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### Dynamics of multiple elements in fast decomposing vegetable residues

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#### HIGHLIGHTS

#### Litter decomposed faster in vegetable farmland than previously studied ecosystems.

- Roots decomposed faster than leaves for studied vegetables.
- As, Cu, Fe, Hg, Mn, and Pb possibly accumulated in the litters after 180 d.

#### GRAPHICAL ABSTRACT



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#### ABSTRACT

Litter decomposition regulates the cycling of nutrients and toxicants but is poorly studied in farmlands. To understand the unavoidable in-situ decomposition process, we quantified the dynamics of C, H, N, As, Ca, Cd, Cr, Cu, Fe, Hg, K, Mg, Mn, Na, Ni, Pb, and Zn during a 180-d decomposition study in leafy lettuce (Lactuca sativa var. longifoliaf) and rape (Brassica chinensis) residues in a wastewater-irrigated farmland in northwestern China. Different from most studied natural ecosystems, the managed vegetable farmland had a much faster litter decomposition rate (half-life of 18-60 d), and interestingly, faster decomposition of roots relative to leaves for both the vegetables. Faster root decomposition can be explained by the initial biochemical composition (more Oalkyl C and less alkyl and aromatic C) but not the C/N stoichiometry. Multi-element dynamics varied greatly, with C, H, N, K, and Na being highly released (remaining proportion < 20%), Ca, Cd, Cr, Mg, Ni, and Zn released, and As, Cu, Fe, Hg, Mn, and Pb possibly accumulated. Although vegetable residues serve as temporary sinks of some metal(loid)s, their fast decomposition, particularly for the O-alkyl-C-rich leafy-lettuce roots, suggest that toxic metal(loid)s can be released from residues, which therefore become secondary pollution sources.

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#### 1. Introduction

Litter decomposition is an important environmental process in regulating carbon (C) sequestration, nutrient release, humus formation,

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and pollutant cycling in ecosystems (Tsui et al. 2008; Berg and McClaugherty 2013; Cotrufo et al. 2015). The decomposition rate reflects the dynamics of element turnover and is believed to be determined by multiple factors, including litter quality (*e.g.*, C/N stoichiometry and molecular-level biochemical composition) and environmental conditions (Manzoni et al. 2008; X. Wang et al. 2012; Keiluweit et al. 2015). Studies about litter decomposition have mainly focused on natural ecosystems including forests (Paudel et al. 2015; Wang et al. 2016; Yeong et al. 2016), grasslands (Suseela et al. 2014), shrublands (Fioretto et al. 2005), and wetlands (Mackintosh et al. 2016; Xie et al. 2017). Although the nutrient and toxicant dynamics in managed agroecosystems are critical to sustainable agriculture, food quality, and human health (Dungait et al. 2012), few studies (Kuzyakov et al. 1997; Chaves et al. 2004) have investigated litter decomposition in the agroecosystems, such as in vegetable farmlands.

It is generally assumed that a large proportion of food crops harvested from vegetable farmlands leave few plant residues for in-situ field decomposition and nutrient cycling. However, a recent study showed that around 1.5 billion tons of vegetables and fruits are produced every year in the world but as much as 45% of them are discarded as wastes (Mazareli et al. 2016). Vegetable waste is generated at all stages of the supply chain, including harvest, retailing, and consumption. In China, about 100 million tons of vegetable waste or residues (including their leaves and roots) are produced per year (Sun et al. 2005), approximately 15–20% of which remains in the vegetable growing farmlands in the stage of vegetable harvest (Gong et al. 2012). A survey also showed that in some areas like Beijing City, ~70% of the 3.33 million tons of vegetable waste generated in 2007 was not reutilized but directly decomposed (Gong et al. 2012). Although vegetable waste digestions have frequently been studied in anoxic reactors (Provenzano et al. 2016; Wu et al. 2016), little is known about the in-situ decomposition of vegetable waste or its residues under natural conditions.

Here, we designed a 180-d field decomposition experiment in a known metal-polluted farmland in northwestern China and quantified the residue mass and multi-elements, including C, H, N, As, Ca, Cd, Cr, Cu, Fe, Hg, K, Mg, Mn, Na, Ni, Pb, and Zn. Among these elements, N, Ca, Cu, Fe, K, Mg, Mn, Ni, and Zn are considered as essential elements for plant growth and reproduction (Barker and Pilbeam 2016). As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, and Zn belong to the category of heavy metal(loid)s, and As, Cd, Cr, Hg, Mn, Ni, and Pb are commonly considered as potentially toxic metal(loid)s. We hypothesized that: 1) vegetable residues decomposed faster than plant litters decomposing in many other ecosystems (e.g., forest and wetland) because vegetable residues have very labile biomass; 2) the decomposition rates of four vegetable residues (including leaves and roots), although highly variable, can be determined by the analysis of C/N stoichiometry and biochemical composition of the initial vegetable residues; and 3) metal(loid)s, especially toxic metal(loid)s, are continuously released from the decomposing vegetable residues, and the changes in their concentrations vary greatly among vegetable residues.

