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© 2020 Elsevier Ltd This study demonstrates the full scale application of iron dosing in a metropolitan wastewater treatment plant (WWTP) and the upstream sewer system for multiple benefits. Two different dosing locations, i.e., the WWTP inlet works (Trial-1) and upstream sewer network (Trial-2) were tested in this study. Both dosing trials achieved multiple benefits such as sulfide control, phosphate removal and improved sludge dewaterability. During Trial-1, a sulfide reduction of >90% was achieved at high dosing rates (>19 kgFe ML\(^{-1}\)) of ferrous chloride in the inlet works and in Trial-2 the in-sewer ferrous dosing had significant gas phase hydrogen sulfide (H\(_2\)S) concentration reduction in the sewer network. The ferrous dosing enhanced the phosphate removal in the bioreactor up to 76% and 53 ± 2% during Trial-1 & 2, respectively. The iron ending up in the anaerobic sludge digester reduced the biogas H\(_2\)S concentration by up to 36% and 45%, respectively. The dewaterability of the digested sludge was improved, with relative increases of 9.7% and 9.8%, respectively. The presence of primary clarifier showed limited impact on the downstream availability of iron for achieving the afore-mentioned multiple benefits. The iron dosing enhanced the total chemical oxygen demand removal in the primary clarifier reaching up to 49% at the high dose rates during Trial-1 and 42 ± 1% during Trial-2. This study demonstrated that multiple benefits could be achieved independent of the iron dosing location (i.e., at the WWTP inlet or in the network). Further, iron dosing at both locations enhances primary settling, beneficial for bioenergy recovery from wastewater.

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Abstract

This study demonstrates the full scale application of iron dosing in a metropolitan wastewater treatment plant (WWTP) and the upstream sewer system for multiple benefits. Two different dosing locations, i.e., the WWTP inlet works (Trial-1) and upstream sewer network (Trial-2) were tested in this study. Both dosing trials achieved multiple benefits such as sulfide control, phosphate removal and improved sludge dewaterability. During Trial-1, a sulfide reduction of >90% was achieved at high dosing rates (>19 kgFe ML⁻¹) of ferrous chloride in the inlet works and in Trial-2 the in-sewer ferrous dosing had significant gas phase hydrogen sulfide (H₂S) concentration reduction in the sewer network. The ferrous dosing enhanced the phosphate removal in the bioreactor up to 76% and 53±2% during Trial-1 & 2, respectively. The iron ending up in the anaerobic sludge digester reduced the biogas H₂S concentration by up to 36% and 45%, respectively. The dewaterability of the digested sludge was improved, with relative increases of 9.7% and 9.8%, respectively. The presence of primary clarifier showed limited impact on the downstream availability of iron for achieving the aforementioned multiple benefits. The iron dosing enhanced the total chemical oxygen demand removal in the primary clarifier reaching up to 49% at the high dose rates during Trial-1 and 42±1% during Trial-2. This study demonstrated that multiple benefits could be achieved independent of the iron dosing location (i.e., at the WWTP inlet or in the network). Further,
Iron dosing at both locations enhances primary settling, beneficial for bioenergy recovery from wastewater.

**Keywords:** Ferrous dosing, sulfide control, phosphate removal, dewaterability, multiple benefits.

**Highlights**

- Demonstration of multiple benefits of ferrous dosing in urban wastewater system
- Ferrous dosing achieved H₂S control, enhanced P removal and sludge dewaterability
- Both ferrous dosing in sewers and WWTP inlet achieved these multiple benefits
- Primary clarifier had limited effects on the downstream availability of iron
1. Introduction

The wastewater generated in households and industries are collected and transferred to wastewater treatment plants (WWTPs) where it is treated before being discharged to the environment. Wastewater treatment induces several key challenges starting from wastewater transportation till the discharge of treated effluents. These challenges include

1) During wastewater transportation in sewers, hydrogen sulfide (H$_2$S), produced by sulfate reduction, transfers to the gas phase and causes serious corrosion, malodour and human health problems (Jiang et al. 2015, Jiang et al. 2016, Li et al. 2017). The H$_2$S induced concrete corrosion possess serious threat to underground sewer infrastructure, an important asset through enormous investment. The asset loss is estimated at US$ 14 billion annually in the USA alone (Pikaar et al. 2014).

2) The phosphorus removal from the wastewater stream is another key challenge in WWTPs. As the main nutrient responsible for eutrophication, the discharge of phosphorus is strictly regulated in many parts of the world (Conley et al. 2009, Tchobanoglous et al. 2014).

3) The sludge generated in the WWTPs are biologically stabilised before being discharged to reduce its volume and mass. The biological stabilisation, usually through anaerobic digestion produces biogas which is a combination of methane and carbon dioxide, but also contains H$_2$S. The H$_2$S concentration even at the concentrations of >2 ppm was believed to cause concrete corrosion in the sewer environment (O'Dea 2007). Hence, the presence of H$_2$S in biogas is undesirable and can cause corrosion in the piping, gas storage tanks and cogeneration engines (Charles et al. 2006).

4) The specific sludge production in WWTP varies widely from 35 to 85 g dry solids per population equivalent per day (Foladori et al. 2010). Mechanical dewatering of the sludge is the common technology employed in WWTPs to separate the solids and...
liquids and to reduce the sludge volume for its easy transportation. Chemicals are being used to enhance the efficiency of dewatering (i.e., increasing the solids separation). The sludge treatment and disposal are expensive that cost 60% of the total operation cost of a conventional WWTP (Low et al. 2000, Zhou et al. 2015).

