Supporting Information

Generation of Reactive Oxygen Species in Water Droplets Levitated in Air

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Experimental section

Generation of microdroplets

The experiments were performed with a single-axis acoustic levitator (Microdroplet Tech Co., Ltd, Tianjin, China) that composes of an emitter and a reflector, working at a frequency of 38.5 kHz. The volume of each levitated droplet is about 2 μL.

All these experiments were carried out under atmospheric pressure. The dynamic process of levitating droplet was recorded by a high-speed camera (PHANTOM T2410, NY, USA) equipped with a 120x optical lens with a frame rate of 30,000 frames per second.

Characterization of levitated droplet

2 μL of deionized water (Milli-Q® IQ 7000 Ultrapure Water System, USA) was levitated under the action of an acoustic levitator, a 50 μm-diameter gold wire was used as a conductive electrode, and an electrostatic meter (6514, keithley, USA) was used to record in real time the charging of the droplets during the suspension process.

For the fluorescence measurement, DI water was spiked with 10 μM of an oxidation-sensitive fluorescent probe (10-acetyl-3,7-dihydroxyphenoxazine). A 530 nm laser was used as the excitation light and a monocular microscope was used to record the resulting fluorescence. The fluorescence was quantified by the Image J software.

A 10 μM aqueous solution of DMPO/TEMPO was suspended for 15 min at a controlled relative humidity of 65% and a temperature of 25 °C under dark conditions. Electron paramagnetic resonance spectroscopy was used to characterize the associated radicals generated in the levitated droplets. EPR spectra were acquired under identical instrumental settings (e.g., gain, microwave power, modulation amplitude) for all samples to allow for direct comparison of signal intensities. The mean signal intensity for each radical adduct was determined by measuring the peak-to-peak amplitude of the characteristic doublet (for DMPO-•OOH/O_2^-) or quartet (for DMPO-•OH) from three replicate samples. The treated aqueous solution was analyzed by mass spectrometry using the nano-electrospray (nESI) ionization.

D_2O and $H_2^{18}O$ were used to characterize the source of free radical material, high-resolution mass spectrometry (E240, Thermo-Fisher, Waltham, MA) was used to analyze the signals of isotopies of the products with DMPO, and an LTQ-XL mass spectrometer

(Thermo-Fisher, Waltham, MA) was used to analyze the signals of the OH--$(H_3O)^+$ radical in the low *m/z* region.

Acoustic field simulation

The commercial finite element software COMSOL Multiphysics 6.1 was utilized to calculate the acoustic pressure. In the calculations, the simulation domain is determined based on the geometry of the levitator. The entire simulation domain was divided using tetrahedral meshing. Rigid boundary conditions were applied at the emitting and reflecting surfaces, and the sidewalls were set as cylindrical wave radiation.

Levitated droplet in an electric field

The levitated droplet was placed in an electrostatic field with a field strength of 30 kV/m, and the levitated droplet was observed with a high-speed video camera (with a 120x macro lens) at 10,000 FPS. For fluorescence measurements, 10 μM of the oxidation-sensitive fluorescent probe (10-acetyl-3,7-dihydroxyphenazine) was added to DI water. A 530 nm laser was used as the excitation light and fluorescence was recorded using a monocular microscope. Fluorescence was quantified by Image J software to compare the effect of the electrostatic field on ROS within the droplet. In addition, PTO color rendering and electron paramagnetic resonance spectroscopy have been used to directly detect the effect of electrostatic fields on ROS of levitated droplets.

