Identifying the Major Reactive Oxygen-Nitrogen Species in a Pulsed Streamer Discharge

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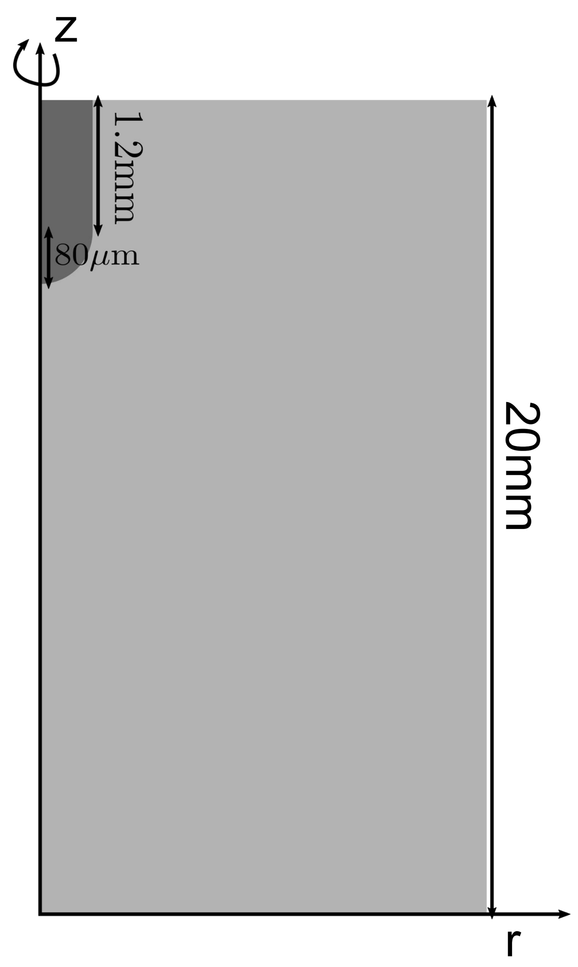
Motivation

Plasma-activated water (PAW) generated by electrical discharges in air contains reactive oxygen-nitrogen species (RONS). Depending on the type of chemical species, plasma-activated water can for example be used for disinfection or to increase the shelf-life of agricultural products. We want to understand how to efficiently produce various RONS by controlling the input conditions of the discharge. Various operating conditions such as voltage amplitude, voltage rise time, pulse shape, repetition frequency, and gas composition influence the yield of different RONS.

In this work, we investigate the effect of varying the water percentage in the gas composition on the streamer discharge properties, and RONS yield for a single pulse+afterglow regime.