These problems in the urban wastewater system (sewers and WWTPs) are often managed separately. The most common technique is the dosage of chemicals separately at different locations in the urban wastewater system. For example, chemicals were commonly used in the control of sulfide in the sewers, including i) chemical oxidation of sulfide by the injection/addition of air, pure oxygen, nitrate salts (Jiang et al. 2009, Ochi et al. 1998, Okabe et al. 2003, Tanaka and Takenaka 1995) ii) pH elevation by the addition of alkali such as Mg(OH)\(_2\) and iii) precipitation of sulfide by forming insoluble metal particles through the addition of metal salts such as iron, zinc, lead and copper salts (Zhang et al. 2009). Each method has its own disadvantages such as high energy and maintenance requirement during the injection of oxygen due to its less solubility that requires multiple injection points (Firer et al. 2008). Similarly, dosing of Mg(OH)\(_2\) requires high dosage due to the strong buffering capacity of wastewater (Jensen 1990) and nitrate addition to the sewers increases the nitrate load to the downstream treatment process (Firer et al. 2008). Iron salt dosing also has disadvantages such as flocculation and settling in the sewer pipes and possible drop in the pH due to high dosage. But among the other methods, iron salt dosing was widely used approach and has greater advantages because of its specificity to H\(_2\)S, less toxicity (i.e. no production of any harmful by-products) and relatively low cost (Firer et al. 2008, Jameel 1989). In Australia, iron salts accounted for 66% of the sewage flow dosed with chemicals for sulfide control (Ganigue et al. 2011). The iron salt dosing in the sewers effectively controls the dissolved sulfide by forming iron sulfide precipitates (Ganigué et al. 2018, Gutierrez et al. 2010, Haaning Nielsen et al. 2005). These studies showed that ferrous salts (Fe\(^{2+}\)) reacts directly with sulfide
forming ferrous sulfide precipitates (FeS), while ferric (Fe$^{3+}$) ions oxidizes sulfide chemically to elemental sulfur, with itself being reduced to Fe$^{2+}$, which subsequently precipitates sulfide. Iron salts either in the form of ferrous or ferric are also dosed in the activated sludge bioreactor for the chemical precipitation of phosphate (Oikonomidis et al. 2010, Singer 1972). Also, a small dosing of iron in the form of ferric to anaerobic digester (27 mgFe L$^{-1}$) showed a complete removal of dissolved sulfide by precipitation, thus reducing H$_2$S emission to the biogas (Charles et al. 2006).

With the usage of chemical dosing at multiple locations of the urban wastewater system, the cost of chemicals is crucial for the wastewater management. The cost of inorganic coagulants (such as aluminum or ferric salts) alone was US$ 1.37 billion and predicted to increase to US$ 1.84 billion by 2023 (BCC Research 2018). To reduce the chemical cost and associated environmental footprint, an integrated approach to corrosion control in sewers, phosphorus removal and sulfide control in the biogas is necessary for the wastewater treatment utilities.

The dosing of ferrous or ferric in the sewer network changes the composition of the wastewater and it could have multiple impacts on the downstream processes in the WWTP (Sharma et al. 2012). Previous short-term laboratory studies found that FeS precipitates formed in sewers were re-oxidised in the aerobic conditions of the activated sludge bioreactors. This enabled phosphorus removal at a ratio of 0.44 mg P (mg Fe$^{3+}$)$^{-1}$ and 0.37 mgP (mgFe$^{2+}$)$^{-1}$ added to the sewer network (Gutierrez et al. 2010). The iron dosing (e.g. 5–20 mgFe L$^{-1}$) to the sewer network was also demonstrated to provide sufficient amount of iron for the desulfurization of H$_2$S from biogas in the anaerobic digester by forming iron sulfide precipitates (Ge et al. 2013). These previous laboratory studies have demonstrated the potential to achieve multiple benefits in the WWTP using iron salts dosed into the upstream sewers, including sewer corrosion and odor control, phosphorus removal and biogas desulfurization. A recent long-term study successfully demonstrated these multiple benefits of sewer dosed iron salts in an integrated
laboratory-scale system comprising of sewer reactors, biological reactor and the anaerobic
digester (Rebosura et al. 2018).

It is important to note that all these studies were carried out in the controlled laboratory
conditions. However, in real life, the sewers are considered as very dynamic systems with
varying flow, retention time and sewage characteristics (Sharma et al. 2013). Salehin et al.
(2019) replaced alum dosing at a full-scale WWTP (for P removal) with in-sewer dosing of
ferrous chloride at a similar dosing rate (190 kgAl d\(^{-1}\) vs. 160 kgFe d\(^{-1}\)), and found that P
removal did not deteriorate. The in-sewer iron salt dosing also reduced H\(_2\)S in biogas from 495
± 10 to 283 ± 4 ppm. The study was complicated by the fact that the treatment plant also
received ferrous chloride dosing at 295 kgFe d\(^{-1}\) to the centrate stream for odour control, which
was recycled to the mainstream treatment process. Further, the treatment plant imported a
substantial amount of sludge from other treatment plants (at ~150 wet tons of sludge/day,
compared to ~120 wet tons of sludge/day produced at the plant) for anaerobic digestion,
making mass balance analysis difficult. Moreover, the treatment plant did not have a primary
clarifier, and hence the impact of a primary clarifier on the multiple benefits remained
unknown. A primary clarifier likely directs a certain fraction of added iron to the anaerobic
sludge digester, thus not available for P removal in the bioreactor. Retaining iron in the primary
clarifier could be accompanied by the removal of more organic fractions, as iron salts are
effective coagulants (Boyd et al. 2003, Carballa et al. 2005, Ternes et al. 2002). This might
lead to the deterioration of biological nutrient removal due to the lack of carbon sources.

This study carried out full-scale field trials in the sewers and the Luggage Point WWTP
located in Brisbane, Australia. Ferrous dosing at the upstream sewer network (~18 km upstream
of the WWTP) and the inlet works of the WWTP were carried out to determine the potential
impacts on multiple processes, i.e. sulfide control in sewage, settling in the primary clarifier,
phosphate removal in the bioreactor, desulfurization of biogas and biosolids dewaterability.

Plant-wide sampling campaigns were conducted to collect wastewater and sludge samples. Also, some long-term monitoring data were gathered from the WWTP data acquisition system. The mass balance of the entire treatment plant during the in-sewer ferrous dosing trial was determined to assess the overall impacts of ferrous dosing. The results obtained would support the further full-scale application of iron dosing for multiple benefits in urban wastewater management.

2. Materials and methods

2.1. Process configuration of the Luggage Point wastewater treatment plant

The Luggage point-wastewater treatment plant (LP-WWTP), shown in Fig. 1A, locates in Brisbane, Queensland, Australia and has been designed with the treatment capacity of 900,000 population equivalent, treating about 60% of Brisbane’s domestic wastewater. The wastewater from different sewer catchments were collected at the Eagle Farm pumping station, Brisbane and then pumped through a long rising main sewer (about 9.5 km) to the inlet works of the LP-WWTP (Fig. 1B). The primary treatment of the LP-WWTP includes a grit chamber followed by a primary clarifier. The average dry weather flow hydraulic retention time (HRT) of the primary clarifier is about five hours. After the primary treatment, the wastewater is mixed with return activated sludge (RAS) at 1:1 mixing ratio and fed to the biological treatment process. The biological treatment has been designed for nitrification/denitrification to remove nitrogen from wastewater. The plant is not designed for biological phosphorus removal. One part of the effluent from the secondary clarifier is subjected to advanced treatment (ultrafiltration/reverse osmosis) for the non-potable reuse, and the remaining effluent was discharged to the Moreton Bay, Australia.
The thickened primary and secondary sludge was anaerobically digested for energy recovery as biogas, for power production with co-gen unit. The digested sludge is dewatered using a centrifuge. The centrate and other plant drains are sent back to the inlet works of the treatment plant.

