A strategy for reducing pollutants at source in order to obtain sustainable agricultural recycling of wastewater sludge

A. Mattsson, J. Mattsson and F. Davidsson

ABSTRACT

The Swedish licensing system for wastewater sludge use in agriculture, REVAQ, sets challenges. These include a maximum nominal accumulation rate of 0.2%/year on farmland, for specified metals, to be reached by 2025. Here a model is suggested, and applied for the Gothenburg regional wastewater treatment plant, Gryaab, to quantify historic sludge quality improvements and necessary future development. Local sampling campaigns covering two decades show a substantial reduction of heavy metals and ecologically harmful organic substances (such as adsorbable organic halogens, nonylphenols, phthalates, naphthalenes and polycyclic aromatic hydrocarbons) from households and society at large. For the metals studied the historic mass flow reduction to sludge varies from 1 to 2%/year for mercury, zinc and copper to 15%/year for silver. Copper needs further reduction, involving water pipes and copper roofing. Silver is rare in soil, and significant reduction from already low levels is needed to reach the accumulation goal. Further reduction of other metals involves addressing storm- and drainage water entering the sewers and the sediments already in the sewers. Fulfilling the goals of REVAQ implies national and local measures affecting public and private stakeholders including property owners, the wastewater collection system, commercial businesses and legislating authorities.

INTRODUCTION

This paper presents the strategy Gryaab AB has adopted in order to obtain the sustainable recycling of wastewater solids, and thus phosphorus, to agriculture. Gryaab owns and operates the Rya wastewater treatment plant (WWTP) and treats wastewater from 650,000 persons in the Gothenburg region, Sweden. Through a long history of upstream pollutant source management, the Rya WWTP is able to produce sludge with relatively low concentrations of heavy metals (Balmér 2001). With regard to most heavy metals Swedish sludge is among the cleanest in Europe (Salado et al. 2008). The exception is copper, likely due to the widespread use of copper plumbing in buildings. The limits with respect to heavy metals in Sweden are stricter than those of the EU. However even stricter limits concerning heavy metals will be implemented on a national basis. Also a certification system has been implemented in Sweden (REVAQ) as a result of cooperation between stakeholders of agriculture, food industry, retailers and sludge management. The certificate implies improvement of sludge quality in order to guarantee safe reuse of sludge and to meet demands from the agriculture and food producer markets. Reaching these levels is feasible. However as only a minor part of the important pollutants come from major point sources, such as industry, a broader approach will be needed. As also shown by Eriksson et al. (2011) a combination of emission control strategies may be needed in order to minimise mass flow of harmful substances to sensitive end points. Substance flow analysis may be useful in order to identify correct measures (for instance Björklund 2010; Chèvre et al. 2011).

Since the 1960s Gryaab and its owner municipalities have had a team of dedicated personnel involved with reducing pollutants of the wastewater (up-stream management). The focus of this work has developed over time. Although initially focusing on reducing the risk of clogging, degenerating sewers, occupational hazards and protecting the processes of the WWTP, a main focus now is on protecting the receiving water and land from pollutants.
Heavy metals are the main concern in agricultural use of sludge and therefore the main scope in this paper. Our opinion is that hazardous organic substances are a bigger issue for receiving waters than for agricultural land. Most organic substances are degradable in soil environments. Exceptions are PCBs, dioxins, brominated flame retardants and some other substances known to have low biodegradability also in soil.

During the last two decades the major focus has been on improving the sludge quality to achieve safe recycling of nutrients in the sludge to agricultural land. The focus has been on phosphorus as a limited resource. Cordell states that food security cannot be reached without better management of phosphorus (Cordell 2010). During the last decade several stakeholders have shown increased interest in sludge recycling as a means to recycle phosphorus from wastewater. The Swedish parliament has set as a target in the environmental goals that 60% of the phosphorus in wastewater should be recycled by year 2015, whereof at least 50% to agricultural land. On the other hand there is public concern and ongoing debate in Sweden about the presence of pollutants in the sludge used in agriculture (Bengtsson & Tillman 2004).

Gryaab has adopted a sludge strategy which involves four main options: (1) to continuously improve sludge quality in order to meet standards set within REVAQ and thereby making it possible to deliver sludge to agriculture; (2) to use the greater part of the sludge production to produce soil for local land reclamation use; (3) to maintain the possibility to dispose of sludge in a rock cavern formerly used for crude oil storage; (4) to monitor development concerning phosphorus reclamation, incineration, etc.

The aim of the current study is to predict further sludge quality improvements in order to be able to predict how well the sludge will meet the standards set by REVAQ and the Swedish authorities in a business as usual scenario (BAU). The BAU scenario includes several upstream measures which has been ongoing for many years. This includes surveillance and improvement of process sewage from large industrial point sources as well as many minor point sources such as car washes, dental clinics and auto repair garages. In order to communicate with households Gryaab offers a two day educational programme for all secondary school students in the Gryaab region. Gryaab performs a comprehensive analysis programme of the influent wastewater and campaigns for instance to reduce or prohibit cadmium in artist paints and silver in household products.

