Figure 2B). When classified by particle size and $\Lambda_H = 5 \times 10^{-6}$ s^{-1}, only the smallest particle class with D_n = 1−100 μm has $\Lambda_H > \Lambda_G$ (Figure 2C). However, as h and Λ_H increase, there is a wider range of particle size classes observed with $\Lambda_H/\Lambda_G > 1$, including the largest particle size class (D_n = 500−1000 μm) (Figure 2C, second and third columns).

Remobilization to the Water Column during Baseflow Conditions. Not all of the microplastics that are delivered to the riverbed by hyporheic exchange are retained. Instead, particles can be transported through porewaters back to the water column under baseflow conditions. For example, in the Toenepi experiment, 57% of the particles were retained for a much longer period of time and remobilized only in response to a stormflow event.[37,38] However, the remaining 43% of particles were retained within the 130 m reach for a relatively short period of time (from minutes to 15 days), indicating remobilization to the water column under baseflow conditions. The downstream delay of these particles was a result of processes such as the slow transport through the hyporheic zone, reversible filtration to sediments, or loose and reversible attachment to submerged vegetation and/or streambed biofilms.[38] Microplastics larger than the pore size of the sediment will be trapped at the sediment−water interface.[40] However, microplastics larger and denser than used in the Toenepi experiment (i.e., particles for which $\Lambda_H/\Lambda_G \leq 1$) are also resuspended during baseflow conditions.[41−43] Without considering baseflow remobilization, microplastic retention may be overestimated and microplastic fate will be inaccurately represented in rivers.

Discussion about Possible Alterations of Microplastic Properties within the Hyporheic Zone. Understanding time scales of microplastic retention in the hyporheic zone is crucial as the passage provides time for alteration of plastics by fragmentation, biofouling, and heteroaggregation (i.e., attachment to fine sediments or bacteria). Hence, knowledge of the duration and size fractions of particles retained in the hyporheic zone improves our understanding of microplastic fate in aquatic environments. For instance, heteroaggregation and other alterations within the hyporheic zone may change the plastic size and density and therefore the settling rates,[12,17] which could potentially increase the level of immobilization within streambed sediments during downstream transport prior to reaching the ocean. Fragmentation and weathering (i.e., surface striations, cracks, pits, and embedded particles) have been observed in microplastics collected from the freshwater environment,[44,45] but it is unclear how this may alter their subsequent fate and transport. Furthermore, polymers may have large proportions of additives or fillers that can leach out of the plastics,[46] changing the density and surface chemistry, which will subsequently alter their settling rates.[47]

The knowledge gained from this study on plastic size fractionation and preferential accumulation in hyporheic sediments is critical for improved predictions of microplastic fate because hyporheic alterations will preferentially affect smaller sizes because of their higher surface area/volume ratios. Hyporheic abrasion or fragmentation of particles that increases surface area may lead to enhanced colonization/attachment of microbes, which could facilitate biodegradation or serve as a vector for pathogenic bacteria or viruses.[48] Heteroaggregation with phytoplankton can alter particle buoyancy,[18,19] with species that produce more extracellular polymeric substances forming larger aggregates. Fractal aggregates with macropores through which water can easily flow can reduce drag and also promote particle settling.[18,19] These alterations that are facilitated preferentially in hyporheic environments may increase particle bioavailability to sediment-dwelling organisms, which have been shown to both ingest particles[4,49] and incorporate them into physical structures.[50−52] Toxicology studies of freshwater chironomid larvae suggest a size-dependent effect of microplastics on survival, growth, and emergency, with the smallest size class of particular concern.[53,54] However, if hyporheic exchange is not considered as a mechanism for the delivery of particles to the streambed, then these particle alterations and biological uptake by invertebrates will also not be considered, which will be especially important for the smallest size class with D_n = 1−100 μm.

Implications. The results presented in this study indicate the following. (1) Hyporheic exchange is important to the retention of microplastics that are <100 μm in diameter and low-density particles of all sizes. Model simulations of microplastic retention will be substantially underestimated by models if hyporheic exchange is not considered. (2) Hyporheic exchange increases overall travel times and retention times of microplastics in rivers but does not always produce long-term retention because of continuous remobilization of particles from riverbeds. As a result, particle retention time scales may be overestimated if remobilization under baseflow conditions is not considered. (3) Overall, hyporheic exchange is expected to substantially increase the retention of microplastics in rivers and facilitate modification of these particles prior to reaching the oceans. Hyporheic exchange facilitates alteration of plastics through the combination of increased retention and riverbed processes such as fragmentation, biofouling, and aggregation.

Future microplastic monitoring programs should sample streambed sediments and quantify in particular smaller microplastic size fractions (D_n = 1−100 μm) to improve our understanding of microplastic fate in stream ecosystems. Furthermore, models that incorporate hyporheic processes

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are needed to appropriately characterize residence times of microplastics in freshwater systems and close the global plastic budget. Finally, because streambed sediments represent accumulation zones where habitats may experience critical exposures, further studies of the impact of microplastic on the ecological function of hyporheic zones are needed.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.0c00595.

Overview of hyporheic exchange processes and importance to microplastic fate in streams, experimental methods for the tracer experiment in Toenepi stream, and details of in-stream solute transport and transient storage models for determining the hyporheic exchange rate in streams (PDF)

AUTHOR INFORMATION

Corresponding Author

Jennifer D. Drummond − *School of Geography, Earth and Environmental Sciences, University of Birmingham, Birmingham B15 2TT, U.K.;* orcid.org/0000-0002-6501-7618; Email: j.drummond@bham.ac.uk

Authors

Holly A. Nel − *School of Geography, Earth and Environmental Sciences, University of Birmingham, Birmingham B15 2TT, U.K.;* orcid.org/0000-0002-0571-2678

Aaron I. Packman − *Department of Civil and Environmental Engineering, Northwestern University, Evanston, Illinois 60208, United States*

Stefan Krause − *School of Geography, Earth and Environmental Sciences, University of Birmingham, Birmingham B15 2TT, U.K.*

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.estlett.0c00595

Notes

The authors declare no competing financial interest.

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