Fig. 1. A) Simplified process flow diagram of Luggage point wastewater treatment plant with sampling points (yellow circles) and the ferrous dosing location B) Simplified catchment
diagram of the Luggage point treatment plant with ferrous dosing and the sampling locations (yellow stars)

A few chemicals are dosed at the LP-WWTP for different purposes. Polymers (purchased from SNF (Australia) pty. Ltd.,) were dosed at the primary sludge thickener (Polymer-FO4358SH, dose rate – 2-4 kg/dry tonne of solids), dissolved air flotation (DAF) unit for secondary sludge thickening (Polymer-FO4400, dose rate – 4-5 kg/dry tonne of solids) and centrifuge (Polymer-FO4650SH, dose rate – 10-12 kg/dry tonne of solids) to enhance the thickening process. Ethanol was dosed at concentrations of 59 L hr⁻¹ in the bioreactor as an external carbon source for maintaining the biological nitrogen performance of the reactor. Ferrous chloride solution (30% concentration, purchased from IXOM, Melbourne, Australia) was dosed at the rate of 60 L hr⁻¹ (i.e., 262.8 kgFe d⁻¹) in the passage of the DAF thickened sludge feeding to the anaerobic digester for H₂S control in biogas.

2.2. Ferrous dosing trials

2.2.1. Trial 1 - Inlet ferrous dosing

The hydrogen sulfide produced as a result of long rising main sewer passage led to serious corrosion issues to the pumps and concrete structures at the inlet works of LP-WWTP. In order to control the emission of hydrogen sulfide, ferrous chloride solution (30%) was dosed at the inlet syphon of the LP-WWTP (Fig. 1A). The ferrous dosing trial lasted for 50 days at different dosing rates, i.e. 4.8, 9.5, 14.3, 19.1, 23.9 and 28.8 kg ML⁻¹ of sewage. The ferrous dosing was intermittent, which lasted for about 6-10 hr each day. Each dosing rate was tested for 1 to 3 days. The detailed information regarding the dosing dates, dosing rate, duration and the sampling dates are presented in Table S1.

2.2.2. Trial 2 - In-sewer ferrous dosing
The average sludge retention time (SRT) in the bioreactor and anaerobic digester was 13 and 30 days respectively. So, after finishing Trial-1, a three-month transient period (3 SRT of digester) was provided to avoid the residual effects from Trial-1 in the LP-WWTP. In Trial-2, the ferrous dosing was shifted upstream to Caswell Street sewer pumping station (SPS), which is ~18 km away from LP-WWTP (Fig. 1B). The location was chosen as it is a hotspot for the H$_2$S concentration and odour complaints. Continuous dosing was provided in the gravity sewer line bypassing the SPS at an average dosing rate of 66 L hr$^{-1}$ (289 kgFe d$^{-1}$ / 2.2 kgFe ML$^{-1}$ of sewage entering the WWTP). Another dosing pump was fixed at the inlet of the wetwell located at the caswell street SPS, providing an instantaneous ferrous dosage, while the pumping station pumps the water from the wet well to the main gravity line. The average run time of this dosing pump is 15 min at an average of 12 times per day. This dose rate is set at 180 L hr$^{-1}$, providing about 98.5 kgFe d$^{-1}$ (0.75 kgFe ML$^{-1}$ of sewage entering the WWTP).

During Trial-2, ferrous dosing rate to the digester was reduced from 262.8 to 131.4 kgFe d$^{-1}$.

**2.3. Sampling campaign**

To assess the multiple benefits and other potential impacts of the ferrous dosing trials, a comprehensive sampling campaign was designed to collect various wastewater and sludge samples. The sampling campaign was divided into four different periods 1) baseline period (no ferrous dosing), 2) Trial-1 period (inlet ferrous dosing), 3) Transient period (interval between Trial 1 and Trial 2), and 4) Trial-2 period (upstream in-sewer ferrous dosing).

During the baseline period, a three-day sampling campaign was conducted before the initiation of any ferrous dosing trials. The whole treatment plant processes were divided into two different zones, i.e., zone-I which includes the liquid handling processes and zone-II that includes the solid handling processes (Fig. 1A). Composite samples (24-hr) were collected at the inlet and the effluent of the WWTP using auto samplers (AS950, Hach, USA). The sample containers were kept in refrigerated conditions inside the auto-samplers. Grab samples were
collected between 8-hr (i.e., 3 samples between 9 AM and 5 PM) at other locations (yellow circles in Fig. 1A) and mixed as the daily composite sample. The collected samples were immediately stored in the refrigerator at 4 °C and at the end of the day taken to the labs for analysis. During the Trial-1 period, a one-day sampling campaign was conducted for each dosing rates. The sampling was done during the mid-day (around 1 to 2 PM) at the sampling locations, allowing the inlet ferrous dosing to distribute throughout the WWTP considering the HRT of each processes. For the dosing rates that lasted for three days, the sampling was conducted at the last day of each dosing rate allowing the time to reach a stable condition. Considering the SRT, the sampling at zone-II (Fig. 1A) was started after the third week of initiating Trial-1.

The transient period was provided between mid-September 2018 and 4th January 2019. Sampling campaign was conducted (weekly) for one month prior to the initiation of Trial-2. During the Trial-2 period another three-day sampling campaign (weekly) was conducted after a month from the initiation of Trial-2. The sampling campaign for the transient period and the Trial-2 period follows the same procedure as that of the baseline period described above. Briefly, samples were collected weekly once at 19 different sampling locations in the WWTP as marked in the Fig. 1A. Along with these samples, additional grab samples were also collected at different locations of the sewer network (Fig. 1B).

2.4. Online monitoring at the Luggage Point WWTP and upstream sewer network

The effluent quality of the treatment plant (such as phosphates, ammonia and nitrate) were continuously monitored during all the periods through online sensors. The data were then extracted from the supervisory control and data acquisition (SCADA) system. The average biogas production in the anaerobic digester and the biogas compositions (H₂S, methane) were also obtained from the SCADA system.
The impact of in-sewer ferrous dosing (Trial-2) on the gaseous H$_2$S concentration in sewers was investigated through online monitoring during both baseline period (no ferrous dosing) and the Trial-2 period (upstream sewer ferrous dosing). The gas phase H$_2$S concentration was measured in the sewer headspace at three different locations, i.e. location A, B, and C (Fig. 1B). H$_2$S gas sensors (App-Tek Odalog® Logger L2) were used for recording the gas phase H$_2$S concentration. The H$_2$S were recorded at an interval of 1 min over a period of 14 and 28 days during the baseline and Trial-2 period in the aforementioned locations. Grab samples were taken weekly once during the trial period at these locations for analysing the pH, total and soluble metal concentrations.