#### 2. Materials and methods

#### 2.1. Study area

Baiyin City, or "Tong Cheng" (which means "Copper City" in Chinese), is located in the Gansu Province in northwestern China. This area has an annual average temperature of 6–9 °C, an annual average precipitation of 180–450 mm, and an annual average evapotranspiration of 2101 mm (Cao et al. 2016). It has been an important nonferrous metal mining and smelting base in China since the 1950s (Nan and Zhao 2000; Li et al. 2006). The suburban farmlands in this city have been subjected to wastewater irrigation for decades, and severe metal pollution has been discovered. Therefore, Baiyin City was listed as the first among 30 priority areas that the Chinese Ministry of Finance and Chinese Ministry of Environmental Protection funded for soil metal

2015 remediation in (http://jjs.mof.gov.cn/zhengwuxinxi/ tongzhigonggao/201506/t20150602\_1248397.html). The city is divided into Xidagou Basin (or "West Big Ditch", 428 km<sup>2</sup>) and Dongdagou Basin (or "East Big Ditch", 368 km<sup>2</sup>) (Nan and Zhao 2000). Farmlands in Xidagou Basin mainly receive treated municipal (or domestic) wastewater, and those in Dongdagou Basin mainly receive treated industrial wastewater (Nan and Zhao 2000; Cao et al. 2016). Soil properties and concentrations of heavy metals in these farmlands are shown in Table S1. These large farmlands provide vegetables for local communities and nearby cities (Cao et al. 2016). The cultivated vegetables mainly include leafy lettuce (Lactuca sativa var. longifoliaf L.), rape (Brassica chinensis L.), carrot (Daucus carota L.), Chinese lettuce (Lactuca sativa L.), Chinese cabbage (Brassica pekinensis L.), tomato (Lycopersicon esculentum L.), zucchini (Cucurbita pepo L.), and eggplant (Solanum melongena L.). High adverse health risks from diets based on vegetables growing in these farmlands have been reported (Cao et al. 2016). Two representative vegetable species, leafy lettuce and rape, growing in a greenhouse in the Dongdagou Basin (36.48°N, 104.31°E; altitude 1746 m) with the highest degree of pollution (Cao et al. 2016), were selected for our litter decomposition experiment.

#### 2.2. Litter bag experiments

Leafy lettuce and rape were planted in the spring of 2014 and harvested in the summer of 2014. After harvesting, the leafy lettuce and rape samples were gently washed with deionized water, divided into leaf and root parts, and then dried at 60 °C for 24 h to determine their mass. In July 2014, litter bags of dimension  $20 \times 20$  cm, with 2 mm mesh size, were filled with a 30-g sample of washed and dried vegetable leaf and then placed on the soil surface for *in-situ* field decomposition. On the same day, litter bags of dimension  $10 \times 10$  cm, with 1 mm mesh size, were filled with a 15-g sample of washed and dried vegetable root and then buried in the soil at a depth of 3–5 cm. After 60, 120, and 180 d, six bags of each tissue type (leaf or root) of each vegetable species (leafy lettuce or rape) were collected from the field. The collected samples were transported to the lab, gently washed with deionized water, dried, and weighed.

#### 2.3. Chemical analyses

The freshly collected (non-decomposed) and decomposed vegetable samples were all dried, ground, and passed through a 2-mm sieve. All samples were analyzed for C, H, and N contents by an elemental analyzer (Vario EL III, Elementar, Germany) (J.J. Wang et al. 2012). Each one of the samples (0.5 g) was mixed with 20 mL concentrated HNO<sub>3</sub> in a 300 mL Teflon beaker and then evaporated at 240 °C to obtain a final volume of 5 mL. The Teflon beaker was again heated at 240 °C for 1.5 h after addition of 10 mL concentrated HNO<sub>3</sub>, 1 mL concentrated HF, and 3 mL concentrated HClO<sub>4</sub>. After cooling to room temperatures, the digested solution was transferred from the Teflon beaker to a volumetric flask, which was then filled with Milli-Q water to make a final volume of 100 mL. Concentrations of As, Ca, Cd, Cr, Cu, Fe, Hg, K, Mg, Mn, Na, Ni, Pb, and Zn in the digested samples were analyzed by inductively coupled plasma atomic emission spectroscopy (using the ICP-AES, IRIS Intrepid XSP equipment, Thermo Fisher Scientific, MA, USA) and inductively coupled plasma mass spectrometry (using the ICP-MS,  $\times 7$ equipment, Thermo Fisher Scientific, MA, USA) following the Chinese standard methods of JY/T 015-1996 (State Education Commission of China 1996) and GB/T 6041-2002 (State Standard of China 2002). Specifically, ICP-MS analyzed the concentrations of As, Cd, Cr, Hg, and Pb, and the concentrations of the other elements were detected by ICP-AES. This detection method showed the recovery of 86.8-116.9% for all the metal(loid)s and standard deviations were lower than 20% in the repeat tests on standards and procedure blanks.

The organic carbon compositions of the non-decomposed vegetables were characterized by <sup>13</sup>C cross-polarization/total sideband

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