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Plasma agents in bio-decontamination by dc discharges in atmospheric air

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Abstract
Bio-decontamination of water and surfaces contaminated by bacteria (*Salmonella typhimurium*) was investigated in two types of positive dc discharges in atmospheric pressure air, in needle-to-plane geometry: the *streamer corona* and its transition to a novel regime called *transient spark* with short high current pulses of limited energy. Both generate a cold non-equilibrium plasma. Electro-spraying of treated water through a needle electrode was applied for the first time and resulted in fast bio-decontamination. Experiments providing separation of various biocidal plasma agents, along with the emission spectra and coupled with oxidation stress measurements in the cell membranes helped to better understand the mechanisms of microbial inactivation. The indirect exposure of contaminated surfaces to neutral active species was almost as efficient as the direct exposure to the plasma, whereas applying only UV radiation from the plasma had no biocidal effects. Radicals and reactive oxygen species were identified as dominant biocidal agents.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Recently non-equilibrium plasmas, thanks to their reactive nature, have found numerous biological and biomedical applications, in particular when operated at atmospheric pressure without the need for costly vacuum equipment. Atmospheric pressure plasmas applied for sterilization and bio-decontamination are mostly generated by radio-frequency (RF) [1–6] discharges, and various plasma jets and afterglows [1–3, 7–11], usually in rare gases (He or Ar) with/without admixtures of O_2 (or H_2O). The plasma jets are typically blown into ambient air where the rare gas plasma entrains air components. Air plasmas at atmospheric pressure have the additional advantages of not requiring special gases and an easy application in ambient environment. Bio-decontamination by atmospheric air plasmas was tested in dc [7, 12], dielectric barrier (DBD) [9, 13–20], RF [1] and pulsed discharges [8–10, 15]. The plasmas can also be generated directly in water or at the water–air boundary [7, 16, 17, 21, 22], which is of considerable interest for water decontamination. Atmospheric pressure plasmas have been tested on a large variety of prokaryotic microorganisms, such as bacteria, spores, viruses, and some eukaryotic yeasts, fungi and microalgae, resulting in partial disinfection (1–2 log reduction in microbial population) up to complete sterilization. Interaction of plasmas with tissues of higher organisms including humans and plasma applications for skin disinfection, wound healing, blood coagulation, dentistry, surgery, inducing apoptosis pathways for cancer treatment, etc are targeted by a revolutionary novel discipline—plasma medicine [23–27].

In bio-decontamination by non-equilibrium plasmas, it is very important to assess the role of various mechanisms involved. Although there is no general consensus among the authors, the significant mechanisms depend strongly on the plasma composition (gas), temperature, treated microorganisms and the environment in which they are present (air, water, surfaces, etc). In atmospheric pressure plasmas, the major role is played typically by radicals and reactive oxygen

species (ROS, e.g. OH, O, O_3) [1, 3, 6–10, 12, 17–19, 22] and charged particles, particularly O_2^- [14, 27], affecting the cell membranes. UV radiation plays a role in atmospheric pressure plasmas only if photons in the UV C germicide region (220–280 nm) or in vacuum UV are produced [18, 22, 28]. UV C comes typically from the NO γ system, which is formed in air at high plasma powers if both O and N radicals are generated. It is typical in RF and microwave discharges and plasma jets blown into the air, and high power DBDs. UV C and VUV radiation are also partially produced in discharges in Ar [3, 4, 28] or in water [22]. In cold (temperature near ambient) atmospheric air discharges (corona, DBDs, pulsed discharges), NO γ and other sources of UV C or VUV radiation are usually not generated, so radicals and ROS are identified as the dominant bio-inactivation agents [6–9, 11, 16, 17, 29].

Our previous bio-decontamination studies with dc discharges in atmospheric air with water were in agreement with this statement and demonstrated the dominant role of radicals and ROS [30]. NO γ radiation was only detected in our dc atmospheric pressure glow discharge [31], which was found efficient for bio-decontamination but consumed large power [30].

In this paper, the biocidal effects of two plasma sources in atmospheric air with water are investigated—a positive dc *streamer corona* (SC) and a novel regime named *transient spark* (TS). Despite a dc applied voltage, these discharges have a pulsed character with nanosecond repetitive pulses. Pulsed plasmas have been demonstrated as an effective source of active species and they find application in pollution control, plasma assisted combustion and bio-decontamination They are, in particular, convenient for penetration into topographically non-uniform surfaces and cavities, which is applicable, e.g. in teeth root canal disinfection [9, 10, 15, 32].