2.5. Chemical analysis

To measure the dissolved concentrations such as phosphates, ammonia, nitrate, and nitrite in wastewater samples were immediately pre-treated by filtering through 0.22 μm filter (Millipore, PES) and then stored in the refrigerator until analysis. Dissolved phosphates, ammonia, nitrate and nitrite were measured using the flow injection analyser (Lachat QuickChem 8000, Lachat instrument, Milwaukee). To measure the dissolved inorganic sulfur species, the samples were immediately filtered (0.22 μm filter) and pre-treated with sulfur antioxidants buffer as described by Keller-Lehmann et al. (2006) and were stored in the refrigerator until analysis. The dissolved inorganic sulfur species (i.e. sulfide, sulfate, sulfite, and thiosulfate) were measured using ion chromatography coupled with an ultra violet and conductivity detector (Dionex ICS-2000). The samples for the analysis of metal concentrations were digested using 10% nitric acid before being analyzed. The total and soluble metal concentration of the samples were analysed using inductively coupled plasma- optical emission spectroscopy (Perkin Elmer optima 7300 DV, Waltham, MA, USA).
Dewaterability analysis was performed following the method described by Wei et al. (2018). Briefly, digested sludge samples were centrifuged at 1757g for 10 min. The liquid was discarded, and the settled solids were mixed with 2 mL of 0.7% polymer collected from the LP-WWTP so that the coagulation occurs. The sample was then strained through belt fibre fabric and then centrifuged again at 1757g for 10 min. The sludge cakes remained were placed in pre-weighed crucibles and then dried overnight at 105 °C, followed by weighing and determination of solid content. Total suspended solids (TSS), volatile suspended solids (VSS), total and volatile solids and sludge volume index (SVI) were measured using the standard methods (APHA 2005). Total and soluble chemical oxygen demand (TCOD and SCOD) were measured using COD cell test kits (Merck, range 25-1500 mgCOD L$^{-1}$). The shear viscosity of the digested sludge was measured using a rheometer (Physica MCR101 Modular Compact Rheometer, Anton Paar, Australia), equipped with cup and four-blade vane geometry of diameter 14 mm. Prior to each measurement, 50 mL sample was kept in the measuring cup where vane is immersed. All measurements were carried out at 25±0.01°C. The temperature was maintained by a Peltier control. The shear viscosity measurement was carried out as a function of time at a shear rate of 250 s$^{-1}$.

3. Results and Discussion

3.1. Impact of ferrous dosing on the sulfide control in sewers and WWTP inlet works

The summary of dissolved sulfide, total and soluble iron concentration at the inlet works of the WWTP during both trials are shown in Fig. 2. In Trial 1, the dissolved sulfide concentration at the inlet works of the treatment plant decreased gradually from 4.9 ± 1.8 mgS L$^{-1}$ during the baseline period to 0.1 mgS L$^{-1}$ at the ferrous dosing rate of 23.9 kgFe ML$^{-1}$ of sewage. The dissolved sulfide concentration reduction gradually increased with the ferrous dosing rate, reaching >90% reduction at the dosing rates of 23.9 and 28.8 kgFe ML$^{-1}$ comparing to the baseline period. Similar gradual reduction in the gas phase H$_2$S concentration
at the inlet works and negligible impact of all concentrations of ferrous dosing on the wastewater pH comparing to the baseline period was observed during Trial-1 (Fig. S1). The sulfide reduction at the inlet works of the treatment plant positively correlated with the amount of iron dosed \((r=0.9088, p=0.0046, \text{two-tailed } t\text{-test})\). Asides the reduction of dissolved sulfide concentration at the inlet works, the inlet ferrous dosing also caused a significant reduction of dissolved sulfide in the effluent of the primary clarifier. A maximum sulfide reduction of 91% was achieved comparing to the baseline period in the primary clarifier at the dose rate of 19.1 kgFe ML\(^{-1}\). After this, a slight increase in the sulfide concentration of the primary effluent was observed (Fig. S2). This could be likely due to the operational fluctuations of the primary clarifier.

**Fig. 2.** A) Impact of ferrous dosing on the dissolved sulfide and iron concentration at the inlet works of the WWTP during Trial-1 (The error bars are the error calculated from triplicates collected at three different days. The error bars were not presented for samples collected at only one day in particular during ferrous dosing period. One day sampling is due to the intermittent ferrous dosing strategy) B) Impact of in-sewer ferrous dosing (Trial-2) on the dissolved sulfide and iron concentration at the inlet works of the WWTP (error bars are the error calculated from triplicates)

The Fe:S molar ratio of each dosing rate has been calculated by considering the baseline sulfide concentration (Table S1). The dissolved sulfide concentration reached <0.4 mgS L\(^{-1}\) at
the Fe:S molar ratio of 2.8:1 and 3.4:1. A higher dosage of ferrous than the theoretical Fe:S molar ratio (1.3:1) was previously suggested to achieve a dissolved sulfide concentration of <0.1 mgS L⁻¹ (Firer et al. 2008). Nielsen et al. (2007) also reported that only a fraction of ferrous added reacts with dissolved sulfide necessitating the higher dosage of ferrous than the theoretical requirement. The molar ratio between ferrous and sulfide in this study is also higher than the previous study in the laboratory scale sewer reactor (1.33Fe :1S) while dosing with ferric chloride (Rebosura et al. 2018).

The total and soluble iron concentrations at the inlet works of the treatment plant during the Trial-1 period showed a linear increase in iron concentrations corresponding to the iron dosage at the inlet works. The difference between the total and soluble iron concentrations suggesting the presence of iron precipitates. The complex iron speciation in wastewater could lead to precipitates such as iron oxides (Cheng 2002), iron sulfides (Gutierrez et al. 2010, Haaning Nielsen et al. 2005, Padival Navnit et al. 1995, Zhang et al. 2009) and iron phosphates (Wilfert et al. 2018). The decrease in the sulfide concentration with the ferrous dosing trials could be attributed to the formation of iron sulfide precipitates, which were observed as black precipitates in the water (Kulandaivelu et al. 2019). The total iron concentrations at the inlet were higher than the dosed amount because of the iron-containing plant drain (containing primary thickener filtrate, centrate and other plant wastes) being recirculated back to the inlet works (Fig. 1A, Fig. S3).

