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Research Article

IMPACT OF COLD ATMOSPHERIC PLASMA JET TREATMENT ON THE PHYSICOCHEMICAL PROPERTIES OF SURFACE WATER

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ABSTRACT

Cold atmospheric plasma jet (CAPJ) is a rapidly emerging multidisciplinary research field that stands out for its chemical-free and eco-friendly characteristics. It has garnered notable interest in recent years owing to its vast potential applications across various domains. One of its intriguing applications is in the treatment of water. Plasma transforms water into an acidic environment, leading to alterations in several key properties, including redox potential, electrical conductivity, dissolved solids, pH, and turbidity. Water treated with plasma, due to its distinct chemical composition compared to regular water, offers a promising alternative for microbial disinfection. In this research, CAPJ is generated using high voltage power supply (5 kV, 20 kHz) at standard atmospheric conditions and room temperature in an argon environment. The discharge generated by this system is thoroughly characterized using electrical and optical methods. Water sample collected from tap and well are treated with cold plasma jet and their physiochemical properties have been characterized.

Keywords: Cold atmospheric plasma jet (CAPJ), chemical properties, physical properties, physicochemical properties, plasma liquid interaction

INTRODUCTION

Nowadays, there has been an increasing emphasis on the interaction between plasma and liquid, referred as plasma-liquid interaction. This increased interest arises from the confirmed potential of plasma technology, as demonstrated by previous research. Moreover, in response to the critical global concerns of water pollution and the scarcity of potable water, a growing body of research has focused on exploring the use of different types of discharge plasmas for water purification and wastewater treatment, with the goal of improving water quality (Bruggeman *et al*., 2016; Rezaei *et al*., 2019; Yin *et al*., 2023). Cold atmospheric plasma jet (CAPJ) is a unique form of plasma characterized by significantly higher electron temperatures compared to ion temperatures, while the bulk plasma temperature stays low. Cold atmospheric plasma, on the other hand, serves as a fundamental tool for facilitating various chemical reactions at lower temperatures. Cold atmospheric plasma jet treatment has proven effective for enhancing the quality of both underground well water and surface drinking water. Bourke *et al.* and Shrestha *et al.* show that it leads to a substantial decrease in the concentrations of biological parameters, including total coliform and *E. coli*, as the treatment duration is extended (Bourke *et al*., 2018; Shrestha *et al*., 2020). Currently, the use of cold atmospheric plasma jet for water treatment is an increasingly active area of research. The diverse mechanisms of action associated with cold plasma offer a fertile ground for innovative solutions.

Water treated by CAPJ yields an acidified solution containing RONS, collectively referred to as plasma-activated water (Thirumdas *et al*., 2018). The study of physicochemical properties of PAW, including conductivity, pH, turbidity, and the comparing of chemical parameters before and after treatment have been done. The simultaneous occurrence of these physio-chemical phenomena sets PAW as a promising method for liquid processing (Bardos & Barankova, 2010; Foster *et al*., 2012). Additionally, cold atmospheric plasma jet alters physical and chemical properties such as pH, dissolved oxygen (DO), turbidity, conductivity, nitrite, nitrate, ammonia etc. It's worth noting that the composition and concentration of reactive species in PAW are influenced by the choice of gases and liquids used in the plasma generation process (Hamdan *et al*., 2018; Svarnas *et al*., 2022). The generation of plasma in water presents an opportunity to introduce advanced oxidation processes into water without raising the temperature significantly. This technology holds the potential to revolutionize drinking water treatment (Hübner *et al*., 2024). Our investigation centres on assessing the transformation of physical and chemical parameters in both surface tap water and underground well water, for investigation of its potential applications. The significant alterations observed in these parameters following plasma treatment underscore the significance of plasma-liquid interaction as a valuable tool for enhancing water quality for irrigation as well as for drinking purpose (Masood *et al.*, 2024; Sanchez *et al.*, 2025).

Materials and Methods

Figure 1 provides a schematic representation of the experimental setup alongside an image of the cold atmospheric plasma jet discharge. To ensure continuous operation and cost-effectiveness, the CAPJ was constructed using readily available local materials. The setup consists of a glass tube with an external diameter of 4 mm and an internal diameter of 3 mm. Argon gas is introduced into the tube from the top, as shown in Figure 1. Two copper tapes, each 3 cm in length, are wrapped around the outer surface of the tube to serve as electrodes, with a 5 cm gap maintained between them. The argon gas flow is regulated at a constant rate of 5 L/min. Water samples (10 mL) from various sources were treated for durations ranging from 2 to 12 minutes, using a system operating at 5 kV and 20 kHz. The cold atmospheric plasma jet length, measured from the nozzle, was 5 cm. For optical characterization of the discharge, emission spectra were recorded using an optical emission spectrometer (USB 2000+, Ocean Optics). The electrical characterization of the discharge was performed using a voltage probe across the electrode and a current probe across an appropriate resistance. It also shows the nature of the discharge. The electrical characterization of the stable cold atmospheric plasma jet, generated using a high voltage power supply, PINTEK HVP-15HF, high voltage probe and TEKTRONIX TDS 2002 two channel digital oscilloscope were used for electrical characterization. To determine electron density and energy dissipation by an electron in each cycle, the power balance method is used.

