

A new approach of nonpoint source pollution/stormwater sludge treatment by an integrated thermal plasma system

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Abstract In this study, we designed and demonstrated the effectiveness of an integrated waste treatment system for contaminated stormwater sludge. The system is comprised of two components: pulsed arc electrohydraulic discharge (PAED) for aqueous-phase treatment and thermal plasma decontamination for solid-phase treatment. PAED provided a reduction of 80 % in total organic carbon (TOC) within a treatment time of 5 min in the aqueous phase. With a prolonged treatment of 30 min, the decontamination of TOC reached over 90 % in the aqueous phase, accompanied with 20 % carbon and 70 % sulfur decreases in weight in the solid phase. A huge weight reduction can be achieved prior to further solid treatment. The solid phase, named as the PAED-treated sludge, was separated by sedimentation and transferred to thermal plasma treatment. The thermal

plasma-treated sludge was slightly melted, and the formation of slag was possible. X-ray energy dispersion solid analysis showed that carbon and sulfur were reduced to zero after treatment. Si, Mg, and Al decreased by 25, 30, and 60 %, respectively, while Ca and Fe enriched by 400 and 25 %, respectively. The emission gas analyses demonstrated a possible energy recovery from gaseous compounds with adequate gaseous treatment devices. This research demonstrates an innovative treatment approach of various sludge types, especially those types that contain high water contents and are severely contaminated by toxic organic compounds.

Keywords Nonpoint source pollution · Stormwater sludge · Thermal plasma · Pulsed arc electrohydraulic discharge · Integrated system

Introduction

Urban stormwater detention/retention ponds are designed and constructed to partly control pollution caused by urban stormwater. With an adequate design, these ponds can achieve removal efficiencies as high as 90 % (Marsalek et al. 1997). Regular removal of sediment is required to ensure the availability of adequate storage capacity and to maintain treatment effectiveness (MOEE 1994). In order to protect the aquatic environment, the disposal of sediment and surface water was strictly regulated by global government agencies (MOEE 1993, 2003; USEPA 2005). Previous studies have intensively examined the quality of stormwater sediment worldwide (Behera et al. 2006; Bishop et al. 1997, 2000;

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Cox and Livingston 1997; De la Torre et al. 2011; Karlsson et al. 2010; Marsalek and Marsalek 1997). Based on these studies, it can be concluded that most of the stormwater sediments are contaminated and cannot be disposed of without treatment (Duke et al. 1998; Graney and Eriksen 2004; Li et al. 2011a; Rochfort et al. 2004). Generally, the most contaminated part is found between the layer of sediment and the bottom of the pond water, a layer of substances referred to as the sludge water (Li et al. 2011b). The treatment is recommended to carry out in two scenarios: (1) when the treatment efficient is below than that of designed range, and (2) when the contamination of sediment is higher than that of regulation. Thus, a feasible and effective stormwater sediment decontamination technology must be able to deal with a greater complexity and can be applied to treat more than one medium, whereas the wide variety of organics and heavy metals contained in stormwater sediments makes bioremediation, conventional thermal, and chemical extraction treatment processes challenging (Berge et al. 2009; Okochi and McMartin 2011; Morris and Barlaz 2011; Rienks 1998; Santiago and Pelletier 2001; Seadon 2006).

A type of liquid-phase thermal plasma process, named pulsed arc electrohydraulic discharge (PAED), can result in concurrent removal of chemical contaminants and inactivation of viruses and bacteria (Angeloni et al. 2007; Bian et al. 2009; Lee et al. 2008; 23–30; Locke et al. 2006; Shi et al. 2009; Vel Leitner et al. 2005; Yantsis et al. 2008; Zastawny et al. 2004). On the other hand, thermal plasma decontamination technology offers near-quantitative destruction of various organic and inorganic contaminants (Bernardo and Dal Maschio 2011; Gomez et al. 2009; Jones et al. 2001; Lemmens et al. 2007; McLaughlin 1999; Morrin et al. 2012; Sakai and Hiraoka 2000; Yang et al. 2010; Zhao et al. 2010). The technique is expected to become increasingly commercially viable in the future (Gomez et al. 2009). In addition, the generated gaseous compounds from the waste can be used for energy recovery (Arena et al. 2012; Byun et al. 2011; Consonni and Vigano 2012; Lombardi et al. 2012).

