Organic substances in sewage sludge intended for agricultural land – what do we know and where are the knowledge gaps?

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Introduction

Sludge application on agricultural land is a prospective dispersal of a broad range of substances on soil intended for food production. The wastewater, and the wastewater sludge, is a mirror of the chemical society, reflecting the substances used, released and discharged. But sludge is also a rich source of nutrients which can be used as an alternative or supplement to mineral fertilizers. The Swedish committee on environmental objectives has proposed that 75% of the phosphorus in waste and wastewater such be a part of a sustainable eco-cycle incorporating both rural and urban areas by the year 2010. Furthermore, the phosphorous recovery can mitigate eutrophication by removing nutrients from the aquatic environment and decrease the burden of mining mineral fertilizers (Naturvårdsverket, 2002). Thus, the use of wastewater and sludge on agricultural land has to be deemed acceptable both in the short- and long-term perspective.

Aim and approach

In the 'Action plan for recirculation of phosphorous from sewage' (Naturvårdsverket, 2002) suggestions and measures for managing organic substances in sludge are addressed. As input Samsø-Petersen (2002) conducted a state-of-the-art survey on the presence and risk associated with organic substances in sludge. However, the action plan is up for revision and it is the aim in this study to review the current sludge situation based on a number of predefined reports and compare with the materials from 2002.

The literature lists consisted of the reports (full references in the reference list):

1. DEFRA (2005) Sources and impacts of past, current and future contamination of soil.
2. DG Environment (2000) Working document on sludge, 3rd draft
3. Eriksen et al. (2009) Risk assessment of contaminants in sewage sludge applied on Norwegian soils.
4. Haglund P. and Olofsson U. (200X) Miljöövervakning av slam. Redovisning av resultat från 2007 års provtagning.
5. Langenkamp H. et al. (2001a) Organic contaminants in sewage sludge for agriculture use.
6. Naturvårdsverket (2002) Aktionsplan för återföring av fosfor ur avlopp.
7. Samsø-Petersen L. (2003) Organic contaminants in sewage sludge. A review of studies regarding occurrence and risk in relation to the application of sewage sludge to agricultural soil.

And PowerPoint presentations:

8. Krogh PH., and Jensen J. (2006) Er slam skadeligt eller gavnligt for jordmiljøet? Kan man forudse nogen miljø- og sundhedsrisici af miljøfremmede stoffer og lægemidler i slam?
9. Langenkamp H., et al (2001b) Organic contaminants in sewage sludge for agricultural use.
10. Zambrzycki B. (2009) Zukunft der Klarschlammverwertung aus sicht der EU-Kommission.

Sludge trends

The use of sludge differs between the member states in the European Union (EU) countries and for example Spain, Ireland, France, United Kingdom and Denmark use over 50% of their sludge on agricultural land. Others, as Sweden, Finland, Slovenia and Czech Republic use less 20% for the same purpose (Zambrzycki, 2009). Sludge generation in EU-15 ("old" member states) increased between 1995 and 2005 from ca. 6.5 million tons of dry matter (dm) to ca. 8.5 million tons dm (Zambrzycki, 2009). The total generation of sludge in the EU will increase further with implementation of the urban wastewater directive (91/271/EEC) in EU-12 ("new" member states) and full implementation in EU-15.

European sludge related regulation

The urban wastewater directive (91/271/EEC) lay down directives for secondary treatment (including biological treatment and secondary settling) of all discharges from cities of > 2000 population equivalents (PE), and even more advanced treatment for >10 000 PE in so-called sensitive areas. Here it is encouraged that sewage sludge shall be re-used whenever appropriate and that the disposal shall minimize the negative effects on the environment. Similarly the sewage sludge directive (86/278/EEC) focuses on protection the environment when sludge used in agriculture but also encourages and regulates the use of sludge. The alternatives in sludge management applied in different EU countries are land filling (1999/31/EC) and incineration (2000/76/EC) but these are now being discouraged in many countries due to their low sustainability, e.g., high carbon dioxide emissions. DG Environment (2000) draw the potential lines for a revised EU sludge directive and states that use on land should be performed whenever possible, according to EU or national legislation, and according to the nutrient need. Sludge treatment types are described and it is stated that the level of treatment will depend on the land type on which the sludge should be spread.