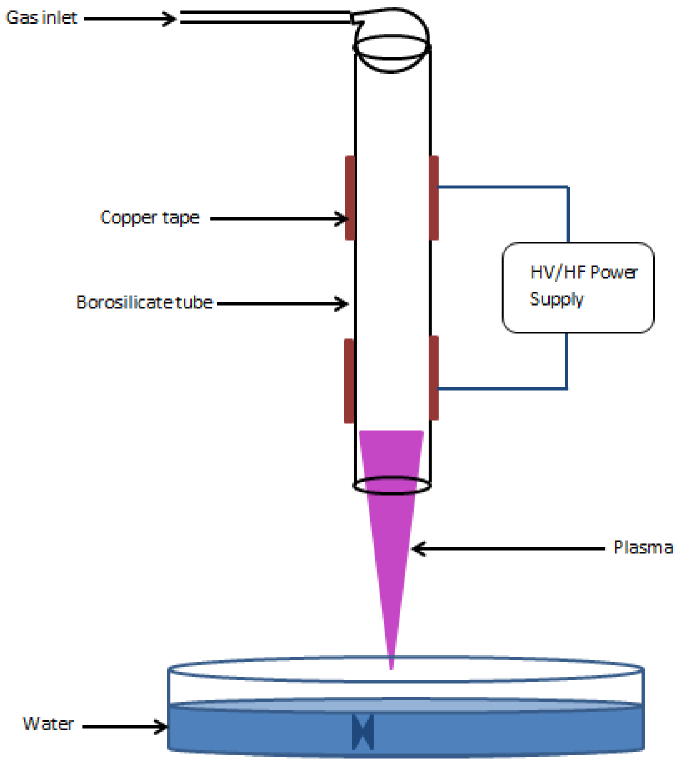

Figure 1. Schematic diagram of the experimental setup

Figure 2 depicts a photograph illustrating the direct interaction between a cold atmospheric plasma jet and water. The water samples were collected from surface drinking water sources, including tap and well water, in Dudhpokhari, Kirtipur, Kathmandu, Nepal. A CAPJ was introduced into the water samples contained in a beaker, and the physical and chemical parameters of the surface drinking water sources were analysed before and after treatment. The treated water samples were examined, and the concentration of chemical species were determined using the UV-Visible spectrophotometer (Baniya *et al.*, 2021).

RESULTS AND DISCUSSION

Discharge temperature

The temperature variation of the plasma plume tip over time at a discharge voltage of 5 kV is shown in Figure 3. It was linear up to 70 seconds then practically remains constant at 25^0 C. Thus, the temperature of plasma jet approximately matches with surrounding temperature. So, this plasma is also called cold atmospheric plasma jet and widely used in investigating on physiochemical parameters of water samples (Maldonado *et al.*, 2019; Baniya *et al.*, 2020).

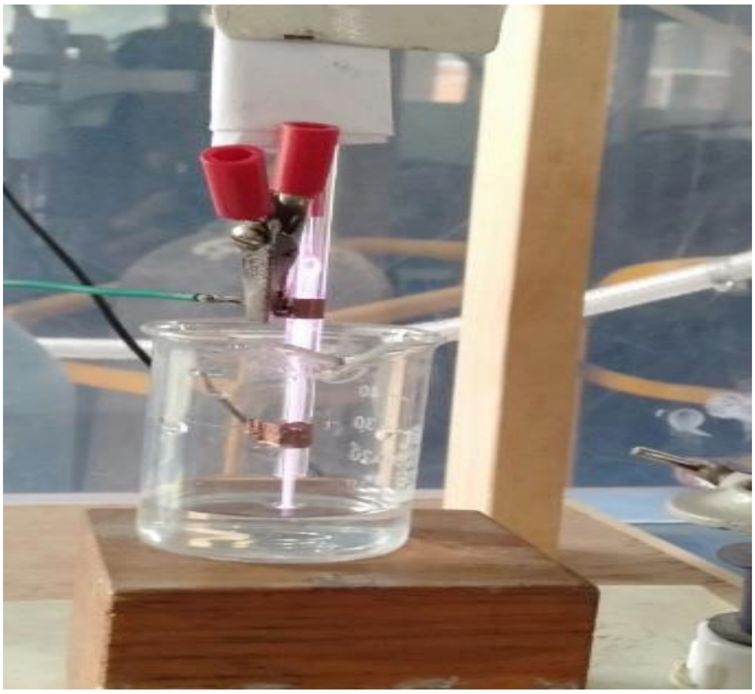

Figure 2. Cold atmospheric plasma jet (CAPJ) interaction with water samples [14]

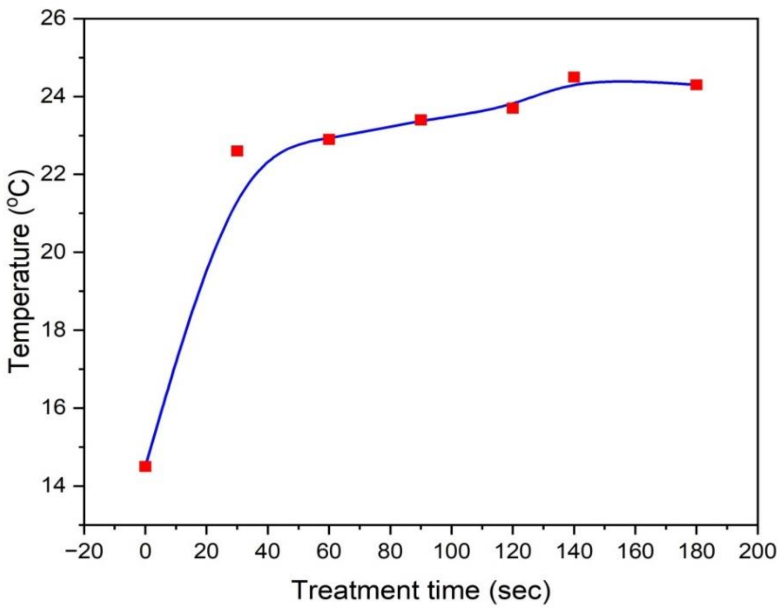

Figure 3. Direct temperature measurement of CAPJ at 5 kV

Electrical Characterization of CAPJ
Power Balance Method

The estimation of electron density (n_e) was evaluated using the power balance technique which assumes that the energy lost by plasma characteristics equals the power given by the source. The equation used to estimate the electron density is given by (Balcon *et al.*, 2017).

$$n_e = \frac{P_{av}}{2Av_bE_{lost}} \quad (1)$$

Where, P_{av} is the average power dissipation on the discharge, A is area of the electrode, E_{lost} represents the energy lost per cycle during the discharge, and v_b be the Bohm velocity.