Based on the promising results of these earlier studies on thermal plasma systems, this study proposes an integrated system for treating urban stormwater sediments. This system is comprised of two main components: (1) PAED for aqueous-phase treatment and (2) thermal plasma decontamination for solid-phase treatment. Sludge water samples collected from a stormwater pond were treated experimentally by the proposed integrated system, and the multimedia detoxification characteristics were examined

and discussed in detail. The experiment was performed with a total five sets of experiment with the same sample. All experiments were carried out at the Department of Engineering Physics, McMaster University, Canada, from year September 2006 to June 2010.

Materials and methods

Sample preparation and treatment train

The sediment samples were collected from the Dartnell stormwater pond, located at the Dartnell Road Interchange of the Lincoln Alexander Parkway in the city of Hamilton, Ontario. The particle sizes ranged from 0.3 to 1,000 μm , and the size distribution was bimodal and has peak particle sizes around 10 and 700 μm . Sediments were screened by a filter with 208 μm opening. The filtrate is taken as sludge water that has a peak particle size around 10 μm . The sludge water was first treated by PAED for 30 min and was further separated into liquid and solid fractions by sedimentation. The solid component (referred to as the PAED-treated sludge) was dried at room temperature and then transferred to thermal plasma treatment for 2 h. The treatment train of the study is shown in Fig. 1.

Pulsed arc electrohydraulic discharge experimental apparatus and procedures

The PAED system consisted of a spark-gap switch type power supply (0.5 kJ) and a 3-L stainless steel reactor with eccentrically configured 6-mm rod-to-rod titanium electrodes. The discharge voltage, current, and electrode gap distances were reported to be 1 kV, 59 kA, and 0.5 mm by Li et al. 2012a. Aqueous samples were obtained for water quality analysis via a liquid sampling valve for treatment time intervals of 5, 15, and 30 min. The schematic of the experimental setup is shown in Fig. 2.

Thermal plasma experimental apparatus and measurement techniques

A schematic of the plasma torch-type sludge treatment system under DC transferred mode is shown in Fig. 3. The ceramic reactor was composed of 99.8 %

pure alumina with 7.5 cm i.d. and attached to a ground electrode at the reactor's bottom. A DC of 10 kW plasma torch was placed vertically 5 cm above the ceramic reactor and the environmental chamber (Beuthe and Chang 1997; Chang et al. 2008; Li et al. 2012b). Pure argon gas (99.995 %) was used as the plasma gas, and air was injected through the environmental chamber in order to produce an oxidation environment. The PAED-treated sludge of 10 g was decontaminated in various air flow rates with a total treatment time of 2 h. The main purposes of the experimental work were to demonstrate the effectiveness of further treatment in solid phase provided by thermal plasma along with the possibility of energy recovery under oxidation and reduction atmospheres. Thus, the system was yet to be optimized for commercial scale.

In the preliminary tests, we have found that the plasma treatment provided the highest efficient with a flow rate of 24 L/min. Also, the emission of the SO₂, CO, and CO₂ would be above the detection limit of gaseous analysis equipment if the air flow rate was greater than 20 % of the argon flow rate. Air flow rates to the environmental chamber were adjusted to 0–20 % of the fixed argon flow rate of 24 L/min, which were correspondingly 0–4.8 L/min. The gaseous emission was sampled at the exit of the water heat exchanger.

Solid, liquid, and gaseous analyses

For the solid products, neutron activation analysis (NAA) and X-ray energy dispersion microanalysis were carried out on the original and PAED-treated sludge for elemental compositions. Scanning electron microscope (SEM) images were taken to analyze the nature of the original sludge and structural changes after plasma treatment. For liquid analysis, water quality indicators including pH, total dissolved solids (TDS), dissolved oxygen (DO), salinity, and conductivity were recorded by a multiparameter water quality monitoring device (Horiba U-21 ex) and TOC was measured by the digestion method (Hach Co., TOC reagent set, ranging from 15–500 mg/L). In gaseous by-product analysis, the concentrations of O₂, CO, CO₂, NO, NO₂, SO₂, C_xH_y, and H₂S were measured by an online combustion gas analyzer (Eurotron Inc., GreenLine 8000).

