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Uptake of silver by brown rice and wheat in soils repeatedly amended with biosolids

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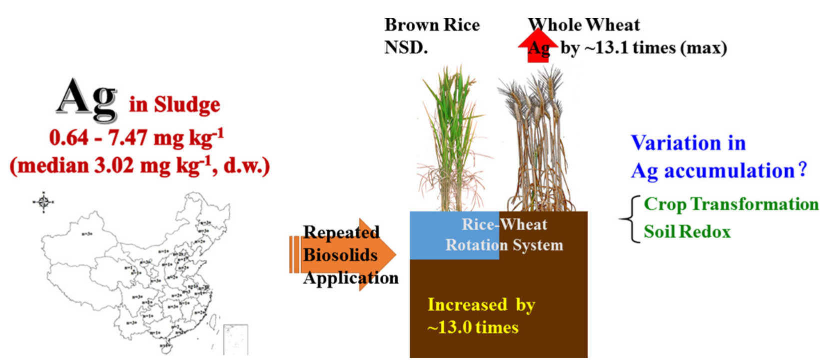
HIGHLIGHTS

- Silver concentrations in biosolids across China range from 0.64 to 7.47 mg kg^{-1}.
- Silver was higher in wheat with biosolids amendment under rice-wheat rotations.
- Silver accumulation in crops is mainly affected by water content and species.

GRAPHICAL ABSTRACT



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ABSTRACT

There have been numerous studies simulating the behaviour and fate of silver (Ag) applied to soils in biosolids in recent decades but the results remain to be verified under actual farming conditions. Here, we report a study of the transfer of Ag along the biosolids-soil-crop pathway with repeated biosolid applications over a four-year period to three contrasting soil types under rice-wheat rotation cultivation. A systematic investigation of Ag concentrations in biosolids throughout China shows Ag concentrations ranging from 0.64 to 7.47 mg kg^{-1} with a mean value of 3.58 mg kg^{-1} and a median of 3.02 mg kg^{-1} on a dry weight basis. Silver concentrations were significantly higher in industrial biosolids than in domestic or mixed flow biosolids. Biosolids application enhanced Ag accumulation in whole wheat. Silver concentrations in whole wheat increased to 20.8, 20.5 and 4.87 μg kg^{-1} after four years of high-metal industrial biosolids application to an acid Typic Ali-Perudic Aragsol, a neutral Typic Hapli-Stagnic Anthrosol, and a calcareous Typic Carbonati-Perudic Ferrosol, respectively. Moreover, the Ag translocation factor also increased in wheat following biosolids application with values of 5.6, 3.1 and 1.4, respectively. However, Ag accumulation in rice was found only in the acid soil with no discernible increase ($p > 0.05$) in the translocation factor. The seasonal redox cycle may contribute to this phenomenon. A seedling incubation experiment confirms the influence of soil water regime on Ag bioavailability with a higher Ag translocation factor during the wheat growing season than the rice growth period. Incorporating a fallow period during the wheat (winter crop) season might be a suitable strategy for repeated biosolids application.

1. Introduction

Silver (Ag) is usually assumed to have relatively low toxicity in soils because of its occurrence as a precipitate and the effects of aging on Ag

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bioavailability (Fjällborg et al., 2005; Ratte, 1999). However, the production and use of Ag nanoparticles (Ag-NPs) continue to increase (Calderon-Jimenez et al., 2017; McGillicuddy et al., 2017; Maillard and Hartemann, 2013), adding to the risk of Ag release into the environment.

Silver can accumulate in biosolids as a result of its passage through wastewater treatment systems (Kaegi et al., 2015; Kaegi et al., 2013; Impellitteri et al., 2013; Kim et al., 2010). There has been increasing concern regarding Ag-NPs since the publication by the United States Environmental Protection Agency (US EPA) of the Targeted National Sewage Sludge Survey Statistical Analysis Report (US EPA, 2009) which indicates that the total concentration of Ag in U.S. publicly owned treatment works (POTWs) might peak at 197 mg kg^{-1} (with one extreme outlier value of 856 mg kg^{-1} obtained). The results were assumed to be associated with the increasing production and use of AgNPs (Kim et al., 2010). There is limited information on Ag in sewage sludges or biosolids because Ag is one of the minor metals, with no potentially toxic element concentration limits as part of the biosolids management strategies in most countries (Shamuyarira and Gumbo, 2014). The limited published data on Ag in biosolids include 5 to 200 mg kg^{-1} in the UK, 1.1 to 33 mg kg^{-1} in Sweden, 0.02 to 3.1 mg kg^{-1} in the Czech Republic, 2.85 to 61.0 mg kg^{-1} in Japan, 1.3 to 15.9 mg kg^{-1} in India and 0.22 to 21.9 mg kg^{-1} in South Africa (Shamuyarira and Gumbo, 2014; Urbanová et al., 2013; Whiteley et al., 2013; Venkateswaran et al., 2007; Eriksson, 2001; Kawasaki et al., 1998). There are few, if any, published data on Ag concentrations in Chinese biosolids.

Land application of biosolids for the utilization of their plant nutrients in agriculture may result in the introduction of Ag into the human food chain. Some studies have detected migration of Ag-NPs in the soil profile (Makselon et al., 2017; Fujita and Kobayashi, 2016; Cornelis et al., 2013; Sagee et al., 2012) and the transformation of Ag-NPs in the natural environment (Hashimoto et al., 2017; Lowry et al., 2016; Sekine et al., 2015; Navarro et al., 2014). Data have also been obtained showing Ag speciation and accumulation in plants following amendment with biosolids (Pradas del Real et al., 2016; Stegemeier et al., 2015; Colman et al., 2013; Rico et al., 2011) but there have been no systematic long-term investigations of exposure to Ag in the biosolids-soil-crop continuum. In addition, soil type and repeated biosolids application may greatly affect Ag bioavailability. Thus, our understanding of Ag transformations in actual field conditions with repeated biosolids applications is fragmentary.