In Trial-2, the in-sewer ferrous dosing had substantial effect on the hydrogen sulfide concentration in sewer network. The average gas phase H₂S concentration over a month duration at different locations of the sewer network is shown in Fig. 3, with long-term data further shown in Fig. S4 – Fig. S9. The average gas phase H₂S concentration was reduced from 47.3 ± 0.2, 12.1 ± 0.4 and 3.9 ± 0.1 ppm during baseline period to 27.3 ± 0.2, 2.6 ± 0.1, and 0.5 ± 0.01 ppm during Trial-2 at location A, B and C, respectively. Relatively, the respective
The lesser gas phase reduction at location A could be due to the accumulated sulfide coming from other sewer locations. The dissolved sulfide concentration at these locations were $0.5 \pm 0.2$, $2 \pm 0.7$, $1.6 \pm 0.3$ mgS L$^{-1}$ respectively. The pH at these locations were in the range of 7.3 and 8. The total and soluble iron concentrations at these respective locations are shown in the Fig. S10. The difference between the total and soluble iron concentration, the reduction in the gas phase H$_2$S and the lower dissolved sulfide concentration at these locations suggests the formation of iron sulfur complexes.

The total iron concentration at the inlet works of the WWTP increased from $1.3 \pm 0.1$ during transient period to $2.8 \pm 0.4$ mgFe L$^{-1}$ during Trial-2 period (Fig. 2B). However, the soluble iron concentration increased from $0.2 \pm 0.1$ to $0.4 \pm 0.01$ mgFe L$^{-1}$, largely due to the upstream reaction and dilution of sewage from different sewer lines. The negligible increase and very low concentration of dissolved iron showed no reduction effects on the sulfide at the inlet works of the treatment plant (Fig. 2B).

![Box and whiskers plot representing the gas phase H$_2$S concentrations at different locations of the sewer network.](image)

**Fig. 3.** Box and whiskers plot representing the gas phase H$_2$S concentrations at different locations of the sewer network (Middle line of the box represents the median, dotted line
represents the mean, the upper and lower lines represent the 5th and 95th percentile, the whiskers extending the box and the outliers represent the data outside the interquartile range)

### 3.2. Role of primary clarifier for the ferrous dosing trials

The fraction of upstream dosed iron retained in the primary sludge and the availability of iron to the downstream processes is a critical information for achieving its beneficial reuse particularly the removal of phosphates in the biological reactor. Fig. 4A & 4B shows the amount of total iron carryover from the primary clarifier to the biological reactor during both Trial-1 & 2 respectively. The retention of total iron (influent concentration – effluent concentration) at the primary clarifier ranged between 19.1% at the low ferrous dose rates (4.8 kgFe ML⁻¹) and 65.5% at the high dose rate (23.9 kgFe ML⁻¹). The amount of total iron retained positively correlated with the amount of iron dosed at the inlet works (r=0.8416, p=0.0356, two-tailed t-test). The total iron concentrations in the primary effluent increased from 0.9 ± 0.1 mgFe L⁻¹ during baseline to 16.4 mgFe L⁻¹ at the high ferrous dose rate of 28.8 kgFe ML⁻¹ of sewage. Interestingly, the total iron concentration retained during the Trial-2 was 38.3 ± 5.8%.

The amount of iron entering the primary clarifier during Trial-2 (in-sewer ferrous dosing) is 40% of the dose rate (4.8 kgFe ML⁻¹) tested in Trial-1 (Inlet ferrous dosing). But, the percentage iron retention during Trial-2 is twice than that of the iron retention percentage at the low dose rate (4.8 kgFe ML⁻¹) during Trial-1.
Fig. 4. A) Summary of total and soluble iron concentration, total iron reduction % and total COD reduction % in the primary effluent during Trial-1 (The error bars are the error calculated from the triplicate samples at three different days. The error bars were not presented for samples collected at only one day in particular during ferrous dosing period) B) Summary of total iron concentration and total COD reduction % during Trial – 2 (error bars are the errors calculated from the triplicate samples)

This clearly emphasizes that while shifting the iron dosing to the upstream sewer location (i.e., ~18 km away from the WWTP), the iron retention in the primary clarifier is doubled comparing to the iron dosage at the inlet works of the treatment plant. This is likely due to the formation of larger aggregates of iron sulfides (FeS) caused by the mixing provided during the transportation of wastewater in the sewers. Iron as a coagulant, forms the aggregates of different sizes depending on the mixing time (Metcalf 2003). This result is similar to the result reported by Gutierrez et al. (2010) where he demonstrated the effect of mixing time (depending on the location of iron dosing) on the settleability of FeS precipitates in the laboratory simulated primary settling tank. Similar to total iron retention in the primary clarifier, the total phosphorus retention ranged between 14.2% at the dose rate of 4.8 kgFe ML⁻¹ to 45.7% at the dose rate of 28.8 kgFe ML⁻¹ during Trial-1 and was about 25.2 ± 9.1 % during Trial-2 period (Fig S3). However, no difference in the phosphate-P (PO₄³⁻-P) was observed instead there is a slight increase in the PO₄³⁻-P concentration in the effluent comparing to the concentration entering the clarifier during both the trials (Fig S3). This could be due to the accumulation of PO₄³⁻-P in the clarifier due to the mixing of centrate at the inlet works of the treatment plant (Fig 1A).

In parallel to iron retention in the primary clarifier, the TCOD reduction in the primary clarifier also increased from 31.2 ± 1.2 % during baseline to 49% at the high ferrous dose rates such as 19.1, 23.9 and 28.8 kgFe ML⁻¹ during Trial-1. Previous research reported the positive
correlation between iron dosage and COD removal (Kumar and Bishnoi 2017). During the transient period, the TCOD removal in the primary clarifier dropped back to 31.2 ± 1.7% and then increased to 42 ± 1% owing to the in-sewer ferrous dosing (Trial-2). Though the amount of iron entering the primary clarifier is lower comparing to the dose rates tested during Trial-1, the TCOD removal during Trial-2 is higher and it corresponds to the iron retainment in the primary clarifier during this period. TCOD is one of the critical parameters in maintaining the performance of biological nitrogen removal of the activated sludge biological reactor.