The effects of SC and TS on selected bacteria in water solutions and on surfaces were tested. Bio-decontamination of water is important from the viewpoint of waste water cleaning or drinking water disinfection and is involved in all biomedical applications and food technology, since cells and most foods contain water. Plasma decontamination of various surfaces is important in medicine, e.g. for the sterilization of endoscopes, implants and other heat sensitive materials. The surfaces can also be potentially used as carriers of bioterrorism agents (e.g. anthrax contaminated letters). The plasmas generated by the studied dc discharges, particularly TS, induce chemical effects that are crucial in bio-decontamination and environmental applications, such as VOC abatement, which have been successfully tested in the past [33].

We focus on the identification of the dominant plasma agents in bio-inactivation by coupling their electrical characteristics, emission spectra and biocidal effects in various regimes. Comparing direct with indirect plasma effects enables separation of various biocidal plasma agents (electric field, charged particles, neutral active species, UV radiation). In addition, measurements of the oxidative stress induced in microbial cells applied for the first time in plasma bio-decontamination enable one to further indicate their respective roles.

2. Experiments

2.1. Experimental set-ups

The experimental set-up for fundamental investigations of dc discharges in point-to-plane geometry, with a high voltage (HV) hollow needle electrode enabling water flow through the discharge zone and a plane or mesh electrode, is depicted in figure 1. The inter-electrode spacing was varied from 5 to 10 mm. A positive dc HV was applied through the ballast resistor R (20 MΩ for SC and ∼5 MΩ for TS). The discharge voltage was measured by a HV probe Tektronix P6015A. The discharge current was measured on a 50 Ω (SC) or a 1 Ω (TS) resistor and by a Rogowski current monitor PEARSON 2877. The current and voltage signals were processed by a digitizing 200 MHz oscilloscope Tektronix TDS 2024. The discharges were photo- and video-documented with digital cameras Olympus E410 or Nikon Coolpix S10.

The emission spectroscopy optical system comprised a dual fibre-optic compact spectrometer Ocean Optics SD2000 with a CCD detector for fast scanning in the UV and VIS–NIR regions (200–500 and 500–1050 nm, resolution 0.6–1.2 nm), filters, fused silica lenses, irises and fibre optics. The discharge set-up was placed in a Faraday cage together with the optical components mounted on translation stages, which enabled lateral and vertical scanning capabilities.

The bio-decontamination effects of the dc discharges were tested on flowing water and on direct/indirect treatment of the contaminated solid medium.

The discharge set-up shown in figure 1 enabled the contaminated water to flow directly through the HV hollow needle electrode, and so through the corona active region in its proximity. The effect of electrostatic spraying (electrohydrodynamic atomization, EHDA) occurred when the HV was applied on the needle electrode [34].

We also used point-to-plane electrode geometry for the comparison of direct and indirect plasma effects on contaminated solid agar surfaces. A needle electrode was placed about 1 cm above the agar surface at the centre of the Petri dish and the discharge was applied for 1 or 2 min. In direct treatment, the agar was grounded with a wire. The indirect plasma effects on the contaminated agar were tested as follows:

(1) by placing the grounded mesh electrode ∼2 mm above the agar, this shielded the electric field and trapped the charged particles, but allowed the neutral particles and partial UV light to reach the agar surface; this concept is similar to [14];
(2) by placing the 3 mm thick quartz window on the agar surface and a grounded ring electrode on its top, this allows only the light emitted from the discharge to reach the agar, including UV. We also tested MgF_2 window transmitting vacuum UV; this concept is similar to [3].

Figure 2 schematically depicts these arrangements. The agar in the direct treatment (figure 2(*a*)) represents a certain electrical resistance between the discharge and the ground; its typical value was 1–2 kΩ depending on the water content. In indirect set-ups (figures 2(*b*) and (*c*)), a small resistor r was inserted