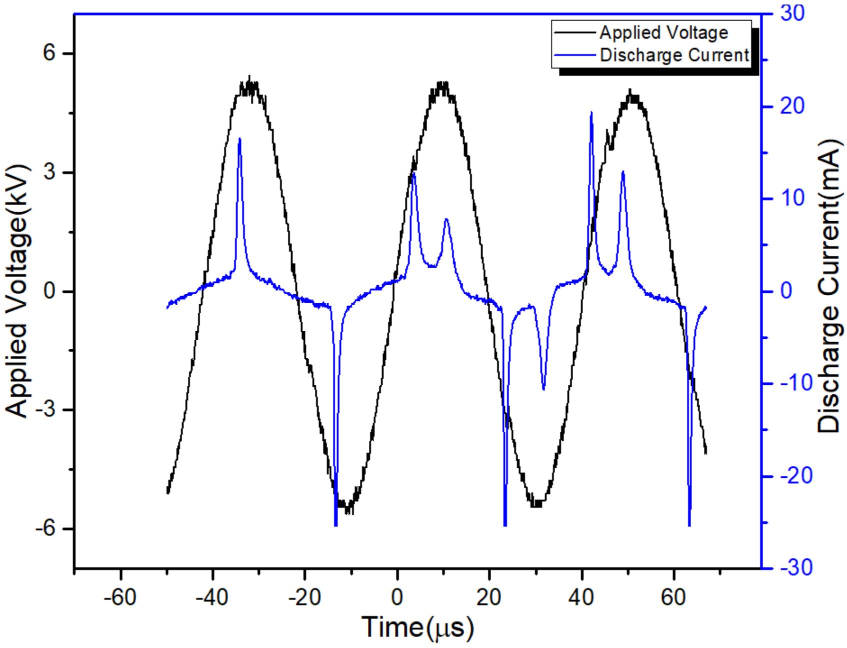

Figure 4. current-voltage waveforms of the discharge

To calculate the electron density (n_e), the following values were used: The electrodes distance was 5 cm apart, applied frequency (f)= 20 kHz, A=12.46× 10^{-6} cm^2, (v_b) = 2×10^3 m/sec, E_{lost} = 50 eV. Using these values in equation 1, the electron density was found to be 1.42×10^{14} cm^{-3}. The average power dissipation on the discharge is measured by the integration of instantaneous voltage $V(t)$ and current $I(t)$ (Kramida *et al.*, 2018) which is given by,

Discharge power, $P_{av} = f\int_0^T V(t)I(t)dt$ (2)

where, f is the frequency and T is the time of the cycle. Using values of V_{rms} = 5 kV and I_{rms} = 20 mA into Equation (2), the power consumed per cycle was found to be approximately 56 watts.

Optical characterization of CAPJ

In optical charcterization, electron temperature is estimated of the discharge. Boltzmann method was used for the estimation of electron temperature (Ohno *et al.*, 2006; Zhang *et al.*, 2019; Thouin *et al.*, 2023):

$$\ln\left(\frac{I\lambda}{Ag}\right) = -\frac{E}{k_B T_e} + ln(C) \quad (3)$$

where, "E, I, g, A, and k_B are the energy of the upper transition state, intensity for a particular wavelength (λ), statistical weight, transition probability, and Boltzmann's constant respectively. Here argon I species are used, and their respective values are taken from the NIST database" (Kramida *et al.*, 2018).

Table 1. Intensities and wavelengths of Ar I lines along with energy level and transition probability

Wavelength (nm)	Intensity (a.u.)	Energy (eV)	Transition probability* Statistical weight [A*g] (s^{-1})
696.54	13885.26	13.22	1.90×10^7
750.39	6609.19	13.47	1.45×10^7
763.51	15359.44	13.47	1.22×10^8
772.38	11635.61	13.32	3.51×10^7

The intensity wavelength graph of the spectra produced from CAPJ using argon gas is shown in figure 5. Wavelengths, intensities and their respective values of energy level and product of transition probability and statistical weights are given in the table 1.

Using the values of Table 1 in equation (3) and plotting $\ln\left(\frac{I\lambda}{Ag}\right)$ versus energy level (E) a graph is obtained as shown in figure 6. The reciprocal of the negative slope of the line provides the electron temperature, which in our experiment was estimated to be 0.16 eV.