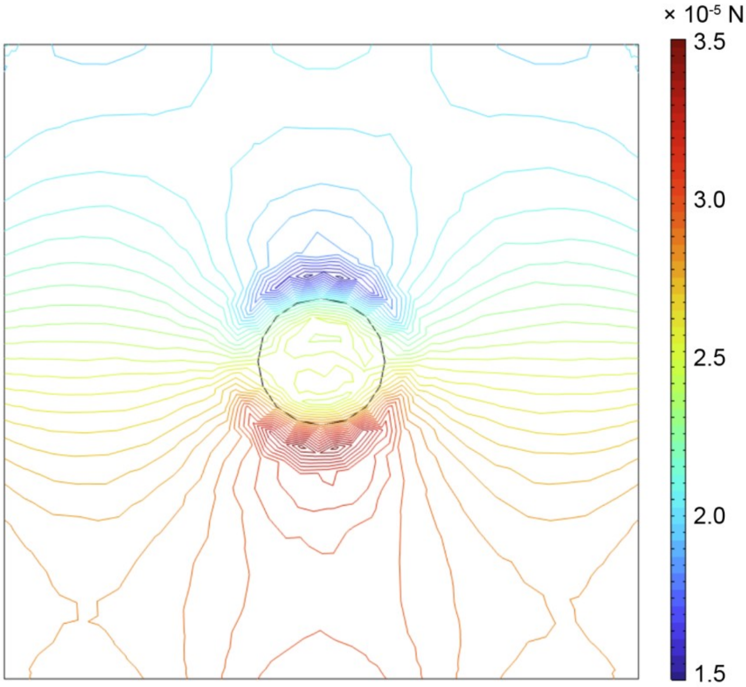

Figure S1. Finite element simulation results of acoustic forces on a levitated droplet.

