Optimization of resource and water recovery from urine

Olivier Lefebvre, Jiangyong Hu, Say Leong Ong and How Yong Ng

ABSTRACT

This study deals with the feasibility and practicality to recover water and nutrients from fresh urine by means of evaporation/condensation. The evaporation process generated two distinct fractions: a condensate and a concentrate. The optimal percentage of evaporation (in volume) was found to be 80%, resulting in optimal condensate quality. Higher percentages of evaporation resulted in a deterioration of water quality, as urea decomposed into ammonia, followed by volatilization of the ammonia which ended up in the condensate. Following evaporation, struvite was recovered from the concentrate at an optimal Mg/N ratio of 1/1. The condensate was purified by filtration into two layers of soil and sand, followed by zeolites. Complete removal of $N-NH_3$ and 91% removal of chemical oxygen demand (COD) could be achieved throughout this process. Finally, the condensate was disinfected by sodium hypochlorite, achieving over 6-log inactivation of MS2 bacteriophage at a dose of 1,200 mg min/L. In conclusion, this study shows that there exist some potential benefits to the production of high-quality water and fertilizer from urine. The value of struvite recovered from the concentrate was found to be equivalent to that of the water from the condensate, showing that both streams deserve equal attention.

Key words | fertilizer, resource recovery, struvite, urine diversion dehydration toilet, used water, water reuse

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INTRODUCTION

For over a decade, there has been an ongoing shift of paradigm in the approach of wastewater treatment and technology (Guest *et al.* 2009). Instead of dissipating the resources, it has been increasingly acknowledged that there is a need to turn to maximum use of resources present in wastewater, which should really be referred to instead as 'used water' (Verstraete *et al.* 2009). Therefore, sewage should be considered as a renewable resource, from which value can be extracted in the form of water, energy and nutrients (mostly nitrogen, N and phosphorus, P). Based on an approximate cost of 0.25 €/m^3, water represents about two-thirds of the total value of wastewater (Verstraete *et al.* 2009), making it cost-effective to implement water reuse in various parts of the world where there is shortage of water, as is the case in many areas and especially in

most of the developing world. Although many experiences in the world have shown the promise of water reuse, it is less technically and economically feasible to recover N and P, which are too diluted in the wastewater matrix. Indeed, urine makes less than 1% of the total volume of domestic wastewater, but contributes 81% of the nitrogen and 50% of the phosphorus (Guest *et al.* 2009).

An approach to overcome this problem is sewage pre-concentration by chemically enhanced primary treatment, using iron and aluminium-based coagulants (Diamantis *et al.* 2013). Another approach consists of the crystallization process, which allows reducing waste production as well as recovering various materials (Aldaco *et al.* 2008). Although both techniques seem promising, they have yet to be applied in full-scale sewage treatment plants. A simpler and most

cost-effective approach may lie in source separation and decentralized treatment of urine and faeces (Kulkarni 2010). One of the limitations commonly associated with source separation of human waste is the need to rethink our sewage infrastructure; however, nearly 2.6 billion people around the world still do not have access to sanitation and a large fraction also lacks access to clean water, especially in rural areas in the developing world (WHO & UNICEF 2010). This means that there is huge potential to implement such practice in the developing world, which, as shown above, would in turn provide valuable and recyclable resources such as energy, urea, salts, minerals and water.

Human urine is a complex aqueous solution that mostly contains water (over 95%), sodium chloride (NaCl) and urea $(CO(NH_2)_2)$ as dominant compounds, but also potassium (K^+), and phosphate (PO_4^{3-}) in lower concentrations (WHO & UNICEF 2010). This makes urine an important source of nitrogen and phosphorus and, in fact, most of the nutrients that people eat and drink are passed on in their urine. The recovery of phosphorus from urine is especially of crucial importance because of the limited availability of phosphate rocks found in nature (Rodehutscord 2008). It has been estimated, indeed, that our reserves of phosphorus will last for less than a hundred years, after which there will be no other choice but to recover it from water bodies, due to the very slow regeneration of phosphate rocks over geological times. Lately it has been suggested that struvite, also known as magnesium ammonium phosphate and consisting of magnesium, ammonium and phosphorus in equal molar concentrations $(MgNH_4PO_4.6H_2O)$, could be obtained from sewage or urine and used in agriculture as a slow-release fertilizer (Guest *et al.* 2009; Verstraete *et al.* 2009).

