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Research article

Use of high metal-containing biogas digestates in cereal production – Mobility of chromium and aluminium

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ABSTRACT

Biogas digestate use as organic fertilizer has been widely promoted in recent years as a part of the global agenda on recycling waste and new sustainable energy production. Although many studies have confirmed positive effects of digestates on soil fertility, there is still lack of information on the potential adverse effects of digestates on natural soil heavy metal content, metal leaching and leaching of other pollutants. We have investigated the release of aluminium (Al) and chromium (Cr) from different soils treated with commercial digestates high in mentioned potentially problematic metals in a field experiment, while a greenhouse and a laboratory column experiment were used to address mobility of these metals in two other scenarios. Results obtained from the field experiment showed an increase in total concentrations for both investigated metals on plots treated with digestates as well as a significant increase of water-soluble Al concentrations. Factors that were found to be mostly affecting the metal mobility were dissolved organic carbon (DOC), pH and type of soil. Metal binding and free metal concentrations were modelled using the WHAM 7.0 software. Results indicated that the use of digestates with high metal content are comparable to use of animal manure with respect to metal leaching. Data obtained through chemical modelling for the samples from the field experiment suggested that an environmental risk from higher metal mobility has to be considered for Al. In the greenhouse experiment, measured concentrations of leached Cr at the end of the growing season were low for all treatments, while the concentration of leached Al from digestates was higher. The high irrigation column leaching experiment showed an increased leaching rate of Cr with addition of digestates.

1. Introduction

For the past few decades, both environmental scientists and policy makers are struggling between addressing the climate change and renewable energy issues and, at the same time supporting the focus on increased food production. Industrial production of biogas through the anaerobic fermentation of different organic material represents just a small part of the 21st century's green industry agenda (Gissen et al., 2014; Hijazi et al., 2016). The importance of biogas industry contribution to the circular economy and environmentally friendly energy production has resulted in increase of the number of biogas plants in Europe where digestates are produced as by-products. According to the report of the European Biogas Association (EBA) there was 17,439 biogas plants registered in Europe by the end of 2017 (EBA, 2017). For most of the biogas production facilities, there is a considerable amounts of biogas digestates produced and regarded as waste that needs sustainable handling and reuse. Increased interest for the research of numerous aspects of biogas digestates have been supported by the European Union through financial support of projects with budgets ranging from 0.5 up to 1.5 million EUR (Up2Europe, 2018). Agronomical use of biogas digestates is today seen as an additional advantage of the biogas industry, since considerable amounts of digestates are made available as fertilizer, thus facilitating an organic waste disposal (Alvarenga et al., 2015) with additional benefits to both companies and farmers. Digestates are often used in agronomic plant production, in either their liquid or a dewatered solid form, and their increasing use has been widely accepted. There are efforts made continuously to use digestates as a basis for manufacturing other types of fertilizers that can be used for more

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specific agronomic production. In Norway one of the best examples of integrated approach to the commercial use of digestates is the Minorga fertilizer company, that is producing several fertilizing products by using biogas digestates acquired from the neighboring biogas plant (Minorga, 2012). Still, problems related to the use of digestates are similar to the use of other type of organic waste based fertilizers. One of the main challenges is the establishment of the fair certification system that would facilitate objective assessment of any organic based fertilizer prior its use. The European directive 2008/98/EC (EC, 2008), also known as Waste framework directive, only provides a frame for the national regulators to define the methodology, processes and limitations for organic waste handling and use in agricultural production. One of the ways to systematically test the quality of biogas digestates is the use of bio certification schemes applied in the United Kingdom (EA, 2014). In addition, implementation of similar certification standards such as PAS100:2014 (BSI, 2014), has supported a fast development of the digestate market in England and Wales and their use as organic fertilizer in agriculture. Norwegian national regulations in general are covering the common heavy metals and set limits to their total concentration in fertilizers (Mattilsynet, 2006). In general, all the organic fertilizers, which are exceeding these limits are usually not allowed to be used in agronomic production, especially organic food production in Norway. Fertilization by using digestates can serve as an input of different heavy metals and can result in soil and environment contamination (Fang et al., 2016; Studer et al., 2017) especially in cases where levels of metals are exceeding the defined regulations. In general, amounts of heavy metals and other pollutants are dependent on the type of substrate used for the biogas production process.

Metals such as nickel (Ni), cadmium (Cd), arsenic (As), mercury (Hg), but also chromium (Cr) and aluminum (Al), have an adverse effect on the environment if their concentrations are elevated in either fertilizers, soil or ground water systems. If the concentrations of heavy metals are elevated in biogas digestates, the risk of soil pollution and leaching and thus contamination of groundwater is increasing. Since digestates are rich in dissolved organic matter (DOM), this factor can enhance the metal mobility and by doing so, increase the release of the added and naturally present metals to the environment, especially to surface and groundwater systems. This study is focused on the potential use of high metal-containing digestates (exceeding the regulated values) for cereal production with a focus on mobility and release of Al and Cr when digestates are used as fertilizers in a usual agronomic practice.

