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Disinfection byproduct precursors in paddy fields under swine manure application: Reactivity, origins and interception

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ABSTRACT

High content of organic matter in paddy fields contributes substantial organic loading to ambient waterways via runoff, which is the potential precursor of disinfection byproducts (DBPs) in downstream water treatment plants. Moreover, submerged conditions of paddy fields pose a high DBP precursor export risk. The major objectives of this study were to characterize and quantify DBP precursors in paddy field floodwater under swine manure application, and to explore the origins and interception strategy of DBP precursors. In spite of high amounts of DBP precursors in paddy fields, the aromaticity and formation reactivity of DBP precursors are relatively low. Parallel factor analysis (PARAFAC) and self-organizing map (SOM) showed protein-like substances contributed the major part of bulk DBP precursors, while humic-like substances have a stronger tendency to yield DBPs than protein-like substances. The highest DBP precursor amounts occurred on the second day after swine manure application, and DBP precursor export potential fell by up to 56.2% within 7 days. Therefore, making one or more weeks' interval between the highest DBP precursor occurrence and a forecasted rainfall event could be an advisable strategy for interception of DBP precursors.

1. Introduction

It is the characteristic of organic rice farming that no chemical fertilizers or pesticides are employed (Sanders, 2006). Thanks to lower cost of production, organic paddies are not inferior to conventional high-yielding chemical-treated paddies in profitability (Dawe et al., 2003). As a form of organic fertilizer, swine manure is widespread in organic paddy production due to its cheapness and resource reuse in developed farming regions (Li et al., 2015a, 2015b) such as Taihu Lake Basin which is also a source of drinking water for approximately 36 million people in southeastern China (Sun and Mao, 2008; Townsend-Small et al., 2007). However, paddy fields under swine manure application is increasingly becoming an environmental concern because paddy fields under submerged conditions have a higher risk of exporting pollutants than dryland cropping systems (Chen et al., 2014b). Rainfall is the major driver of the pollution export from submerged paddy fields, and runoff is the carrier to transport contaminants to adjacent waterways. Additionally, irrigation and fertilization have effects on timing and magnitude of the pollution export, in that the irrigation water flows over soil surfaces and manure fertilizers rich in nitrogen, phosphorus and organic matter. Nitrogen and phosphorus will lead to eutrophication of waterways; organic pollution derived from manure fertilizers will rise the chemical oxygen demand (COD) and introduce hormones (Gall et al., 2011; Leet et al., 2012) and antibiotic resistance on microorganisms (Heuer et al., 2011; Tang et al., 2015) of adjacent waterways.

Among various organic pollutants, rise of the risk of disinfection byproducts (DBPs) formation is of particular human health concern because dissolved organic matter (DOM) is the precursors of DBPs. DBPs are formed during disinfection processes when oxidants (e.g. chlorine, chloramine, chlorine dioxide) react with organic and/or inorganic precursors in raw water. DBPs and their precursors are directly related to human health effects due to their genotoxicity and carcinogenicity. It is proved that cancer and adverse reproductive and developmental effects are pertinent to drinking/dermal/inhalation exposure. For instance, risks for bladder and rectal cancers are shown to be associated specifically with trihalomethane (THM) levels. Additionally, it is shown that haloacetic acids (HAAs) might induce liver and lung tumors (Richardson et al., 2007). Among halogen-containing DBPs, iodinated DBPs are generally the most toxic and chlorinated species the least, with brominated analogs of intermediate toxicity (Krasner, 2009; Wang et al., 2014; Zhang et al., 2015). In addition to common carbonaceous DBPs (e.g. THMs, HAAs), emerging nitrogenous (e.g. haloacetonitriles (HANs)) and oxygenated (e.g. Chloral hydrate (CH)) DBPs

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pose more cytotoxic and genotoxic effects than regulated carbonaceous DBPs.

Previous researches investigated different sources of DBP precursors in watersheds such as soils (Chow, 2006b), plant litter (Chow et al., 2011; Wang et al., 2012a, 2012b), and detritus (Wang et al., 2015a). These studies used pure water to extract DOM to test DBP formation propensity of the extract water under laboratory condition rather than raw water from field study. In addition, these studies focused on single source of DBP precursors rather than a specific environment or system as DBP precursor source. Hereby, we chose paddy field under manure fertilization as our study case because it is a specific ecosystem field comprising soils, plants, manure and other single DBP precursor sources. Moreover, precursors came from raw water for in situ field study that is more representative of reality compared with extract water under laboratory conditions.

In spite of a variety of water treatment processes that can remove some precursors from source water (Chu et al., 2015; Drikas et al., 2008; Wang et al., 2015b; Xie, 2003), controlling precursors at their sources such as paddy fields in our research might be a more effective and economical approach (Díaz et al., 2008; Díaz et al., 2009). The objectives of this study were to (1) characterize and quantify DBP precursors in paddy field floodwater affected by swine manure application and (2) determine origins of DBP precursors in paddy field under swine manure application. To our best knowledge, this is the first study to quantify the effects of swine manure application on DBP formation potential from floodwater DOM of paddy fields and to explore origins of DBP precursors in paddy fields (Supplementary material Section S1).

2. Materials and methods

2.1. Study area

The study was conducted in 2014 at the long-term rice field experiment station Shuangqiao Farm (120°40′E, 30°50′N). The site is characteristic of a subtropical monsoon climate with an average temperature of 28 °C in summer and an average annual rainfall of 1200 mm (http://data.cma.cn/data/cdcindex/cid/6d1b5efbdcbf9a58.html). The dominant soil type at the site is a gleyed paddy soil (clay loam, mixed, mesic Mollic Endoaquepts) which is the typical soil in the Taihu Lake Region of Southeast China (http://gis.soil.csdb.cn/).