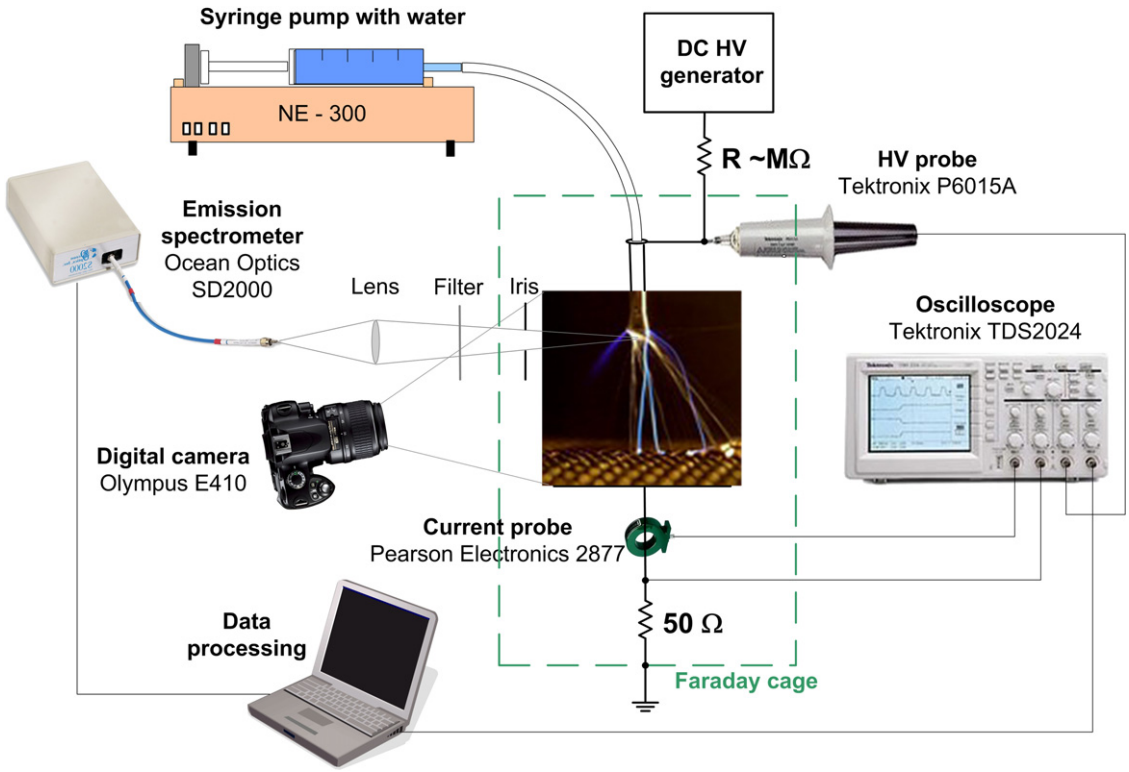

Figure 1. Experimental set-up for dc discharges, with a HV hollow needle electrode enabling water flow through the discharge zone and a plane or mesh electrode.

between the mesh (or ring) and the ground to simulate the agar's resistance. Its exact value was set empirically from case to case to make discharge pulses of about the same amplitude and shape as in the direct treatment on agar. This ensured the same discharge properties in all three set-ups.

2.2. Treated microorganisms, microbial handling and cultivation procedure

The bio-decontamination effects of investigated dc discharges were tested on selected Gram-negative bacteria *Salmonella typhimurium* (*Salmonella enterica*, serovar Typhimurium, strains TA 98 and 100) in water with initial populations from 10^3 to 10^7 colony forming units per ml (CFU ml^{-1}), or directly spread on the solid nutrient medium (agar, Roth Ltd) on a Petri dish, about 10^6 per dish. *S. typhimurium* is a pathogen causing salmonellas diseases; its inactivation is important from the viewpoint of drinking water and food disinfection.

The microbial cultivation was carried out in a sterile environment in the following steps: an overnight bacterial culture was first prepared in a shaker with sterile liquid nutrient. A hot sterile nutrient medium—agar—was poured into sterile Petri dishes, on which the bacteria were grown, and solidified. Cultivated bacteria in the liquid nutrient were compared with McFarland turbidity scale to assess their initial population per millilitre. They were then diluted in water to obtain the desired concentrations. The plasma experiments were performed with both discharges, at various parameters and treatment times and repeated 5–8 times. Usually, 50 μl of both plasma treated and reference (control) samples were spread on Petri dishes with agar. Three to four Petri dishes from each sample were taken for statistical evaluation. These were incubated for 12–24 h in a thermostat at 37 °C. The grown CFUs on the treated and the reference samples were counted and evaluated.

