



Assessing the sorption and leaching behaviour of three sulfonamides in pasture soils through batch and column studies



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HIGHLIGHTS

- Sulfonamides exhibited a mobility pattern similar to that of conservative tracer.
- Considerable retardation was observed for the Hamilton soil.
- No resident concentration of sulfamethazine was detected at any soil depth.
- Model estimated and the experimental retardation factors deferred.

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ABSTRACT

We investigated the sorption potential and transport behaviour of three sulfonamides, namely, sulfamethoxazole (SMO), sulfachloropyridazine (SCP) and sulfamethazine (SM), and a conservative bromide tracer (Br^-) in two undisturbed soil columns collected from the dairy farming regions in the North Island of New Zealand. Based on the low $\log K_{oc}$ values obtained from the sorption study, all three sulfonamides are likely to have high mobility, making them a potential threat to surface and ground water. Soil column studies also showed that the mobility of the sulfonamides varied among soils and antibiotic type. Sulfonamides exhibited a mobility pattern similar to that of conservative Br^- tracer. Considerable retardation was observed for the Hamilton soil, and the delayed peak arrival time (or maxima) was due to the role of sorption-related retention processes under saturated flow conditions. Residual antibiotic concentrations for SMO and SCP were detected in all soil sections including at 18 cm depth, while no resident concentration of SM was detected at any depth in the entire length of the core for both soils. The deterministic, physical equilibrium model (CXTFIT) described the peak arrival time as well as the maximum concentration of the antibiotic breakthrough curves reasonably, but showed some underestimation at the advanced stages of the leaching process. There was a significant difference in the model estimated retardation factors obtained from column study and the experimental retardation factors obtained from the conventional batch sorption experiments.

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1. Introduction

Sulfonamides are one of the most commonly used veterinary antibiotics for therapeutic and sub-therapeutic purposes (Sukul and Spittler, 2006). According to the New Zealand Food Safety Authority, the sulfonamide group contributes ~10% of total antibiotic usage (Srinivasan et al., 2014), and is a common class of antibiotics widely used in livestock industries in New Zealand (NZ). Land-application of animal waste effluent is a permitted activity in NZ as long as the farmers follow the prescribed conditions set out by the respective regional councils (Sarmah et al., 2006). This, along with the direct excretal inputs from the grazing

animals, constitutes a direct source of exposure for antibiotics in the pasture environment and makes the assessment of its retardation and mobility in soil environment necessary to establish their risk to humans and environment. Recent fate studies conducted on sulfonamides in pasture soils collected from dairy farming region of NZ suggest that sulfonamides have a medium to high potential to leach through the soil profile and be transported to the groundwater, drainage water, and by surface runoff to surface waters (Srinivasan et al., 2014).

Widespread occurrence of sulfonamides in aquatic and terrestrial environments has been reported in a number studies (Batt et al., 2006; Garcia-Galan et al., 2011; Kim et al., 2011). For example sulfamethoxazole antibiotic, which belongs to this group, was detected at a concentration of up to $0.48 \mu\text{g L}^{-1}$ in surface water samples in Germany (Hirsch et al., 1999). SMO was also detected in groundwater at a concentration of $0.22 \mu\text{g L}^{-1}$ (Lindsey et al., 2001) and in monitoring well samples (Barber et al., 2009), and streams (Kolpin et al., 2002) in the USA. These

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studies show that there is a possibility that these chemicals may be encountered in drinking water supply.

The transport of veterinary antibiotics in soils depends on several factors such as the chemical properties of the antibiotics, temperature, moisture content, ionic strength (Chen et al., 2011), soil physico-chemical properties (Strauss et al., 2011), soil charge density, and contact time (Kurwadkar et al., 2011). Chemical properties such as water solubility, dissociation constants, sorption–desorption processes, stability, binding to the soils, and the partitioning coefficients at various pH values can all affect the mobility of antibiotics in the soil environment (Kurwadkar et al., 2011; Tolls, 2001; Unold et al., 2010). Other factors that can influence the mobility of veterinary antibiotics are the timing of manure application, prevailing weather conditions, preferential flow via desiccation cracks and worm channels as recently demonstrated in a UK field study (Kay et al., 2004; 2005a). In addition to sorption, the fate and transport of sulfonamides in subsurface environments can also be governed by other abiotic reduction due to the presence of reduced sulfur compounds (e.g., bisulfide and polysulfides) in soil/sediment pore waters (Zeng et al., 2011).

Results obtained from overseas studies cannot be extrapolated to reflect the behaviour of the antibiotics in New Zealand conditions. This is primarily due to differences in pedo-climatic conditions, rainfall pattern, rainfall intensity and other temporal and spatial variability of soils including carbon content, mineralogical characteristics (such as clay and silica content) or soil pH. The predominance of volcanically-derived soils with a high percentage of allophonic clay mineral having high surface area, and high organic carbon content is known to strongly influence the fate of other organic contaminants such as pesticides (Sarmah et al., 2005) and veterinary antibiotics (Srinivasan et al., 2014). However, to date there is no published information available on the transport behaviour of sulfonamides in NZ soils.

