



Sulfadimethoxine and sulfaguanidine: Their sorption potential on natural soils

Anna Białk-Bielińska^{a,*}, Joanna Maszkowska^a, Wojciech Mroziak^b, Agata Bielawska^a, Marta Kołodziejka^a, Richard Palavinkas^c, Piotr Stepnowski^a, Jolanta Kumirska^a

^a Department of Environmental Analysis, Faculty of Chemistry, University of Gdańsk, ul. Sobieskiego 18, 80-952 Gdańsk, Poland

^b Medical University of Gdańsk, Department of Inorganic Chemistry, Al. Gen. J. Hallera 107, 80-416 Gdańsk, Poland

^c Federal Institute for Risk Assessment BfR, Thielallee 88-92, D-14195 Berlin, Germany

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ABSTRACT

Sulfonamides (SAs) are one of the oldest groups of veterinary chemotherapeutic agents. As these compounds are not completely metabolized in animals, a high proportion of the native form is excreted in feces and urine. They are therefore released either directly to the environment in aquacultures and by grazing animals, or indirectly during the application of manure or slurry. Once released into the environment, SAs become distributed among various environmental compartments and may be transported to surface or ground waters. The physicochemical properties of SAs, dosage and nature of the matrix are the factors mainly responsible for their distribution in the natural environment. Although these rather polar compounds have been in use for over half a century, knowledge of their fate and behavior in soil ecosystems is still limited. Therefore, in this work we have determined the sorption potential of sulfadimethoxine and sulfaguanidine on various natural soils. The influence on sorption of external factors, such as ionic strength and pH, were also determined. The sorption coefficients (K_d) obtained for the sulfonamides investigated were quite low (from 0.20 to 381.17 mL g⁻¹ for sulfadimethoxine and from 0.39 to 35.09 mL g⁻¹ for sulfaguanidine), which indicated that these substances are highly mobile and have the potential to run off into surface waters and/or infiltrate ground water. Moreover, the sorption of these pharmaceuticals was found to be influenced by OC, soil solution pH and ionic strength, with higher K_d values for soils of higher OC and lower K_d values with increasing pH and ionic strength.

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1. Introduction

Sulfonamides are among the most commonly consumed veterinary antibiotics in the EU (García-Galán et al., 2009): they are used in agriculture to prevent disease and to treat illness in livestock. Following their administration, substantial quantities may be excreted as the parent compounds and/or metabolites and leak into the environment while animals are grazing or during the spreading of manure. Moreover, during the storage of manure, the excreted acetyl conjugates can be cleaved back to the parent compound (Sukul and Spiteller, 2006). Therefore, once released into the environment, SAs are distributed among different environmental compartments and can be transported to surface and ground waters. They can also enter the food chain and impact on natural environment and human health. The physicochemical properties, applied dosage and the nature of the environmental components with which they interact govern the whole process. However, knowledge of the behavior and fate of these rather polar pollutants in soil ecosystems is still limited.

Previous studies revealed that these compounds are present in soils and manure in concentrations of up to 15 µg kg⁻¹ of soil and 20 mg kg⁻¹ of liquid manure (Sukul and Spiteller, 2006). As soil is the most exposed to pollution by these substances, the degree to which they disperse in this matrix needs to be assessed. It is also important to determine the mobility of these substances in soils with different physicochemical properties in order to gain a better understanding of the general risk accompanying the entry of sulfonamides into surface and ground waters, and thus to assess the scale of threats to the environment and human health. Such studies are necessary as far as the ecotoxicological potential of sulfonamides is concerned. Our recent studies have shown that these compounds can pose a real risk to aquatic organisms, especially to higher plants like duckweed *Lemna minor* and algae (Białk-Bielińska et al., 2011). It is therefore of the utmost importance to evaluate their sorption potential and to determine their behavior and fate in the soil environment.

Although SAs have been the subject of many investigations, to date only a few studies have focused on the behavior and fate in soils of these rather polar pharmaceuticals. Moreover, most studies have dealt with sulfadiazine (Thiele-Bruhn et al., 2004; Burkhardt et al., 2005; Stoob et al., 2007; Sukul et al., 2008; Schauss et al.,

* Corresponding author. Tel.: +48 58 5235448.

E-mail address: abialk@chem.univ.gda.pl (A. Białk-Bielińska).

2009; Unold et al., 2009), sulfamethazine (Thiele-Bruhn et al., 2004; Burkhardt et al., 2005; Gao and Pedersen, 2005; Kurwadkar et al., 2007; Stoob et al., 2007; Lertpaitoonpan et al., 2009; Figueroa-Diva et al., 2010), sulfathiazole (Burkhardt et al., 2005; Kahle and Stamm, 2007a,b; Kurwadkar et al., 2007; Stoob et al., 2007) and sulfachloropyridazine (Boxall et al., 2002; ter Laak et al., 2006). Sorption studies have shown that all of the investigated sulfonamides are characterized by a low sorption potential and are adsorbed less in soil containing manure. Even so, the environmental fate of SAs is still difficult to predict since field and plot studies have yielded an inconsistent picture of SA mobility, which depends on soil and experimental conditions (Boxall et al., 2002; Burkhardt et al., 2005; Kahle and Stamm, 2007a,b; Stoob et al., 2007). Therefore, identification of the factors affecting sorption is essential for a reliable assessment of SA mobility, and hence, their bioavailability. Knowledge of SA sorption remains limited, but existing data suggest that these compounds behave in a complex manner.