The further aim of the study is to identify additional measures necessary in order to meet these standards and thus on a long term basis obtain a sustainable agricultural recycle of wastewater sludge.

**METHOD**

The basis of the predictions used in this study is historical data on heavy metals, phosphorus and other substances in the sludge of the Rya WWTP from 1998 to 2009. The concentrations are difficult to compare as they vary depending on process conditions such as improved digestion of sludge, changed conditions at the WWTP or weather conditions. The mass flow of the conservative substances, such as phosphorus and heavy metals, however was found to be more stable and thus useful in order to establish a historic trend. For most of the substances the best fit was obtained assuming a certain annual percentage reduction of substances to the sludge. This approach has the advantage of being conservative. The mass reduction in the coming year will be lower than the reduction the previous year.

Predicting future reduction is of course not as easy as modelling the past. In this study future development of mass flows of heavy metals in the BAU scenario is assumed to be the same or lower than the historic development. How much lower future reductions are assumed to be is based on knowledge of what has caused the historic reduction and how much potential of further reduction can be expected. This approach cannot be expected to give definite answers as to which mass flows are reached in a number of years. However it is a useful tool in identifying substances which will probably not meet target levels in a certain time in a BAU scenario.

For these substances extra measures need to be taken. In order to evaluate relevant measures the different sources of the target substances are identified. This information can then be used in order to plan and compare the efficiency of the different measures available in order to reduce the concentrations of the substances to the WWTP. Many stakeholders must be involved in order to obtain improvements, including local and national legislators, the local water sector and other local and national private and municipal owned companies. Thus the WWTP does not alone control the problem or the solutions. Clear information is important in order to motivate stakeholders to do their share of the necessary improvement work.

In coming years the procedure will be repeated regularly in order to determine if the measures taken are sufficient or additional measures must be taken.
GOTHENBURG REGION CASE STUDY

How has the heavy metal content changed and why?

When Rya WWTP was commissioned in 1971 Gothenburg was a typical industrial city with heavy industry, shipyards, oil refineries and manufacturing industry such as Volvo, SKF, surface treatment and pulp and paper, etc. The air was polluted both by local emissions and from other countries. Since then there has been an ongoing effort to reduce both water and airborne emissions. At the same time the industrial structure in the region has changed. Today there is less heavy industry and more service sector businesses. A drastic reduction in point source emissions as well as bans on the use of cadmium, mercury and lead in most applications have contributed to a better quality of the influent wastewater. Air quality affects the stormwater quality through deposition (Dembélé et al. 2010; Wicke et al. 2010). Improved air quality has resulted in less deposition of metals as well as less corrosion of buildings and other structures. This has improved the quality of the stormwater entering the WWTP, as well as stormwater going to receiving waters (Måansson et al. 2009). As a consequence of this broad positive development, sludge quality has improved. The concentration of six of the seven regulated heavy metals has decreased to between one-tenth and one-third of the concentrations in the 1970s. The exception is for copper where no major change has occurred.

The contribution of heavy metals from households has also decreased. Gryaab has conducted many investigations over the years to identify the most important sources of undesirable contaminants in the influent water. In 1988 and 2006/2007 studies of domestic sewage from two residential areas in Gothenburg have been performed. In one area the inhabitants live in detached houses or terraced houses (700 inhabitants) and in the other, inhabitants live in apartments or small terraced houses (2,400 inhabitants). In both studies 24 h flow proportional composite samples were collected during four different days. These samples were analysed together with the corresponding 24 h composite influent samples at the Rya WWTP for 175 different parameters (2006/2007, fewer in the study of 1988). These included organics, metals, oil and fat, trace elements, surfactants, LAS, phthalates, aromatics, phenols, pesticides, brominated flame retardants, tin organics and dioxins. The results were used to estimate the total domestic discharge for the catchment area served by the Rya WWTP (Table 1).

Most of the priority substances in focus have decreased during the 20-year period from 1988 to 2006/2007 regarding the total amount reaching the Rya WWTP as well as for the contribution per capita from households. Significant decreases were noted for mercury, cadmium, adsorbable organic halogens, nonylphenol, phthalates, naphthalenes and polycyclic aromatic hydrocarbons. Due to an increasing population, by 85,000 persons over 18 years, and a decrease in industrial activity together with a reduced load from

