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The Utilization of Struvite Produced from Human Urine in Agriculture as a Natural Fertilizer: A Review

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Abstract
Most of the nutrients in municipal wastewater originate from urine. Nevertheless, chemical fertilizers are commonly used in the agriculture instead of urine. There are some problems related to the direct utilization of urine, such as micropollutants present in urine, odour and storage of large volume of urine. In wastewater, phosphorus may contribute significantly to the pollution of the aquatic systems. Therefore, wastewater treatment techniques are mainly focusing on removing phosphorus. Phosphorus is collected in the sludge either by a chemical or by a biological process. With the growing concern of micropollutants present, which are in the sludge, the use of sludge in agriculture has been gradually decreasing. It means that the phosphorus content in sludge is not recycled efficiently whereas the use of limited mineral phosphorus resources is growing. To overcome these issues, urine could be collected separately and struvite could be produced. This may recover about 90 % of phosphate in urine. In this paper, the use of human urine and struvite as a fertilizer in the agriculture and the production of struvite is discussed. Results showed that the struvite could be an effective natural fertilizer.

Keywords
human urine, struvite, agriculture, dry toilets, urine-diversion dry toilets, sustainable development

1 Introduction
With the wide-spread use of flush toilets, the human faeces and urine have become a waste to be removed from the sewer system, especially in the developed world. Despite the significant improvement offered by this system, the flush toilets have a lot of harmful environmental effects [1, 2]. The utilization of faeces and urine in agriculture may reduce the amount of chemical fertilizers used, which may result in decreasing energy demand and therefore decreasing CO₂ emission [3]. The basic elements of artificial fertilizers like phosphorus and potassium are estimated to be exhausted. The potassium stocks may be available for hundreds of years, while phosphorus may last about 25 – 100 years. The world’s phosphorus and potassium stocks are finite, and they are non-renewable resources [4]. The wastewater treatment process is complex, expensive and energy demanding. If human excreta, especially urine is separated from the system, the wastewater treatment would be simpler, and the energy demand would decrease [5]. Although stored urine has been used directly in the field [6], it might contain some harmful micropollutants. It is not feasible to transport a large volume of urine or apply it to the fields. Still, the separation of urine and faeces from the sewage system may prevent the harmful effects of flush toilets i.e. high water consumption, the high costs of wastewater treatment for nutrient removal, eutrophication and nitrogen contamination to the groundwater in the case of lacking nutrient removal. Moreover, separation facilitates recycling nutrients for agricultural purposes, which helps to avoid wasting valuable materials in human urine and faeces [7].

There are different methods for using human waste directly as fertilizer such as the use of stored urine, use of dried faecal matter or produce compost from human waste. Urine offers many benefits, but very few farmers
know how to use urine since they are only familiar with humanure which is a mixture of faeces and urine [8]. Moreover, urine is not allowed to be used as fertilizer in the EU. Therefore we have to seek for technics which allow the legal utilisation of urine or nutrients in it. Struvite made of human urine may be a good solution to this problem. The objective of this paper is to introduce struvite and struvite production as an alternative fertilizer. For recovered struvite there is a proposed EU fertilizer regulation criteria [9].

2 Using human urine as fertilizer
2.1 The composition of urine
The daily amount of urine produced by a person accounts only for 0.4-1 % of the daily sewage water volume, but it contains most of the nutrients, such as nitrogen (~ 80 %), phosphorous (~ 50 %) and potassium (~ 60 %) [10-12]. Therefore, the recovery of nutrients is more feasible if urine is separated from the rest of the liquid waste streams in households such as kitchen and shower wastewater. Nitrogen in the urine is mainly in the form of urea, while phosphorus is in the form of superphosphate and potassium is in ionic form, with a ratio of 18:2:5 NPK [6], which is extremely useful for plants. A typical NPK ratio in normal commercial fertilizer can be 9:6:17 [13]. Moreover, human urine contains essential macro and micronutrients (Table 1, Table 2). The nitrogen content of fresh human urine is up to 9 gN/l [14].