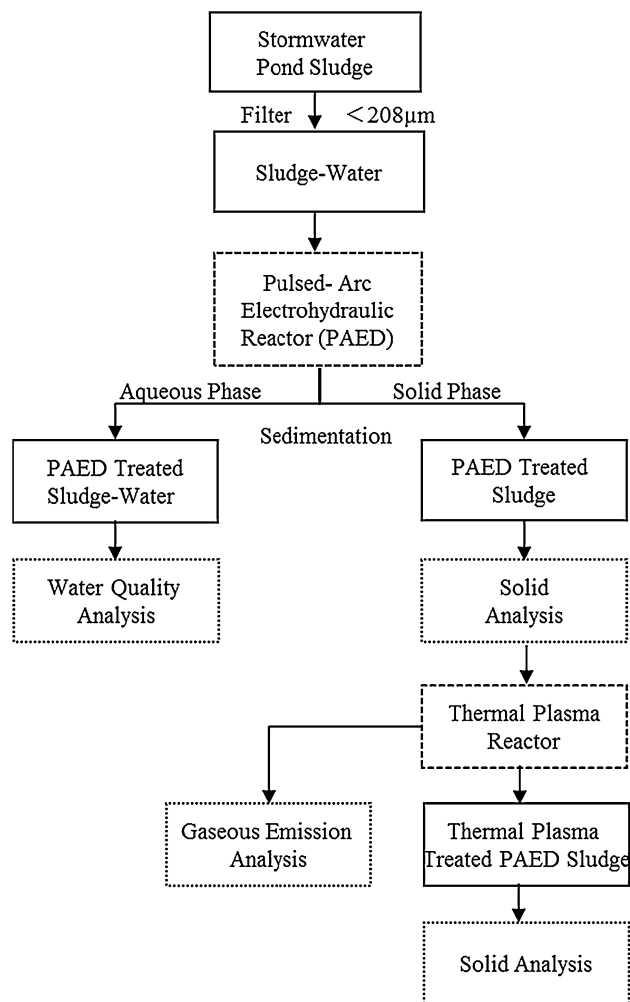


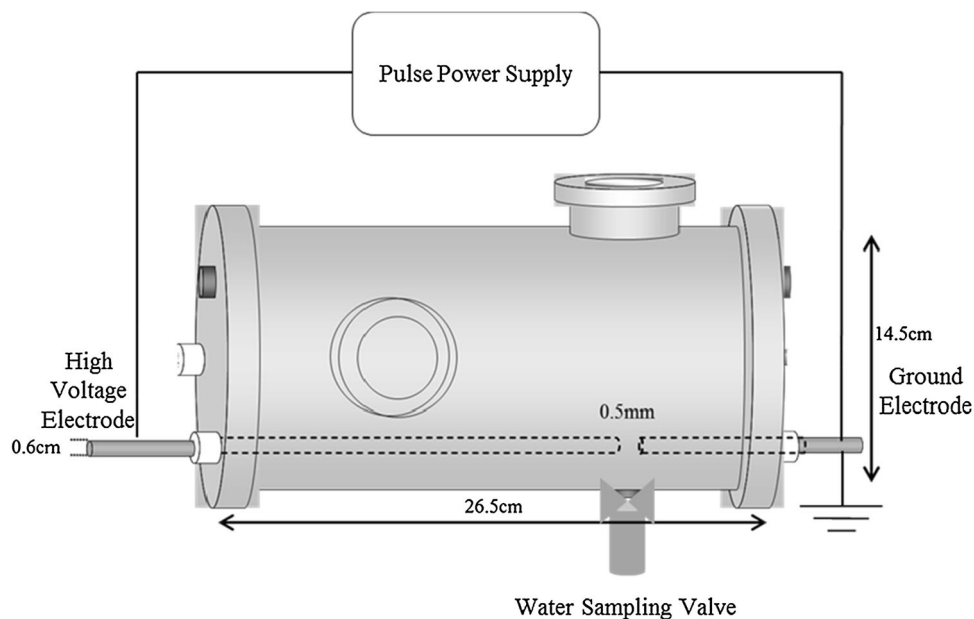
Fig. 1 Treatment train of the proposed integrated thermal plasma system

Results and discussion

Treatment of the aqueous phase of sludge water by pulsed arc electrohydraulic discharge

For water quality analyses, the liquid portion was separated from the solid fraction by sedimentation. PAED treatment formed O[•] and O₃[•] radicals during liquid discharges, and these radicals can degrade TOC to gaseous compounds. For stormwater with low TOC concentration (TOC < 50 mg/L), most of the TOC were degraded within 10 min and the pro-longed PAED treatment slightly increased DO concentrations. The continuous

Fig. 2 Schematic of pulsed arc electrohydraulic (PAED) treatment for aqueous phase of sludge water



generation of $O^{\cdot -}$ and $O_3^{\cdot -}$ radicals resulted in increasing of DO concentration.