Research on biosolids management is required to elucidate the final fate of biosolid-borne Ag. Limited Ag transfer has been observed in a pot experiment with biosolids amendments conducted for four weeks (Pradas del Real et al., 2016). However, the trend may differ over longer time scales. Hirsch (1998) reported Ag accumulation in tissues of corn, lettuce, oat, soybean, and turnip with application of biosolids containing elevated levels of silver. Repeated application maintains continuous inputs of Ag and dissolved organic matter may contribute to the release of Ag into the labile pool. Moreover, nutrient effects can enhance the uptake of trace elements by crops.

Furthermore, there have been few studies of Ag lability under rice-wheat rotations with their typical alternate drying and wetting cycles worldwide. In addition, the availability of trace metals and metalloids will change with changing soil redox potential. For example, arsenic exists chiefly in the form of arsenite (AsIII) under reduced conditions and this is more bioavailable than arsenate (AsV) (Sun et al., 2016). Oxidation of sulphur in Ag_2S might cause Ag^+ release (Dale et al., 2013). From another perspective, Ag (0) will control free Ag^+ activity under reducing conditions (Maurer et al., 2012). The sensitivity of different plant species to toxic metals and their accumulation vary widely (Clemens, 2006). Rice is very sensitive to cadmium (Cd) or mercury (Hg) pollution (Zhang et al., 2010; McLaughlin et al., 1999). In addition, the proportion of labile Ag will decrease in high pH conditions (Li et al., 2016; Zhou et al., 2016b; Sekine et al., 2015). Metal cations will have higher activity when there is a large number of H^+ ions present (Smith, 2009). The behaviour of biosolid-borne Ag in rice-wheat rotation systems therefore merits investigation.

The first part of the present study comprised a survey of the concentrations of Ag in biosolids collected from wastewater treatment plants in provincial capital cities throughout China. In the second part, a pot experiment was conducted to investigate the accumulation and transfer of Ag from soil to crops with repeated biosolids applications to rice-wheat rotation systems. Finally, a seedling experiment was designed to verify the influence of soil-water regime on the extractability and plant uptake of Ag by rice and wheat. The aim was to determine the Ag concentrations present in Chinese biosolids and the translocation of silver in the biosolids-soil-crops continuum after repeated application of Ag-containing biosolids to agricultural soils.

2. Materials and methods

2.1. Experimental design

2.1.1. Part 1: investigation of Ag concentrations in biosolids throughout China

The total of 58 dewatered biosolid samples involved in this study were collected in 2013 for our previous work which investigated the total concentrations of trace metals (excluding Ag) and the occurrence of antibiotics in biosolids from provincial capital cities throughout China (Cheng et al., 2014). The freeze-dried (FreeZone Freeze Dry System, Labconco Company, Kansas City, MO) dewatered biosolid samples were passed through a 2-mm sieve and stored in brown glass vials at −20 °C. Three types of biosolids were collected, namely domestic, industrial and mixed flow biosolids. Sources and sampling details have been published previously (Cheng et al., 2014).

2.1.2. Part 2: repeated biosolid amendment experiment

A pot experiment was carried out at the Institute of Soil Science, Chinese Academy of Sciences, Nanjing, Jiangsu province, east China, from September 2009 to October 2013 with repeated applications of air-dried biosolids, and then from October 2013 to October 2015 without further biosolid amendment. This involved three soils of contrasting pH status and soil type (acid, Typic Ali-Perudic Aragsol; neutral, Typic Hapli-Stagnic Anthrosol; and calcareous, Typic Carbonati-Perudic Ferrosol) and two types of biosolids (domestic biosolids, low metal loading; and industrial biosolids, high metal loading). The experiment simulated a rice-wheat rotation system with wheat (*Triticum aestivum* cv. Yangmai 19, grown from October to May) and rice (*Oryza sativa* subsp. *japonica* cv. Suxiangjing 1, grown from June to October). The selected seeds were all provided by Suzhou Academy of Agricultural Science. Planting and biosolids amendment details are shown in Table S2. Shoots were collected when the crops were fully mature and the roots were returned to the soil. The plant samples were washed with tap water, cleaned with deionized water 3–4 times, dried at 70 °C to constant weight, and divided into two parts, the brown rice or whole wheat (the edible part) and the remains of all collected materials (the inedible part). Soil samples were collected once each year after the rice harvest and air-dried. Soil and plant samples were passed through a 0.15-mm nylon sieve and stored in paper bags prior to analysis.

2.1.3. Part 3: seedling experiment

The effect of soil water regime on Ag accumulation by wheat and rice was investigated at soil water contents of 50 and 70% of soil water holding capacity (WHC) and under flooded conditions. The neutral soil (Typic Hapli-Stagnic Anthrosol) was mixed with Ag-NPs (10 nm, 99.9% purity, cat. no. 100421, XFNANO, Nanjing, Jiangsu, China) two weeks before the rice or wheat seeds were sown. Silver was added to achieve a total target concentration of 2 mg kg^{-1}. Wheat and rice were grown in a glasshouse at a controlled temperature of 30 °C throughout the day and illumination of 10,000 lx for 16 h per day. Plants were grown in unamended soil as blank controls and there were four