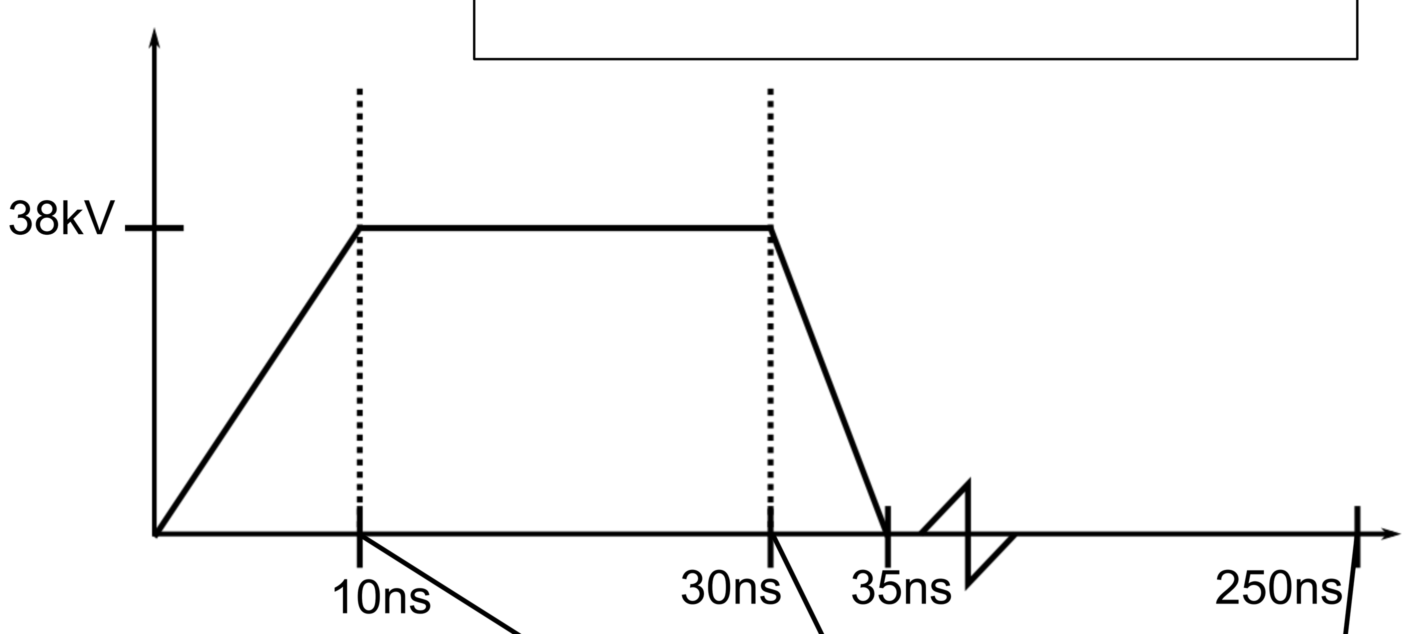
Simulation set-up



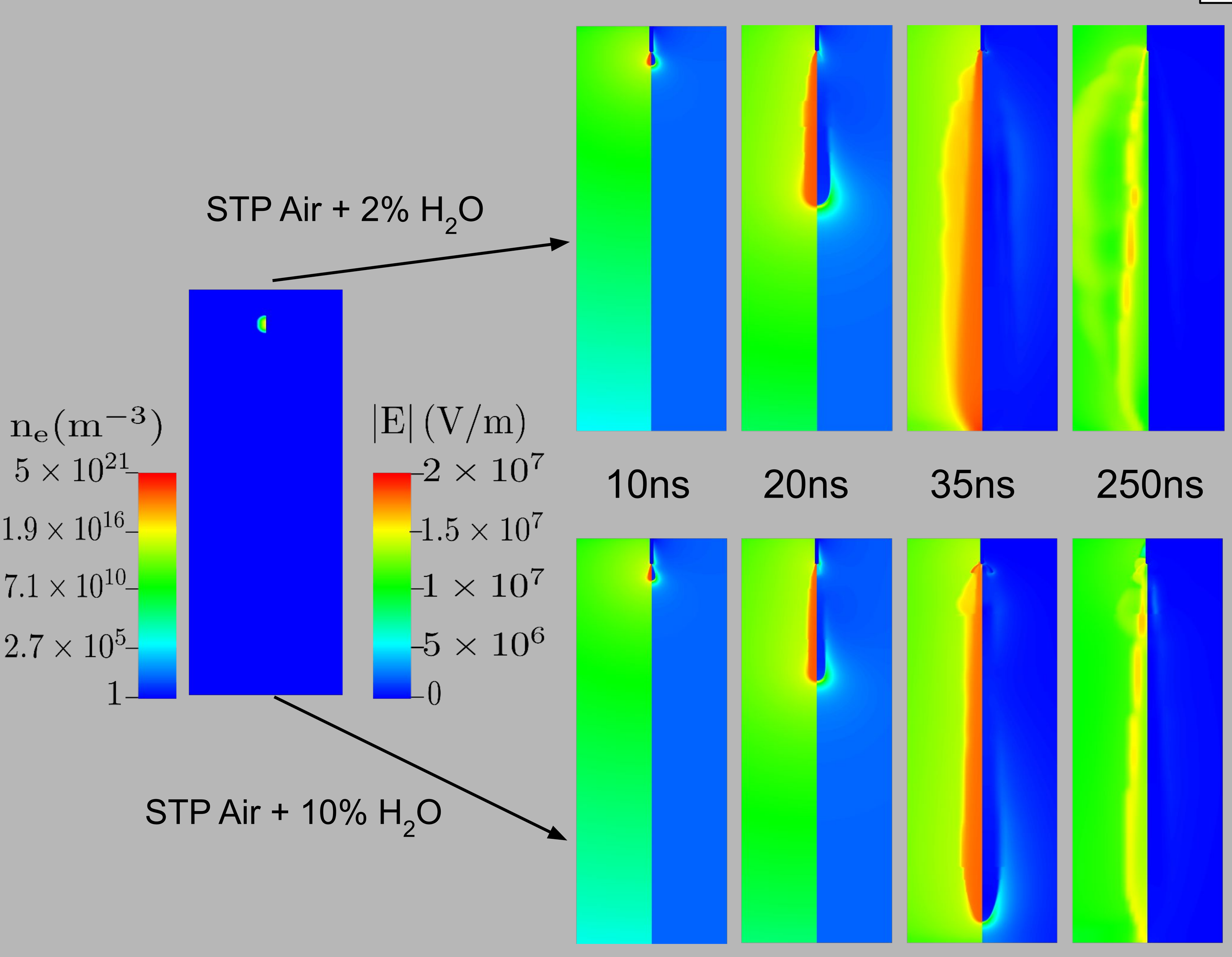
- Drift-diffusion equations, with local field approximation were solved using an in-house code.
- A total of 50 species, and 131 reactions were used.
- Gas: STP Air + varying H_2O %

Neutral	N_2, O_2, H_2O, N, O, N_2O, O_3, H2, H, OH, H_2O_2, HO_2, NO, NO_3, NO_2, N_2O_4, N_2O_5, HNO_2, HNO_3, ONOOH
Positive	N_2^+, H_2O^+, $N_2^+(B^2\Sigma)$, $N_2O_2^+$, N_4^+, O_2^+, O_4^+, OH^+, O^+, H_2^+, H^+, $O_2^+\cdot(H_2O)$, H_3O^+, $H_3O^+\cdot(H_2O)$, $H_3O^+\cdot(H_2O)_2$, $H_3O^+\cdot(H_2O)_3$
Negative	e^-, O_2^-, O_3^-, O^-, OH^-, H^-, $O_2^-\cdot(H_2O)$, $O_2^-\cdot(H_2O)_3$,
Excited	$O_2(a^1\Delta_g)$, $N(^2D)$, $O(^1D)$, $N_2(B^3\Pi_g)$, $N_2(C^3\Pi_u)$, $O(^3P)$.