3.3. Impact of ferrous dosing on the phosphates in the biological reactor

Fig. 5A shows the impact of different ferrous dose rates on the phosphate-P (PO$_4^{3-}$-P) removal in the bioreactor effluent. The PO$_4^{3-}$-P reduction % calculated based on the PO$_4^{3-}$P entering the bioreactor and during Trial-1, the reduction % increased from 25% during baseline period to 76% at the ferrous dose rate of 28.8 kgFe ML$^{-1}$. During the transient period, the PO$_4^{3-}$-P reduction dropped back to 23.1 ± 3.5% and it increased to 53.4 ± 2.3 % due to the upstream sewer iron dosing (Fig. 5B). This impact of upstream ferrous dosings on the effluent phosphate concentration is also clearly evident from the long term effluent phosphate concentration trend (Fig. S11 & Fig. S12). During Trial-1, the phosphate concentration decreased during the dosing dates and then slowly increased during the non dosing dates (Fig. S11). In the transient period, the phosphate concentrations increased back to the concentrations similar to the baseline period. Then a decreasing trend was observed due to the upstream sewer ferrous dosing (Fig. S12).

The PO$_4^{3-}$-P reduction during the baseline and transient period was likely due to the bacterial P uptake for their growth. The enhanced PO$_4^{3-}$-P removal during both the iron dosing trials could be explained by the precipitation with iron that was re-oxidized from iron sulfides formed in the upstream, or the soluble iron. Due to the aerobic conditions prevailing in the bioreactor, the iron sulfides gets re-oxidised and the iron was readily available for binding with
dissolved PO$_4^{3-}$P, enhancing its removal from the liquid phase (Gutierrez et al. 2010). This was also evident from the slow accumulation of iron and phosphorus in the activated sludge (Fig. 5A & 5B). The total iron and phosphorus in the activated sludge during Trial-1 were positively correlated with the ferrous dose rates with the correlation coefficient of 0.9599 ($p=0.0006$, two-tailed $t$-test) and 0.8301 ($p=0.0208$, two-tailed $t$-test) respectively.

The iron concentration carryover from the primary clarifier reached the activated sludge bioreactor enabling the removal of dissolved PO$_4^{3-}$P (Section 3.2). Due to the short ferrous dosing time (6-10 hr d$^{-1}$) and the long HRT (11.3 hr) in the bioreactor, it is hard to determine the accurate phosphate removal ratio. The phosphate removal ratio ranged between 0.07 – 0.14 mgP (mgFe$^{2+}$)$^{-1}$ dosed at the inlet works and 0.11-0.33 mgP (mgFe$^{2+}$)$^{-1}$ entering the bioreactor. This phosphate removal ratio is lower than the removal ratio achieved during Trial-2, i.e., 0.3 mgP (mgFe$^{2+}$)$^{-1}$ dosed and 0.5 mgP (mgFe$^{2+}$)$^{-1}$ carry over to the bioreactor. The higher phosphate removal ratio during Trial-2 could be likely due to the continuous dosing of iron in the sewer network, and the longer HRT (11.3 hr) in the bioreactor. However, these values are lesser than the previously reported phosphate removal rates for example, 0.37 mgP (mgFe$^{2+}$)$^{-1}$ (Gutierrez et al. 2010) and 0.63 mgP (mgFe$^{3+}$)$^{-1}$ dosed to the sewers (Rebosura et al. 2018). As these studies didn’t consider the primary clarifier as part of the treatment process, the lower phosphate removal in our study could be attributed to the amount of iron retained in the primary clarifier. Also, the complex chemistry of iron in wastewater in real life conditions could also lead to the formation of several other precipitates like iron oxides and iron – organic complexes (Cheng 2002).

Apart from the phosphates removal in the bioreactor, the impact of upstream ferrous dosing on the sludge characteristics is critical. During Trial-1, due to the intermittent dosing, the impact of the ferrous dosing on the SVI of the activated sludge could not be measured accurately. But during Trial-2 where continuous ferrous dosing was provided in the upstream
sewer, the SVI of the activated sludge decreased from 83.9±1 during the transient period to 69.4±0.8 during Trial-2 period. Similar decrease in the SVI due to the upstream iron dosing was previously reported in a lab reactor (Rebosura et al. 2018). This could be likely due to the coagulating nature of ferrous ions enabling the conversion of filamentous flocs into the compact structures resulting in improved sludge settleability (Agridiotis et al. 2007). The long-term effluent nitrate, ammonia and total nitrogen concentrations were shown in Fig. S13 & Fig. S14. During Trial-1, the effluent ammonia level was well below the effluent discharge standard (4 mgN L⁻¹), the nitrate and the total nitrogen concentration were also not affected. The ammonia reduction efficiency during all these dose rates were in the range of 98.6 – 99.6% comparing to the influent ammonia concentration. However, at higher ferrous dose rates (>19.1 kgFe ML⁻¹), where the primary clarifier TCOD reduction reached 49%, the ethanol dosage to the biological reactor was increased from 59.6 L hr⁻¹ to 69.6 L hr⁻¹ as a precaution. Similarly, during Trial-2, no negative impact was observed on the performance of the biological system and is evident from the effluent ammonia, nitrate and total nitrogen concentration (Fig. S13 & S14).

Fig. 5. A) Impact of inlet ferrous dosing (Trial-1) on the phosphate reduction and on the composition of activated sludge (The error bars are the error calculated from the triplicate samples at three different days. The error bars were not presented for samples collected at only one day in particular during ferrous dosing period) B) Impact of in-sewer ferrous dosing (Trial-
on the phosphate reduction and on the composition of activated sludge (error bars are the error calculated from triplicates. For some points, the error bars are shorter than the height of the symbol and thus are not shown)

3.4. Impact of ferrous dosing on the anaerobic digester

During the baseline period, ferrous chloride (30%) was dosed directly to the anaerobic digester at the concentration of 262.8 kgFe d⁻¹ for H₂S control in the biogas. The total iron concentration in the anaerobic digester sludge during this period was 16.1 ± 0.4 mgFe g⁻¹ of dry sludge. Trial-1 (inlet ferrous dosing) leads to the accumulation of dosed iron in the anaerobic digester through primary and DAF thickened sludge (Fig. S15). The total iron concentration of the sludge in the digester increased to 21.6 ± 0.7 mgFe g⁻¹ after 3 weeks of the initiation of Trial-1 (Fig. 6A). This leads to the decrease of dissolved sulfide concentration from 8.9 ± 0.6 mgS L⁻¹ (baseline) to 2.3 ± 0.3 mgS L⁻¹, i.e., a reduction of 77.6 ± 3.6%. The average gas phase H₂S concentration in the biogas dropped from 303.7 ± 1 ppm during baseline to 194.4 ± 0.6 ppm during Trial-1 period, i.e., a 36% reduction (Fig. 6B).