### Table 1 | Results of studies of household wastewater in the Gothenburg region

<table>
<thead>
<tr>
<th></th>
<th>Unit for per capita load</th>
<th>1988</th>
<th>2006/2007</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Per capita load</td>
<td>Household share of total%</td>
</tr>
<tr>
<td><strong>BOD7</strong></td>
<td>g/d</td>
<td>45</td>
<td>51</td>
</tr>
<tr>
<td><strong>COD</strong></td>
<td>g/d</td>
<td>115</td>
<td>49</td>
</tr>
<tr>
<td><strong>Nitrogen</strong></td>
<td>g/d</td>
<td>8.6</td>
<td>62</td>
</tr>
<tr>
<td><strong>Phosphorus</strong></td>
<td>g/d</td>
<td>2.2</td>
<td>63</td>
</tr>
<tr>
<td><strong>Zinc</strong></td>
<td>mg/d</td>
<td>33</td>
<td>38</td>
</tr>
<tr>
<td><strong>Copper</strong></td>
<td>mg/d</td>
<td>9.6</td>
<td>29</td>
</tr>
<tr>
<td><strong>Nickel</strong></td>
<td>mg/d</td>
<td>&lt;2.5</td>
<td>23</td>
</tr>
<tr>
<td><strong>Lead</strong></td>
<td>mg/d</td>
<td>&lt;1</td>
<td>15</td>
</tr>
<tr>
<td><strong>Chromium</strong></td>
<td>mg/d</td>
<td>&lt;1</td>
<td>21</td>
</tr>
<tr>
<td><strong>Mercury</strong></td>
<td>μg/d</td>
<td>61&lt;sup&gt;b&lt;/sup&gt;</td>
<td>40</td>
</tr>
<tr>
<td><strong>Cadmium</strong></td>
<td>μg/d</td>
<td>86</td>
<td>25</td>
</tr>
</tbody>
</table>

<sup>a</sup>Only the area with 700 inhabitants.

<sup>b</sup>If the local dentistry is excluded 20 μg/d.

<sup>c</sup>If the local dentistry is excluded 7 μg/d.
stormwater due to reduced air pollution, the relative contribution in load from households increased by 10–25% between the two investigations. During the same period the number of inhabitants connected to the WWTP has increased by 16%. Considering the fact that people contribute wastewater at work, at school, etc. the contribution of the population to the total mass flows is even greater. For organic matter and nutrient (biochemical oxygen demand (BOD₅), chemical oxygen demand (COD), N and P) the population is estimated to contribute 90% of the total load or more.

Some ecologically harmful organic substances which have come into use during the last 20 years were detected in the samples from 2006/07. Among these were perfluorooctane sulfonate (PFOS), triclosan, tin organics (TBT), brominated flame retardants and dioxins. The total load to the Rya WWTP of these substances was 10–60 kg per year, except for the dioxins which were approximately 10,000 times less.

Where do the pollutants come from?

Stormwater, drainage- and leakage water, larger industries, operators like hospitals, small industries and landfills as well as households, all contribute to the supply of pollutants. Gryaab has conducted comprehensive studies in order to identify the main sources for the seven heavy metals regulated in Sweden. This is exemplified by the relative contribution of different sources of cadmium and copper in Figure 1.

For cadmium, the contribution from households is the dominating source. Apart from when the inhabitants are at home in their residential areas they also contribute at work, in school and so on. However, other sources are equally important, altogether contributing about half of the total load. Most of the other sources have the disadvantage of having a high cadmium/phosphorus ratio, due to their low concentration of phosphorus. Domestic sewage, on the other hand, is rich in nutrients leading to a low and favorable cadmium/phosphorus ratio around 20 mg Cd/kg P as compared with urine 0.6 and faeces 20 (Jönsson et al. 2005). In order to reach a more favorable ratio between nutrients and pollutants the reduction of loads from other sources poor in nutrients is important.

What further improvement is necessary?

The REVAQ certification system sets several criteria regarding which chemical elements should be given priority in the up-stream source management. Apart from criteria related to regulatory limits for agricultural use, any non-essential chemical element (of 60 specified) which accumulates in a standard soil at a rate exceeding 0.2%/year at a given sludge application rate should be given priority. Also the ratio between cadmium and phosphorus should be reduced to a level comparable with that of water from toilets by the year 2025 (17 mg Cd/kg P). For the Rya WWTP sludge, five of the seven presently regulated metals and silver (which is proposed to be regulated) fulfil the criteria to be given priority in the up-stream management (Table 2). Most of these six metals show substantial reductions during the period from 1998 to 2009 (see Table 2 and Figure 2). For cadmium, lead and silver the observed reduction exceeds 5% per year. Zinc, copper and mercury show less reduction. By analysing the mechanisms of the historic reduction a future reduction in a BAU scenario is proposed. Here factors such as how much of easily obtained improvements have already been exhausted have been considered.