2.2. Field operation and irrigation and fertilization strategy

Treatments compared four different swine manure (organic matter: 15%; N: 0.56%; P: 0.43%; K: 0.40%) application rates (i.e., SM0, SM1, SM2 and SM3) with three repetitions, representing 0 (Control), 714.1 (Low), 1428.2 (Middle), and 2142.3 (High) kg C ha^{-1}, respectively. It is a field experiment that could simulate the practical paddy cultivation. The average hydraulic retention time was 5 days because the floodwater level declined by about 1 cm/day. Swine manure was applied at transplanting (July 1) and incorporated into the top 20 cm soil by raking. The plots (4 × 5 m) were separated by field bunds and bordered on one side by a 1-m-depth trench. To reduce edge effects, non-experimental guard plots planted with paddy were established around the entire experiment. Phosphorus (0–19 kg P_2O_5 ha^{-1}) and potassium (57–99 kg K_2O ha^{-1}) were applied in all plots to ensure that these nutrients were not limiting crop growth. These fertilizers were broadcast by hand and incorporated with the 0–5 cm soil layer by puddling. Twenty-eight-day-old rice seedlings were transplanted (Jul. 1) to the fields. As is typical agricultural practices in the region, paddy fields were flooded under a 5 cm deep water layer kept constant by irrigation. A ruler fixed to a brick flushed with the soil surface was used to measure the floodwater depth. The air temperature was measured during sampling. The variation of temperature over the observation period is given in Fig. S1.

2.3. Sample collection

Floodwater samples were collected one day before swine manure application and every two days the first week after application. Later, samples were collected at 5- to 7-d intervals. Water samples were immediately filtered through precombusted 0.45-μm nominal pore size GF/F filters in the field and were preserved in iced-coolers for less than 1 h during their stay in the field and their way back to lab. As soon as we returned to the laboratory, the dissolved organic carbon (DOC) and ultraviolet absorbance at 254 nm (UVA_{254}) measurements and DBP incubation were conducted. Filtered samples were preserved at 4 °C through the course of analyses.

2.4. Chemical analysis

DOC concentration was determined by a MultiN/C2100TOC/TN analyzer (Analytik Jena AG). UVA_{254} was determined with a Persee TU-1810 spectrophotometer (Beijing Purkinje General Instrument). As a simulation of chlorination in drinking water treatment plants, DBP formation potential (DBPFP) tests were conducted with the dose-based method (Wang et al., 2015a). First, water samples were diluted to a DOC concentration of 3 mg L^{-1}; second, samples' pH was adjusted to 8.0 with H_3BO_3/NaOH buffer; third, samples were chlorinated with freshly prepared NaOCl/H_3BO_3 solution at a DOC/Cl_2 ratio of 1/3. All samples were preserved in 40 mL borosilicate amber vials and sealed without headspace. The vials were incubated for 24 h at 25 °C. Thereafter, residual chlorine was quenched by 10% Na_2SO_3 solution. DBPs were extracted and then determined by a 7890A Gas Chromatography (Agilent) with electrolytic conductivity detector following EPA method 551.1 and 552.2 (USEPA, 1995). Four THMs (trichloro-, dichlorobromo-, dibromochloro-, and tribromomethanes) and Five HAAs (chloro-, dichloro-, trichloro-, bromo-, and dibromoacetic acids) were determined.

The slope of the linear regressions between UVA_{254} and DOC concentrations is considered as specific UVA_{254} ($SUVA_{254}$). $SUVA_{254}$, in general, is proportional to the amount of aromatic carbon per unit of DOC and has also been widely considered as a surrogate for indicating DBP precursors (Abouleish and Wells, 2012; Hua et al., 2015). Similar with $SUVA_{254}$, the slope of the linear regression line of DBPFP versus DOC is considered as specific DBPFP (SDBPFP). SDBPFP means the DBP reactivity of DOC, namely the amount of DBP formation per unit of DOC.

Fluorescence excitation-emission matrices (EEMs) were measured by an F-4500 fluorescence spectrophotometer (Hitachi) with 0.050-s integration time and 5-nm band pass. Fluorescence intensity was measured at excitation wavelengths of 230 to 450 nm at 5-nm intervals and emission wavelengths of 300 to 600 at 5-nm intervals at room temperature (25 °C) in a 1-cm quartz cell. All fluorescence intensity data was calibrated to values in Raman Unit (RU).

2.5. Statistical design and data analysis

The field plots were a completely randomized block design with three replicates (Fig. S2). One-way analysis of variance (ANOVA) with least significant difference (LSD) set at $p < 0.05$ level was conducted to test differences of DBP precursor parameters among the four swine manure application rates. Correlation and regression analyses (two-tailed test of significant difference set at $p < 0.05$) of DBP precursor parameters, parallel factor analysis (PARAFAC) components, sample time and swine manure application rate were performed. One-way ANOVA and correlation and regression analyses were performed using IBM SPSS Statistics 19 (https://www.ibm.com/products/spss-statistics/). Four distinct algorithms for multiple or multivariate linear regression, *viz.* ordinary least square, principal component, partial least square and ridge regression, were employed for solving multicollinearity in independent variables. Canonical correlation was employed for simplify