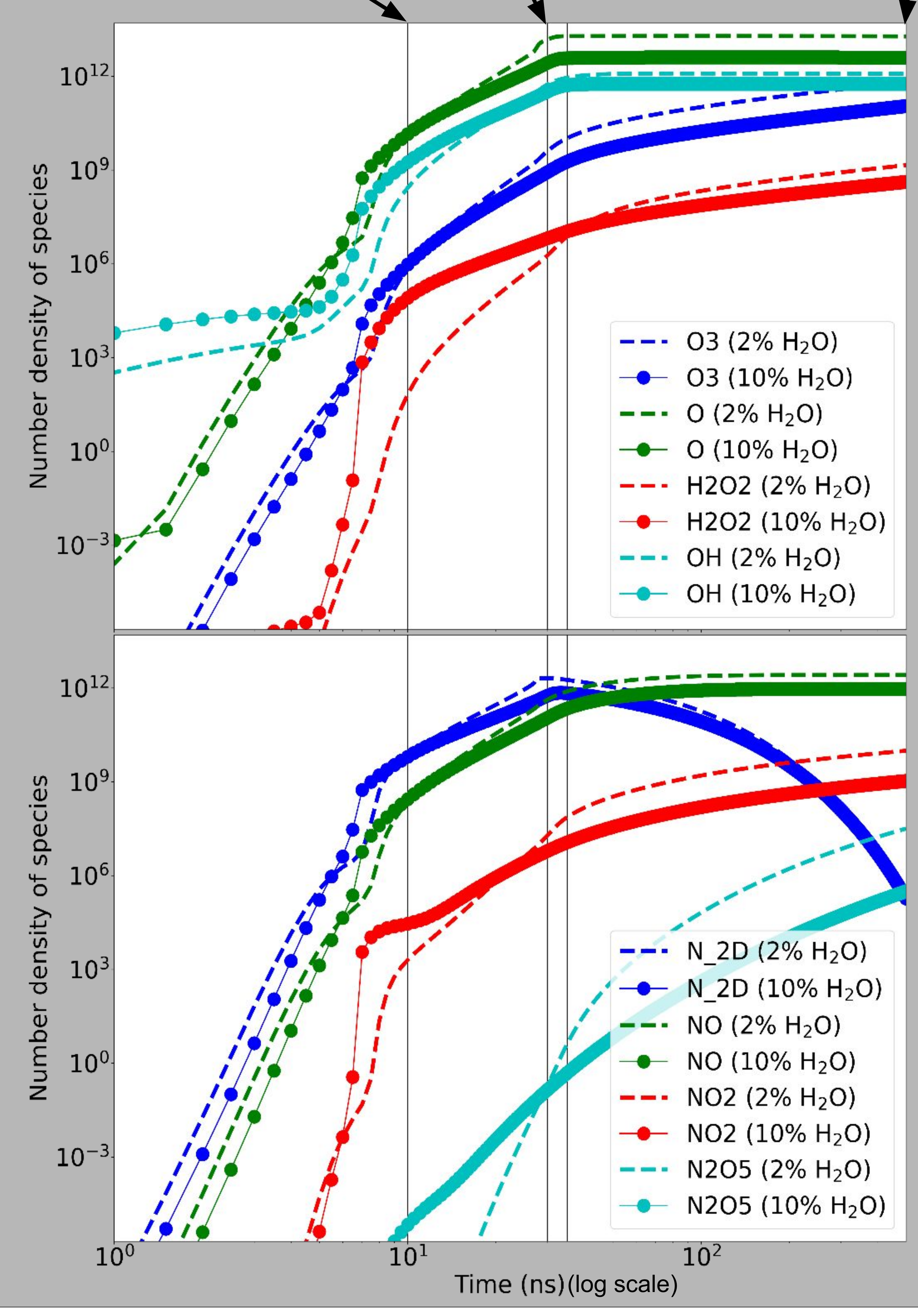
Scan this to access our simulation code



Results

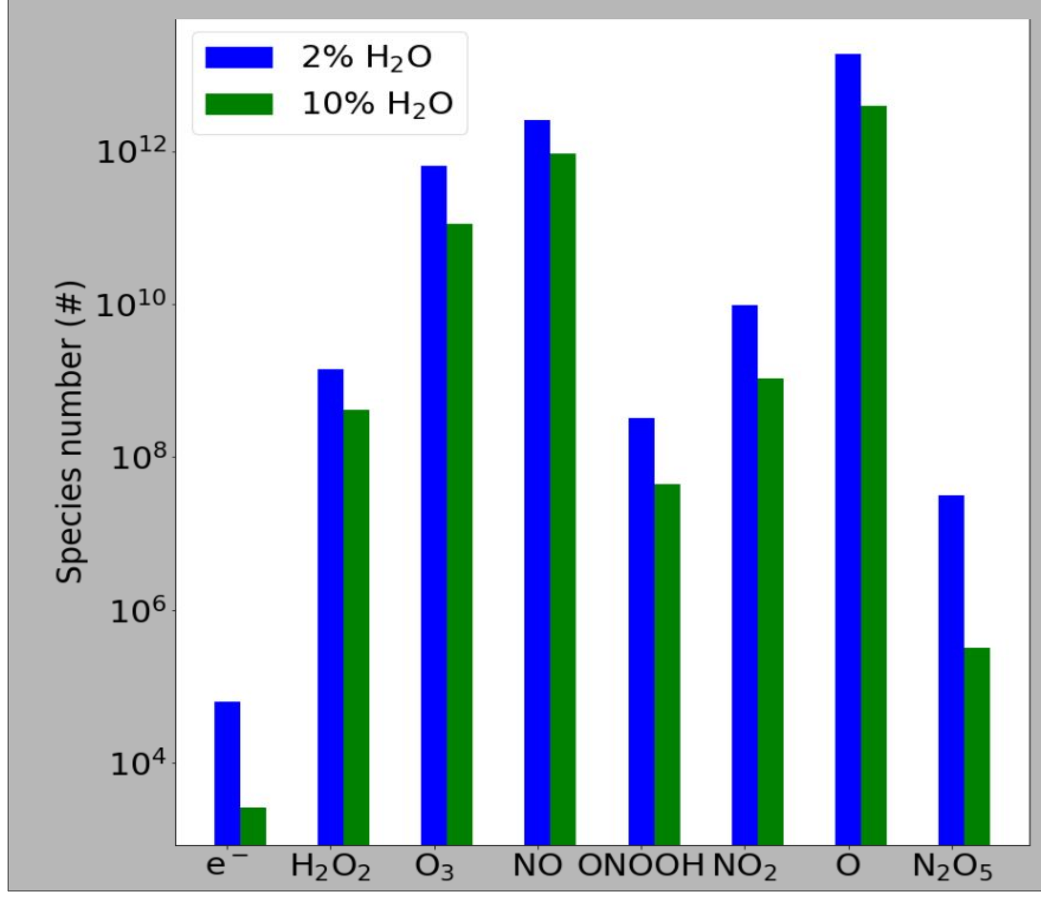


Increasing the water percentage gave rise to a slightly slower, and thinner streamer



- Most of the RONS are produced when the pulse is on.
- By the end of 250ns, more RONS are produced for 2% H_2O than 10 % H_2O.

Important species at the end of the (pulse + afterglow) phase

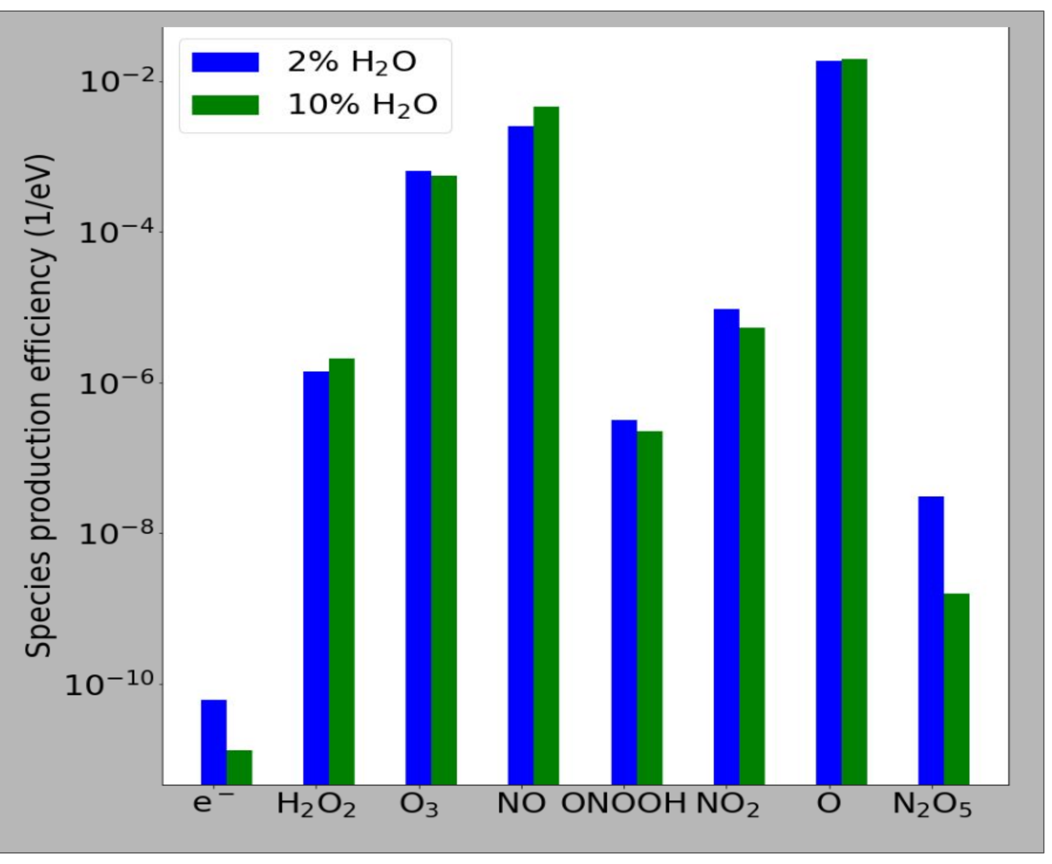


- Higher electron density in 2% H_2O due to the reduced effect of water clustering reactions.

Species	Decrease ratio
e	0.04
H_2O_2	0.29
O_3	0.17
NO	0.36
ONOOH	0.14
NO_2	0.11
O	0.21
N_2O_5	0.01

Species production efficiency = Species number / j.E (in eV)

- In 10% H_2O, RONS are produced less efficiently, except for H_2O_2 and NO.



Acknowledgements

This work was supported by the Dutch Research Organisation NWO under the project "Plasma for Plants".

Conclusions and Further Questions

- Increasing humidity from 2% to 10%:
  - Minor change in discharge properties.
  - Chemistry: decreased overall electron density, small percentage decrease in RONS

How will the species trend change during subsequent pulses?