In our previous report (Lefebvre *et al.* 2013), we disclosed a novel, sustainable and low-cost decentralized sanitary system that uses biochar or animal dung to (i) dry faeces, such that it can be combusted and converted into ashes and (ii) extract water from urine by boiling it under reduced-pressure conditions. Building on that experience, the objective of this work is to provide an overview of the feasibility and practicality to recover water and nutrients from fresh urine by means of evaporation/condensation, followed by distinct treatment processes for both the concentrate (to produce struvite) and the condensate (to recover clean water).

MATERIALS AND METHODS

Urine evaporation

Urine evaporation was conducted in the laboratory with a rotary evaporator (IKA RV 10, Singapore). For each experimental batch, 500 mL of urine, freshly collected from a panel of ten individuals, were evaporated at 78 °C and −200 mbar, and water samples were collected at different time intervals, when the volume of the condensate (and therefore of urine evaporated) reached 10%, 50%, 60%, 80%, 90%, 95% and 100%, respectively. The evaporation process therefore generated two distinct fractions: a condensate and a concentrate.

Struvite formation from the concentrate

Struvite formation was induced by addition of magnesium oxide (MgO) at different Mg/N molar ratios ranging from 1/1 to 3/1 into 100 mL of the concentrate fraction collected after 80% of the original volume of urine was evaporated. The mixture was allowed to stir for 40 min and then the crystals of struvite were recovered by filtration through a 0.45-μm glassfibre filter, followed by drying and weighing.

Condensate filtration and disinfection

Filtration experiments were carried out on the condensate fraction by gravity in an open tank (total volume of approximately 20 L). Two layers of filtration materials were used in the form of (i) a top layer (20-cm height) of soil obtained from a local plant nursery (dark-brown colour; density around 1.1 g/cm³, sandy loam texture) and (ii) a bottom layer (13-cm height) of two types of sands, consisting of silica fine sand (10-cm height, light-brown colour, diameter below 1 mm, density around 1.7 g/cm³) over coarse sand (3-cm height, grey-brown colour, diameter 2–5 mm, density around 2.0 g/cm³). Following this filtration tank was another tank of similar dimensions, where additional gravity-driven post-filtration treatment was provided in the form of a layer (15 cm) of zeolites (clinoptilolite, light-green colour, diameter 1–2 mm, density around 1.1 g/cm³).

Following filtration, disinfection studies were carried out on the condensate using sodium hypochlorite. MS2

coliphage ATCC 15597-B1 was chosen as a model of virus in water. Incubation with *Escherichia coli* was conducted at 37 °C and 120 rpm for 20 hours. The MS2 suspension was then centrifuged at 3,600 g for 10 min and the supernatant was passed through a 0.22 μm sterile filter. Detection of MS2 coliphage relied on the standard double agar overlay technique, following the method of Butkus *et al.* (2004).
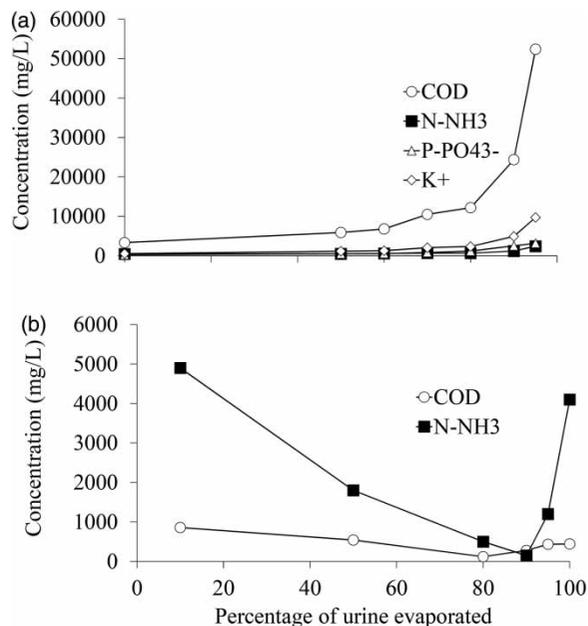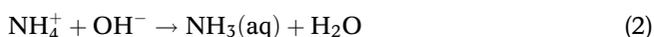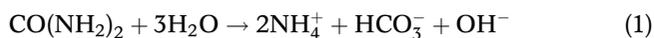
Analyses

pH and conductivity were determined according to *Standard Methods* (APHA 2005). Chemical oxygen demand (COD) was determined by the closed reflux titrimetric method, ammonia (N-NH$_3$) was analysed by spectrophotometry, and potassium (K$^+$) and phosphates (P-PO$_4^{3-}$) were determined by ion chromatography (Dionex, USA), after filtration using 0.45-μm glassfibre filters.