In this study, the concentration of DO in pond water, on the other hand, decreased 27 % after treatment. This might be due to the fact that more reactions were carried out with ions and radicals in the contaminated pond water with higher concentration of TOC (TOC = 310 mg/L). From previous study, the DO concentration will eventually increase after the TOC degradation was terminated. Degradation of TOC accompanied with the detailed mechanism of PAED has been reported by the authors in other paper (Li et al. 2012a). The summary of the change of water quality is shown in Table 1. The standard deviation of all data was within 3 %. A reduction of 80 % of TOC was achieved within a treatment time of 5 min, and the decontamination increased over 90 % with a treatment time prolonged to 30 min. The cumulative energy input per volume (CEIV) of 5- and 30-min treatment was respectively, 2.7 and 16.2 kWh/m³. Due to the detection limit of the TOC measurement technique, concentration below 20 mg/L cannot be confirmed. The concentration of TDS dropped from 4 to 1 g/L with 30 min of treatment. It is supposed that the mechanisms in PAED encouraged sedimentation of TDS.

Other water quality parameters stayed relatively constant with increasing treatment time, which indicated that the reactions occurred in PAED did not have negative effects on these parameters. By optimizing the performance

of the PAED reactor, it is believed that the treated aqueous phase of the sludge water can be disposed safely into surface water bodies. The PAED system is currently installed in a wastewater treatment plant in pilot scale. This system can easily be expanded to treat stormwater sludge in the range of million per day. This approach has a great significance because the aqueous phase contributed more than 70 % by weight of sludge water, thus a huge volume reduction can be achieved prior to solid treatment by thermal plasma.

Treatment of the solid phase in sludge water by pulsed arc electrohydraulic discharge

In this study, we focused on examining the reduction of TOC and volatile metal concentrations in the solid phase of sludge water after PAED treatment. Since significant sedimentation was observed after PAED treatment, the solid phase was able to be separated by gravity and was dried under a fume hood for solid analyses. Figures 4a, b, and c showed the NAA elemental comparisons of the original and PAED-treated sludge for major elements (>10,000 ppm), minor elements (100 to 10,000 ppm), and trace elements (<100 ppm), respectively. In addition, elements such as C, O, Si, Cu, and S were determined by XRF, as shown in Fig. 4d. Two of the most abundant



elements, Al and Ca, decreased 1 and 2.5 % in mass, respectively. The concentrations of volatile elements such as Br and K decreased by 90 and 20 %, respectively. As shown in Fig. 4d, the weight of carbon was reduced by 20 % while the weight of sulfur decreased by 70 %. The standard deviation of all data was approximately 5 %. Since the PAED discharge in liquid solution generates oxygen and hydroxyl radicals, these radicals react and convert to their corresponding gaseous compounds such as CO, CO₂, and SO₂ (Li et al. 2012c). On the contrary, the weight percentage of oxygen increased more than 5 %

after treatment. This increase in oxygen concentration might be contributed by O₃ and O radicals after PAED.

Treatment of PAED-treated sludge by thermal plasma

The PAED-treated sludge was dried in room temperature and transferred to the thermal plasma reactor for further treatment. This treatment of dried PAED-treated sludge was conducted at an argon flow rate of 24 L/min for 120 min. In order to observe the effect