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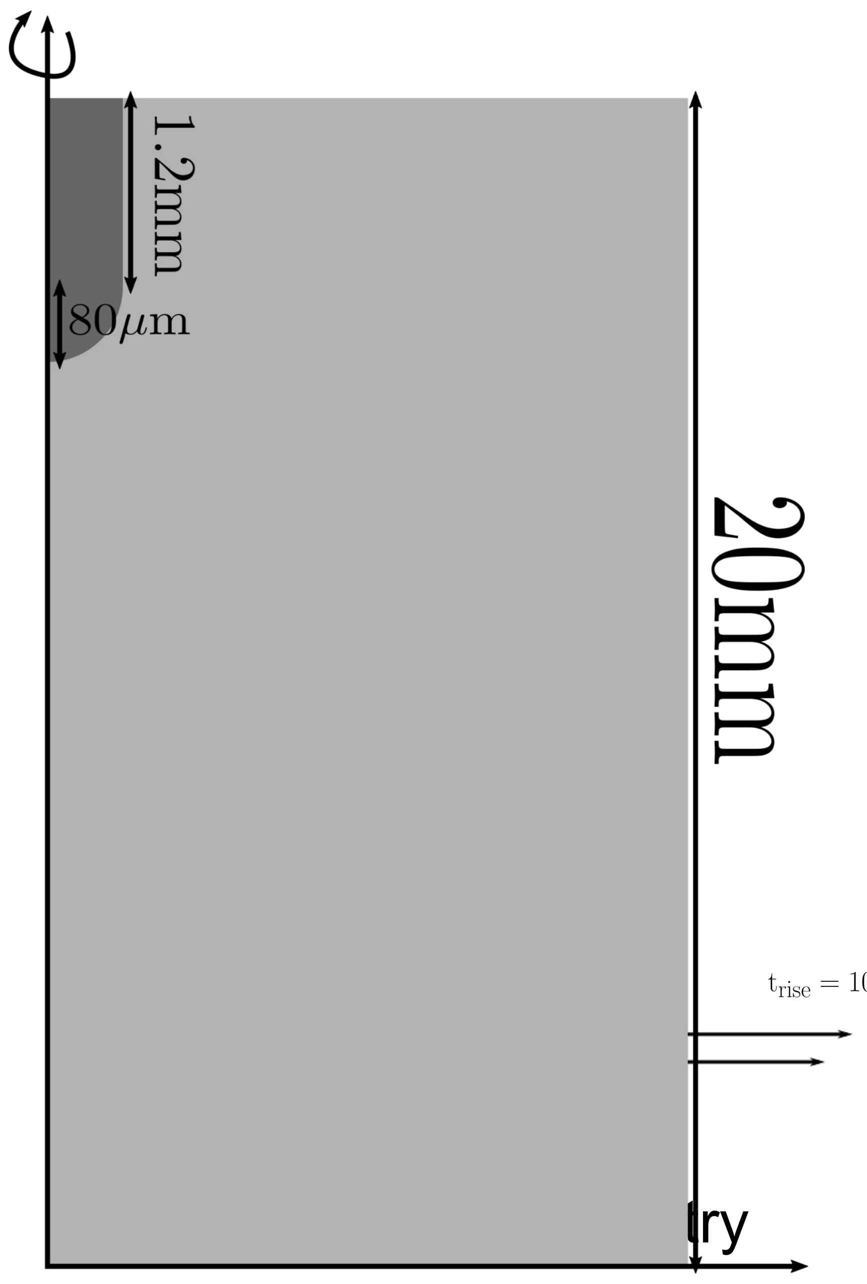
Motivation

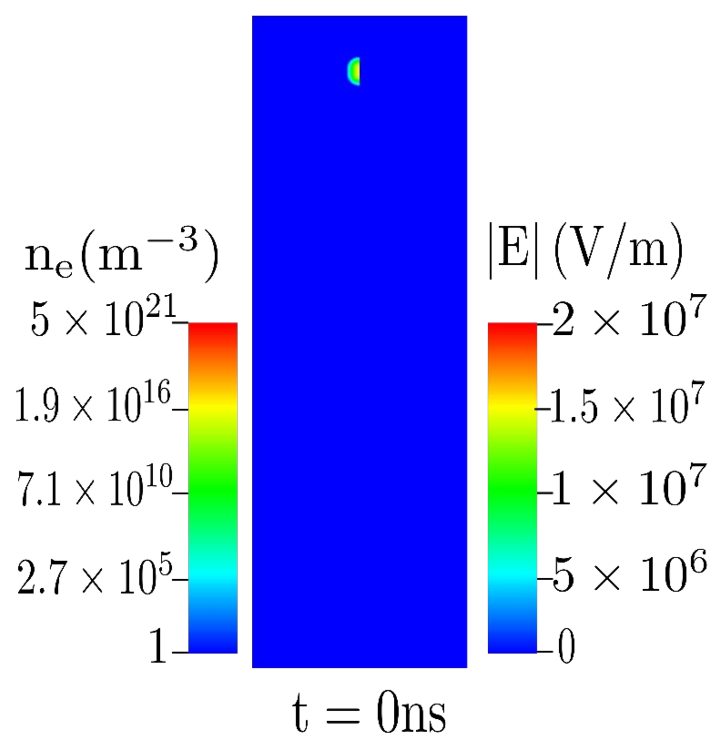
Plasma-activated water (PAW) generated by electrical discharges in air contains reactive oxygen-nitrogen species (RONS) [1]. Depending on the type of chemical species, plasma-activated water can for example be used for disinfection or to increase the shelf-life of agricultural products. It is not fully understood how various RONS are produced/consumed on a nano-second timescale. Various operating conditions such as voltage amplitude, voltage rise time, pulse shape, repetition frequency, and gas composition influence the yield of different RONS.