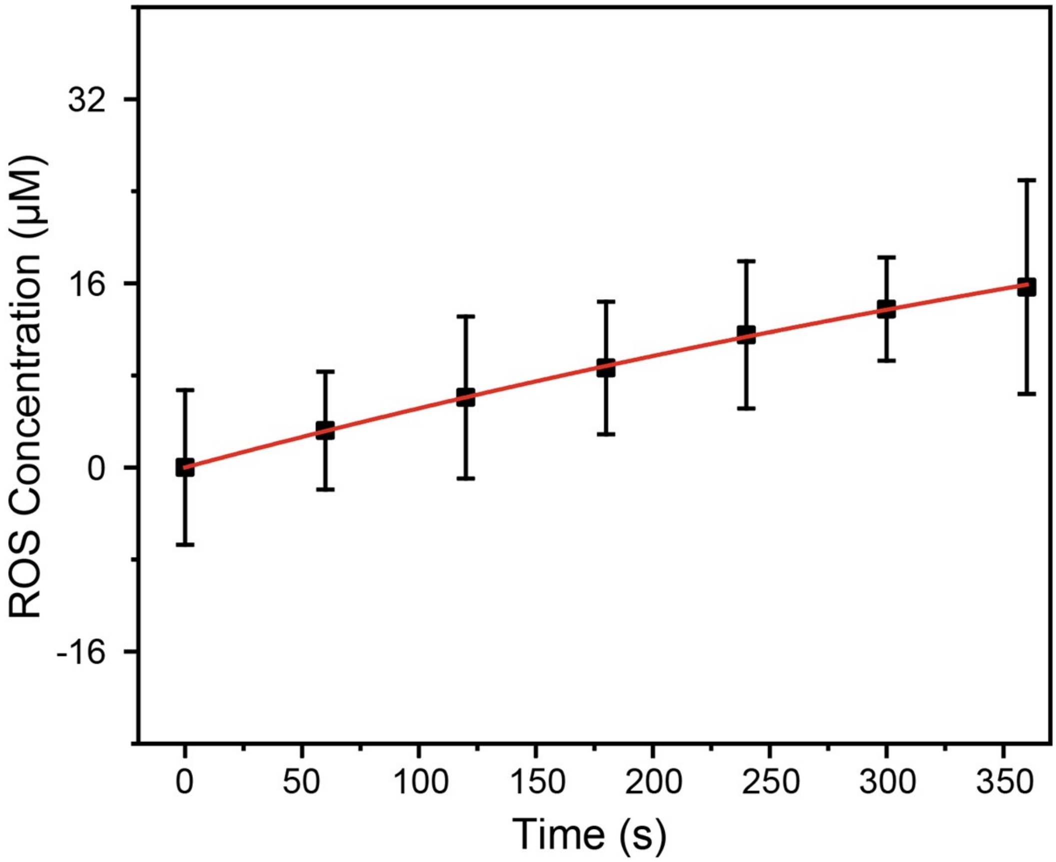

Figure S2. The relationship between ROS concentration a suspension time

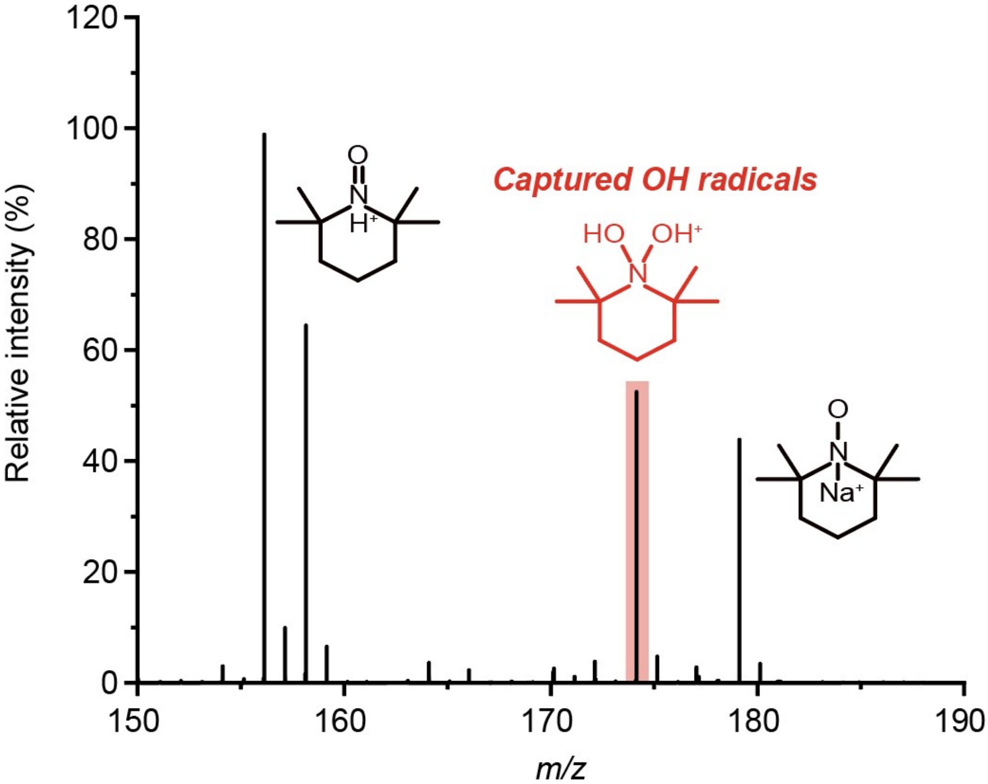

Figure S3. Hydroxyl radicals were confirmed by recording the mass spectrum of the levitated droplets.

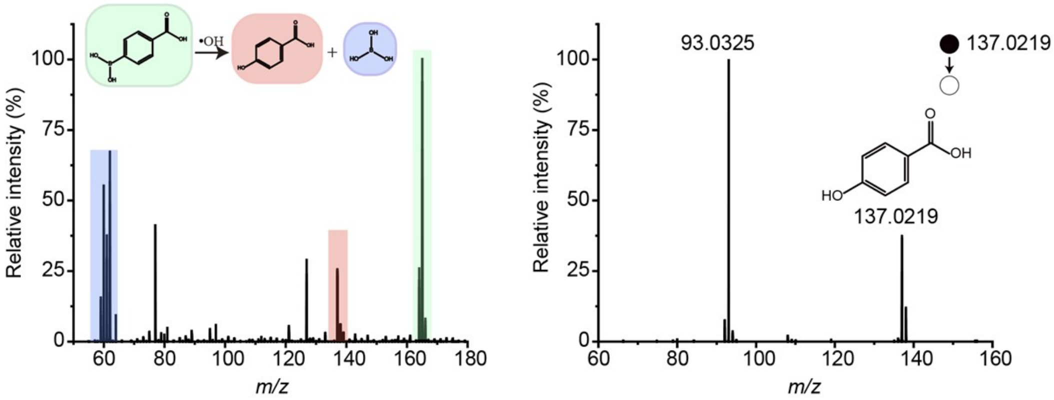

Figure S4. Mass spectrometric results of the reaction of levitated droplets with 4-carboxyphenylboronic acid.