Fig. 3 Schematic of thermal plasma treatment for solid phases of sludge water

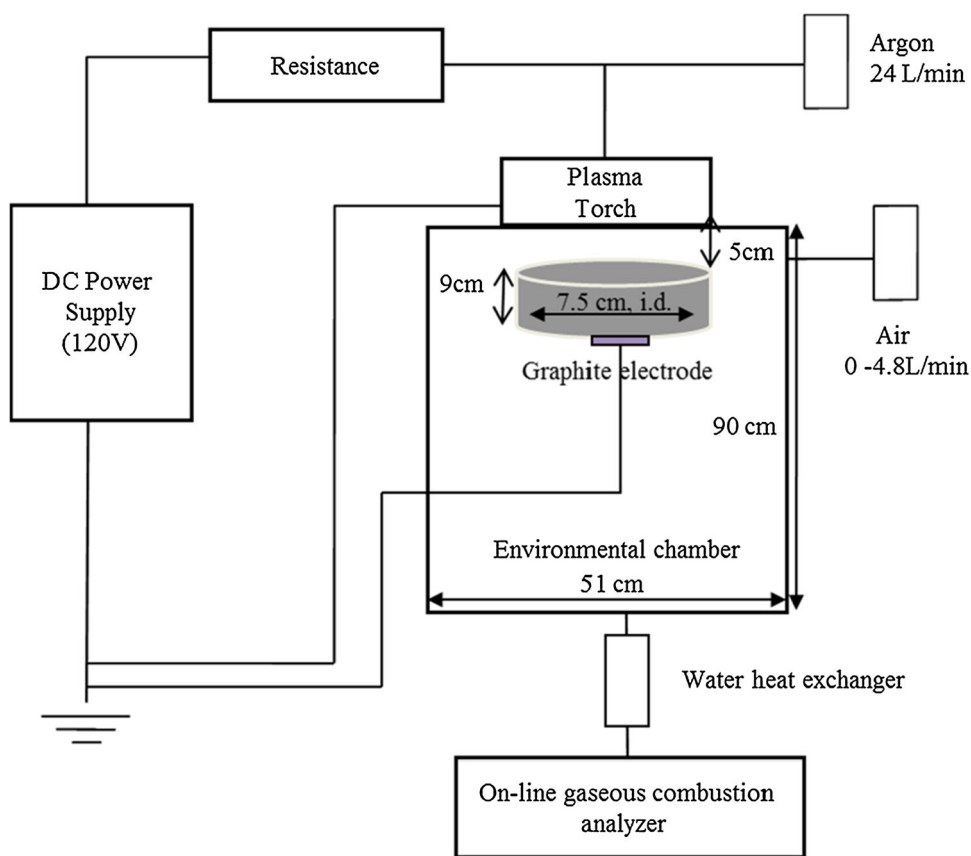


Table 1 Summary of the change of water quality by PAED treatment

	pH	Conductivity (mS/m)	Dissolved oxygen (mg/L)	Total dissolved solid (TDS) (g/L)	Salinity (%)	Total organic carbon (TOC) (mg/L)
Original sludge water	7.49	637	1.8	4	0.3	310
PAED treatment for 5 min	7.5	626	1.3	1.2	0.3	53
PAED treatment for 15 min	7.51	625	1.2	1	0.2	40
PAED treatment for 30 min	7.53	623	1.3	1	0.2	<20

of oxygen level on the concentration of gaseous by-products, various air flow rates were applied to the system at different treatment times. No air flow rate was applied into the environmental chamber at the start of the experiment. The air flow rate was increased to 2.4 L/min after 5 min and then gradually increased to 4.8 L/min. After 60 min, the air flow rate was reduced to 1.2 L/min and adjusted to 0 L/min after 90 min.

Figure 5 shows the images of dried PAED-treated sludge before and after thermal plasma treatment, respectively. The thermal plasma-treated sludge was slightly melted and the formation of slag might be possible (Chang et al. 2008). Figure 6 shows the X-ray energy dispersion solid analysis results of PAED-treated sludge after thermal plasma treatment. Carbon and sulfur were reduced to zero after treatment; Si, Mg, and Al decreased 25, 30, and 60 %, respectively, while Ca and Fe enriched 400 and 25 %, respectively. The standard deviation of all data was approximately 5 %. Depending on the plasma

temperature and other parameters, thermal plasma is capable of immobilizing inorganics inside the melted slag [31, 33, 35, 40, 46].