During the transient period between Trial-1 and Trial-2, the dissolved sulfide concentration in the digester increased back to 6.9 ± 0.2 mgS L⁻¹ and the average gas phase H₂S concentration increased to 235.7 ± 0.5 ppm. The iron concentration during this period dropped to 16.9 ± 0.6 mgFe g⁻¹ of dry sludge. After one month of the initiation of Trial-2, the dissolved sulfide concentration in the anaerobic digester decreased to 0.3 ± 0.05 mgS L⁻¹, i.e., a 96.2 ± 0.7% reduction due to in-sewer ferrous dosing. The reduction in the dissolved sulfide during both the trials is likely due to its precipitation with Fe ions that were regenerated from the ferric phosphate precipitates (previously formed due to the re-oxidation of FeS in the activated sludge process) (Ge et al. 2013, Rebosura et al. 2018). Also, the average gas phase H₂S concentration decreased to 129.1 ± 0.3 ppm, a 45.2% decrease compared to the transient period. The long
term \( \text{H}_2\text{S} \) concentration of biogas and the impact of ferrous dosing during both trials has been included in Fig. S16. During Trial-2, the ferrous dosing rate to the digester was decreased from 262.8 to 131.4 kgFe d\(^{-1}\). Though this dosing rate was reduced, the iron concentration in the digester during Trial-2 period was 19.5 ± 0.2 mgFe g\(^{-1}\) and is still higher compared to the transient period. The possible reason could be the accumulation of upstream sewer dosed iron in the anaerobic digester. The gas phase \( \text{H}_2\text{S} \) reduction observed during Trial-2 is lesser than the previously reported 82.3 ± 3.7% biogas \( \text{H}_2\text{S} \) reduction, while dosing ferric chloride in the sewer reactor of laboratory integrated system (Rebosura et al. 2018). The lesser reduction observed in this study is due to the lesser concentration of iron dosed in the upstream sewer comparing to the previous lab-scale study (10 mgFe L\(^{-1}\)) (Rebosura et al. 2018).

The average biogas production over all these periods were not deviating from each other significantly (Fig. S17). This shows that iron precipitates in the anaerobic digesters, either in the form of FeS (created by regenerated Fe\(^{3+}\) in the digesters) or ferric phosphate precipitates (previously formed during FeS re-oxidation in the bioreactor), did not have a significant impact on the anaerobic digestion process. The similar finding was reported in previous studies (Ge et al. 2013, Novak and Park 2010, Rebosura et al. 2018). Due to the increase in the primary sludge production by the iron dosing, the average biogas production was expected to increase but was not observed in this study, likely due to the intermittent dosing pattern and thus limited change of sludge in the anaerobic digester. The methane composition of the biogas during all these periods were also not impacted (Fig. S18). Previously, the inhibition of methanogenic activity of the rising main sewer reactor by about 52-80% was reported after two months of Fe\(^{3+}\) addition (Zhang et al. 2009). However, no inhibition of methanogenic activity was observed in this study which involved a lower iron dosing concentration than the previous study (21 mgFe\(^{3+}\) L\(^{-1}\)).
Fig. 6. A) Summary of dissolved sulfide and total iron concentrations in the anaerobic digester (The error bar are the error calculated from triplicate samples collected at different weeks) B) Box and whiskers plot showing the gas phase H$_2$S composition of biogas and the impact of ferrous dosing during Trial-1 and Trial-2 (Middle line of the box represents the median, dotted line represents the mean, the upper and lower lines represents the 5$^{th}$ and 95$^{th}$ percentile, the whiskers extending the box and the outliers represents the data outside the inter quartile range)

3.5. Impact of ferrous dosing on the dewaterability of the digested sludge

The impact of inlet ferrous dosing on the dewaterability performance was tested using the batch tests and has been shown in Fig. 7A. The inlet ferrous dosing (Trial-1) had a significant impact on the dewaterability of the sludge. The percentage of total solids in the dewatered sludge increased from 17.6 $\pm$ 0.3$\%$ of total dry solids to 19.3 $\pm$ 0.1$\%$ due to the inlet ferrous dosing (Trial-1). This leads to the relative increase of 9.7$\%$ of total solids in the dewatered sludge comparing to the dewatered sludge from the baseline period. The increase in total solids positively correlated with the amount of iron present in the dewatered solids ($r=0.9773$, $p=0.0008$, two-tailed $t$-test) (Fig. S19).

Also, the upstream sewer ferrous dosing (Trial-2) led to the relative increase in total solids of 9.8$\%$ comparing to the transient period without sewer ferrous dosing. These results are comparatively smaller to the previously reported 17$\%$ increase in the total solids of the dewatered sludge due to ferrous dosing in a lab-scale reactor system (Rebosura et al. 2018).
The intermittent dosing patterns for limited durations in the field trials vs. the constant ferrous dosing at 10 mgFe L\(^{-1}\) for the lab study is potentially responsible for the difference. The reason for the enhanced total solids in the dewatered sludge could be explained by the sludge destabilisation provided by iron in the sludge. This destabilisation could be attributed either to the bridging of negatively charged sludge particles with positive iron hydroxide species, or to an enmeshment of flocs in the precipitate of ferric hydroxide (Deneux-Mustin et al. 2001). The formation of ferric phosphate hydroxide species in the digester due to the sewer iron dosing has been reported recently (Salehin et al. 2019). The improvement in the dewaterability of the sludge is also supported by the sludge viscosity (Fig. 7B). The sludge viscosity of the dewatered sludge at the applied shear rate of 250 s\(^{-1}\) decreased from 246.1 ± 0.04 during the transient period to 156.8 ± 0.2 mPa.s due to the upstream sewer ferrous dosing. Decrease of sludge viscosity is a critical parameter supporting the increased dewaterability (Ratkovich et al. 2013, Xiao et al. 2019).