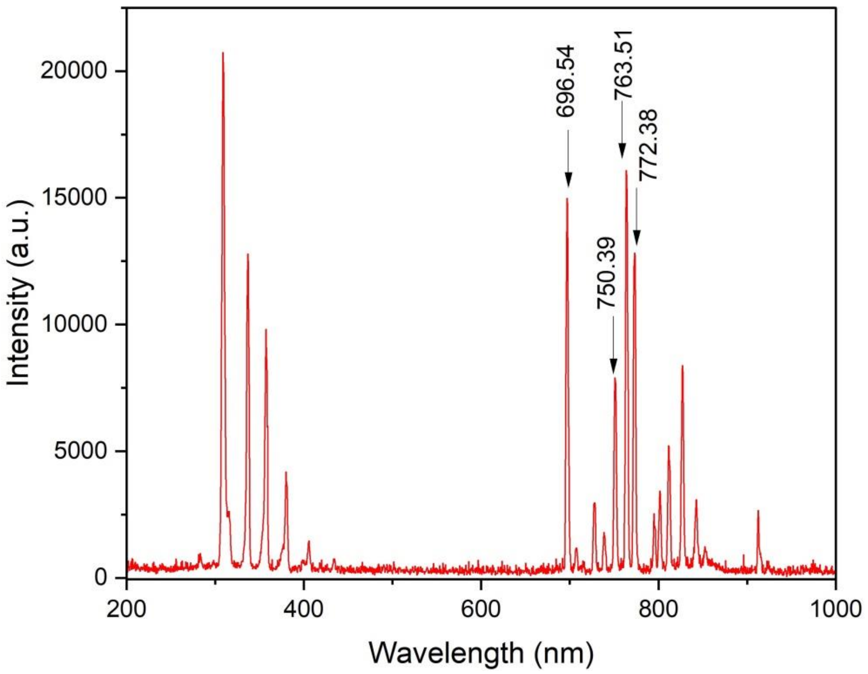

Figure 5. Spectra of the argon discharge

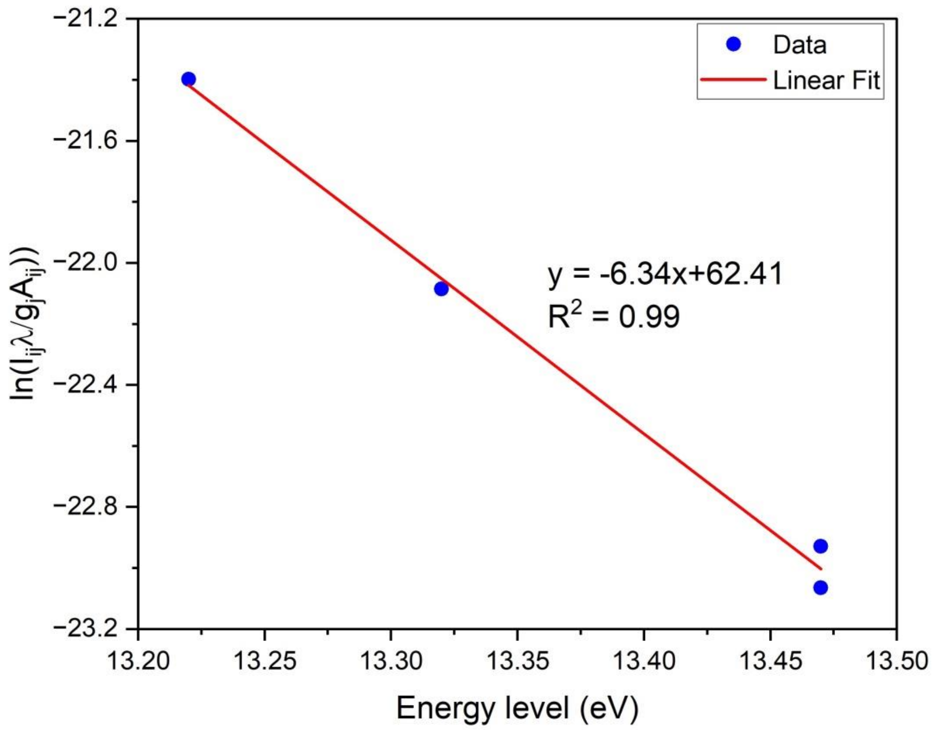

Figure 6. Graph showing Boltzmann's plot for estimation of T_e

Chemical Parameters

Nitrite/Nitrate ions (NO_2^- / NO_3^-)

Figures 7 and 8 illustrate the dependence of nitrate and nitrite ion concentrations on the plasma exposure duration. Initially, untreated tap water contains a negligible amount of nitrate, while well water has a nitrate concentration of 6.2 mg/L. As the exposure duration increases, the nitrate concentration rises in both tap water and well water. In the case of nitrite, its concentration shows a slight increase in well water, but a significant increase is observed in tap water.

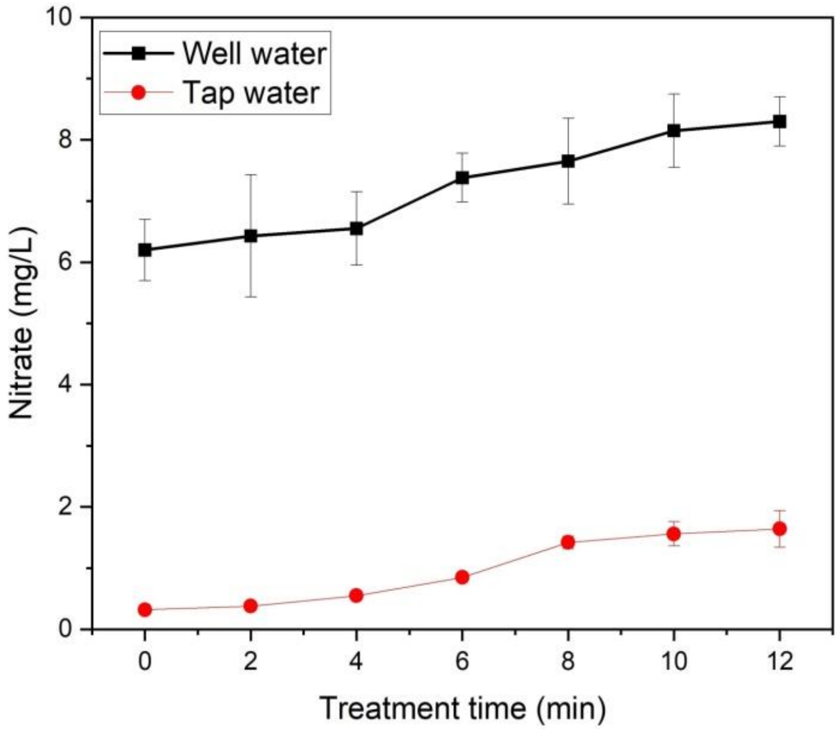

Figure 7. Variation in nitrate concentration with treatment time for tap and well water.