Figure 1 | Characteristics of (a) concentrate and (b) condensate fractions of urine at various percentages of urine evaporated.

RESULTS AND DISCUSSION

Optimization of evaporation rate

Urine was evaporated so as to collect two fractions: a condensate and a concentrate. Different percentages of urine volumes were allowed to evaporate and the quality of both fractions was analysed by conventional water quality parameters, as a function of the fraction of urine which had been evaporated. The pH of the concentrate and condensate remained stable at 6.7 ± 0.3 (close to that of raw urine) and 9.9 ± 0.3, respectively. In the concentrate, the COD, N-NH$_3$, P-PO$_4^{3-}$ and K$^+$ concentrations increased regularly as the fraction became more concentrated, which was expected (cf. Figure 1(a)). The higher pH of the condensate could be explained by the presence of ammonia, resulting from the volatilization of urea as well as its decomposition into ammonia at high temperature, followed by volatilization of the ammonia, as displayed in Equations (1)–(3).

$$CO(NH_2)_2 + 3H_2O \rightarrow 2NH_4^+ + HCO_3^- + OH^- \tag{1}$$

$$NH_4^+ + OH^- \rightarrow NH_3(aq) + H_2O \tag{2}$$

$$NH_3(aq) \leftrightarrow NH_3(g) \tag{3}$$

Indeed, a large fraction of the ammonia was found in the condensate and this was maximal at the beginning of the evaporation process. Figure 1(b) clearly shows that the concentration of N-NH$_3$ was maximal (4,900 mg/L) when the percentage of urine evaporated attained 10%. It was then reduced, showing that the quality of the condensate recovered from urine after this initial fraction improved. In other words, a large amount of ammonia was released initially upon heating and the subsequent fraction of water evaporated was of better quality. When 90% of the urine was evaporated, the concentration of N-NH$_3$ was at its lowest at 150 mg/L. However, further evaporation of urine provoked another increase of ammonia concentration in the condensate and this could be explained by further degradation of urea at high temperature, a phenomenon previously described by Liu *et al.* (2008). The results on ammonia were confirmed with the COD, which reached a minimum of 120 mg/L at 80% of water recovery and then increased when more condensate was recovered (Figure 1(b)). Again, this can be explained by urea volatilization ending up in the condensate fraction.

In conclusion, these results show that the COD and nutrient content (N, P and K) of the concentrate increased as water was withdrawn from this fraction, making it richer in nutrients and a more suitable fertilizer than raw urine. An optimal water recovery percentage of 80% led to the optimal quality of the condensate recovered. It should be noted here that the time required to evaporate 80% of the urine was 1.5 h under our experimental conditions.

The reduction in volume would certainly facilitate storage and transportation. One of the potential drawbacks to storing and transporting the concentrated urine could again be the potential losses of ammonia by evaporation, following Equations (1)–(3). Yet, this is not a concern that is specific to our process, as fresh urine is also known to experience hydrolysis of urea within a day of storage. This phenomenon, known as ureolysis, typically leads to an increase in pH and to the production of gaseous ammonia and bicarbonate ions, by Equations (1)–(3) (Hellström *et al.* 1999). Therefore, it would be useful in further studies to compare the losses of ammonia during storage and transportation of concentrated urine with that of fresh urine, possibly by using our decentralized sanitary system in the field (Lefebvre *et al.* 2013). In practice, Hellström *et al.* (1999) reported that such losses are generally small in the field.

Struvite production from the concentrate stream

Despite the high nutrient content of the urine concentrate, its salinity would be too high for agricultural and landscape purposes, as previously demonstrated by Beler-Baykal *et al.* (2011), who used raw urine to grow a variety of *Ficus elastica*. Indeed, the conductivity of the condensate increased from 0.25 to 1.16 S/m (about 1/5 that of seawater) after 80% of the urine was evaporated (data not shown). Therefore, in our experiments, struvite was formed by addition of MgO into the concentrate collected after 80% of the urine was evaporated, corresponding to the optimal quality of the condensate recovered. Struvite was formed according to Equation (4) and the results are shown in Figure 2.

$$Mg^{2+} + NH_4^+ + PO_4^{3-} + 6H_2O \rightarrow MgNH_4PO_4(H_2O)_6 \quad (4)$$