Urine is normally free from pathogens when releasing from a healthy person. Some pathogens such as Schistosoma can be found in urine excreted from infected individuals [19]. Even the pathogens are present in urine, they usually die during storage [8], and do not cause any threat to further utilization of urine applied to the soil [20]. The hormones and pharmaceuticals appearing in human urine and faeces are not removed completely with wastewater treatment processes [21-24]. As 60 % of the pharmaceuticals are in urine, urine separation itself is not a solution for the problem of micropollutant. Several techniques exist for removing micropollutants and pharmaceuticals, such as electrodialysis, nanofiltration, ozonation, activated carbon filtration and advanced oxidation [16].

2.2 Urine-diversion toilets
The separation of human urine from other wastewater streams can be implemented by using dry toilets. Several types of dry toilets are available commercially, such as composting toilets or ECOSAN toilets (which handle faeces and urine together), urine-diversion dry toilet (UDDT – separate urine from faeces), incinerating toilets, etc. [25]. The incinerating toilet can produce electrical energy. During the combustion process, a significant amount of valuable nutrients are lost, although elemental analysis of sewage sludge ash showed that it comprised 6-10 % P and 7-18 % Ca [26]. Urine can be collected and managed separately by using urine-diversion dry toilets which toilets are

<table>
<thead>
<tr>
<th>Total-P (g/l)</th>
<th>K (g/l)</th>
<th>SO₄²⁻ (g/l)</th>
<th>Na (g/l)</th>
<th>Cl (g/l)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stored 0-3 months</td>
<td>0.210</td>
<td>0.875</td>
<td>0.225</td>
<td>0.982</td>
<td>2.500</td>
</tr>
<tr>
<td>Stored 6 months</td>
<td>0.200</td>
<td>1.150</td>
<td>0.175</td>
<td>0.938</td>
<td>2.235</td>
</tr>
<tr>
<td>Stored, undiluted</td>
<td>0.540</td>
<td>2.2</td>
<td>0.505</td>
<td>2.6</td>
<td>3.8</td>
</tr>
<tr>
<td>Fresh</td>
<td>0.8-2.0</td>
<td>2.737</td>
<td>1.315</td>
<td>3.45</td>
<td>4.97</td>
</tr>
<tr>
<td>Fresh</td>
<td>0.367</td>
<td>2.170</td>
<td>0.748</td>
<td>2.670</td>
<td>3.830</td>
</tr>
<tr>
<td>Stored</td>
<td>0.076</td>
<td>0.770</td>
<td>0.292</td>
<td>0.837</td>
<td>1.400</td>
</tr>
<tr>
<td>Fresh</td>
<td>0.388</td>
<td>1.870</td>
<td>0.878</td>
<td>3.240</td>
<td>6.620</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Ca (mg/l)</th>
<th>Mg (mg/l)</th>
<th>Mn (mg/l)</th>
<th>Fe (mg/l)</th>
<th>B (mg/l)</th>
<th>Al (mg/l)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stored 0-3 months</td>
<td>15.75</td>
<td>1.63</td>
<td>0</td>
<td>0.205</td>
<td>0.435</td>
<td>0.210</td>
</tr>
<tr>
<td>Stored 6 months</td>
<td>13.34</td>
<td>1.5</td>
<td>0</td>
<td>0.165</td>
<td>0.440</td>
<td>0.185</td>
</tr>
<tr>
<td>Stored, undiluted</td>
<td>0</td>
<td>0</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Fresh</td>
<td>0.233</td>
<td>0.119</td>
<td>0.0019</td>
<td>-</td>
<td>0.097</td>
<td>-</td>
</tr>
<tr>
<td>Fresh</td>
<td>129</td>
<td>77</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Stored</td>
<td>28</td>
<td>1</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Fresh</td>
<td>89.2</td>
<td>45.4</td>
<td>-</td>
<td>-</td>
<td>-</td>
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</tr>
</tbody>
</table>
available commercially [27]. Urine-diversion toilet systems can be considered a sustainable alternative to wastewater management because they allow nutrient recycling and reduce water use [28] (Fig. 1). These types of toilets have been manufactured in several countries including Germany, Finland and Sweden [29].