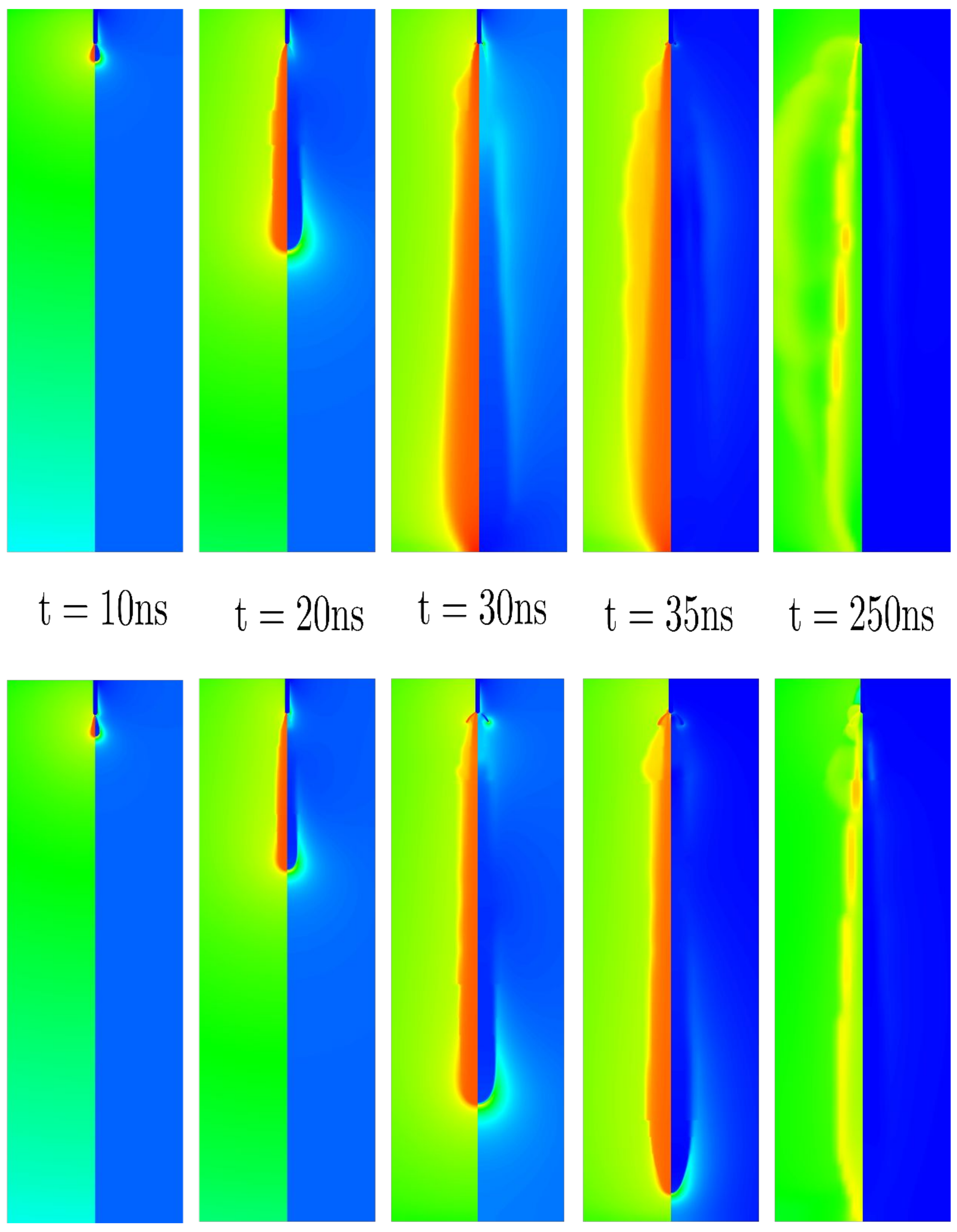
In this work, we investigate the effect of varying the water percentage in the gas composition on the streamer discharge properties, and RONS yield for a single pulse+afterglow regime.

Simulation Model

- Drift-diffusion equations are solved using afivo-streamer [2].
- Gas: $N_2 + O_2 + H_2O$, at 300K, 1bar.