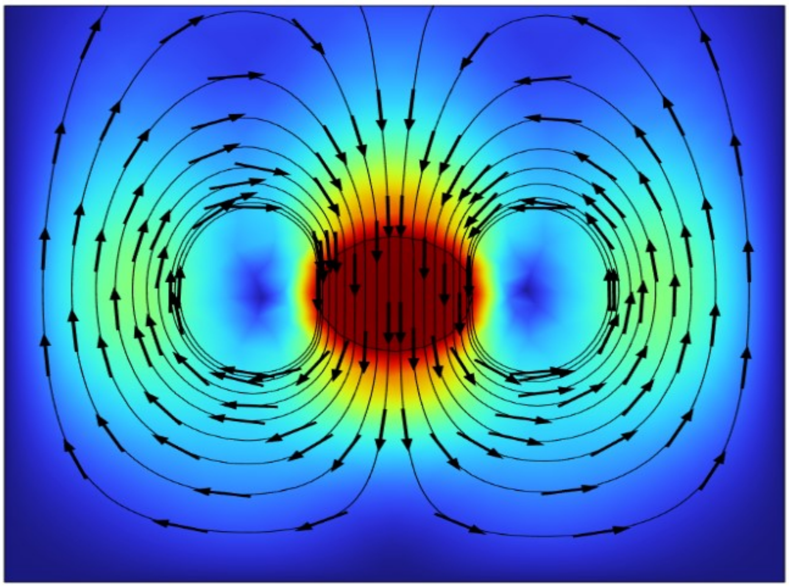

Figure S5. Finite element simulation results of direct interaction between airflow and a levitated droplet in the presence of an acoustic field.

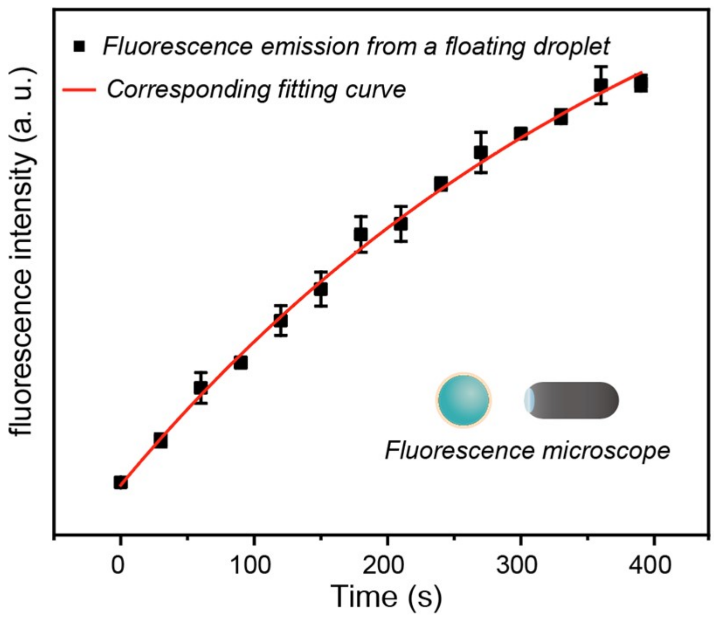

Figure S6. Dependence of the fluorescence intensity on the suspension time.

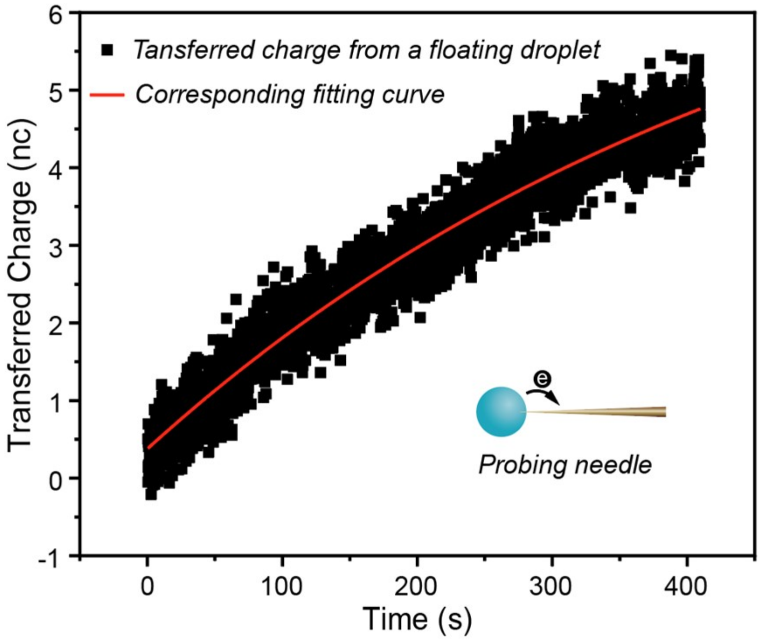

Figure S7. The transferred charge when a probing needle is inserted into the droplet.