The urine-diversion toilet has one compartment for faeces and one for urine. The urine flows through separate pipes to a storage tank. Storage is decentralized and uses large tanks that are periodically emptied. For a small flush, the urine-diversion toilet needs 0.0-0.2 litre water, while a modern water-saving toilet uses 2-3 litre. The front compartment of the toilet is used for urine and the back is used for faeces. The mechanism works the following way: if the back compartment is flushed, the front compartment outlet is closed by a valve. One version of the source separated urine system is being explored in the Novaquatis project at Eawag (Swiss Federal Institute of Aquatic Science and Technology). This transdisciplinary research project was concerned with urine source separation to control water pollution by reducing inputs of nutrients and micropollutants and to close the nutrient cycles [31]. Urine stored in-house was released into the sewers at controlled times and was transported to the wastewater treatment plant, where it could be diverted for separate treatment.

Storage is an important factor when using human urine [32]. The storage of urine should last at least one month when it is used as fertilizer for food crops that are not consumed untreated and even six months when it is used for all plants. The container should be airtight during urine storage because nitrogen in urine (in the form of ammonia) is volatile and may lost due to evaporation [33, 34].

3 Using struvite made from human urine in agriculture as fertilizer

3.1 The characteristics of struvite

To overcome the challenge of the direct use of human urine, struvite can be produced. Although urine needs to be transported also in the case of struvite production, its utilization has a lot of advantages. The struvite molecule consists of magnesium, ammonium and phosphate, combined with six water molecules (MgNH₄PO₄·6H₂O: magnesium ammonium phosphate hydrate, M-A-P). The molecular weight of struvite is 245.43 g mol⁻¹. Struvite contains 15.4 % Mg, 4.3 % NH₄⁺, and 35.6 % PO₄³⁻. It is a crystalline material precipitated from human urine or other waste [35, 36]. Struvite also exists as a natural mineral. It is a white, odourless powder (Fig. 2). Struvite is easily storable, transportable and applicable, especially in granulated form [37]. The struvite precipitation process is an attractive method because it can remove and recover simultaneously P and N [38]. Moreover, during struvite precipitation, more than 98 % of hormones and pharmaceuticals remain in the solution [39]. Heavy metals in struvite precipitated from normal urine have not being detected.

3.2 The production of struvite

Phosphate recovery as struvite is based on a single chamber microbial electrolysis cell [42], but controlled struvite recovery from wastewater or from human urine can be achieved with chemical reaction too. Crystal precipitation occurs when concentrations of Mg²⁺, NH₄⁺ and PO₄³⁻ exceed the solubility limit for struvite formation. Urine contains phosphate (PO₄³⁻) and ammonium (NH₄⁺) (Fig. 3(a)), if magnesium is added to the urine (Fig. 3(b)), than the phosphate, ammonium and magnesium react and form crystalline struvite (Fig. 3(c)). This crystal can be filtered, collected and turned into fine powder (Fig. 3(d)) [40]. The formation of struvite by chemical addition is illustrated in Fig. 3.

The struvite crystals are formed in alkaline conditions according to Eq. (1) [43]:

$$\text{Mg}^{2+} + \text{NH}_4^+ + \text{PO}_4^{3-} + 6\text{H}_2\text{O} \rightarrow \text{MgNH}_2\text{PO}_4 \cdot 6\text{H}_2\text{O}$$

