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Q1 Potential for natural and enhanced attenuation of 2 sulphanilamide in a contaminated chalk aquifer

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A B S T R A C T

Antibiotic compounds in the environment are of concern as they are biocidal and have the potential to drive the development of antibiotic resistance in microbes. Understanding antibiotic biodegradation is important to the appreciation of their fate and removal from the environment. In this research an Isotope Ratio Mass Spectrometry (IRMS) method was developed to evaluate the extent of biodegradation of the antibiotic, sulphanilamide, in contaminated groundwater. Results indicated an enrichment in $\delta^{13}\text{C}$ of 8.44‰ from -26.56 (at the contaminant source) to -18.12‰ (300 m downfield of the source). These results confirm reductions in sulphanilamide concentrations (from 650 to 10 mg/L) across the contaminant plume to be attributable to biodegradation (56%) vs. other natural attenuation processes, such as dilution or dispersion (42%). To understand the controls on sulphanilamide degradation *ex-situ* microcosms assessed the influence of sulphanilamide concentration, redox conditions and an alternative carbon source. Results indicated, high levels of anaerobic capacity (~50% sulphanilamide mineralisation) to degrade sulphanilamide under high (263 mg/L), moderate (10 mg/L) and low (0.02 mg/L) substrate concentrations. The addition of electron acceptors; nitrate and sulphate, did not significantly enhance the capacity of the groundwater to anaerobically biodegrade sulphanilamide. Interestingly, where alternative carbon sources were present, the addition of nitrate and sulphate inhibited sulphanilamide biodegradation. These results suggest, under *in-situ* conditions, when a preferential carbon source was available for biodegradation, sulphanilamide could be acting as a nitrogen and/or sulphur source. These findings are important as they highlight sulphanilamide being used as a carbon and a putative nitrogen and sulphur source, under prevailing iron reducing conditions present in the aquifer.

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Q5 Introduction

55 Advances in analytical techniques have highlighted emerging
 56 organic contaminants, such as pharmaceutical and personal care
 57 products, in multiple environmental media (Lapworth et al., 2012;

Pal et al., 2010). One particular concern is the occurrence of antibiotic compounds; as these have the potential to interact with microorganisms in the environment and through this interaction to perpetuate the development of antibiotic resistance (Kümmerer, 2009a). Thus, there is concern that increasing

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63 levels of antibiotics, found in the environment, may promote
64 antibiotic resistance in microbes and potentially render anti-
65 biotics ineffective in treating human and veterinary infections
66 (Kümmerer, 2009a). It has been reported that between 100,000
67 and 200, 000 tons of antibiotics are used worldwide each year;
68 with approximately 50% used for human consumption and the
69 remainder for animals, agriculture and aquaculture (Kümmerer,
70 2009b). Due to their extensive use, these compounds are readily
71 released into the environment, from sources such as: wastewater
72 treatment plants; hospital effluents; livestock activities and
73 manure application to soil, and; indirectly through ground and
74 surface water exchange (Lapworth et al., 2012; Michael et al., 2013;
75 Rizzo et al., 2013). Antibiotics and antibiotic resistant genes have
76 been detected in wastewater discharges and have been reported
77 to persist in wastewater following its treatment (Michael et al.,
78 2013). Indeed, the biological processes employed at wastewater
79 treatment plants have been reported to promote the develop-
80 ment and transfer of antibiotic resistant genes (Michael et al.,
81 2013; Larcher and Yargeau, 2012).

82 Of the reported persistent pharmaceutical products,
83 sulphonamides are widely detected in groundwater across
84 Europe (Lapworth et al., 2012), the United States of America
85 (Barnes et al., 2008) and China (Sui et al., 2015). Since the 1930s,
86 over 5000 sulphonamide compounds, (all derivatives of
87 sulphanilamide) have been developed, with approximately 100
88 used as antibiotics (Holm et al., 1995). The sulphonamide class
89 of antibiotics can inhibit gram-positive and gram-negative
90 bacteria, as well as protozoa and as a consequence are among
91 the most frequently used antibiotics for human, veterinary and
92 agriculture purposes (Larcher and Yargeau, 2012; Brown, 1962;
93 Liao et al., 2016). It is estimated that 9.3 million kg of anti-
94 microbials are used annually in the USA, with 70% used in animal
95 feed as growth promoters. In the UK, sulphonamides are the
96 second most commonly used veterinary antibiotic, making up
97 21% of the annual consumption (448,000 kg) of antibiotics in the
98 UK (Sarmah et al., 2006).