In treated water highly reactive species nitric oxide may react with oxygen in aqueous solution to produce nitrite (NO_2^-). It decreases pH of the solution. Nitrogen species are formed by the dissolution of nitrogen oxides formed in plasma discharge. Nitrous acid (HNO_2), major sources of Nitrite anions. In acidic medium, it also decomposes rapidly into nitrogen dioxide which reacts with hydroxyl radicals in plasma discharge and lead to the formation of peroxynitrous acid and finally converts into stable nitrate anions (NO_3^-). Also, in acidic condition, nitrite (NO_2^-) react with hydrogen peroxide forming peroxynitrite to nitrate (NO_3^-) (Braun *et al.*, 1988; Dhali & Sardja, 1991).

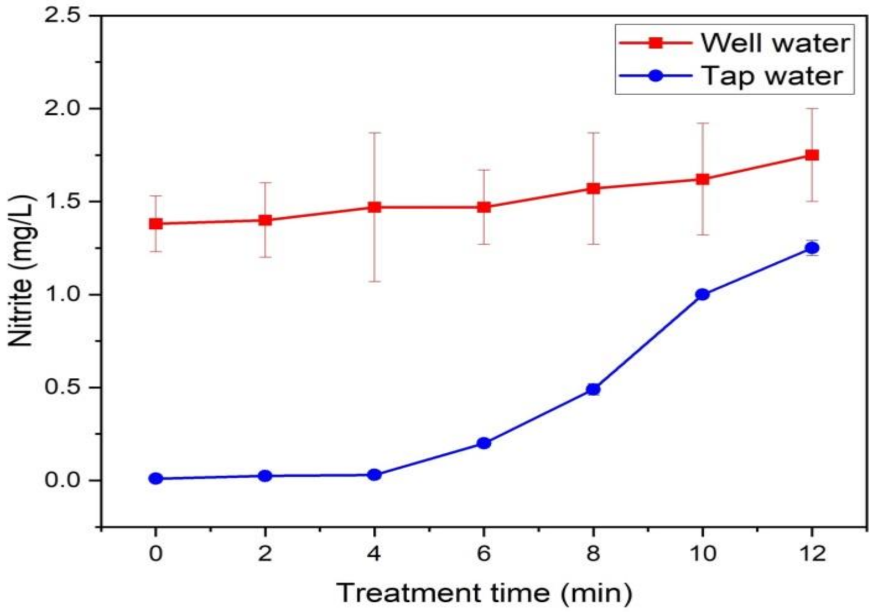

Figure 8. Variation in nitrite concentration with treatment time for tap and well water.

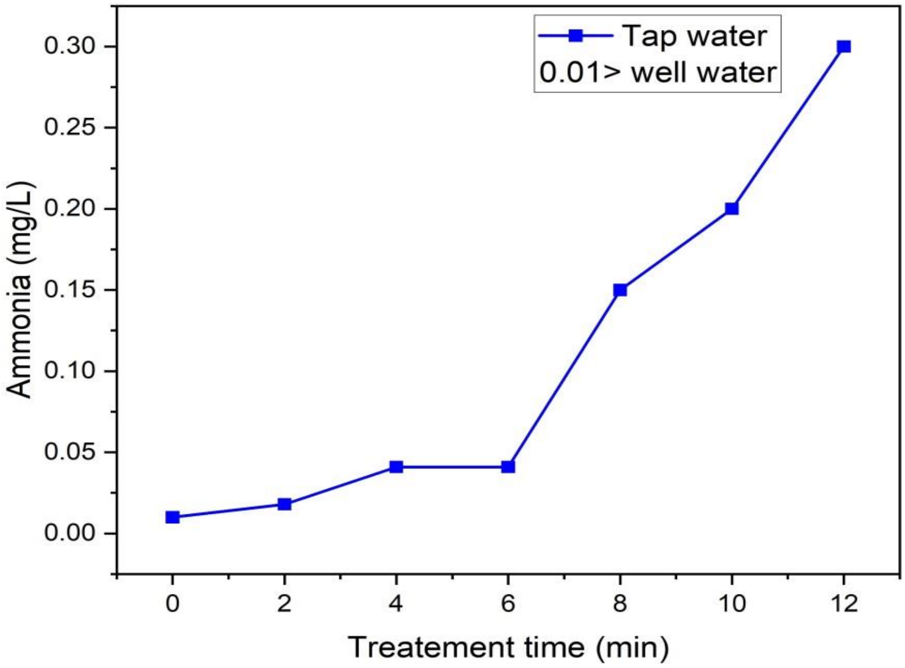

Figure 9. Variation in ammonia concentration with treatment time for tap water.

Ammonia

Figure 9 shows the graph showing role of plasma treatment time with concentration of ammonia. It has been found that very negligible amount (less than 0.01mg/L) of concentration of ammonia is found in well water. On the other hand, tap water contain measurable amount of ammonia which is increases on increasing the treatment time from 0.015 to 0.30 mg/L. From both the findings it can be concluded that cold atmospheric plasma jet plays significant role to increase the concentration of nitrite, nitrate and ammonia. CAPJ treatment involves the application of non-thermal plasma to water, generating reactive species that can potentially lead to the degradation or conversion of contaminants, including ammonia. The treatment may involve processes such as electron impact dissociation, chemical reactions, and the generation of reactive species that can react and slightly change in ammonia ions. The concentration of ammonia is still below the standard value of ammonia specified by WHO (1996).

Chromium

Figure 10 depicts the variation of concentration of chromium in water with plasma treatment time. It shows that cold atmospheric plasma jet has reduced the chromium concentration by 46% in well water and 62% in the case of tape water after 12 minutes of treatment time.