In a review by Langenkamp et al (2001b) is was noted that the limit values (LVs) for sludge in the EU is restricted to a few substances or groups of substances (adsorbable organic halides (AOX), di(2-ethylhexyl) phthalate (DEHP), linear alkyl benzene sulphonate (LAS), nonylphenol and nonylphenol ethoxylates (NP/NPE), polycyclic aromatic hydrocarbons (PAH), polychlorinated biphenyls (PCB) and dioxins/furans (PCDD/F)) and mainly focussed in northern and central Europe (Denmark, Sweden, Austria, Germany and France as well as suggested LVs in the EU (DG Environment, 2000)). In Denmark were the sludge regulated revised in 2006 and addresses LAS, PAH, NPE and DEHP (Retsinfo, 2006). Many countries or regions have threshold values or quality criteria for substances in soil such as Germany (Langenkamp et al., 2001b) and in the counties of North-Zealand (2004) but the focus is often on metals, PAHs, oil and pesticides. There is no apparent harmonisation between sludge and soil priority substances and LVs. Pharmaceuticals and biocides are not regulated in sludge but should be assessed for their environmental impacts according to other EU directives (2001/83/EC; CMHP, 2006; European Commission, 2009) and other substances will fall under REACH (2006/121/EC).

Sludge quality

The quality of the sludge is highly dependant on the use of substances in the society. Differences in different countries due to consumer habits, e.g. LAS is lower in Norway then in Denmark because of the more extensive use of eco-labelled detergents in Norway (Tørsløv et al., 1996 and Paulsrud et al., 2000). In Sweden mitigation measures to reduce NP and NPE use have been successful but levels in sludge have not continued to decline but rather stabilized (Hök et al., 2007) and it was found that textiles; towels and children's clothing (Hök et al., 2007 and Testfakta, 2007), imported from non-EU countries contribute with substantial amounts. The number of studies on sludge quality has increased since 2000 which presumable is due to the publication of the 3rd draft on sludge from the DG Environment. Nonetheless it have been possible to note that preventive measures have been efficient and levels of AOX in German sludge has degreased over time and by 2000 were 90% below the limit of detection (UMK-AG 2000). Within the framework of the Swedish National Environmental Monitoring, where sludge from eight wastewater treatment plants (WWTPs) sampled and analyzed. The results were compared with previous studies yielding a data set covering four years (2004-2007). Similar to continental Europe flame-retardants, phthalates, dioxins as well as chlorinated phenols and benzenes were measured but also other substances as antibiotics, fluorinated substances, and phosphate esters (Haglund and Olofsson, 200X). Generally are the pollutants in the sludge found in the same order of magnitude for all the WWTPs and the dataset is too short to see any trends over time. But as time goes by it will be possible to note the effects by e.g. substitution. For example a shift of phthalates in wastewater has been noted in Malmö, as industrial wastewaters contain primarily di-isononyl phthalate (DiNP) and only low levels of DEHP whereas domestic wastewater is dominated by DEHP (Flygare, 2008).

Sludge applications and associate risks

Pollution of agricultural soil may derive from many point and diffuse sources including wet and dry deposition, mineral fertilizers, manure, and deposition from floods as well as sludge (DEFRA, 2005). The Priority Organic Pollutants (POPs) such as PAHs, PCBs, and PCCD/Fs primarily originate from atmospheric deposition (Langenkamp et al., 2001a; DEFRA, 2005) but levels are dropping in both deposition and sludge owing to mitigating actions. Thus comparison with contributions from other sources and the fate in soil are of importance (Samsø-Petersen, 2002). The input rate of the organic substances to agricultural soil should not exceed the rate of degradation, which makes the local factors of high importance when deciding the application volume and frequencies (Langenkamp et al., 2001a)

