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Review

Implications of stillage land disposal: A critical review on the impacts of fertigation

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ABSTRACT

Stillage is the main wastewater from ethanol production, generated specifically in the step of distillation. Regardless the feedstock, stillage contains high concentrations of organic matter, potassium and sulfates, as well as acidic and corrosive characteristics. Currently almost the entire volume of stillage generated in Brazilian distilleries is directed to the fertigation of sugarcane fields, due to its fertilizer character. However, the polluting potential of stillage characterizes its land disposal as problematic, considering probable negative impacts on the soil structure and water resources in case of excessive dosages. Since the literature lacks critical content describing clearly the cons related to the reuse of stillage in agriculture in the long-term, this review aimed to assess the real polluting potential of stillage, and the implications of its land disposal and/or discharge into water bodies. Evidence from the literature indicate that the main obstacles to reuse stillage *in natura* include risks of soil salinization; clogging of pores, reduction in the microbial activity and the significant depletion of dissolved oxygen concentrations in water bodies; contamination per nitrates and eutrophication; soil structure destabilization due to high concentrations of potassium and sodium; and, possible acidification of soil and water resources, considering the low pH of stillage (~4,5). Toxic metals, such as cadmium, lead, copper, chromium and nickel, were also identified in concentrations above the recommended limits in stillage samples, increasing risks to human health (e.g. carcinogenic potential) and to crops (e.g. productivity loss). In short, although some studies report benefits from the land application of stillage, its treatment prior to disposal is essential to make fertigation an environmentally suitable practice.

1. Introduction

Ethanol represents one of the main alternative energy sources developed in the attempt to reduce the dependence on fossil fuels, whose trade often faces political and economical barriers due to the instability in the major oil-producing countries (Gunaseelan, 1997; Pant and Adholeya, 2007; Báez-Vásquez and Demain, 2008). The main uses of ethanol regard the automotive industry, so that in comparison with other technologies employed in the production of biofuels some important advantages may be highlighted. Firstly we point out the technological consolidation of ethanol worldwide, as well as the large variety of convertible raw materials that can be produced in different climatic conditions (Wilkie et al., 2000; Hill et al., 2006; BNDES and CGEE, 2008; Cavallet et al., 2012). Additionally, some important environmental benefits result from its production and use: a renewable character, due to its biological origin (Hill et al., 2006), and an intrinsic potential to reduce the emission of greenhouse gases, based on carbon sequestration by the crops and on cleaner combustion (BNDES and CGEE, 2008; Macedo et al., 2008; Khatiwada and Silveira, 2011). However, the holistic characterization of ethanol as a self-sustaining technology also depends on the proper management of stillage, the main wastewater from distilleries.

Stillage, also named vinasse or distillery wastewater, is a dark-brown high-strength wastewater (HSW) whose organic content may be 100 times higher than the ones found in domestic sewage. It also presents acidic and corrosive characteristics, as well as appreciable concentrations of macro- and micronutrients (Pant and Adholeya, 2007; Strong and Burgess, 2008; Mohana et al., 2009; España-Gamboa et al., 2011; Fuess and Garcia, 2013). Regardless the feedstock used, ethanol plants usually generate 10–15 L of

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stillage per liter of produced ethanol (Pant and Adholeya, 2007; Satyawali and Balakrishnan, 2008; Mohana et al., 2009; Oliveira et al., 2013). Assuming an average rate of 13 L of stillage per liter of ethanol (BNDES and CGEE, 2008; Wilkie et al., 2000), a single relatively large-scaled distillery (365,000 m^3 per year, Dias et al., 2011) could produce annually up to 4.7 billion liters of stillage.

The available technological approaches applied to the management and/or treatment of stillage include mainly evaporation and concentration to produce animal feed and return to agricultural fields through fertigation, which have been used extensively over the last decades (Sheehan and Greenfield, 1980; Willington and Marten, 1982; Macedo, 2005; Pimentel et al., 2007; Smeets et al., 2008; Christofoletti et al., 2013). Currently several works describe the application of treatment technologies aiming at reducing the polluting load of stillage, which include: [i] anaerobic and aerobic conventional processes (Wilkie et al., 2000; Nandy et al., 2002; Pant and Adholeya, 2007; Acharya et al., 2008; Agler et al., 2008; Satyawali and Balakrishnan, 2008; Tondee et al., 2008; Mohana et al., 2009; Ferreira et al., 2011; Robles-González et al., 2012); [ii] phytoremediation (Billore et al., 2001; Valderrama et al., 2002; Singh et al., 2005; Olguín et al., 2008; Sohsalam and Sirianuntapiboon, 2008); [iii] conventional physicochemical methods (e.g. adsorption and coagulation–flocculation) (Zayas et al., 2007; Ryan et al., 2008; Liang et al., 2009, 2010; Rodrigues et al., 2013); and even [iv] advanced oxidative processes (Sangave and Pandit, 2004, 2006; Sangave et al., 2007; Yavuz, 2007; Asaithambi et al., 2012). However, most of the available data still refer to bench-scale systems, so that special attention should be given to pilot- and full-scale plants, in order to investigate the real potential applications of such technologies to stillage. Additional approaches for the management of stillage include its use as a source of nutrients in soil bioremediation (Mariano et al., 2009; Crivelaro et al., 2010; Christofoletti et al., 2013) and cultivation of microalgal biomass (Doušková et al., 2010; Yen et al., 2012).