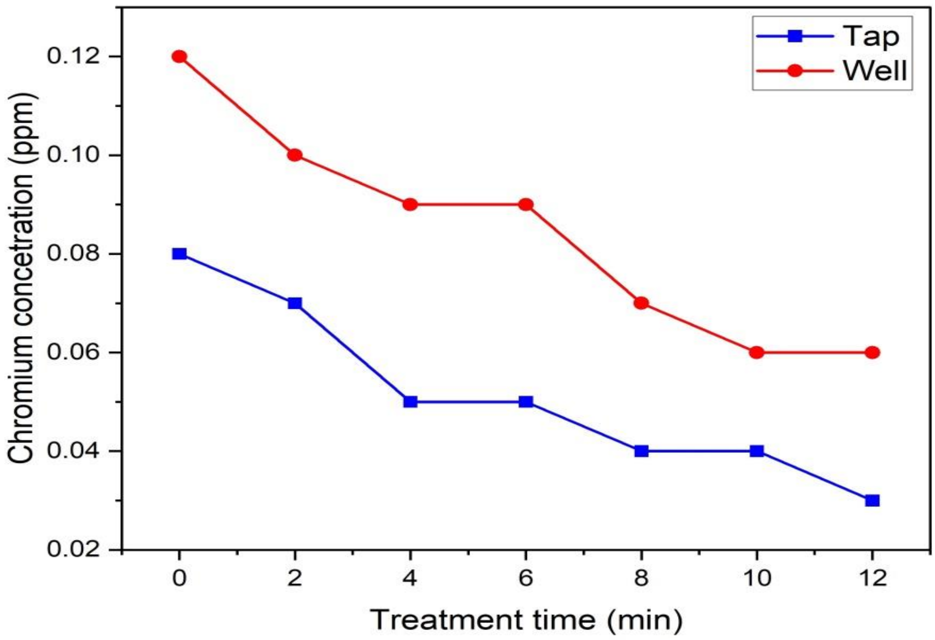

Figure 10. Variation in chromium concentration with treatment time for well and tap water.

Chromium in early days was thought to be non-toxic, so it is widely used for different applications such as alloy manufacture, Chromate production, metal plating and welding. Later on it is found as acute irritant, allergies, also as carcinogen (Dayan & Paine, 2001). Chromium (IV) plays vital role for causing cancers to the animals' cells including human. It is safe to drink water having less than 0.1 mg/lit chromium in any form.

Dissolved Oxygen (DO)

Figure 11 shows the variation of DO in samples of tap and well water over time of treatment. The figure shows that DO level in untreated well water (2.7 mg/L) is less than that of tap water (3.5 mg/L) but after plasma treatment its value increases with treatment time. After 4 minutes of treatment time DO in well water exceeds the value of tap water and finally reach the value of 4.81 mg/L. The level of DO in case of tap water increases slightly with treatment time and reach up to 4.0 mg/L. Plasma treatment can activate oxygen molecules present in the water, resulting in the formation of reactive oxygen species such as ozone and hydrogen peroxide. These ROS are highly reactive and can oxidize organic matter present in the water, breaking it down into simpler compounds. This breakdown process releases additional oxygen into the water, thus increasing the dissolved oxygen concentration (Busco *et al.*, 2018).

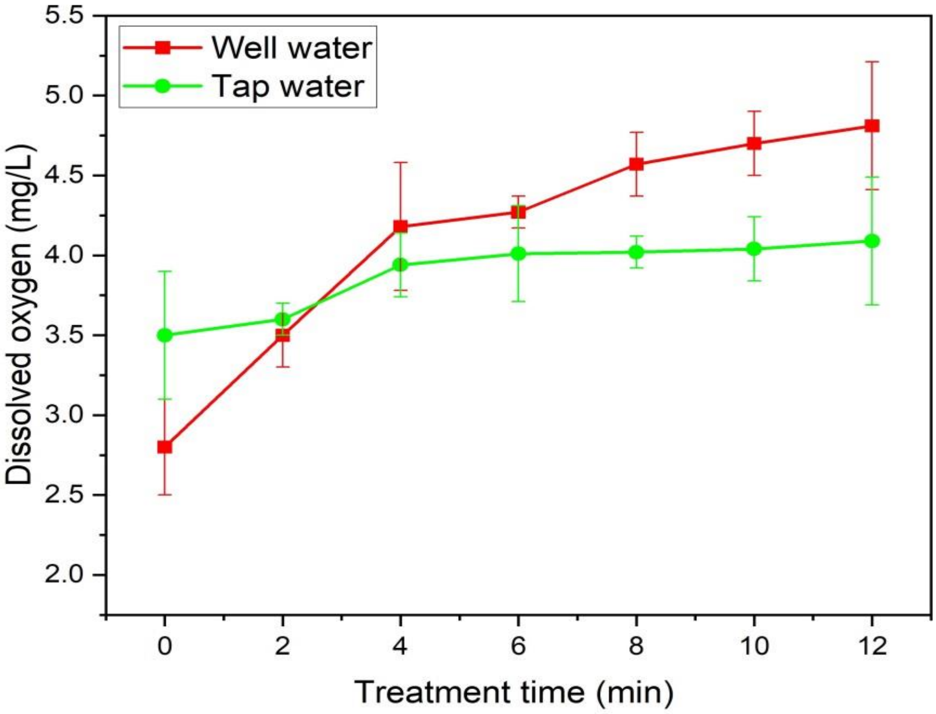

Figure 11. Variation in dissolved oxygen (DO) levels of well and tap water with treatment time

Physical Parameters

Conductivity

Electrical conductivity measures the total ions in a solution and reflects the solution's capacity to transmit electrical current. Figure 12 demonstrates the variation of conductivity of well water and tap water with treatment time. The conductivity of control sample of tap and river water is found to be 770 µS/cm and 301 µS/cm respectively. As the plasma treatment time increases, the conductivity increases in both sources of water. After 12 minutes of treatment time, the conductivity of the well water becomes 925 µS/cm and that of tap water becomes 389 µS/cm.