2.3. Measurements of the oxidative stress

The interaction of ROS with the bacterial cell membranes results in the peroxidation of membrane lipids. The final product of lipoperoxidation is malondialdehyde (MDA), quantifiable by spectrophotometry after the reaction with thiobarbituric acid (TBA) at 90–100 °C [35]. This method of *thiobarbituric acid reactive substances* (TBARS) was applied for the first time to measure the oxidative stress induced in bacteria in distilled water exposed to SC and TS. We assigned the TBARS concentrations from the absorbance of MDA at 532 by using Lambert–Beer's law with an absorption coefficient of 1.57×10^5 mol^{-1} l cm^{-1} [35].

3. Results and discussion

3.1. Applied dc discharges

Two types of dc discharges of both polarities operating in atmospheric air with water were investigated: a well-known SC, and a novel TS. These discharges generate non-equilibrium plasmas inducing various chemical and biological effects important in bio-decontamination. Their electrical parameters and emission spectra were documented in detail in our previous works [31, 36]. The typical voltage and current waveforms of SC and TS discharges in a 10 mm gap with an electro-spray

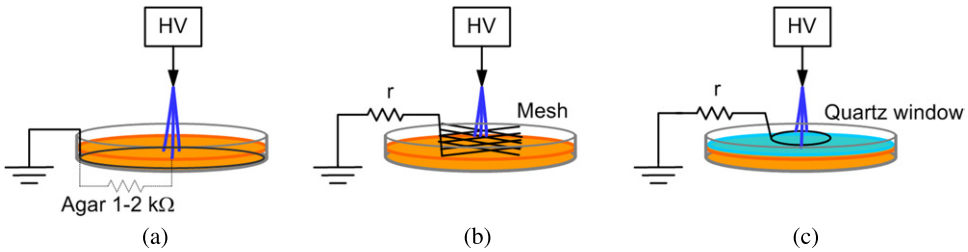

Figure 2. Schematics of electrode arrangements for (*a*) direct and (*b*), (*c*) indirect plasma treatment of contaminated agar plates. (*b*) Mesh electrode ∼2 mm above agar surface trapped the charged particles and shielded the electric field. (*c*) Quartz (or MgF_2) window transmitted only light from the discharge (including UV).

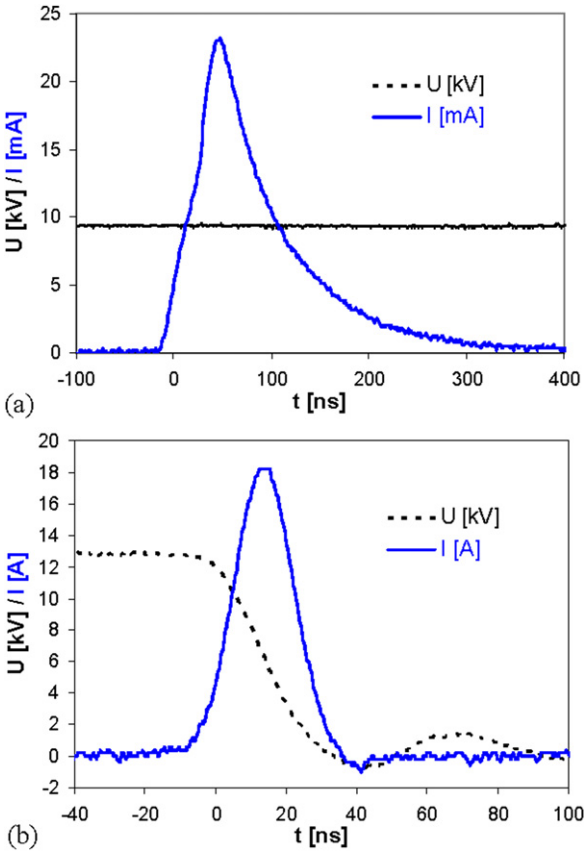

Figure 3. Typical voltage and current waveforms of (*a*) SC and (*b*) TS discharges with electro-spray of water, 10 mm gap.

of water are shown in figure 3. When a HV of a few kilovolts is applied to the point electrode, SC appears, typically with small current pulses of streamers (∼10 mA) with a repetitive frequency of 10–30 kHz, during which the discharge voltage remains fairly constant and generates a cold plasma (∼300 K).

As the voltage is further increased (to ∼12 kV in the 10 mm gap), the streamers establish a conductive channel that gradually heats up, thus enhancing the reduced electric field E/N, which eventually leads to a spark breakdown with excessive current pulse [37].