Focusing on the conventional methods of concentration and fertigation, some considerations should be made. The production of animal feed through stillage concentration comprises the main technological approach to manage stillage used in the corn-to-ethanol industry from USA (Agler et al., 2008; Cassidy et al., 2008). In short, the liquid fraction remaining from distillation (whole stillage) is firstly separated from the insoluble solid fraction, usually through centrifugation. The solids are then dried to 10–12%, in order to produce distillers dried grains with solubles (DDGS), which present increased shelf-life, as well as high concentrations of soluble proteins (33% w/w), raw fat, fibre and elements such as phosphorus and potassium (Liu et al., 2008; Nichols et al., 2008; España-Gamboa et al., 2011). Although some calculations indicate an energy output from DDGS production and use of approximately 1.86 MJ L_{EtOH}^{-1} (Pimentel et al., 2007), due to the energy savings in conventional animal feed production processes, the main limitation associated with the concentration of stillage still comprises the high energy consumption. In this case, water removal demands energy amounts as high as 2.88 GJ (800 kWh) for each ton of evaporated water (Murphy and Power, 2008).

Considering fertigation, although some benefits should be highlighted, such as the reductions in the use of fresh water and mineral fertilizers (Macedo, 2005; BNDES and CGEE, 2008; Smeets et al., 2008), the direct land application of stillage may be problematic, since its low pH and high concentrations of sulfate and organic matter may compromise the soil structure and the surrounding water bodies, besides reducing crop productivity (Pant and Adholeya, 2007; Mohana et al., 2009). In Brazil most of the stillage is directly recycled through fertigation. This scenario is adequately represented by the ethanol industry in the State of São Paulo, which concentrates 55% of the Brazilian ethanol plants and where only 8 out of 165 distilleries employ alternative processes, such as anaerobic digestion, to manage stillage (Cruz, 2011). In global terms, the volume of stillage annually disposed on the ground may reach up to 325 billion liters in Brazil (Fuess, 2013). Considering the organic load supplied to the soil, each hectare might receive about 4.2 tons of organic matter (as chemical oxygen demand – COD) in sugarcane crops – based on an average rate for the application of stillage equal to 140 m^3 ha^{-1} (BNDES and CGEE, 2008), as well as on an average COD of 30 g L^{-1} (Wilkie et al., 2000; Fuess, 2013) for stillage. Furthermore, although scarcely quantified, side effects from the illegal discharge of stillage into water bodies should be considered, especially the depletion in the concentrations of dissolved oxygen.

Despite the adverse effects potentially associated with fertigation, the literature does not present a critical content that clearly assesses such impacts. In fact, a few works describe some impacts, usually positive ones (Pathak et al., 1999; Jain et al., 2005; Kaushik et al., 2005; Hati et al., 2007; Jiang et al., 2012; Previna and Saravanan, 2013; Silva et al., 2014), resulting from using stillage as a fertilizer. However, most of the experiments are conducted in the short-term (i.e. 2–3 years), so that the literature (regarding technical and legal aspects) still lacks consistent data relating the cons from reusing stillage in agriculture in the long-term. In this context, the main objective of this review was to assess the polluting potential of stillage, in order to point out the implications of its improper land disposal and/or discharge into water bodies. Data from literature were compiled and scenarios regarding the fate of stillage in environment were discussed. Finally, some alternatives for the proper management of stillage were assessed, with emphasis on the energy generation prior to its land disposal.

2. Stillage characterization: qualitative and quantitative aspects

Ethanol production, regardless the feedstock, is roughly summarized in two basic processes, which comprise the fermentation of the sugar source and the distillation of the alcoholic media formed during the fermentative process. Stillage is generated specifically in the distillation step, so that each type of unit processes and operations used to produce ethanol results in specific qualitative characteristics for stillage (Satyawali and Balakrishnan, 2008; Mohana et al., 2009; España-Gamboa et al., 2011; Christofoletti et al., 2013). Another important factor is related to the feedstock processed. For instance, in production chains based on the processing of cereal grains, stillage usually presents a higher protein content, and as a consequence, a higher nitrogen content is observed (España-Gamboa et al., 2011). In distilleries where sugar-based feedstocks are used, such as sugarcane, beet and sweet sorghum, high concentrations of sulfate are usually found in stillage (Fig. 1) (Ensinas et al., 2009), due to the broth's pH correction prior to fermentation with sulfuric acid (H_2SO_4), mainly in cases based on the use of molasses as feedstock. As a direct consequence, the concentrations of sulfate tend to increase by the end of the sugarcane season due to the continuous application of H_2SO_4 in the fermentation vessels.

Fig. 1 depicts the composition of stillages from different feedstocks in comparison with domestic sewage. Besides the influence of sulfuric acid, the low pH usually observed in stillage (~4.0–4.5) is strictly related to the formation of organic acids during the step of fermentation. Boopathy and Tilche (1991), de Bazúa et al. (1991) and Nasr et al. (2011) have reported volatile fatty acids concentrations of up 44.8, 18.8 and 12.3 g L^{-1} in stillages from beet molasses, sugarcane molasses and corn, respectively. Studies also have indicated the presence of significant amounts of phenolic (e.g. tannic