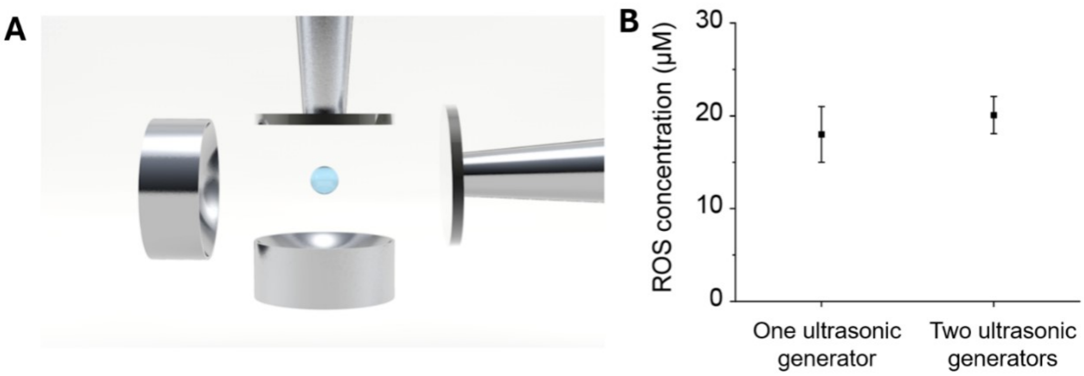

Figure S8. (A) Two ultrasonic generators, one parallel to the direction of gravity, the other perpendicular. (B) ROS concentrations generated from droplets in the two different experimental setups

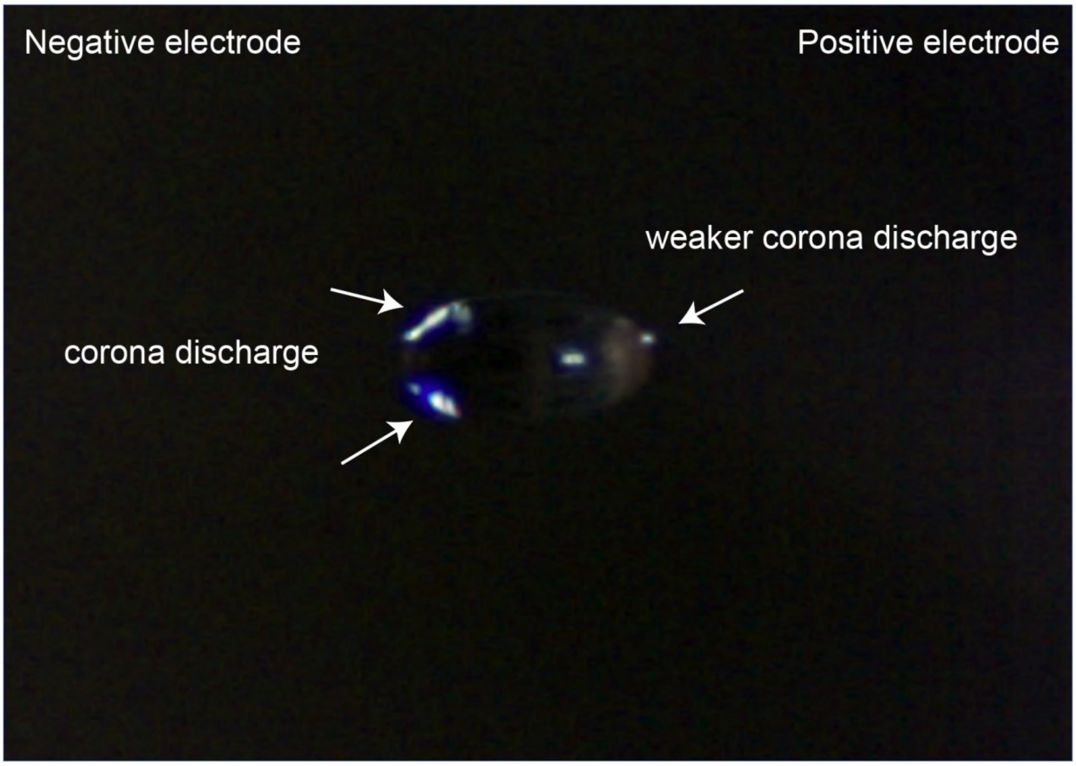

Figure S9. A weaker corona discharge on the side of the droplet near the positive electrode.