In our case, the spark pulse current is limited by (1) the ballast resistor R that drops the voltage as the current increases and (2) the capacity C between the electrodes that is small (order of 10 pF). C is a sum of the internal capacity of the discharge gap (C_{int} ∼ 1 pF) and the capacities of the HV cable (C_{cable} ≈ 5–20 pF depending on the length) and the voltage probe (C_{probe} = 3 pF). Thus, when the spark forms, it is only transient since the discharged energy is small (0.1–1 mJ). After the pulse, C is recharged by a growing potential on the stressed electrode and triggers a new pulse. This TS then becomes a repetitive streamer-to-spark transition discharge, with each spark pulse (∼1 A) preceded by one or a sequence of streamer pulses. The repetitive frequency of pulses is 0.5–10 kHz, and increases with the applied voltage. Thanks to the very short pulse duration (∼10–100 ns) given by the small C and a limiting R, the plasma cannot reach equilibrium conditions and remains at a relatively low gas temperature, depending on frequency, i.e. dissipated power (∼500–1500 K).

3.2. Optical emission spectroscopy (OES)

OES in the UV–VIS region is a powerful technique of plasma diagnostics, because it gives valuable information on excited atomic and molecular states, enables one to determine the rotational and vibrational temperatures and thus the level of non-equilibrium in the plasma, and gives insight into ongoing plasma chemistry [31, 38]. We employed OES together with the comparison of the biocidal effects of the dc discharges to better understand the dominant mechanisms involved in bio-decontamination. Both SC and TS generate cold, non-equilibrium plasmas (300–550 K) in the discharge channel. OES characteristics of the applied discharges described in detail in [31] showed that electrons with the highest energies were present in TS. These electrons initiate dissociations, ionizations and excitations of various species. Atomic O, N and H radicals, and N_2^+ ions were detected only in TS, and there were a number of OH radicals. A part of O radicals reacts with air O_2 and forms ozone, O_3. There was no UV C radiation detected from SC and TS.

3.3. Flowing water treatment through the stressed electrode

The contaminated water flowed directly through the HV hollow needle electrode, and so through the plasma active zone in its proximity, which substantially improved the volume efficiency compared with our previous set-ups for water treatment [30]. This idea was first introduced for polluted gas treatment through HV needle electrodes [39], and we applied it for the first time for water treatment. The effect of electrostatic spraying (EHDA) occurred when the HV was applied on the needle electrode [34]. The electrostatic spraying of

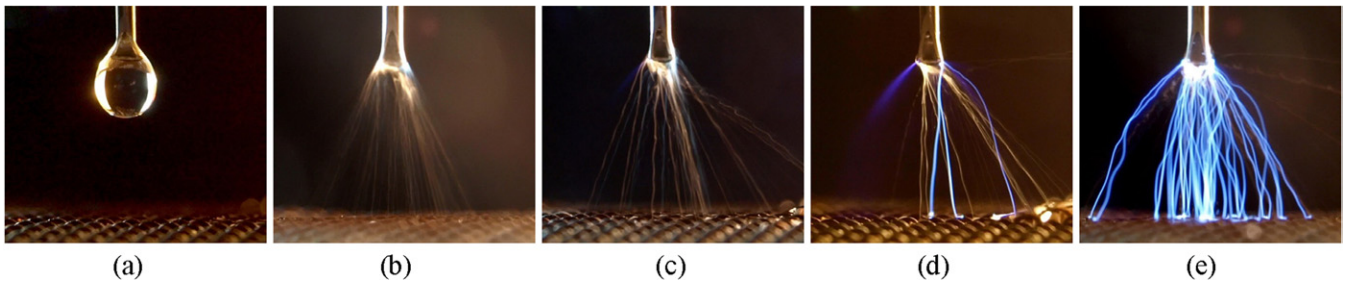

Figure 4. Photographs of the EHDA of water in 8 mm gap, water flow rate 0.5 ml min^{-1}: (*a*) droplet without a HV, (*b*) EHDA spray with a HV applied, 5.5 kV, (*c*) EHDA together with SC, 6.5 kV, (*d*) transition SC–TS, 7.8 kV, (*e*) spray with TS, 9 kV.