**Fig. 7.** A) Impact of iron dosing on the dewaterability of the sludge B) Impact of iron dosing on the shear viscosity of the digested sludge (error bars are the errors calculated from the triplicate samples. For some points, the error bars are shorter than the height of the symbol and thus are not shown)

### 3.6. Economic analysis
The economic analysis for the both ferrous dosing trials was carried out based on the ferrous chloride solution cost at 340 $ ton\(^{-1}\). During Trial-1, in order to achieve >90% sulfide removal about 23.9 kgFe ML\(^{-1}\) was required leading to the dosing of 131 L of ferrous chloride solution per ML of wastewater. By considering the baseline sulfide concentration (4.9 kgS ML\(^{-1}\)), the chemical cost is estimated as 13 $ kgS\(^{-1}\) removal or 0.06 $ (m\(^3\))\(^{-1}\) of wastewater. Similarly, during Trial-2, to achieve 88% gas phase H\(_2\)S reduction at the downstream eagle farm SPS (Location-3) (Fig. 1B), around 2124 L day\(^{-1}\) of ferrous chloride was dosed. The cost is estimated as 1062 $ day\(^{-1}\) or 0.01 $ (m\(^3\))\(^{-1}\) of sewage entering the treatment plant. Further, in Trial-2, the ferrous dosing to the anaerobic digester was halved (from 60 to 30 L hr\(^{-1}\)). The ferrous dosing to the anaerobic digester was reduced due to the increase in the iron concentration of the wastewater entering the WWTP. The total iron concentration in the wastewater influent increased from 165.3 ± 20.8 kgFe day\(^{-1}\) to 372.7 ± 52.7 kgFe day\(^{-1}\) (Fig. S20 & Fig. S21) due to upstream sewer ferrous dosing during Trial-2. This reduced chemical dosage to the digester reduced the chemical cost up to 360 $ day\(^{-1}\). The high dissolved phosphate removal achieved during Trial-1 and Trial-2 were about 76% and 53.2% respectively (Section 3.3). This leads to the reduction of around 200 and 100 tonnes of dissolved phosphorus discharge per annum during Trial-1 and 2 respectively. Such phosphorus reduction in the WWTP discharge was not included for the economic analysis. Further, the benefits such as corrosion management in sewers, co-gen maintenance cost due to the H\(_2\)S reduction in the biogas, and improvement in the sludge dewaterability were not monetized.

4. Conclusion

Overall the upstream dosed iron at both locations had significant positive impact on the downstream treatment processes. The key conclusions are:
The dosage of iron both at the inlet works and at the sewer network showed significant reduction of dissolved sulfide in sewage thus reducing the gas phase H\textsubscript{2}S concentration.

The phosphate removal ratio ranged between 0.07 – 0.14 mgP (mgFe\textsuperscript{2+})\textsuperscript{-1} dosed at the inlet works and 0.11-0.33 mgP (mgFe\textsuperscript{2+})\textsuperscript{-1} entering the bioreactor. This phosphate removal ratio is lower than the removal ratio achieved during Trial-2, i.e., 0.3 mgP (mgFe\textsuperscript{2+})\textsuperscript{-1} dosed and 0.5 mgP (mgFe\textsuperscript{2+})\textsuperscript{-1} entering the bioreactor.

The ferrous dosing at both the locations reduced the biogas H\textsubscript{2}S concentration in the anaerobic digester. The reduction in gas phase H\textsubscript{2}S concentration up to 36% and 45% was observed during Trial-1 & 2 respectively. The biological performance of the digester was not affected during both trials.

The dewaterability test of the digested sludge shows that there is 9.7% and 9.8% relative increase in the total solids of the dewatered sludge during Trial-1&2 respectively.

The iron retention in the primary clarifier during Trial-1 ranged between 19% - 65.5% depending on the iron dose rates. During Trial-2 the iron retention was doubled when compared to the low ferrous dose rate (4.8 kgFe ML\textsuperscript{-1}) during Trial-1. The TCOD removal % increased from 31.2 ± 1.2 % during baseline to 49% at high ferrous dose rate during Trial-1 and 42 ± 1% during Trial-2. The primary clarifier showed limited impact on the availability of iron for the downstream processes.

This demonstration of beneficial reuse of upstream dosed iron in a full scale treatment plant enhances the confidence towards the integrated management of sewer network and the treatment plant.

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References


Supplementary information

**Table S1.** Ferrous dosing schedule at the inlet works of the treatment plant, the dosing duration, Fe:S molar ratio and the sampling dates

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*Sampling dates
S1. Impact of ferrous dosing on the gas phase hydrogen sulfide concentration during trial-1 and trial-2

**Fig S1.** The long term gaseous hydrogen sulfide concentration and the ferrous dosing rate at the inlet works of the treatment plant

**FigS2.** Composition of the plant drain mixing with the influent at the inlet works of the STP
S2. Long term gas phase H₂S concentration at different locations of the sewer network

**Fig S3.** Long term gas phase hydrogen sulfide concentration in the sewer headspace at location A (Lytton road pump station) during the baseline period

**Fig S4.** Long term gas phase hydrogen sulfide concentration in the sewer headspace at location A (Lytton road pump station) during the trial-2 period
Fig S5. Long term gas phase hydrogen sulfide concentration in the sewer headspace at location B (riding road pump station) during the baseline period

Fig S6. Long term gas phase hydrogen sulfide concentration in the sewer headspace at location B (riding road pump station) during the trial-2 period
Fig S7. Long term gas phase hydrogen sulfide concentration in the sewer headspace at location C (Eagle farm pump station) during the baseline period.

Fig S8. Long term gas phase hydrogen sulfide concentration in the sewer headspace at location C (Eagle farm pump station) during the trial-2 period.
**Fig S9.** Total and soluble iron concentration at the different locations of the sewer network during trial-2 period

**Fig S10.** Impact of upstream ferrous dosing on the biological performance of the reactor during trial-1 & trial-2
S3. Long term effluent phosphate concentration from the SCADA system

Fig S11. Long term effluent phosphate concentration and the impact of different ferrous rates at the inlet works of the LP-STP

Fig S12. Long term effluent phosphate concentration and the impact of upstream sewer ferrous dosing
**Fig S13.** Impact of inlet ferrous dosing on the composition of the primary and the DAF thickened sludge fed to the anaerobic digester
S4. Impact of upstream ferrous dosing (both inlet and sewer ferrous dosing) on the performance of the anaerobic digester

**Fig S14.** Long term average biogas production in the anaerobic digester

**Fig S15.** Long term gas phase hydrogen sulfide concentration of the biogas and the impact of inlet and sewer ferrous dosing
Fig S16. Long term gas phase methane concentration of biogas and the impact of inlet and sewer ferrous dosing

S5. Dewaterability of the digested sludge

Fig S17. Percentage increase in the total solids of dewatered sludge and the iron concentration
S7. Mass balance analysis and impact of sewer ferrous dosing in Luggage point sewage treatment plant

**Fig S18.** Plant wide mass balance analysis of the Luggage point sewage treatment plant during the transient period (the period between trial-1 and trial-2). Data shown are average ± standard mean error.
Fig S19. Plant wide mass balance analysis of the Luggage point sewage treatment plant during the trial-2 period (sewer ferrous dosing trial).

Data shown are average ± standard mean error