The main objective of this study was to investigate the transport behaviour of three sulfonamides, namely SMO, SCP and SM in two different undisturbed soil cores under saturated conditions. In an earlier study it was observed that the mobility of mixed sulfonamides was not impacted by competitive sorption (Kurwadkar et al., 2011), which meant that the mobility of all the three sulfonamides could be evaluated simultaneously relative to a conservative Br⁻ tracer. Sulfonamide sorption affinity onto these two soils was also investigated through a series of batch sorption experiments by extracting the residues from solid phase using a solvent extracted scheme. In order to test the applicability of the column test to other polar contaminants, results from the batch and column studies were compared against each other.

2. Materials and methods

2.1. Chemicals

SMO, SCP, and SM (Table 1) of >99% purity, and calcium chloride dihydrate (CaCl₂·2H₂O > 99% purity) were obtained from Sigma Aldrich, Australia. Acetonitrile (Mallinckrodt ChromAR, ≥99.8% purity) and

dichloromethane (Mallinckrodt UltimAR, ≥99.9% purity) were obtained from Thermo Fischer Scientific Ltd. New Zealand. HPLC grade deionized water was obtained from an Arrium® 61316 high performance reverse osmosis system (Sartorius Stedim Biotech GmbH, Germany). Nitrogen gas oxygen free (gas code 152) was purchased from BOC (New Zealand).

2.2. Soils

Two top soils (0–5 cm) from the North Island of New Zealand, Hamilton clay loam from the Waikato dairy farming regions, and Matawhero silt loam from the Gisborne region were freshly collected, air-dried to room temperature and sieved (<2 mm). Some important physico-chemical properties of the soils used in this study are summarized in Table 2. Detailed physico-chemical properties of these soils can be found in an earlier study (Srinivasan et al., 2014).

2.3. Batch sorption studies

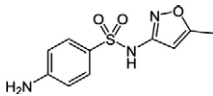
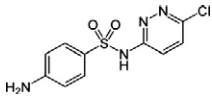
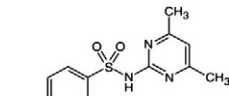
A stock solution of SMO, SCP and SM antibiotic each at a concentration of 1000 mg L⁻¹ was prepared in methanol. By adding an appropriate amount of the stock solution separately to 5 mM CaCl₂ solution, six different initial aqueous solution concentrations were prepared in duplicate. Batch studies were performed using a similar experimental protocol earlier adopted by Sarmah et al. (2008). Briefly, duplicate samples of air-dried soils (2 g) were weighed into glass centrifuge tubes (35 mL) with Teflon-lined screw caps. Aliquots (30 mL) of six concentrations of each sulfonamides (1.5, 3, 5, 7.5, 10 & 15 mg L⁻¹) were added to the respective tubes, wrapped in aluminum foil, placed in the dark and shaken in an end-over-end shaker for 24 h. The solution pH for the batch sorption studies ranged from 5.22 to 5.41 for Matawhero and from 5.60 to 6.03 for Hamilton soil. Preliminary investigation carried out on the sorption kinetics showed that an apparent equilibrium time of 24 h was sufficient for the batch sorption studies performed (Srinivasan et al., 2014).

2.4. Preparation of the soil lysimeters

The soil lysimeters used in this study were made of polyvinylchloride (PVC) with a length of 20 cm and an internal diameter of 7.5 cm. Undisturbed soil cores of Matawhero and Hamilton soil were obtained in situ by hand carving from the ground surface. The internal cylindrical surface of the lysimeter casings was coated with petroleum jelly and the casings were slowly pushed into the soil under pressure. When the desired length of core was reached (18 cm) the lysimeters were carefully cut off from the bottom with a sharp metal blade.

Immediately after excavation, both lysimeters were covered with polythene sheet and PVC end caps were attached to prevent water loss. The lysimeters were transported to the laboratory and the bottom of the cores was cleaned and secured with a fly screen (ø 1 mm) to allow leachate percolation, and sealed with a PVC cap containing a number of holes that acted as sampling ports for leachate collection. On the outer

Table 1
Selected chemical properties of the antibiotics used in the study.

Properties	SMO	SCP	SM
Molecular formula	C ₁₀ H ₁₁ N ₃ O ₃ S	C ₁₀ H ₉ N ₄ ClO ₂ S	C ₁₂ H ₁₄ N ₄ O ₂ S
Molecular weight (g mol ⁻¹)	253.28	284.72	278.3
Log K _{ow}	0.89 ^a	0.31 ^b	0.89 ^a
pK _{a1} /pK _{a2} at 25 °C	1.83 ^a /5.57 ^a	1.87/5.5 ^b	2.28 ^a /7.42 ^a
Structure			

^a Figueroa-Diva et al. (2010).

^b Srinivasan et al. (2014).

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