Plasma can break down molecules in the water, leading to the formation of ions such as H+, OH-, and various other charged species. Furthermore, Plasma treatment can generate radicals such as hydroxyl radicals (OH•) and hydrogen radicals (H•) which can react with water molecules to form ions, thus increasing conductivity. Our result has good agreement with the results presented by Sailaja *et al.* (2015) and Lawaju *et al.* (2018).

pH

pH is the acidity or alkalinity of a fluid by showing the concentration of hydrogen ions in the liquid. A low pH signifies a higher concentration of hydrogen ions, whereas a high pH indicates a lower concentration of hydrogen ions. Figure 13 shows the variation of pH values of different sources with treatment time. Initially, the pH values well water and river water are found to be 8.6 and 8.5 respectively. Both sources of water have high pH values than pure water. But after 12 minutes of treatment, it becomes 7.5 and 7.3 for well water and tap water respectively. Its means basicity of water decreases, and its pH tends to reach drinkable range.

The main reason behind the decreasing in pH value is due to the generation of reactive species like hydroxyl (OH) ions, ozone (O_3) and hydrogen peroxide (H_2O_2) ions (Subedi *et al.*, 2012).

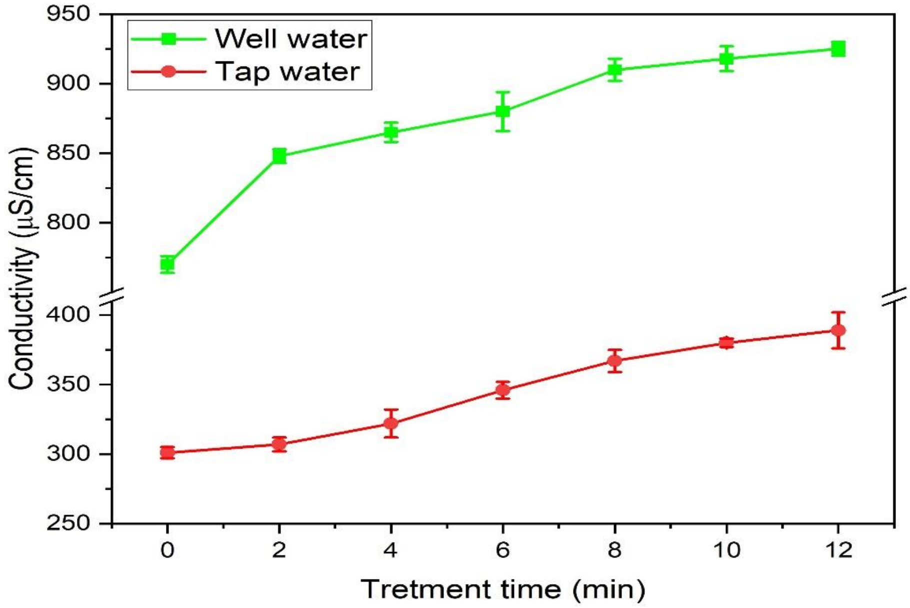

Figure 12. Variation in conductivity of well and tap water samples with treatment time

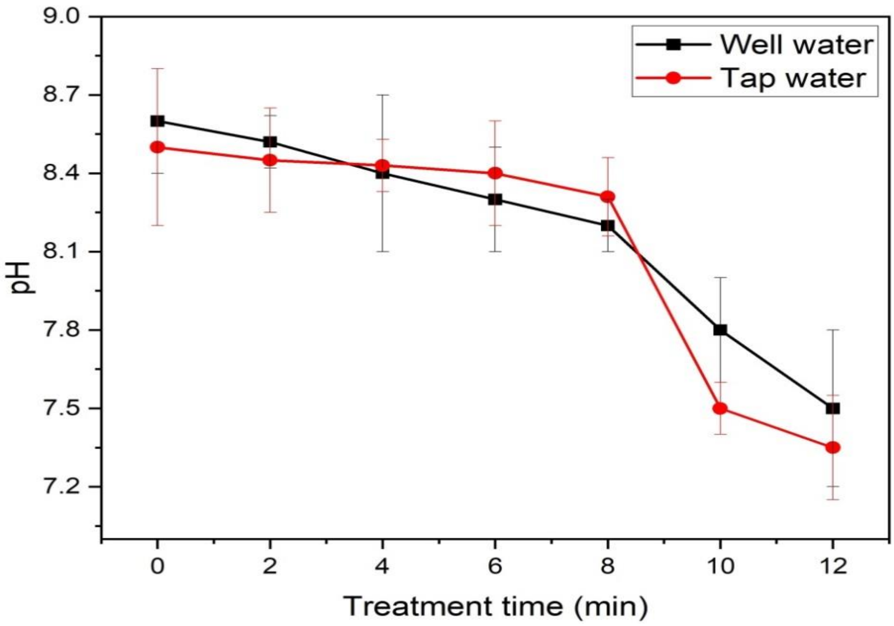

Figure 13. Variation in pH of well water and tap water samples with treatment time

Turbidity

Turbidity is influenced by several factors, including the presence of dissolved and suspended solids, as well as the shape and size of the particles. The experiment study about relation between turbidity and treatment time is shown in figure 14. Initially, the average turbidity of the collected well water and tap water samples was found to be 4.1 NTU and 3.0 NTU, respectively. As the exposure duration rises the values turbidity decreases in both the sources of water. After the 12 minutes of plasma treatment the values of decreases to 2.75 NTU and 1.28 NTU respectively. The decrease in turbidity represents that the cold plasma has capacity to destroy microbes or pathogens and other agents present in water samples. Ozone generated during plasma discharge exerts a direct influence on the microorganisms, leading to the destruction of pathogens in the treated samples. This observation agrees with the findings reported by Dyas *et al.* (2012).