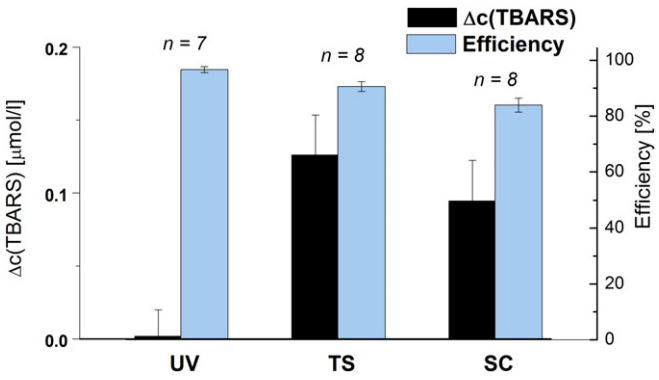

Figure 5. TBARS concentration gains and decontamination efficiencies of *S. typhimurium* in water treated by SC and TS with electro-spray, compared with 1 min exposure to UV C (shown with standard error of the mean, *n*—number of repeated experiments).

contaminated water through the corona active zone contributed to the improved efficiency of bio-decontamination. Figure 4 shows the photographs of the EHDA of water when a HV is applied, EHDA with SC, a transition SC–TS perturbing the EHDA and then TS discharge with water sprayed.

The temperature of the treated water did not change in SC and was increased by a maximum of 10 K in TS. The lethal heat effect of the discharges on bacteria cannot be considered at all.

The electro-spray water treatment resulted in very short D values (0.1–0.4 s in both SC and TS), and low energy costs (0.7–20 J ml^{-1} per log reduction) in SC. Further improvements of the electro-spray and EHDA–discharge interaction will expectantly enhance the inactivation at lower energy costs.

3.4. Oxidative stress induced in bacteria

Figure 5 shows the TBARS concentration gain Δc(TBARS) correlated with the bio-decontamination efficiency of SC and TS applied to the electro-sprayed water. The same bacterial samples were irradiated by biocidal UV C radiation (Hg lamp, 254 nm, 1 min) for comparison.

UV C radiation induced almost no Δc(TBARS) despite its efficiency being very high. Obviously, UV dominant biocidal mechanism is not peroxidation of cell membranes, albeit radiation induction of ROS in cell nuclei is possible [27]. In contrast, SC and TS plasma treatments significantly enhanced the TBARS concentration. This indicates that oxidations of cell membranes by ROS are important in microbial inactivation. More ROS is linked with the higher efficiency.

3.5. Direct versus indirect plasma treatment

A comparison of direct and indirect plasma effects on contaminated agar surfaces was aimed at the separation of various biocidal plasma agents. We compared direct SC and TS plasma treatment with two types of indirect exposures obtained by

(1) filtering the charged particles and electric field from neutral radicals and excited species; and
(2) applying only UV radiation from the plasma.

The corresponding schematics of the electrode arrangements are shown in figure 2. Figure 6 shows the photographs of the Petri dishes with contaminated agar after direct and both indirect treatments together with the untreated control sample, and a sample treated with a 50 μl drop of liquid ethanol (96%) for comparison.

The effects of plasma (and ethanol) on contaminated agar are clearly visible as dark voids, whereas the control sample is homogeneously covered by cultivated bacteria (bright). Dark voids with bright spots represent incomplete decontamination (e.g. with ethanol); the spots are CFUs grown from single bacteria. With respect to the total number of bacteria spread on one Petri dish (10^6), a few tens of surviving bacteria that cause these bright CFUs in the voids are quite negligible.

Both direct plasma and indirect exposures to neutral reactive species (and partial UV) caused apparent bio-decontamination (voids). SC resulted in a larger treated area on the Petri dish, likely due to the electric wind that drives the active species from the point electrode towards and along the agar surface in one preferential direction. TS treatment is more localized at the centre of the dish but more intense. The heat effect of TS on contaminated agar is possible in the very small area under the discharge. TS usually resulted in a tiny hole (1–2 mm diameter) of dried agar. Nevertheless, the area of decontamination (void) was always much larger than this tiny hole at its centre. The void maintained the ambient temperature after treatment, so we can neglect the heat effect on bacteria. In both indirect treatments, the TS heat was led out through the mesh (ring) electrode which was not in direct contact with the agar surface: the heat effect can be excluded.

