

Investigating the Effects of Atmospheric Pressure Air DBD Plasma on Physio-Chemical and Microbial Parameters of Groundwater

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Abstract

Cold atmospheric pressure plasma generated at atmospheric air produces high energetic electrons, ions, UV radiations, reactive oxygen, and nitrogen species (RONS) which has a wide range of applications in biomedicine, agriculture, textiles, water treatment, etc. Recently, many researches have been carried out on ozonation through DBD (Dielectric barrier discharge) plasma in water treatment. However, direct plasma treatment on water hasn't been extensively studied yet. In this study, we examined the change in physical, chemical and biological parameters of groundwater by the means of direct DBD plasma activation. The significant reduction in the concentration of biological parameters such as total coliform and E. coli with increasing treatment time was found. Besides that, DBD plasma also lowered down the concentration of physical parameters such as pH, turbidity, total suspended solids, total dissolved solids, and increased electrical conductivity and dissolved oxygen. In addition, the significant drop in the concentration of chemical parameters such as sodium, phosphorus, and calcium was found along with the reduction in heavy metals concentration such as iron, cadmium, lead, chromium, manganese, and zinc. However, the concentration of nitrite, nitrate, and sulfur augmented gradually. Our results revealed the great ability of direct plasma in water treatment.

Subject Areas

Computational Chemistry

Keywords

Plasma Treatment, RONS, Physio-Chemical, Microbial, Water

1. Introduction

While access to safe drinking water is essential for the health and well-being of all living beings [1], most of the people are unable to access adequate and safe drinking water, especially in developing countries [2] [3]. Groundwater is water contained in an aquifer matrix located beneath the surface in the saturated zone [4] [5] and the quality of groundwater is interrelated with factors such as climate, slope, drainage conditions, water-rock interaction, and anthropogenic activities [6]. The dramatic shored up in water pollution especially due to the rapid population growth and unsustainable urbanization exacerbated the quality of water, which is the leading cause of humans to have dreadful health problems [7] [8] [9]. Many factors such as inter person transmission, food contamination, poor sanitation, and water contamination through fecal-oral route surge the waterborne infectious disease outbreaks [10] [11].

One of the contemporary researches in the field of quality of water is the treatment of water by cold plasma [12] [13] [14] [15]. Among the various methods adapted, dielectric barrier discharge (DBD) plasma has high efficiency in removing contamination [16]. In general, the cold plasma at atmospheric pressure in ambient air produces high energetic electrons, ions, reactive oxygen and nitrogen species (RONS), and ultraviolet radiation which when treated with water changes its parameters out of which, ozone is the stable, long-lived species [17] whereas some of the hydroxyl ions and nitrogen species are short-lived species [18] [19] [20]. Various researches have been carried out on water purification through ozone produced by DBD plasma [21] [22], however, direct plasma discharge treatment in water has yet not been studied in detail. In this method, not only ozone but other short-lived reactive species get to interact with water [20]. The reaction mechanism during the plasma discharge is as follows:

$$O_2 + e^- \rightarrow O + O + e^{-1}$$
$$O_2 + O \rightarrow O_3$$
$$3O_3 + H_2O \rightarrow 2OH^* + 4O_2$$
$$e^- + H_2O \rightarrow OH^* + H^* + e^{-1}$$

The main objective of this study was to identify the ability of direct plasmatreated discharge treatment in enhancing the water quality on the basis of the research question; what are the effects of direct plasma-treated discharge in changing the groundwater quality parameters?

2. Experimental Procedure

2.1. Experimental Setup

In the experimental setup for plasma treatment, the reactor system was covered by a transparent glass cylinder and consists of two electrodes of dimension (5.10 cm diameter and 1 cm thickness) made up of brass inside. A petri dish of borosilicate glass acts as a dielectric barrier, with water of volume 40 ml was kept between the electrodes. The distance between the upper electrode and surface of the water is 1.8 cm and the distance between two electrodes is 2.2 cm. A small hole at the top is made to allow external airflow. The two terminals of the high voltage probe are connected to the upper and lower electrodes of the plasma reactor.

The current probe is attached across the shunt resistor which is then fed into the oscilloscope (Tektronics TDS 2002 digital oscilloscope) for current measurement. A high voltage of 13.71 KV at line frequency of 50 Hz was applied. The power consumed by the discharge was 32.96 Watt (**Figure 1**).

2.2. Sample Collection

The water sample was taken from groundwater well of one residential house (common well) of Panauti municipality of Kavreplanchowk District of Bagmati Province of Federal Democratic Republic of Nepal ($27^{\circ}34'59.8''N 85^{\circ}30'23.9''E$) in June 2019. The samples were stored at $4^{\circ}C$ in the refrigerator.