(1)
Due to the production of struvite, 90% of phosphorus can be recovered from urine [40]. A good example of a small-scale process suitable e.g. for rural areas is described in [40]. Production takes place in a stirred reactor, which can be built easily, using locally available materials. Below the reactor valve, a filter-bag hangs to collect the struvite [40]. To start the process, urine and magnesium are mixed in the reaction tank for 10 minutes. After this, the valve is opened and the suspension is draining into the filter bag. The filter bag retains the struvite while the effluent passes through. The filter bag has to be air dried for one or two days in the sun and then the struvite is ready to use (Fig. 4). According to the field experiments, this type of reactor was able to recover 90% of total phosphate from urine [40]. As struvite also precipitates naturally from urine, any precipitate in the collection system should be incorporated into the final product, in order to maximise the nutrient recovery. It is important that magnesium has to be in soluble form, in sufficient quantity and at an affordable price to operate a profitable struvite producer plant [40].

There are several struvite producing companies e.g. Ostara, Multiform harvest, Nuresys in Belgium, Sustec and Paques in the Netherlands and Suez and Veolia in France. For example; Ostera produce struvite from wastewater using the same method. Phosphorus recovery happens during controlled struvite precipitation. Dewatering liquors are added to the reactor together with magnesium. The struvite granules grow in diameter resulting in a pure fertilizer sold as Crystal Green®. The treated effluent is discharged from the top of the reactor and returned to the plant. Once granules have reached the desired size, they are removed from the reactor. Granules are washed and they are conveyed to dewatering sieve, where being dried using hot air. Thereafter they are delivered to classifying screen, before being deposited in silos. The product is then ready for bagging in bags and loading onto trucks for transport to fertilizer costumers [45].

Many factors have a significant role in the efficiency of struvite precipitation, such as pH, the concentration and molar ratios of Mg²⁺, NH₄⁺ and PO₄³⁻, temperature, aeration rate and presence of Ca²⁺ in the reacting media [46-49]. Struvite can precipitate between 7.0 and 11.5 pH, but the most appropriate pH is between 7.5 and 9 [48]. The pH has an effect on the growing speed of crystals as well as the quality of the precipitated crystals [50, 51].

3.3 The utilization of struvite in the agriculture

Struvite acts the same way as diammonium-phosphate fertilizer (DAP) does [40]. It releases nutrients slowly, which can be favourable in the agriculture. As the solubility of struvite is low (0.033 g 100 ml⁻¹ in weakly acidic water) [47], its leaching from soil is limited. The application of human urine has to face with low social acceptance, but the odourless struvite product made of human urine usually has a good acceptance among farmers [40]. Struvite has been successfully used in the case of turf grass, tree seedlings, ornamentals, vegetables, flower boards and garden grass as [49, 52-54] fertilizer, and it was deemed a great success. Moreover, struvite may be the most efficient fertilizer for crops that need magnesium, like sugar beet [55]. It could be very effective also for those crops, which need slow-soluble fertilizers. However, its slowness is usually a problem in the case of some crops [56].

The agricultural utilisation of struvite made of human urine has many advantages. Struvite is odourless, almost free of hormones and pharmaceuticals due to the precipitation process and it has no heavy metal content.
Its production technology is very simple and can be implemented and operated almost anywhere. Due to its reduced weight and volume and its granular form, the transportation, storage and application is easier compared to urine. The social acceptance of urine as fertilizer is low, but struvite is well accepted. According to field experiments, struvite is a very effective fertilizer, which releases slowly. However, there are some disadvantages concerning the agricultural utilization of struvite. The cost of the establishment of the reactor and the urine collector system is significant and additional soluble magnesium source is necessary for the production. Because of struvite's low solubility, intake of struvite is slow for many crops, but it ensures stable nutrient supply. Crop yield may be lower than in the case of conventional fertilization [40, 57].

4 Conclusions
In our opinion, using human urine in the form of struvite for agricultural purposes is reasonable. This is not only because the harmful effects of inappropriate wastewater treatment on freshwater bodies can be reduced, but also because the basic materials of artificial fertilizer are going to be exhausted in the future. Struvite production from human urine offers an appropriate and suitable solution to take back the nutrient content of urine into the natural cycles.

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