The gaseous by-products were sampled and analyzed online during thermal plasma treatment. Figure 7a, b, and c show the online emissions of carbon, nitrogen, and sulfur compounds with respect to oxygen concentrations. Figure 7a shows that emission of hydrocarbons was continuous and relatively constant throughout the treatment. The concentrations of C_xH_y slightly increased when the CO concentration started to decrease. CO and CO_2 were at maximum at a treatment time of 5 min. CO decreased slowly after it reached the peak level and stayed at 50 ppm after 60 min. The concentration of CO decreased to 7 ppm when the oxygen level was reduced to zero after 90 min. It may be concluded that the organic compounds were decomposed and formed CO within the first hour of treatment, and constant CO concentration was generated by thermal plasma operation, regardless of whether or not the organic compounds were

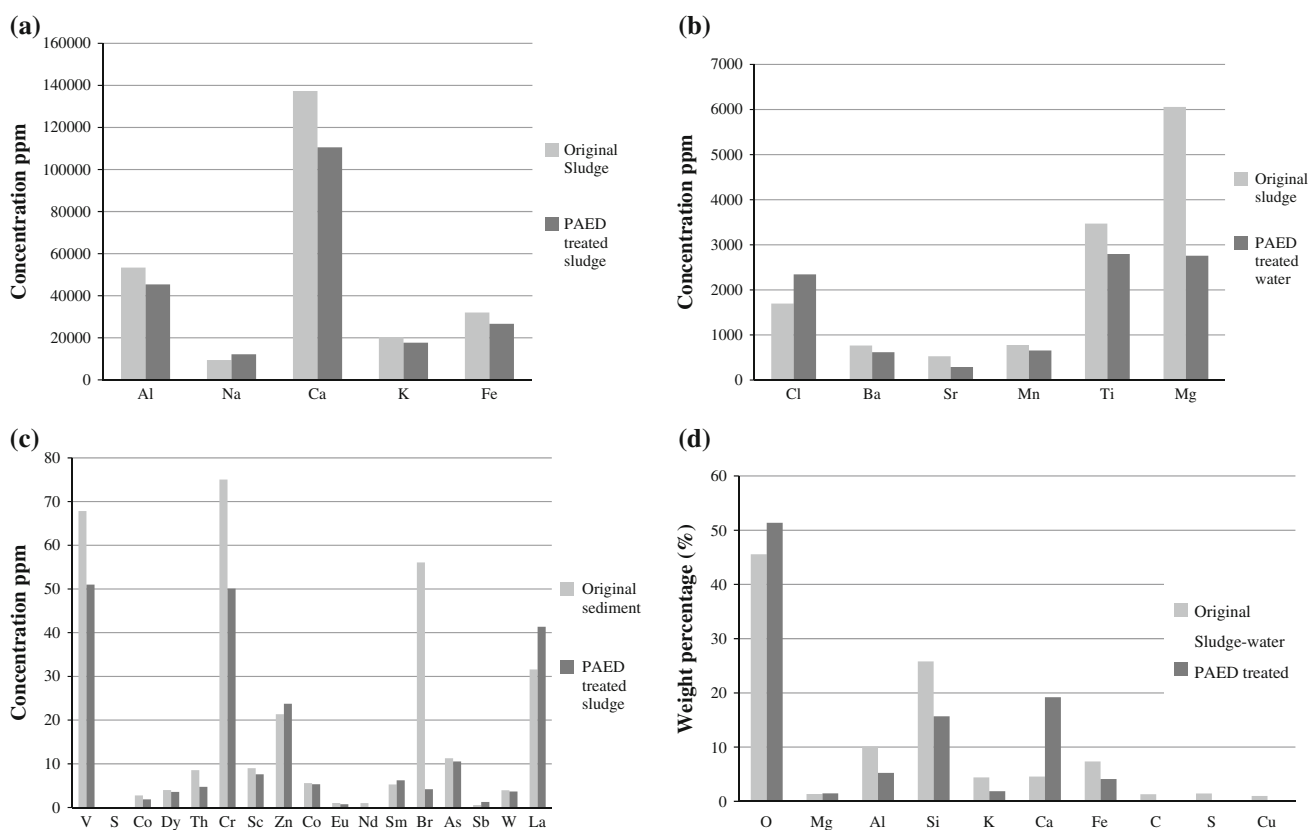


Fig. 4 **a** Comparison of major elements between original and PAED-treated sludge water observed by neutron activation analysis (NAA). **b** Comparison of minor elements between original and PAED-treated sludge water observed by neutron activation analysis (NAA). **c** Comparison of trace elements between original and PAED-treated

sludge water observed by neutron activation analysis (NAA). **d** Comparison of element weight percentages between original and PAED-treated sludge water observed by X-ray energy dispersion analysis



Fig. 5 SEM images of PAED-treated sludge before and after thermal plasma treatment (magnification of 4,800)