2.3. Analytical Methods

The standard methods were used to determine physiochemical and biological parameters [23]. Physical parameters; pH, electrical conductivity (EC) and dissolved oxygen (DO) were measured using standard multi-probe (HANNA edge with HI764080 and HI11310 probe) whereas the turbidity was determined by turbidity meter (HANNA HI88703-Turbidimeter). The total dissolved solids (TDS), and total suspended solids (TSS) were analyzed by the gravimetric method. Biological parameters; total coliform and *E. coli* were determined by the membrane filter technique.

The chemical parameters; nitrate (NO_3^-) , nitrite (NO_2^-) , total iron (Fe), chromium (Cr), manganese (Mn), sulfur (S) and phosphorus (P) determination were carried out by UV-Spectrophotometer (Shimadzu UV-1800). Whereas sodium (Na), lead (Pb), Cadmium (Cd), calcium (Ca), and zinc (Zn) were determined by atomic absorption spectrometry (SavantAA, GBS).



Figure 1. Schematic diagram of the experimental setup: Electrodes (1, 2), Plasma discharge (3), Petri dish with sample water (4), Air pump (5), Power supply (6), Transparent cylinder (7), Ballast resistor (8), High Voltage supply (9), High Voltage Probe (10), Shunt resistor (11), Personal Computer (12)

2.4. Statistical Analysis

All the experiments were carried out triplicate and data were presented in mean \pm SD. Pearson's correlation analysis was done for physio-chemical parameters data analysis with 95% confidence interval.

3. Results and Discussions

3.1. Physical Parameters

pH is responsible for the corrosivity of water [24] and the low pH of water might cause severe ailment like gastrointestinal disorder [25]. Our results revealed the gradual declination of pH with an increase in plasma treatment time (Table 1). The responsible factors for the decrease in pH might be several RONS species which are produced during plasma treatment on water and plasma acids [26] [27]. Electrical conductivity is one of the indicators of dissolved inorganic ions in water [28]. The electrical conductivity (EC) and dissolved oxygen (DO) in plasma-treated waters were increased linearly (Table 1) and this result is consonant with the previous works [12] [29] [30]. Due to the presence of particulates such as clay, silt, organic matter, algae, and other microorganisms water loses its transparency and known for turbid [31] which can cause severe health disorder [25]. Our results revealed a decrease in turbidity with the increase in plasma treatment time which is related to the decrease in total dissolved and suspended solids on the treated water (Table 1). Total dissolved solids (TDS) is the sum of inorganic salts and small amounts of organic matter present in water which usually consists of sulfates, calcium, magnesium, nitrate, nitrites, etc. [32]. Plasma treatment shows significant decrease of TDS in water (Table 1). The total suspended solids (TSS) are the dry weight of the non-dissolving particles in water. Plasma treatment shows significant changes with decrease in TSS as well; which is similar to the results of the previous investigators [12]. The overall results of physical parameters showed a significant negative correlation with treatment time (p < 0.05) *i.e.* the concentration of parameters decreased significantly with an increase in plasma treatment time whereas DO and EC showed positive correlation with the treatment time (p < 0.05).

3.2. Biological Parameters

Reactive species such as OH, H_2O_2 , NO_3^- , and O_3 in the liquid have potent effects on microorganisms [33] [34]. *E. coli* and coliform bacteria are the fecal pollution indicators that are used to evaluate the health of aquatic ecosystems and the potential for health effects among individuals within those environments [35]. Safe drinking water requires the stability of the microbial community to minimize the release of pathogen [36] and environmental conditions that favor an increased load of pathogens in water also are crucial factors contributing to outbreaks of waterborne diseases [37] [38]. Our results revealed a significant decrease in the colony forming unit (CFU) per 100 mL sample for *E. coli* as well as total coliform with the increase in plasma treatment time (**Table 1**). Since, the

Parameters		Treatment Time			
		Untreated	5 minutes	10 minutes	15 minutes
Physical	рН	8.90 ± 0.00	8.80 ± 0.00	8.70 ± 0.00	8.60 ± 0.00
	Turbidity (NTU)	1.40 ± 0.00	1.30 ± 0.00	0.85 ± 0.00	0.70 ± 0.00
	Dissolved Oxygen (mg/L)	6.50 ± 0.00	6.61 ± 0.00	6.70 ± 0.02	6.96 ± 0.02
	Conductivity (µS/cm)	262.00 ± 0.00	264.00 ± 0.00	266.00 ± 0.00	268.00 ± 0.00
	Total Dissolved Solid (mg/L)	150.00 ± 0.06	50.00 ± 0.00	25.00 ± 0.00	0.00 ± 0.00
	Total Suspended Solid (mg/L)	75.00 ± 0.05	50.00 ± 0.00	25.00 ± 0.00	0.00 ± 0.00
Biological	Escherichia coli (CFU/100 mL)	>100	>50	0	0
	Total Coliform (CFU/100 mL)	>100	>50	0	0

 Table 1. Results on physical and biological parameters.

major factor for the increase in bacterial inactivation might be the increase in the applied voltage which increases the production of reactive species; by means of electroporation, cell membrane permeability increases and the reactive species kills the microorganism by deactivating the cell [15]. This significance positive effects of plasma treatment on controlling *E. coli* and total coliform are advantageous in water treatment.