99 Despite the anti-microbial properties of sulphonamides,
100 studies suggest that microbial communities can adapt to their
101 presence; with microbes developing resistance to the antibiotic
102 becoming more dominant and evolving to have the capacity to
103 degrade antibiotics (Collado et al., 2013; Herzog et al., 2013).
104 Early work by Walker (1978) and Balba et al. (1979) demonstrat-
105 ed sulphanilamide to be biodegradable. Sulphonamides have
106 since been reported to degrade under both aerobic (Larcher and
107 Yargeau, 2012; Liao et al., 2016; Collado et al., 2013; Herzog et al.,
108 2013; Drillia et al., 2005; Müller et al., 2013; Reis et al., 2014;
109 van Haperen et al., 2001) and anaerobic (Carballa et al., 2007; Lin
110 and Gan, 2011; Mohring et al., 2009) conditions, and, in both soil
111 and sediment environments (Walker, 1978; Baumgarten et al.,
112 2011). Interestingly, microbes have been reported to utilise
113 sulphonamides as a source of carbon, nitrogen and/or sulphur,
114 depending on the nutrient and environmental conditions they
115 are exposed to (Herzog et al., 2013; Drillia et al., 2005; Müller
116 et al., 2013; Reis et al., 2014; van Haperen et al., 2001).

117 However, there are limited accounts of sulphanilamide
118 biodegradation in groundwater environments and, information
119 regarding their fate and degradation, as controlled by their
120 concentration and prevailing redox conditions, is very limited.
121 Thus, new insight is needed regarding how these controlling
122 factors influence sulphonamide degradation. In addition, if we

are to engineer solutions to mitigate elevated concentrations of
sulphonamides in the environment, then we need a better
understanding of how manipulation of electron acceptors in
groundwater might influence sulphonamide degradation.

The purpose of this research was to investigate the influence
of sulphonamide concentration and redox conditions on the
sulphonamide biodegradation. Significantly, our research
focused on sulphonamide biodegradation in a contaminated
chalk aquifer located below an industrial facility. The site
is unique in that it has been exposed, following chemical
release in 1970, to extreme concentrations of sulphonamide
(sulphanilamide ≤ 650 mg/L). These concentrations are unprec-
edented in the environment; by way of context wastewater
antibiotic concentrations are typically ≤ 0.001 mg/L (Michael
et al., 2013). This location provided sampling transects that
enabled the following controls on sulphanilamide degradation
to be evaluated: i) the interplay of sulphonamide concentration
and redox condition, and ii) the interplay of sulphonamide
concentration, redox condition and the co-presence of alterna-
tive carbon sources (specifically toluene). An Isotope Ratio Mass
Spectrometry (IRMS) method was developed to evidence carbon
isotope fractionation during in-situ sulphanilamide biodegrada-
tion. To assess the potential to enhance sulphonamide degra-
dation, ex-situ microcosms were supplemented with electron
acceptors (sulphur and nitrogen) to evaluate their influence on
sulphonamide degradation.

1. Material and methods

1.1. Site description

This research considered a chalk aquifer situated beneath a
chemical plant in the United Kingdom. The groundwater within
the aquifer contained high levels of sulphanilamide (≤ 650 mg/L)
and toluene (≤ 275 mg/L) (Fig. 1). Partial degradation of these
organic compounds has exhausted dissolved oxygen in the
aquifer and has given rise to anaerobic conditions, dominated
by Fe(III)- reduction (Eh values, reported in 69 sampled boreholes
across the site, from 270 to -50 mV) (SI Fig. 1) and sulphate-
reduction (Eh values, reported in 8 sampled boreholes, from 70
to -130 mV) (SI Fig. 1). Sulphanilamide is mobile within the
aquifer and its movement has resulted in the development of a
solute plume that extends approximately 300 m down gradient
from the source zone (Figs. 1 and 2), 10–18 mbs (meters below
surface). Across the plume sulphanilamide concentrations
range from 1 to 133 mg/L, with movement of the plume
being estimated at ≤ 0.01 m/d (Fig. 1). Beneath the “toluene
works” (Fig. 1), there exists a toluene plume, approximately
190 m long, at 8–12 mbs, with concentrations ranging from 7
to 275 mg/L. The direction of groundwater flow at the site is
from a north to south-westerly direction (Figs. 1 and 2). Thus
the sulphanilamide plume and toluene plume converge at
approximately 140 m down-gradient of the sulphanilamide
source zone (Fig. 1), where an area of mixing exists.

1.2. Chemicals

A radiolabelled analogue of sulphanilamide [ring- ^{14}C (U)]
was obtained from American Radiolabelled Chemicals Inc.,

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