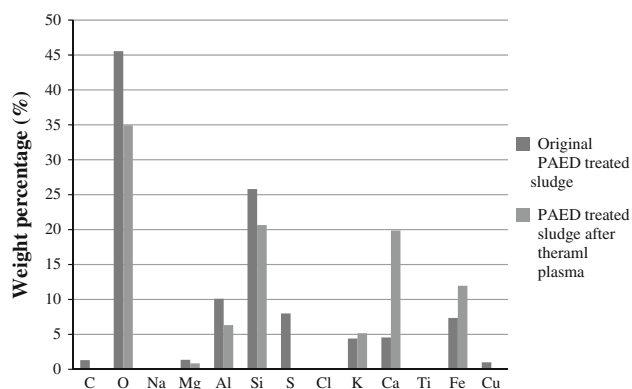
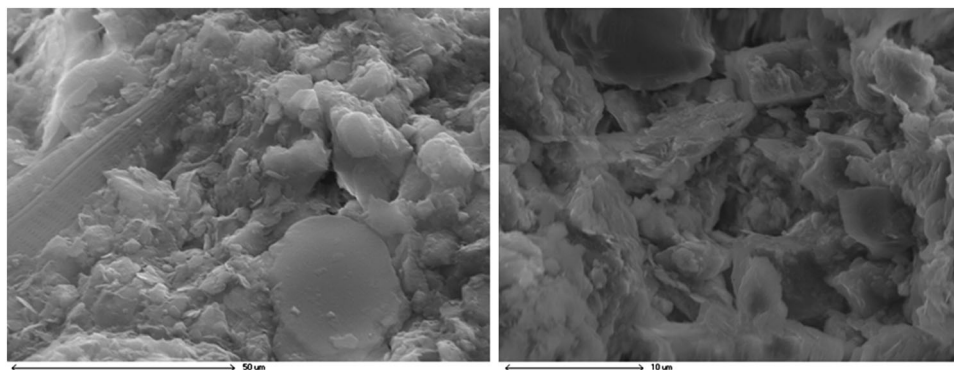


Fig. 6 X-ray energy dispersion solid analysis results of wet sludge and PAED-treated sludge by thermal plasma treatment ($P = 2.1$ kW) with argon flow rate of 24 L/min

present in sludge (Ara et al. 2005). Some organics were converted into CO_2 when the oxygen level increased to 4 % at a treatment time of 15 min. However, the generation of CO was more preferable due to limited supply of oxygen. Oxidation and combustion of organics were mostly completed at the treatment time of 90 min.

Figure 7b shows that the generations of NO and NO_2 were dependent on the oxygen levels as expected. Formation of NO was continuous, while NO_2 was generated in small amounts when oxygen level reached 4 %. This result agreed with previous observations of NO and NO_2 production in argon thermal plasma under air environment by Ara et al. (2005). With the same oxygen level of 4 %, the concentration of NO at the beginning was higher than the concentration at 60 min. It is believed that NO was generated by nitrate compounds in s during the first 10 min, and NO emitted after 60 min was solely

produced by thermal plasma in the argon-air environment. Figure 7c shows that SO_2 was mostly generated in higher oxygen levels and H_2S was produced under oxygen depletion conditions. Since the generation of H_2S was still continuous in small amounts (between 1 and 3 ppm) after 100 min, it suggested that decomposition of sulfur compounds in sludge need longer time as compared to organic and nitrate compounds. The accumulated concentrations of C_xH_y , CO, CO_2 , NO, NO_2 , H_2S , and SO_2 with 120 min of treatment were 7,500, 11,500, 11,200, 119, 260, 56, and 106 ppm, respectively. The standard deviation of total concentration of accumulated gas was approximately 15 %.

Conclusion

Reported in this paper is the design of an integrated treatment system and preliminary experimental results for the innovative treatment of urban stormwater sediments. It was shown that PAED reduced the concentration of TOC in the aqueous phase by 80 and 90 %, respectively, with a treatment time of 5 and 30 min. Thermal plasma treatment provided a complete reduction of TOC and sulfur contents in the solid phase. The concentrations of volatile elements such as Br, S, K, and C decreased approximately 90, 70, 30, and 20 %, respectively. It is summarized that the integrated plasma treatment system provided decontamination and disinfection in the aqueous phase by PAED, and completely destroyed toxic organics in the solid phase by thermal plasma treatment.