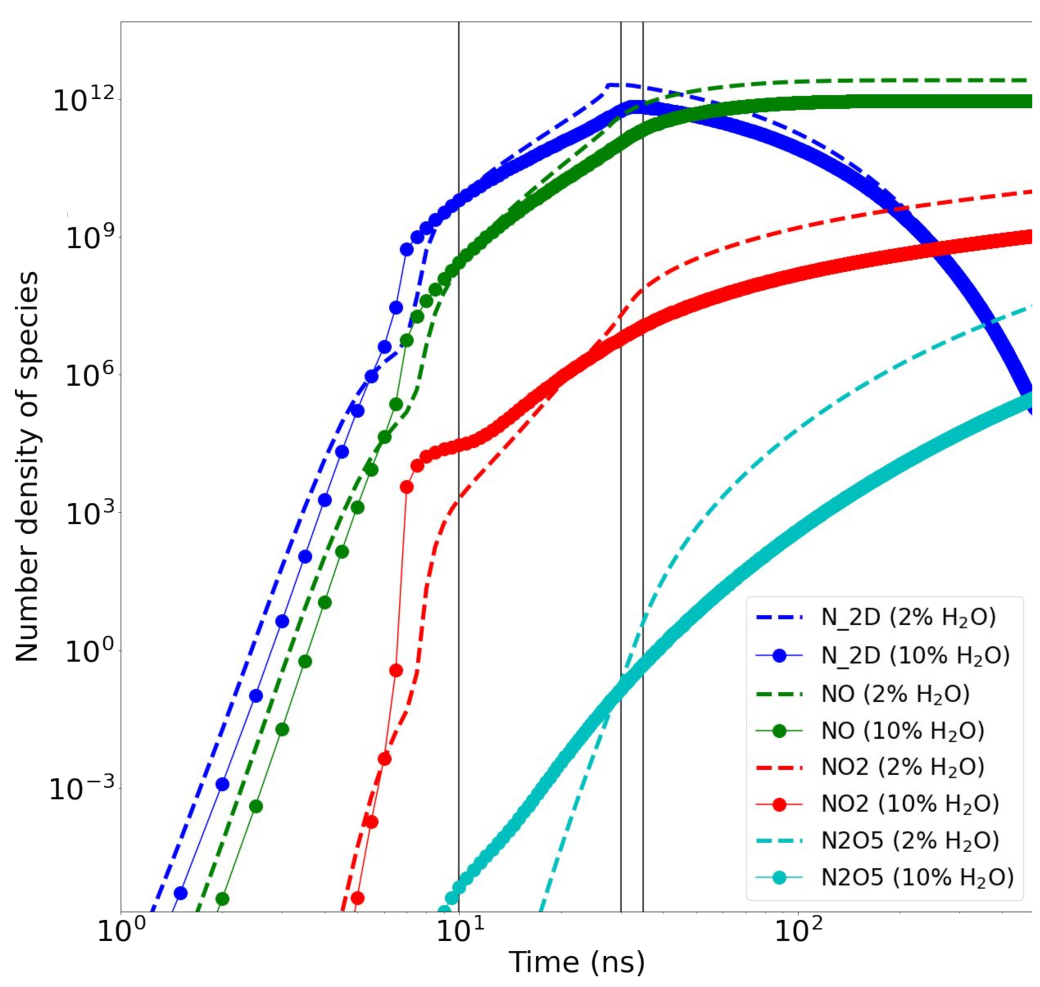


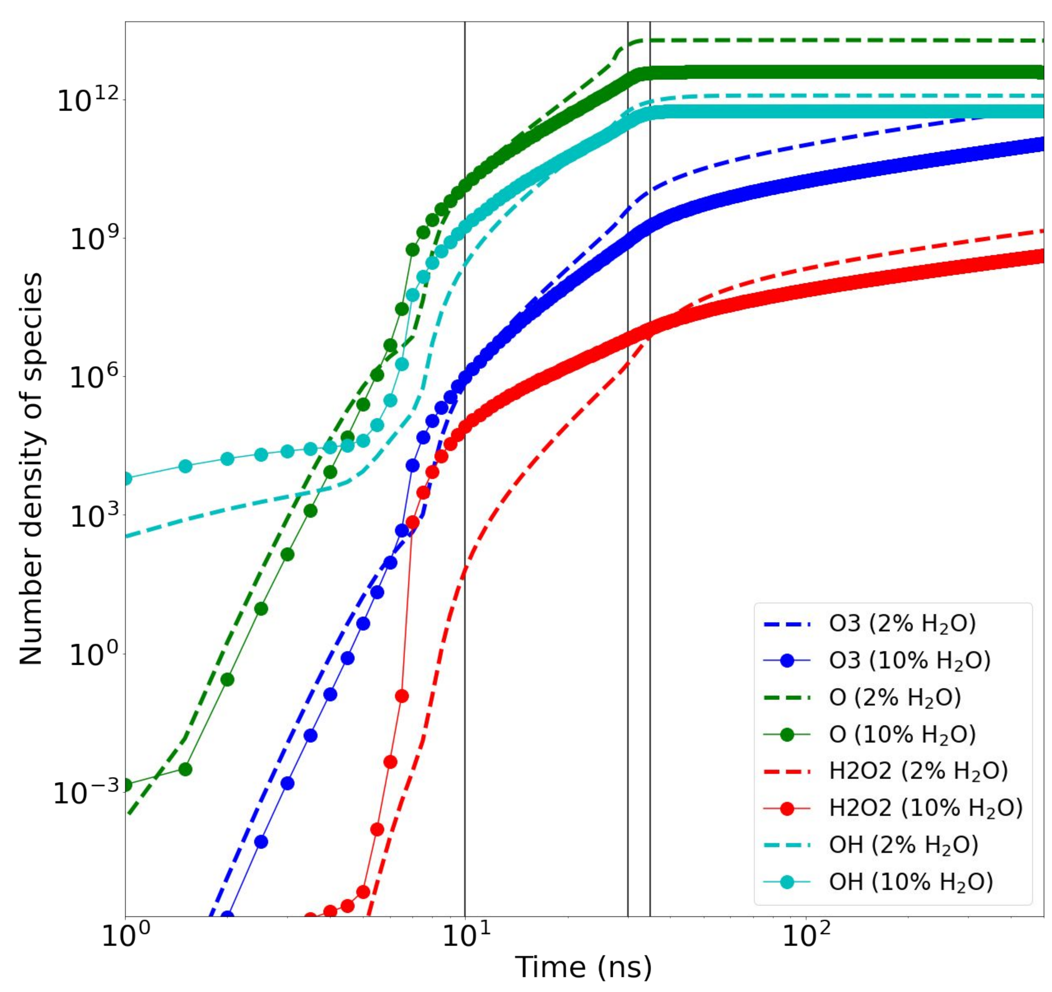


	N_2%	O_2%	H_2O %
Case 1	78.4	19.6	2
Case 2	72	18	10

Neutral	N_2, O_2, H_2O, N, O, N_2O, O_3, H2, H, OH, H_2O_2, HO_2, NO, NO_3, NO_2, N_2O_4, N_2O_5, HNO_2, HNO_3, ONOOH
Positive	N_2^+, H_2O^+, $N_2^+(B^2\Sigma)$, $N_2O_2^+$, N_4^+, O_2^+, O_4^+, OH^+, O^+, H_2^+, H^+, $O_2^+{\cdot}(H_2O)$, H_3O^+, $H_3O^+{\cdot}(H_2O)$, $H_3O^+{\cdot}(H_2O)_2$, $H_3O^+{\cdot}(H_2O)_3$
Negative	e^-, O_2^-, O_3^-, O^-, OH^-, H^-, $O_2^-{\cdot}(H_2O)$, $O_2^-{\cdot}(H_2O)_3$
Excited	$O_2(a^1\Delta_g)$, $N(^2D)$, $O(^1D)$, $N_2(B^3\Pi_g)$, $N_2(C^3\Pi_u)$, $O(^3P)$.

A total of 50 species, and 131 reactions.





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Pancheshnyi, Sergey. "Effective ionization rate in nitrogen–oxygen mixtures." Journal of Physics D: Applied Physics 46.15 (2013): 155201.

[2] Itikawa, Yukikazu, and Nigel Mason. "Cross sections for electron collisions with water molecules." Journal of Physical and Chemical reference data 34.1 (2005): 1-22.

[3] Norberg, Seth A., Eric Johnsen, and Mark J. Kushner. "Formation of reactive oxygen and nitrogen species by repetitive negatively pulsed helium atmospheric pressure plasma jets propagating into humid air." Plasma Sources Science and Technology 24.3 (2015): 035026.

[4] https://gitlab.com/MD-CWI-NL/afivo-streamer

Conclusions and Further Questions

- Increasing humidity from 2% to 10%:
  - caused streamer branching
  - slower streamers
  - decreased overall electron density
  - small percentage decrease in RONS.

BUT

Will the same specie trend be exhibited in subsequent pulses?

Identifying

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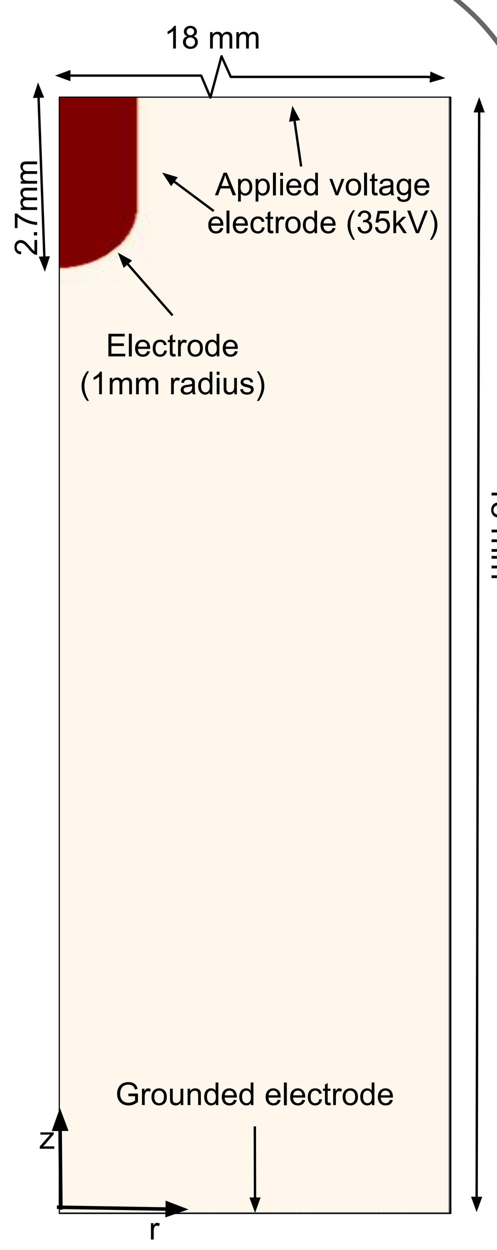
Research Questions