Widely used in veterinary medicine, sulfonamides, sulfadimethoxine (SDM) and sulfaguandine (SGD) were selected for the present work. SDM was chosen mainly because it was recently shown to have the strongest phytotoxic potential of all sulfonamides ever investigated (Białk-Bielińska et al., 2011) and because the sparse data regarding its sorption potential (Thiele-Bruhn et al., 2004; Stoob et al., 2007; Sanders et al., 2008; Figueroa-Diva et al., 2010). It is thus a matter of urgency to determine its mobility in the environment, as it can pose a real threat to the whole ecosystem once it reaches the aquatic environment. On the other hand, sulfaguandine was selected mainly because, according to the 'Analysis of Antimicrobial Agents' Treatment of Swine in Poland in 2010', it is the most frequently used sulfonamide in medicated feeds (Krasucka et al., 2010), and because there are no studies describing its behavior in soils. Furthermore, even though these two compounds belong to the same group of pharmaceuticals, they differ in their physico-chemical properties (Table 1). SGD has an extremely high second dissociation constant compared with SDM and other sulfonamides (Carda-Broch and Berthod, 2004), so it may behave differently in the soil from other sulfonamides.

In the present study, the mechanism of sulfonamide (SDM and SGD) sorption onto three natural soil types differing in their organic content (OC), cation exchange capacity (CEC) and particle size distribution was investigated in detail (Table 2). Isotherms were

employed to describe sorption phenomena. The influence of external factors such as ionic strength and pH on sorption was also determined.

2. Materials and methods

2.1. Chemicals

Standards of sulfaguandine and sulfadimethoxine sodium salt as well as trifluoroacetic acid 99% (TFA) were purchased from Sigma-Aldrich (Steinheim, Germany). Deionized water was produced by the HYDROLAB System (Gdańsk, Poland). Acetonitrile (ACN), hydrochloric acid (HCl), potassium chloride (KCl), calcium chloride (CaCl₂) and potassium hydroxide (KOH) were purchased from POCH (Gliwice, Poland).

Standard stock solutions of sulfaguandine and sulfadimethoxine (800 µg mL⁻¹) were prepared by dissolving the pure compounds in 0.01 M CaCl₂ with the optional addition of HCl or KOH (1 M solutions) to achieve the appropriate pH (pH-dependent sorption experiments). CaCl₂ in concentrations of 0.001 M or 0.1 M was used in the ionic strength sorption experiments. The solutions were sonicated for 15 min to ensure complete dissolution. The spiked solutions (8 points) were prepared from stock solutions in accordance with the serial dilution method in the appropriate CaCl₂ solution.

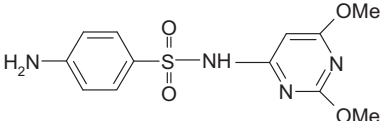
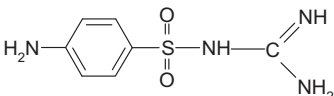
2.2. Soils

The experiments were carried out using three soils with different physicochemical properties (Table 2). The soils were sampled from the region of Pomerania in northern Poland. Afterwards they were air-dried, ground in a mortar and passed through a 2 mm sieve, then re-ground in a mortar with a small rubber pestle. The soil pH was determined with a glass electrode in a 1:2.5 soil/water suspension using deionized water and a 1 M KCl solution. Soil OC was determined by loss-on-ignition. CEC was determined using the BaCl₂ Compulsive Exchange Method.

2.3. Sorption studies

Batch sorption experiments were performed according to the OECD Technical Guideline 106 (OECD, 2000). All samples were pre-

Table 1
Structures and properties of the sulfonamides investigated.

Substance [CAS]	Structure	M.w. ^a (g mol ⁻¹)	pK _{a1}	pK _{a2}	log P ^a
Sulfadimethoxine (SDM) [122-11-2]		310.3	2.5 ^c	5.9 ^c	1.63
Sulfaguandine (SGD) [57-67-0]		214.3	2.8 ^b	12.1 ^b	-1.22

^a Data obtained from <http://www.vclab.org/lab/alogps/start.html> (ALOGPS 2.1).

^b http://web.squ.edu.om/med-Lib/MED_CD/E_CDs/A%20Practical%20Guide%20to%20Contemporary%20Pharmacy%20Practice/pdf/pKa-table.pdf.

^c Lo and Heyton, 1981; Sukul and Spittler, 2006.

Table 2
Physico-chemical properties of soils.

Soil	pH (H ₂ O)	pH (KCl)	OC (%)	CEC (cmol(+) kg ⁻¹)	Clay fraction (<0.01 mm) (%)
Sandy-clayey silt (CA1)	5.80	5.27	24.50	27.0	94.0
Alluvial soil (R13)	7.21	6.65	19.43	85.6	16.7
Beach sand (CA3)	7.38	7.38	0.14	3.0	0.2

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