Long-term studies are important to determine the accumulation and degradation of organic substances in sludge amended soils. In rural United Kingdom were sewage sludge applied 25 times to agricultural land from 1942 to 1961. During the application period the concentrations of PAHs rose but fell again after cessation of the applications. In 1984 the short-chain PAHs had almost fallen to the pre-application levels whereas the long-chain PAHs had approximately had their value since 1960 (Wild et al., 1990). Thus, the inherent properties of the substances have profound effect on the fate in the soil. Wild et al (1990) also noted that the high sludge input to rural soil did not raise the contamination above what is frequently observed in soils in urban areas. In France NP/NPE, LAS, DEHP and PAHs were included in a long-term study of sludge applied bi-annually between 1974 and 1992. The soil was tested 12 years afterwards and NP, NPE and LAS had been decreased too less than 30% of the recorded concentrations during the spreading period whereas DEHP and PAH remained in high concentrations. All but NP were after 12 years increased compared to the control soil but inferior to the Predicted no-effect concentrations (PNECs) (Patureau et al., 2007). In Denmark, a long-term field test on the effect of sewage sludge on microorganisms and soil-dwellers was conducted 1996-2003 using 'worst case scenario' sludges with respect to the substance content. No statistically significant effects could be seen for microorganisms. For earthworms did application of sludge not differ from application of fertilizers or manure, though differences in responses were seen for individual species. The sludge and the manure showed to be beneficial for the soil environment and no long-term effects could be seen on microbial processes or soil-dwelling organisms (Krogh and Jensen (2006)).

The human exposure via direct use of pharmaceuticals, cosmetics and household products are presumably substantially greater then the exposure obtained via food and water (Krogh and Jensen (2006)), due to the route of exposure. Alike, in a review of sludge pollution in 2001 the authors found that organic substances are not expected to cause major health problems to the human population. This is based on the inherent properties of the substances which sorb onto sludge, which generally are lipophilic and have low water solubility, rendering them a low bioavailability to plants (Langenkamp et al., 2001a).

Problematic substances as outlined by Krogh and Jensen (2006) are substances which in 'normal doses' have toxic effects on utility plants and animals; cause accumulation in soil or in food chains; and which may pollute surface or ground waters. Such are, suggested by Samsø-Petersen (2002) PCBs, PAHs, PCDD/Fs, and brominated flame-retardants (BRFs), and possibly nitro-musks, chlorinated paraffin's, and polyelectrolytes. LAS, NP, and DEHP were considered to be of "low concern". Contradictory for POPs (PAHs, PCBs, and PCCD/Fs) it is suggested that the environmental impacts exerted by these substances via application of sludge to agricultural land are insignificant. Primarily due to their origin (atmospheric deposition) (Langenkamp et al., 2001a; DEFRA, 2005) but also that levels are dropping in both deposition and sludge owing to mitigating actions. In Norway phthalates, NP/NPE, octylphenol and ethoxylates (OP/OPE), LAS, PCB, PAH as well as some selected pharmaceuticals were investigated for their environmental risk. However, it was concluded that, in this context sludge is not expected to constitute a significant risk to the aquatic environment nor to food producing animals. Risk quotes (RQs) were above 1 for OP and NP for soil-dwelling organisms and LAS (if sludge are applied in park areas) and in surface waters due to sludge application where pyrene and indeno[1,2,3-cd]pyrene had an RQ just above 1. It was found to unlikely that antibiotic resistance may be enhanced when applying sludge as a fertilizer (Eriksen et al., 2009).

Pollutant mitigation and follow-up

The management paradigm for wastewater in Sweden is based on environmental aspects as well as from a financial point of view (1998/99:63) which means that both aspects need to be considered for viable options, which affect the choices of mitigation measures that can be implemented.