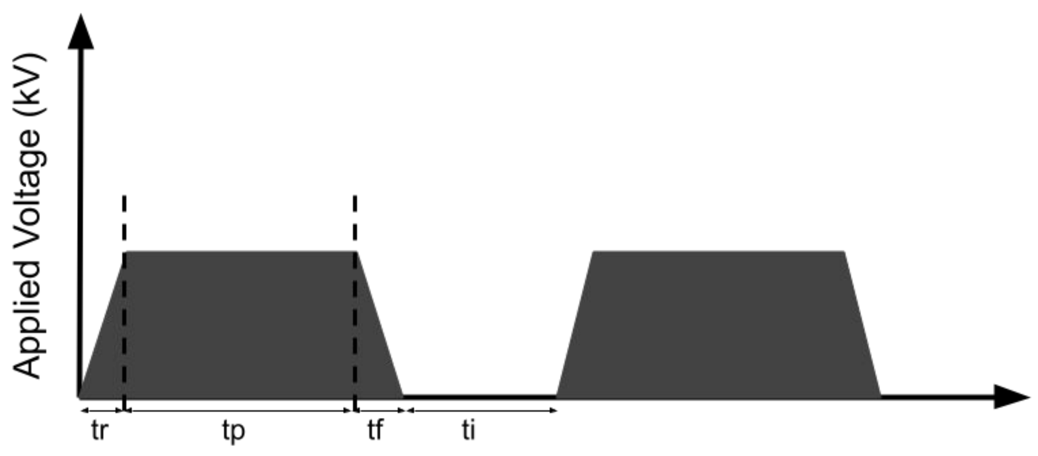
We perform numerical simulations of streamer discharges under repetitively pulsed conditions. It was observed in [1] that the pulse repetition rate influences the evolution of a streamer channel. Incorporating a chemistry model, and a gas heating model, we investigate the following questions:

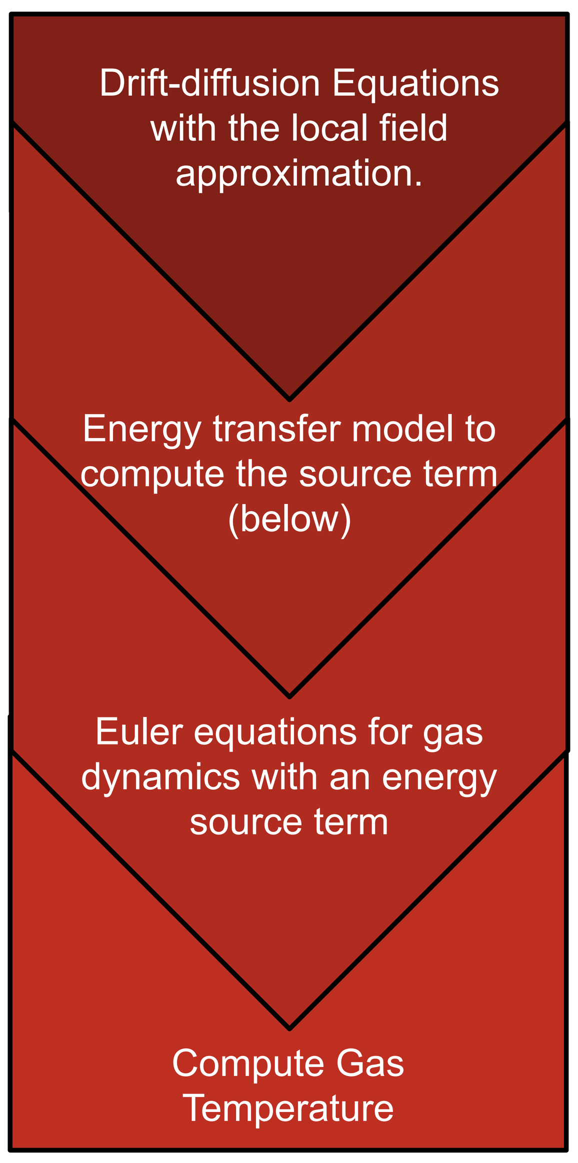
- **How does the pulse repetition rate influence the evolution of a streamer channel in air?**
- **How is the energy (thus temperature) deposited in the streamer channel?**

Simulation Model

- Open source afivo-streamer code.
- Air chemistry (including ionization, attachment, recombination, detachment).
- Air at STP with photoionization
- Initial background ionization:
  - 10^{10} m^{-3} electrons and O2^+ ions
  - Ions are immobile
- Rise time (tr) = 3ns
- Fall time (tf) = 3ns
- Pulse time (tp) = 10ns
- Inter-pulse time (ti) = 20ns OR 80ns