This research demonstrates a new approach of stormwater sediment treatment, especially those types that have high water contents and are severely

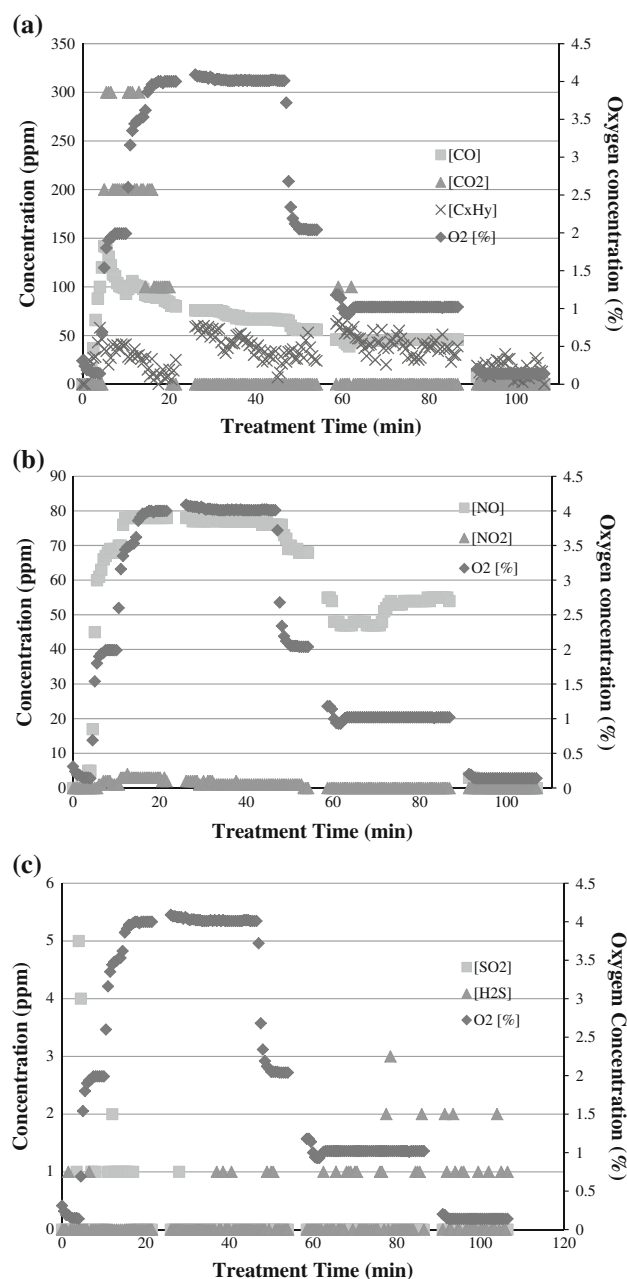


Fig. 7 a CO, CO₂, and C_xH_y generated by thermal plasma treatment of PAED-treated sludge with argon flow rate of 24 L/min and various air flow rates. b NO and NO₂ generated by thermal plasma treatment of PAED-treated sludge with argon flow rate of 24 L/min and various air flow rates. c H₂S and SO₂ generated by thermal plasma treatment of PAED-treated sludge with argon flow rate of 24 L/min and various air flow rates

contaminated by toxic organic compounds. A huge weight reduction can be achieved by PAED, which decontaminates the TOC in liquid phase prior to thermal plasma treatment. For the solid-phase treatment, the toxic organic compounds are converted into gaseous compounds while the inorganics are

immobilized inside the melted slag. From the gaseous emission analyses, it contained majorly (more than 99 %) hydrocarbons and carbon-related gaseous compounds. These gases contain energy value and might be reused as energy resources (Belgiorno et al. 2003; Consonni and Vigano 2012; Moustakas et al. 2005). For sediment contained higher amount of TOC, the treatment system might be energy-sustainable by converting gaseous emission into electricity energy to operate the thermal plasma reactor. A nonthermal plasma reactor is proposed as a post-treatment for gaseous pollution control to remove impurities of NO, H₂S, and SO₂ in the emission gas stream. It has been thoroughly studied elsewhere already (Urashima et al. 1998; Urashima and Chang 2000). With an adequate gaseous treatment device, the generated gases can be transferred to energy source and compensate a major portion of energy consumption of the system.

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