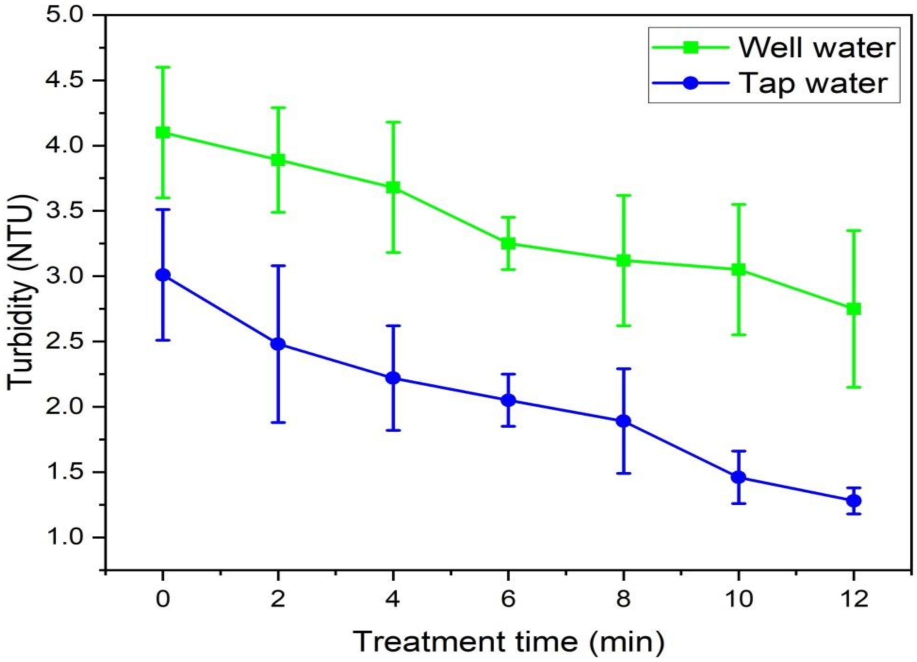

Figure 14. Variation in turbidity of well and tap water samples with treatment time.

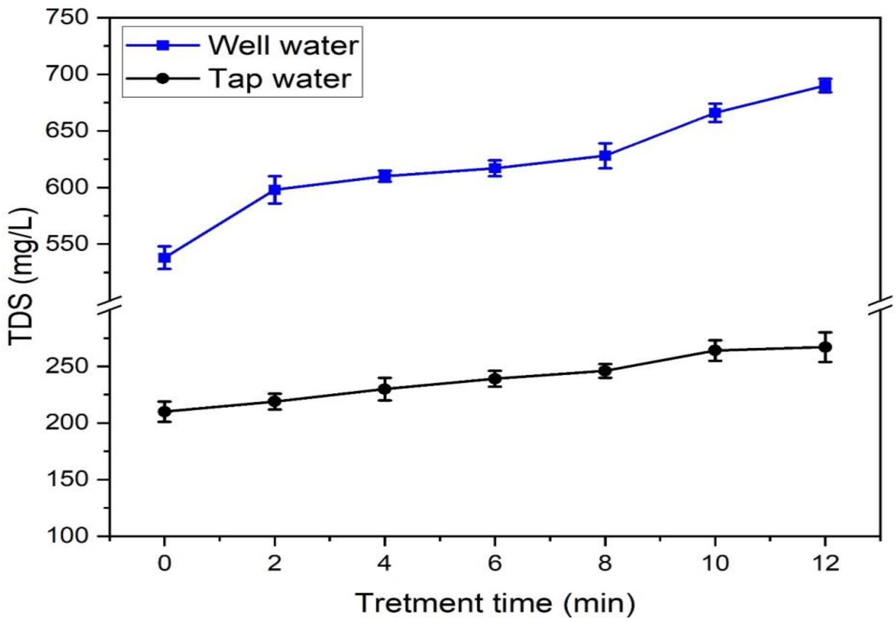

Figure 15. Variation in TDS of well and tap water with treatment time

Total Dissolved Solid (TDS)

The figure 15 depicts the variation of TDS of well and tap water with treatment time. The figure shows that well water contains more total dissolved solid than tap water and there is more increase in TDS values with treatment time in case of well water than tap water. Initially, values of TDS of untreated well water and tap water were 538 mg/L and 210 mg/L respectively. After 12 minutes of treatment time, the values of TDS of well water and tap water found to be 690 mg/L and 267 mg/L respectively. Plasma treatment can introduce new species into the water through chemical reactions with the plasma or with reactive species generated during plasma treatment. These new species can contribute to the total dissolved solids in the water. Furthermore, Plasma exposure can ionize molecules present in the water, resulting in the formation of ions. These ions contribute to the total dissolved solids in the water (Islam *et al.*, 2016).

CONCLUSIONS

A cold atmospheric plasma jet system at atmospheric pressure has been developed, with potential applications in plasma-liquid interactions. The CAPJ temperature was measured at approximately 25°C, confirming its classification as cold plasma. This system has been extensively used to analyze the physical and chemical properties of drinking water before and after plasma treatment. The cold atmospheric plasma jet was characterized by using electrical and optical methods. At an applied voltage of 5 kV, the electron temperature and density were determined to be 0.16 eV and 1.42×10^{14} cm^{-3}, respectively, through Boltzmann's plot and power balance methods. Reactive species generated by the CAPJ significantly influenced the physical and chemical properties of water samples. Notably, the concentration of metal ions in the water increased linearly after treatment, attributed to the role of reactive species, such as ozone, in oxidizing nitrite ions into nitrate. After CAPJ treatment, water samples showed slight increases in electrical conductivity and total dissolved solids (TDS), while pH levels decreased due to the presence of radicals like hydroxyl (OH) and hydrogen peroxide. Turbidity was effectively reduced, suggesting a decrease in suspended particles. Changes in parameters such as pH, turbidity, chromium concentration, and dissolved oxygen highlight the potential of CAPJ for water purification and improve the quality of drinking surface water.

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AUTHOR CONTRIBUTIONS

HBB: resources, writing-original draft, visualization, revision, editing and finalizing the manuscript; SD, AKS, SS, and NK: Conceptualization, investigation, software, data curation; RPG: Methodology, optical characterization, and revision the manuscript.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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