Biogas digestates can contain high concentrations of Al because of the use of precipitating agent (Drosg et al., 2015), such as aluminium sulphate, $Al_2(SO4)_3$ usually applied for waste water processing and production of sewage sludge. One of the biogas plants involved in this study uses aluminium sulphate as explained earlier. As aluminium silicates are a major constituent of the soil forming rocks, Al is typically present in soil as insoluble Al compounds and the potential threat from leaching of Al is usually low. Still under highly acidic conditions in soil, some of the Al is solubilized (Zhaohuai et al., 1995) as positively charged cations, particularly as Al^{3+}, $Al(OH)_2^+$ and $AlOH^{2+}$. These compounds can become mobile and cause significant problems for both plant and soils (Zioła-Frankowska and Frankowski, 2017). In addition, digestates contain high concentrations of dissolved organic carbon (DOC), which can mobilize soil Al. Application of digestates on agronomic soils may influence the soil pH as well and further enhance the rate of Al leaching. In agricultural soils, the soluble Al forms, especially Al^{3+}, can limit plant growth by affecting the root growth and in extreme cases, they can be toxic to terrestrial plants (Kochian, 1995; Samac and Tesfaye, 2003). If Al is released from soil, it can be leached to groundwater, lakes and rivers, where it can cause severe damage to biological organisms, especially in its mobile form.

High concentration of Cr in biogas digestates is not common (Kupper et al., 2014), but in this specific study both of the used commercial digestates had a total Cr content higher than the limitations (100 mg/kg dry digestate) defined in the Norwegian regulation for organic fertilizers for class 0, I, II and III of organic fertilizers (Mattilsynet, 2006). The behavior of Cr in soil depends on soil properties such as pH, organic matter content, cation exchange capacity and soil redox condition (Banks et al., 2006). Since Cr mobilization can cause adverse effects to the environment, especially for aquatic organisms in the form of Cr(VI), the retention of Cr in soil is important, and it can be controlled by mechanisms such as sorption, precipitation and nucleation (Bradl, 2004). High concentration of DOM in digestates may affect Cr mobility of naturally present soil Cr and it can also influence the Cr redox equilibrium (primarily Cr(III)/Cr(VI)) and therefore limit the role of soil as a sink (Kyziol et al., 2006).

Main hypothesis of this study is that high metal-containing digestates will have an adverse effect on to the soil environment and further to the groundwater system. Mobility of Al and Cr were investigated in different soils fertilized with high metal-containing biogas digestates, estimating the potential for the use of these type of digestates with a focus on metal release from agricultural soils by using a field experiment. The results obtained with digestates were compared to animal manure and unfertilized soils (controls). Use of animal manure and control soils for comparison has facilitated in assessing the influence of digestate application and factors such as pH, DOC and soil texture on the release of investigated metals. Beside the field experiment, a leaching study in a greenhouse experiment and high irrigation column leaching experiment were also used to additionally investigate the mobility of mentioned metals in two different scenarios. Measured values of Al and Cr were assessed using the latest regulation on maximum allowed metal concentrations for aquatic organisms and fresh waters.

2. Materials and methods

2.1. Biogas digestates and soils

In the field, greenhouse and column experiments, two digestates were used. The commercial digestates (CDIG) were produced at two Norwegian biogas plants, which use different mixtures of substrates and different technological process. The CDIG1 was produced from with application of pretreatment (steam explosion) of food waste. The second used digestate, CDIG2, was produced using a mixture of municipally collected food waste and sewage sludge (ratio 50:50) as substrate for the anaerobic digestion process. The total concentrations of Al and Cr in digestates and manure are given in Table 1. Besides the total concentrations, also the water-soluble concentrations were analyzed as explained in paragraph 2.4 and 2.5.

Beside the use of treatments with different digestates a treatment with manure (animal manure, AM) and a control treatment without addition of organic or mineral fertilization was used for the comparison purposes. The AM treatment was a cattle manure from the University farm produced in Ås (Norway). Before chemical analysis all the samples were stored at 4 °C. Selected characteristics of digestates and manure are given in Table 2. Calculated and added amounts of different treatments were applied after using the data for inorganic nitrogen (N) content and were equivalent to a dose of 100 kg N ha^{-1}, which is a common amount of N fertilization for cereal production in Norway.

For the purpose of this study, soils from the upper layer (0–20 cm) from three different sites in Norway (southeast part)