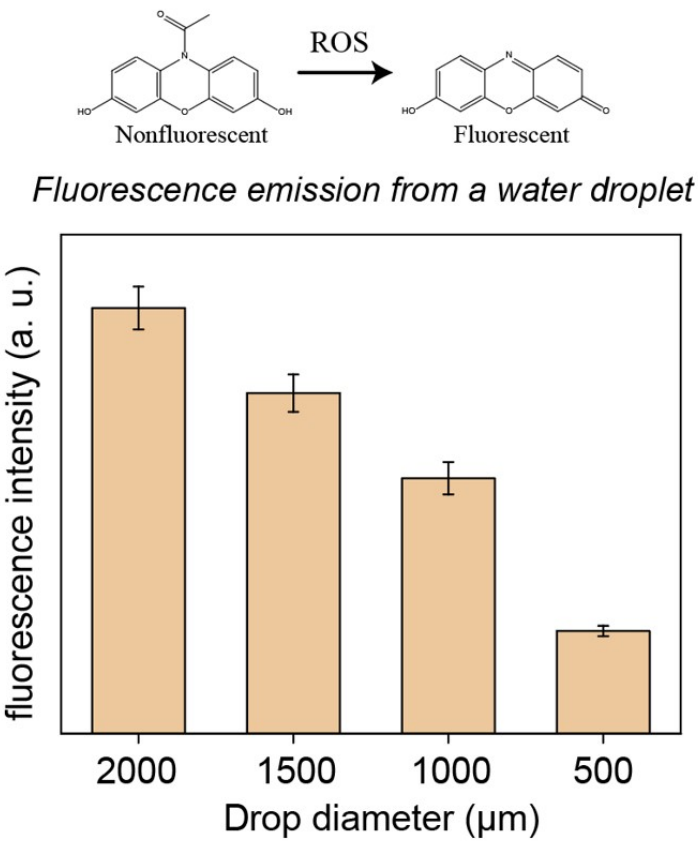

Figure S10. ROS generation in droplets with different diameters.

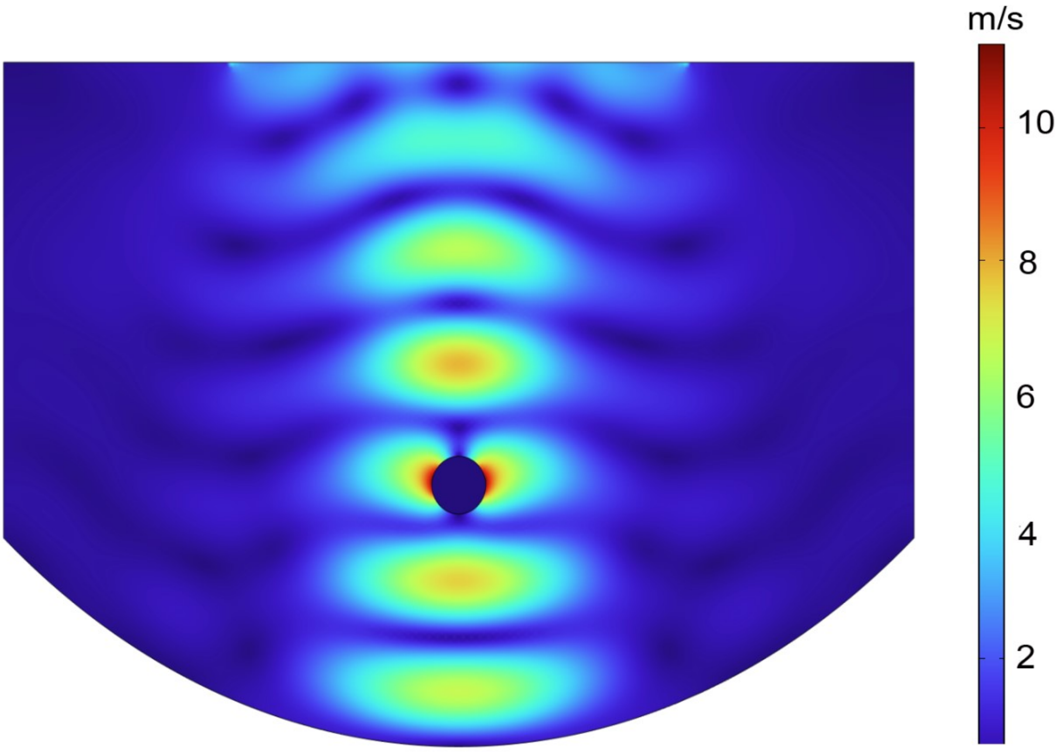

Figure S11. Finite element simulation results of air flow rate surrounding a levitated droplet.