It is apparent from Figure 2 that the optimal molar ratio of Mg/N equalled 1:1. In these conditions, the P-PO$_4^{3-}$
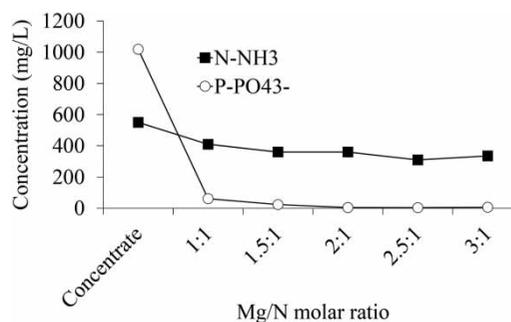
Figure 2 | Water quality of the concentrate following struvite precipitation induced by various molar ratios of Mg/N.

concentration in the concentrate was reduced from 1,018 to 61 mg/L, i.e., a 94% reduction. At the same time, a moderate reduction of N-NH$_3$ was also observed from 550 to 410 mg/L, i.e., a 25% reduction. Increasing the dose of MgO did not significantly alter these results and it is evident that phosphate was the limiting factor. This finding is in accordance with Equation (3) (Mg, N and P should be in equimolar quantities) and also with the literature, where P-recovery as struvite in the range of 90–97% has indeed been reported from digested sludge under a similar Mg/N equimolar ratio (Arakane *et al.* 2006; Shimamura *et al.* 2006). In the optimal conditions, 4.7 ± 0.7 g-struvite/L-concentrate could be recovered in the typical form of a white crystalline substance. Based on an MgO cost of 150 €/tonne and a struvite value of 250 €/tonne, the potential benefit derived from struvite could be estimated at 0.9 €/m³ of concentrate or 0.2 €/m³ of raw urine. Of course, this estimate does not take into account capital and operational costs and, in comparison, others have estimated the total costs of producing struvite in the range of 220–2,750 €/tonne (Doyle & Parsons 2002; Neethling & Benisch 2004).

Water recovery from the condensate stream

Even though the condensate obtained after 80% of the urine was evaporated displayed the optimal quality, additional treatment is still required if water reuse or recycling is to be achieved. A polishing treatment in the form of filtration was therefore applied. Table 1 shows the water quality of the raw condensate and the quality of the final product after sequential filtration by soil, sand and finally zeolite

Table 1 | Water quality of the condensate throughout the sequential filtration treatment

Parameters	Raw condensate	Sequential filtration treatment		
		Soil	Sand	Zeolite
COD (mg/L)	220	90	60	20
N-NH$_3$ (mg/L)	1,160	10	2	bdl
pH	11	8	8	8

bdl = below detection limit.

(clinoptilolite). Complete removal of N-NH$_3$ and 91% removal of COD could be achieved through this process, which is in accordance with the literature where up to 97% of the ammonium in stored urine could be transferred onto clinoptilolite through ion exchange (Beler-Baykal *et al.* 2011). It should be noted here that, once the filtration materials (soil, sand and zeolite) have adsorbed the nitrogen from urine, they could potentially be used as a soil conditioner. The application of the condensate on the soil would indeed be expected to raise the soil pH and increase its value as a fertilizer. However, the impact of such fertilization would require further investigation, with respect to crop production and pollution.

Finally, the product water was disinfected by sodium hypochlorite, showing over 6-log inactivation of MS2 at a dose of 1,200 mg min/L of sodium hypochlorite solution, corresponding to an effective chlorine concentration of 10–15% (Figure 3). This is higher than the 4-log inactivation of MS2 recommended by the World Health Organization
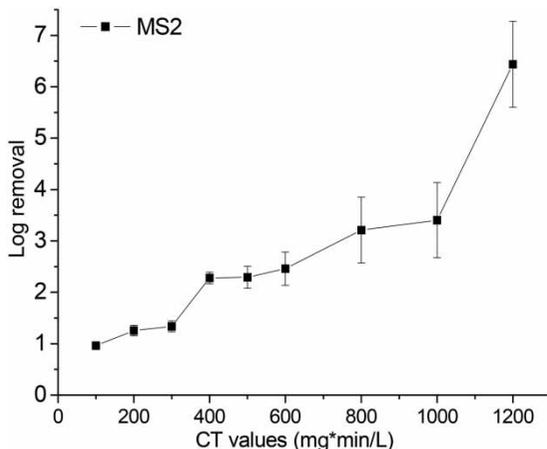
Figure 3 | Sodium hypochlorite inactivation of MS2 bacteriophage.

for water reuse. Based on a water cost of 0.25 €/m^3 and a percentage of water recovery of 80%, there is a benefit from water reuse to be made of about 0.2 €/m^3 of urine, similar to the benefit made from struvite recovery.

