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Long-term amendment of urban and animal wastes equivalent to more than 100 years of application had minimal effect on plant uptake of potentially toxic elements

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ABSTRACT

Urban waste products such as sewage sludge and organic household waste compost have a potential as fertilizers in agriculture, but evidence from historical field experiments performed in England and Germany with sewage sludge as fertilizer pointed towards negative effects of sludge-derived heavy metal accumulation on soil microbiota and plants. However, sewage sludge has improved in quality during the last half century and source separation of organic household waste has been shown to decrease the concentration of heavy metals in composts made from organic waste. This alongside a growing awareness on the benefits of an increased degree a recycling of nutrients from urban areas to agricultural soils led to the establishment of the CRUCIAL field trial in 2003 where different urban and animal wastes have been applied each year. In the present study we investigated the effect of the different animal and urban waste treatments in CRUCIAL on oat yield after ten years of experimentation on concentrations of heavy metals in oat grain harvested in the field and pea plants grown in a pot trial using soil from the CRUCIAL field trial. We found that all animal and urban waste fertilizers were suitable for fertilization purposes, although some of these will result in unbalanced nutrient inputs if used as single sources. Furthermore, we found that of the elements studied only Zn and Cu were significantly elevated in soil concentration in the urban waste treatments. In oat grain Cd was significantly elevated in a sewage sludge treatment corresponding to more than 200 years' legal application, but even in this extreme case the concentration of Cd in grain did not exceed the relevant EU limit for Cd content. In pea plants only the concentrations of Zn and Mo were significantly higher in plants grown in soil that had received large amounts of urban wastes compared to an unfertilized control and the build-up of Zn could be regarded as a beneficial side-effect of using sewage sludge as a fertilizer due to the status of Zn as an essential element.

In conclusion, our data indicates that contemporary urban waste products can be used for supplementary fertilization purposes in agriculture with no substantial risks related to potentially toxic elements.

1. Introduction

Recycling of urban organic waste materials as agricultural fertilizers may represent a resource efficient and sustainable alternative to chemical fertilizers (European Commission, 2011). Sewage sludge is recognized as a soil amendment for its potential as a source of phosphorous (P) and to a lesser extent of nitrogen (N) and organic matter (Magid et al., 2001). According to Hargreaves et al. (2008) composting of municipal solid waste has potential as a beneficial recycling tool. Green manures and animal slurries are organic wastes traditionally applied for increasing the soil content of organic carbon (C) and N (Kirchmann et al., 2013). However, possible undesirable effects on soil quality from all these organic manures and waste materials, such as the accumulation of xenobiotics (Smith, 2009a,b) and heavy metals (Giller et al., 1999) have been the cause of concern. The European Sewage

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Sludge Directive [Council Directive 86/278/EEC (European Commission, 1986)] seeks to encourage the use of sewage sludge in agriculture and to regulate its use in such a way that prevents harmful effects on soil, vegetation, animals and man. Regardless of the international support for recycling sludge to land, the acceptance of this practice among different European countries varies considerably and has declined markedly in some cases (Smith, 2009a,b), e.g. in Switzerland public concern has led to a banning of the use of sludge in agriculture since 2006, despite official recognition that there is no conclusive scientific evidence that the practice of using contemporary sewage sludge is harmful in any way (Eriksen et al., 2009; Smith, 2009a,b). The scepticism towards agricultural use of sewage sludge was in part based on solid evidence that potentially toxic elements (PTE) could disrupt microbial function in soils (Giller et al., 1999, 2009), e.g. the ability of native rhizobium populations to fix nitrogen after entering into symbiosis with clover plants. This was based on observations from long-term experiments in England and Germany that had received substantial amounts of highly polluted sewage sludge from the Second World War and post war years (Ken Giller, personal communication).

In recent years, waste streams in society have changed considerably, due to implementation of environmental regulations, outsourcing of production, and changing waste management technologies (Poulsen et al., 2013b; Magid et al., 2006a,b). This has resulted in a general improvement in the quality of the sewage sludge produced. Thus, there has been a 3–5-fold decline in the content of PTE such as Cadmium (Cd), mercury (Hg), lead (Pb), zinc (Zn) in sewage sludge since the seventies (Amlinger et al., 2004; Anon, 2006). The improvement in waste stream quality, alongside with increasing concern for depletion of non-renewable resources, has led the British Soil Association to propose that amendments should be made to EU regulation No. 889/2008 (European Commission, 2008) to permit the use of sewage sludge on organic certified land subject to certain quality criteria and appropriate restrictions, including maximum concentrations of heavy metal and organic contaminants (Soil Association, 2010).