3.3. Chemical Parameters

Calcium is abundant in drinking water; mainly helps in maintaining bones and essential for the functioning of muscles and nervous system [39] and lack of this ion causes cardiovascular disease [40]. However, a high level of calcium consumption leads to nausea, vomiting, weakness, and muscle cramp and bone pain [41] [42]. The hexavalent chromium is one of the dangerous heavy metals found in well water [43]. The chromium reacts with oxygen at room temperature to form oxides and also with other reactive species like hydroxides and plays an important role in the maintenance of normal glucose metabolism and acts as a cofactor for insulin [44]. Manganese is a pernicious heavy metal found in well water; although, a high level of manganese causes nervous system disorder, sperm damage, impairments fertility, nephritis, and nephrolithiasis [45] [46] [47]. Although naturally present phosphorus in water is not harmful to humans, high concentration can cause dreadful diseases such as diarrhea, hardening of organs and soft tissues [48]. The fluctuating trend of phosphorus in plasma treated water is pronounced in the previous works [29]. Our results revealed the decrease in calcium, chromium, manganese, and phosphorus concentration with an increase in plasma treatment time (Figures 2(a)-(d)).

The excessive intake of nitrate with drinking water causes thyroid, colorectal cancer, breast cancer, methemoglobinemia [45] [49]. Since, plasma discharge produces reactive nitrogen species like NO_2^- and NO_3^- , which results in the increase of concentration of nitrate and nitrite in water with the increase in treatment time (Figure 3(a) & Figure 3(b)). These results are comparative with the



Figure 2. Results on calcium, chromium, manganese and phosphorus.

results from previous investigators [12] [14]. Very low sodium on diet is recommended by the Environmental Protection Agency (EPA); the higher concentration of sodium intake could cause severe health impacts on living beings [50]. Our results showed the decreasing trend of sodium with increasing treatment times (**Figure 3(c)**). Sulfur doesn't create any great health hazards but exposure to excessive concentration might create a temporary laxative effect such as irritation to the skin, eye, and respiratory problems [51] [52] [53]. The concentration of sulfate ion in control and plasma-treated samples shows the increasing trend (**Figure 3(d**)), which is a disadvantage of the plasma treatment.

Iron is the heavy metals found in drinking water which is generally good for health. However, the large concentration of iron in water is not suitable; which causes hemochromatosis, leading to effects on the liver, pancreases etc. [46]. The elevated level of iron imparts smell, taste, and stain on clothes [54]. In our result, the concentrations of iron in the control sample and plasma-treated water are varying in degree with the astonishing decrement (**Figure 4(b)**). The decreasing trend for iron concentration could be explained by the fact that iron reacts with ozone and hydroxides to form iron oxides and hydroxides which get precipitated



Figure 3. Results on nitrate, nitrite, sodium and sulfur.

and settles at the bottom of the water [47]. Cadmium is a significant contaminant of water with high toxicity due to its easy solubility [55]. Despite low abundance in nature cadmium poses an increasing environmental and health risk with profound biological effects on human beings such as dysfunction of renal and lung [56]. Lead causes damage to the central nervous system [57] and also damage to the kidney, liver, reproductive system, and basic cellular processes [58]. Most zinc is introduced into water by artificially with increased anthropogenic activities and an excess amount of zinc in drinking water causes nausea and vomiting [59]. Our results showed the exceptional decrease in heavy metal content of water with the plasma treatment (Figures 4(a)-(d)). The overall results showed the significant negative correlation between the concentration examined chemical parameters and plasma treatment time (p < 0.05) except the significant positive correlation for nitrate, nitrite and sulfur (p < 0.05).

4. Conclusion

From this study, we can conclude that the direct plasma treatment in water has



Figure 4. Results on cadmium, total iron, lead and zinc.

great significance in water quality enhancement. This study shows the potential of direct plasma treatment in the water for the disinfection of bacteria in the water and significant reduction of heavy metals content such as concentration of chromium, manganese, cadmium, iron, lead, and zinc. The overall result gave us the plethora of scientific evidence that the treatment of water with the direct atmospheric air pressure DBD plasma is beneficial for the treatment of water for different physical, chemical, and biological parameters whereas for some chemical parameters, this treatment is counterproductive. Ten minutes of plasma treatment are the best in augmenting overall parameters of groundwater. Moreover, the effects of plasma treatment on wastewater can be studied in future that would aid in the purification of water targeting a wider range of people.

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Author's Contribution

DPS, RS, RPG, SPP and BPP conceptualized and performed preliminary works.

RS and SPP did the laboratory works, data generation, analysis and verification. RS, SPP, BPP, DPS wrote, edited and reviewed the manuscript.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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