CONCLUSION

In this study, we have shown that there exist some potential benefits to the production of high-quality water and fertilizer from urine. The monetary benefit that can potentially be made from crystallizing struvite from the concentrate was found to be equivalent to that made from recovering water from the condensate, showing that both streams deserve equal attention. However, the experiments made in the laboratory should be confirmed at full-scale using our prototype urine-diversion dehydration toilet. In particular, it is necessary to verify that the energy needed to evaporate the urine can either be offset by the value of the products (struvite and water) or can be provided by the burning of the faeces. Moreover, urine characteristics are known to change significantly throughout storage as the predominant form of nitrogen shifts from urea to ammonium and, in the field, it may not be practical to deploy a continuous system where urine is not allowed to settle. Finally, there is, of course, a need to identify the various other barriers to adoption, which could be political and social.

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REFERENCES

Aldaco, R., Garea, A., Fernandez, I. & Irabien, A. 2008 Resources reduction in the fluorine industry: fluoride removal and recovery in a fluidized bed crystallizer. *Clean Technol. Environ. Policy* **10**, 203–210.

APHA 2005 *Standard Methods for the Examination of Water and Wastewater*. American Public Health Association, Washington, DC, USA.

Arakane, M., Imai, T., Murakami, S., Takeuchi, M., Ukita, M., Sekine, M. & Higuchi, T. 2006 Resource recovery from excess sludge by subcritical water combined with magnesium ammonium phosphate process. *Water Sci. Technol.* **54** (9), 81–86.

Beler-Baykal, B., Allar, A. D. & Bayram, S. 2011 Nitrogen recovery from source-separated human urine using clinoptilolite and preliminary results of its use as fertilizer. *Water Sci. Technol.* **63** (4), 811–817.

Butkus, M. A., Labare, M. P., Starke, J. A., Moon, K. & Talbot, M. 2004 Use of aqueous silver to enhance inactivation of coliphage MS-2 by UV disinfection. *Appl. Environ. Microbiol.* **70**, 2848–2853.

Diamantis, V., Verstraete, W., Eftaxias, A., Bundervoet, B., Vlaeminck, S. E., Melidis, P. & Aivasidis, A. 2013 Sewage pre-concentration for maximum recovery and reuse at decentralized level. *Water Sci. Technol.* **67** (6), 1188–1193.

Doyle, J. D. & Parsons, S. A. 2002 Struvite formation, control and recovery. *Water Res.* **36**, 3925–3940.

Guest, J. S., Skerlos, S. J., Barnard, J. L., Beck, M. B., Daigger, G. T., Hilger, H., Jackson, S. J., Karvazy, K., Kelly, L., Macpherson, L., Mihelcic, J. R., Pramanik, A., Raskin, L., Van Loosdrecht, M. C. M., Yeh, D. & Love, N. G. 2009 A new planning and design paradigm to achieve sustainable resource recovery from wastewater. *Environ. Sci. Technol.* **43**, 6126–6130.

Hellström, D., Johansson, E. & Grennberg, K. 1999 Storage of human urine: acidification as a method to inhibit decomposition of urea. *Ecol. Eng.* **12**, 253–269.

Kulkarni, S. 2010 Urine-diversion Dehydration Toilets (UDDTs). In: *SSWM Expert Training Course*, Pune, India.

Lefebvre, O., Wang, M. & Ng, H. Y. 2013 Tackling the sanitation challenge with water and energy reuse from human wastes. In: *9th IWA International Conference on Water Reuse*, Windhoek, Namibia.

Liu, Z., Zhao, Q., Wang, K., Lee, D., Qiu, W. & Wang, J. 2008 Urea hydrolysis and recovery of nitrogen and phosphorous as MAP from stale human urine. *J. Environ. Sci. (China)* **20**, 1018–1024.

Neethling, J. B. & Benisch, M. 2004 Struvite control through process and facility design as well as operation strategy. *Water Sci. Technol.* **49** (2), 191–199.

Rodehutscord, M. 2008 Approaches for saving limited phosphate resources. *Arch. Tierz.-Arch. Anim. Breed.* **51**, 39–48.

Shimamura, K., Hirasawa, I., Ishikawa, H. & Tanaka, T. 2006 Phosphorus recovery in a fluidized bed crystallization reactor. *J. Chem. Eng. Jap.* **39**, 1119–1127.

Verstraete, W., Van de Caveye, P. & Diamantis, V. 2009 Maximum use of resources present in domestic 'used water'. *Bioresour. Technol.* **100**, 5537–5545.

WHO & UNICEF 2010 *Progress on sanitation and drinking-water: 2010 Update*. UNICEF/WHO, New York/Geneva.