Interestingly, there was very little difference between the direct and indirect plasma treatments with both discharges,

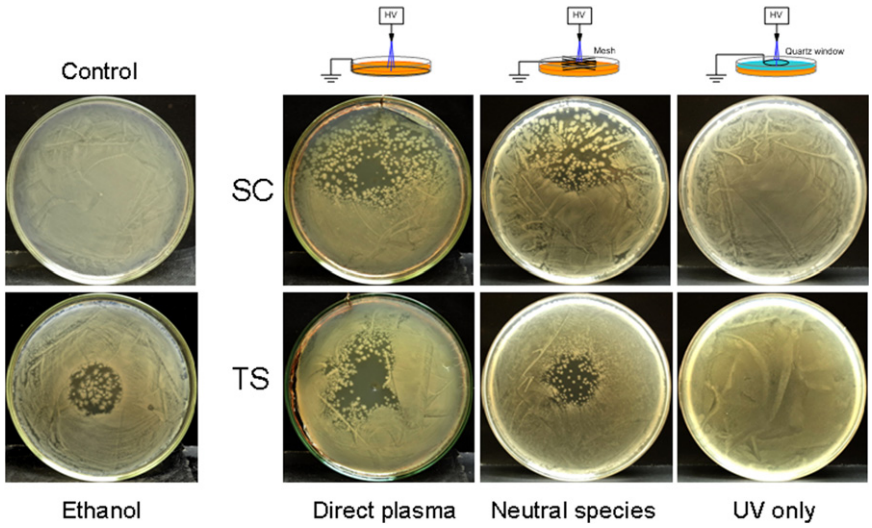

Figure 6. Photographs of Petri dishes with contaminated agar; left side: untreated control and treated by ethanol, right side: treated by SC (upper row) and TS (lower row) direct; and neutral species only; and UV light only.

which indicates that neutral reactive species are crucial even in the direct exposure.

Exposure to the UV light alone, transmitted by quartz or MgF_2 windows, demonstrated no visible decontamination; the dishes were almost identical to the control. This is correlated with the fact that the emission spectra lack any UV C or VUV radiation in SC and TS.

The similarity of the effects of direct plasma and indirect treatments by reactive neutrals agrees with the emission spectra, oxidation stress measurements and our previous findings [30]. Apparently, radicals and active species (O, N, H, OH, O_3, O_2^-), in particular ROS, represent the dominant biocidal mechanism in atmospheric air SC and TS discharges.

4. Conclusions

Bio-decontamination of water and surfaces contaminated by bacteria (*S. typhimurium*) was tested in two types of positive dc discharges in atmospheric pressure air in point-to-plane geometry with one electrode submerged in water or in a new arrangement with water sprayed through the plasma zone. The bacteria were handled and their population evaluated by standard microbiology thermostatic growth cultivation procedures.

The SC with small current pulses (∼10 mA) and 10–30 kHz repetitive frequency generates a cold (300–350 K) non-equilibrium plasma. With increasing applied voltage, the streamers transit to the novel regime TS with short (<100 ns) current pulses (∼1–10 A) of 0.5–10 kHz repetitive frequency and very limited energy. Thanks to the very short spark pulse duration given by the small internal capacity of the discharge system and the limiting series resistor, the TS plasma remains relatively cold (∼500 K). The emission spectra of the discharges and the measured temperatures indicate that both SC and TS generate non-equilibrium plasmas with various excited species (N_2^*), and molecular (OH) and atomic (O, N, H) radicals.

Both SC and TS were found to be very efficient when the treated water was sprayed directly through the HV needle electrode and thus through the active discharge zone. Considerable efficiencies at short treatment times resulted in very short D values and relatively low energy costs. The EHDA effect occurring with the corona discharge in this regime applied for the first time enhanced the efficiency of the process.

The comparisons of direct and two types of indirect exposures of contaminated agar plates to the plasma of SC and TS enabled the separation of the various biocidal agents. It was demonstrated that the direct exposure and indirect exposure to only active neutral species generated in the plasma had almost the same effect on bacteria. On the other hand, separated plasma radiation, including UV, had no significant effect. These investigations, together with the emission spectra of the discharges, indicated the major role of radicals and ROS (O, OH, O_3). Their role in the plasma treatment was confirmed by the absorption spectroscopic detection of the products of microbial cell membrane oxidation in the TBARS method.

In summary, we demonstrated that cold atmospheric air dc discharges can be efficiently used for bio-decontamination of water and surfaces. The dominant biocidal plasma agents are radicals and ROS, which agrees with the findings published by many other authors.

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