<table>
<thead>
<tr>
<th>Table 2</th>
<th>Annual reduction of mass flow with sludge at the Rya WWTP (%/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lead (Pb)</td>
<td>5</td>
</tr>
<tr>
<td>Cadmium (Cd)</td>
<td>5.5</td>
</tr>
<tr>
<td>Copper (Cu)</td>
<td>2</td>
</tr>
<tr>
<td>Mercury (Hg)</td>
<td>1</td>
</tr>
<tr>
<td>Zinc (Zn)</td>
<td>1.5</td>
</tr>
<tr>
<td>Silver (Ag)</td>
<td>15</td>
</tr>
</tbody>
</table>
The reduction needed in order to meet the criteria of REVAQ was calculated. For lead, zinc and silver, reductions in the BAU scenario can be expected to meet the criteria. For silver it is worth noting that future necessary reductions are quite high. No increase can be accepted. Thus it is a concern that silver seems to be used increasingly as a bactericide in some consumer products, including clothing. This development could endanger a favourable development concerning silver. For cadmium, reduction may be enough if the present reduction rate is upheld. However if the reduction rate decreases, as assumed in the BAU scenario, the criteria will not be met. For copper and mercury, the present reduction rate is low and will not meet the criteria. For these three elements further improvement will be needed.

How can the necessary improvements be obtained?

For cadmium a few point sources are being addressed in the region, such as deicing water from the airports and leachate from landfill sites. However, the total amount of cadmium from known major point sources is low, due to decades of up-stream work, and actions concerning these sources can not be expected to make a great impact on the cadmium concentration. In order to meet the targets atmospheric deposition must continue to decrease and the amount reaching the WWTP with stormwater and drainage must decrease further. Also the continued effect of the cadmium ban in Sweden in most applications is likely to further decrease cadmium flow to the WWTP. There have been some exceptions from the cadmium ban. An important exception has been sale of artist paints containing cadmium. Estimates have shown that approximately 10% of the cadmium to Swedish WWTP’s come from artist paints. Since 2009, the rules for the use of cadmium are the same in Sweden as in the rest of the European Union.

Where mercury is concerned, the current decrease of 1%/year is clearly insufficient as the decrease needs to be nearly 6%/year in order to meet the targets for 2025. One component in meeting this target is the proactive and safe removal of mercury that remains ‘stored’ in sewers connected to dentistry and hospitals. The main hospital in Gothenburg contributes about 3% of the total mercury load to the Rya WWTP.

For copper, the sources are known, as are the methods of reducing copper even if they are costly and take a long time to implement. Most of the copper in the wastewater comes from copper pipes used for drinking water distribution within buildings. Copper corrosion is further increased by the raised alkalinity during drinking water production which is intended to decrease the corrosion of the iron pipes of the municipal water distribution system. Choice of pipe materials as well as drinking water quality are keys to success. Of the remaining copper, much of it comes from copper in brake pads for vehicles and from copper roofs via stormwater to combined sewer systems. This may also be solved through the choice of construction materials and sewer system design, in theory simple solutions, which however are costly, time consuming and concerning many different stakeholders with agendas of their own. National regulations will probably be necessary in order to reduce the use of copper piping in buildings, primarily for all new construction and in future for existing installations as well. Due to the leakage of copper to stormwater, some Swedish municipalities have recommendations on the use of new copper roofing.
CONCLUSIONS

The historic trend shows good reduction of heavy metal contributions to the Rya WWTP. This development is the result of ambitious up-stream management in combination with good national legislation.

A method of evaluating historic and modelling future reductions of substances has been developed.

Copper, mercury and cadmium have been identified as substances for which a higher ambition concerning up-stream management and national legislation is needed if the targets are to be met.

The necessary improvements concern several different stakeholders with agendas and interests of their own. It will be a challenge for national and local authorities and water boards to reach these targets. It is also important to consider if the ambition of the targets is appropriate. For this work it is necessary to have clear directives from national and local government concerning the priority of recycling the phosphorus of wastewater sludge to agriculture.

Some important measures which need to be considered are:
- Reducing the pollutants from stormwater
- Reducing atmospheric deposition on surfaces dewatered to the sewage system
- Removing or renovating contaminated sewers (from hospitals/dentistries)
- Further reduction in household contaminants and other non-point sources
- Banning the use of silver in household products
- Change of materials for plumbing and roofing from copper to other materials.

DISCUSSION

Today there are different opinions at the Swedish national authorities regarding whether it is safe or even appropriate recycle sludge to agriculture. The Swedish Environmental Protection Agency has had the task to produce a proposal for a new regulation for reuse of sludge in agriculture. The proposal was presented for consideration in April 2010. Other authorities, such as the Swedish Chemicals Agency and the National Food Administration, have been critical of the proposal. So far there are no new regulations in sight. It will be difficult to get other stakeholders to do their share of the necessary improvement work if the national authorities do not agree on the long-term goals. It is probably essential that the authorities clearly show the way in order to obtain a successful sustainable reuse of sludge.

REFERENCES


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