Even though a number of trace elements such as Zn, copper (Cu), molybdenum (Mo) and boron (B) in waste materials are essential plant nutrients and thus constitute important inputs for crop production it is none the less advisable to limit the input of these elements to agricultural soils. Organic farmers have stressed the importance of treating organic waste materials before their application to agricultural soils, providing an integrated ecological sanitation and quality analysis. One possible solution is to provide separated fractions (for instance human urine) instead of merely delivering the composite that may contain undesirable compounds and thereby reduce the need for purification of sewage water (Magid et al., 2006a,b; Mihelcic et al., 2011). Human urine contains the greater part of nutrients in excreta, and far less PTE than solid waste, though it is not allowed at the moment in EU for organic farming (Jönsson, 2001; European Commission, 2007, 2008). The application of human urine has been gaining popularity as fertilizer in selected countries for agricultural practices (Karak and Bhattacharyya, 2011), but the source separation systems need to be further tested and developed to integrate technical and socioeconomic aspects (Magid et al., 2006a).

Several factors could contribute to PTE in composted household waste, such as household pesticides, paint residues, batteries etc. (Smith, 2009b). Source separation of the household waste before composting of the organic fraction is considered to be the best option for decreasing the PTE content of the compost (Hargreaves et al., 2008; Smith, 2009b). Animal waste may contain salts, pathogenic organisms and PTE, mainly Zn, Cu and Mn that could contribute to agricultural nonpoint pollution (Reddy et al., 1981) and increase the uptake in plants cultivated in the amended soils (Moral et al., 2006).

Long-term experiments can give a realistic overview of the accumulative effects on soil properties and plant quality when applying waste materials. This is both relevant to consider regarding plant nutrients and PTE uptake. The residual effect from repeated applications that are incompletely mineralized in preceding years must be taken into consideration when evaluating the effect of these materials on soil properties and plant uptake (Schröder, 2005). Most of the publications regarding the soil accumulation and plant uptake of PTE after organic waste applications have mainly been evaluating the effect after short-term applications (Zheljakov and Warman, 2004) and have to a large extent focused on the possible fertilization with micronutrients (Fe, Mn, Zn, Cu) (Moral et al., 2006; Nikoli and Matsi, 2011; Fan et al., 2012) but have not dealt with other possible toxic metals. Sewage sludge and animal manures are the most studied waste materials regarding the plant uptake of PTE. Human urine has been studied in the recent years due to its potential as a N and P fertilizer and low concentration of PTE (Karak and Bhattacharyya, 2011). Only a few studies have reported the effect of consecutive applications of organic wastes under field conditions. Cambier et al. (2014) analysed the trace metal soil solution concentration and leaching after the long-term application of urban waste composts and farmyard manure in a field site in France and a prediction model was reported by Legind et al. (2012) simulating the Cd and Pb uptake in plants and leaching to groundwater at the same site. Hamnér et al. (2013) studied the nickel concentrations in topsoil and wheat after long-time applications of cattle manure and sewage sludge in a Swedish field site and concluded that Ni was not accumulated significantly in soil or was deposited in the crop. Borjesson et al. (2014) have recently reported an increase in crop yield and soil organic carbon and a limited effect on metal uptake after the repeated application over 14–53 years of sewage sludge on four Swedish soils.

Despite the common findings of elevated PTE concentrations in waste materials described above, to the best of our knowledge, a comparative study on the accumulation in soil and the plant uptake of several PTE after long-term application of several waste materials under field conditions is not available.

The need for interdisciplinary studies on integrated ecological waste management systems have promoted the establishment of a long-term field trial in Denmark (the 'CRUCIAL' project) to demonstrate and understand the effects of fertilization with urban and animal waste materials (Magid et al., 2006b). The soils have been treated annually since 2003 with different urban and animal organic materials at normal and accelerated N levels as well as NPK, giving a possibility to obtain a realistic picture of their impacts on soil properties and crop quality. Previous results from the CRUCIAL project have shown that application of urban and agricultural organic fertilizers has significant effects on soil organic matter, respiration and the microbial biomass (Poulsen et al., 2013a) and that repeated organic waste applications reduces draught force and fuel consumption for soil tillage (Peltre et al., 2015).

Here we evaluate the effect on soil properties in the CRUCIAL field trial, oat yield and uptake of PTE after long-term applications of waste materials after 11 years repeated application. This corresponds to 100–200 years of legally acceptable application in the case of accelerated sewage sludge and composted household waste that is regulated by its content of P. Considering the above discussed changes in contemporary waste quality, we hypothesized that this long-term use would be safe considering risks from PTE. In parallel, we evaluate biomass production and metal accumulation in pea (*Pisum sativum*) under controlled conditions in a green-house experiment based on soil from the CRUCIAL field