Eco-labelling has been pointed out as promotion for enhanced sludge quality by e.g., furthering readily biodegradation of the included substances such as LAS (Tørsløv et al., 1996 and Paulsrud et al., 2000). Source control, by removing unwanted substances from cosmetics, household chemicals and commodities will yield an improved sludge quality. For industrial substances this will be addressed by REACH (2006/121/EC). In the foregoing review Samsø-Petersen (2002) outlined a strategy for risk reduction on both short- and medium-term as well as long-term measures; e.g. on the short/medium-term scale to include treatment; limits to application; identification and assessment of 'new' substances; LVs for sludge, LVs for industrial wastewater; source tracking of selected substances; and implementation of a monitoring programme and on the long-term scale phase-out of substances; revisits with monitoring and source separation of wastewaters. In part this has been addressed and implemented in the ReVAQ project which aimed at safe use of sewage sludge in agriculture by quality control and pro-active measures. Subsequent, WWTPs conforming to ReVAQs approach and can document the effects in the sludge quality (Hugmark, 2007) and be certified by a certification developed by ReVAQ in co-operation with the Technical Research Institute of Sweden.

It has been stated that monitoring must be related to the origin of the sludge as the pollution levels are closely linked the pollutant sources, such as industrial wastewater, municipal wastewater, and stormwater (Langenkamp et al., 2001a). This is also important from source control point-of-view where it is of vital importance to know the sources, uses and emissions in order to make best use of the measures. One identified aspect is the drawback that most of the reviewed studies only base their monitoring on legislation and only a few substances are generally measured, which may result in ignorance to the most important pollutants. In the United States of America (US) a review from 2006 showed that a total of 516 organic substances had been measured in US sludges in 113 studies (Harrison et al., 2006), though the vast majority of the substances had only been measured in one single study. A vast number of substances can also be present in wastewater and sludge as they are used by industry connected to the municipal sewage system. In Eslöv it was found that 42 small and medium size businesses discharged 346 organic substances (Eriksson et al., 2007)

Data availability

The data availability for risk assessment of sewage sludge is generally sparse (see e.g. Zambrzycki, 2009; Eriksen et al., 2009). Literature reviews show knowledge gaps in relation to presence of less known pollutants, these substances' impacts, sludge routes and impact of sludge on soil (Zambrzycki, 2009). Also soil concentrations after application of sludge and on data from long-term periods is in some studies based on estimations due to the lack on measured data (Eriksen et al., 2009). Furthermore, degradation rates are not always measured under comparable environmental conditions so the local situation may be hard to foretell (Eriksen et al., 2009). Aquatic toxicity data are not always present in literature but for the vast majority of the substances in society are the knowledge of the toxicity responses on soil-dwelling organisms/terrestrial organisms limited or lacking (Krogh and Jensen (2006), which may hamper further assessment (Eriksen et al., 2009). Thus, it is important to work according to the precautionary principle where scientific data are insufficient to conduct a full environmental risk assessment (Krogh and Jensen (2006)). Uptake in crops also needs to be further investigated.

Conclusions

The number of studies reporting levels of pollutants in sludge and the fate of organic substances in sludge amended soils has increased since the turn of the millennium. The concentrations are seemingly dropping for many of the studied substances thanks to already implemented mitigating measures. Long-term studies show that pollutant levels increase during the application period but begin to decline when the application stops. The levels have not been found to return to the background levels within the studied time-frames but have not exceeded the levels usually found in urban areas or the predicted no-effect concentrations. In a 'worst case' field test no adverse effects on microorganisms, microbial processes or soil-dwelling organisms could be confirmed. The application of sludge and manure were beneficial for the soil environment. The reviewed studies are in agreement that the risks due to exposure of organic substances in sludge amended soils are insignificant, though there are some discrepancies with regard to individual substances. The conducted risk assessments point out that they are faced with a shortage of environmental data and standardised effect data for terrestrial organisms which have contributed to the level of uncertainty in the studies. Thus, the risk may be underestimated or totally left out as we are ignorant the presence or effect of a certain substance or mixtures of substances.

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