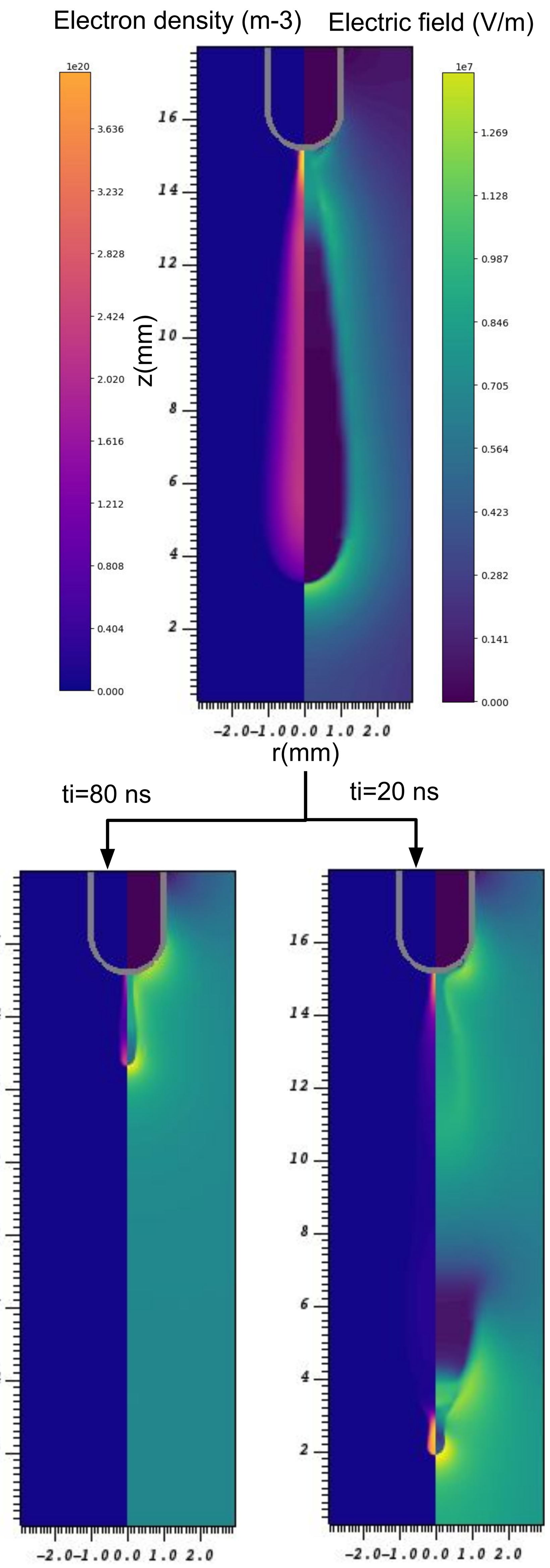


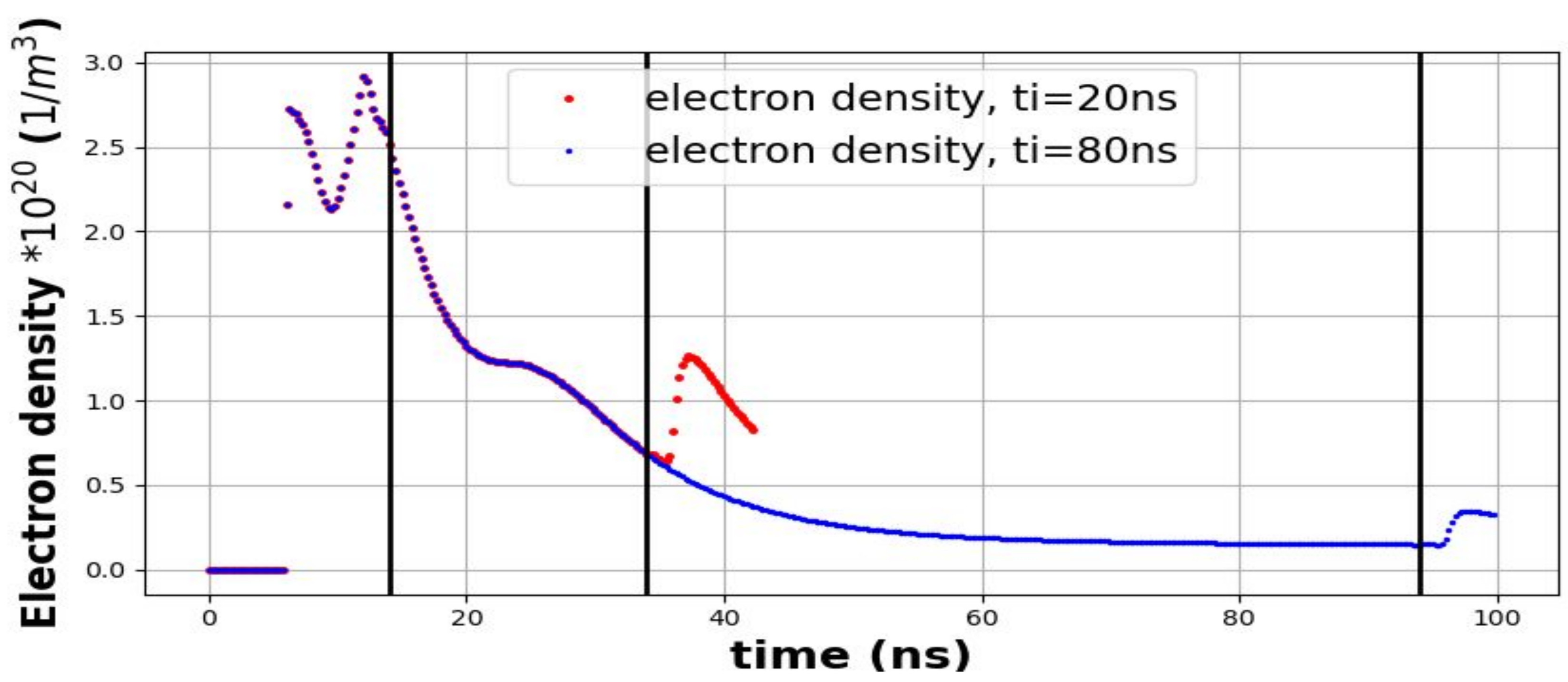


$$
\begin{cases}
S_{\text{Joule}} = \mathbf{j}\cdot\mathbf{E} \approx (e\mu_e n_e \mathbf{E})\cdot\mathbf{E} \\
\dfrac{\mathrm{d}S_{\text{slow}}}{\mathrm{d}t} = \dfrac{1}{\tau_{\text{slow}}}\left(\eta_{\text{slow}} S_{\text{Joule}} - S_{\text{slow}}\right) \\
S_{\mathcal{E}} = \eta_{\text{fast}} S_{\text{Joule}} + S_{\text{slow}}
\end{cases}
$$

Fast Heating
Instantaneous transfer, due to chemical processes like ionization, attachment, electronic-excitation, detachment

Slow Heating
A fraction of the total energy goes into vibrationally exciting nitrogen molecules, which is then slowly converted into heat at a characteristic time scale.





Conclusions and Further Questions

- During the pulse-off phase, certain chemical reactions dominate and consume electrons. This determines where the subsequent discharge starts. Which reaction set [2] or set of reaction/species are important?
- For the time scale considered here (<150 ns), we observe at most a 14K rise in gas temperature. How and when does the gas heating lead to gas motion, which in turn starts affecting the discharge physics?

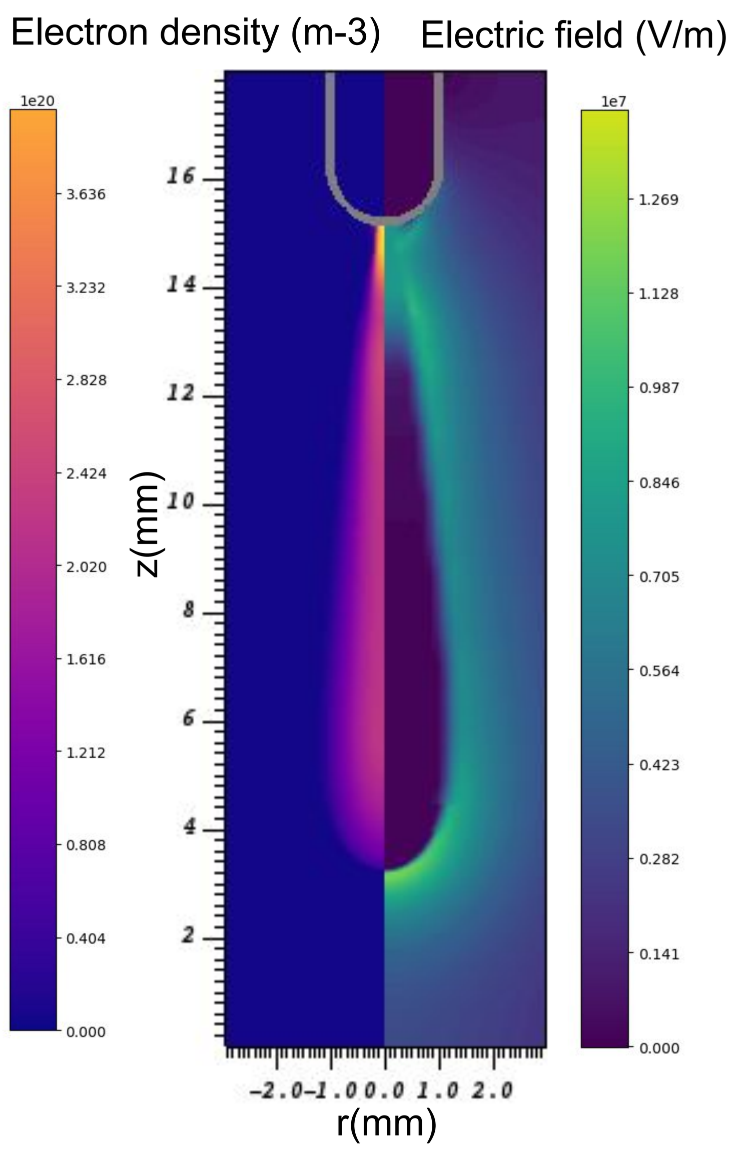
Issues

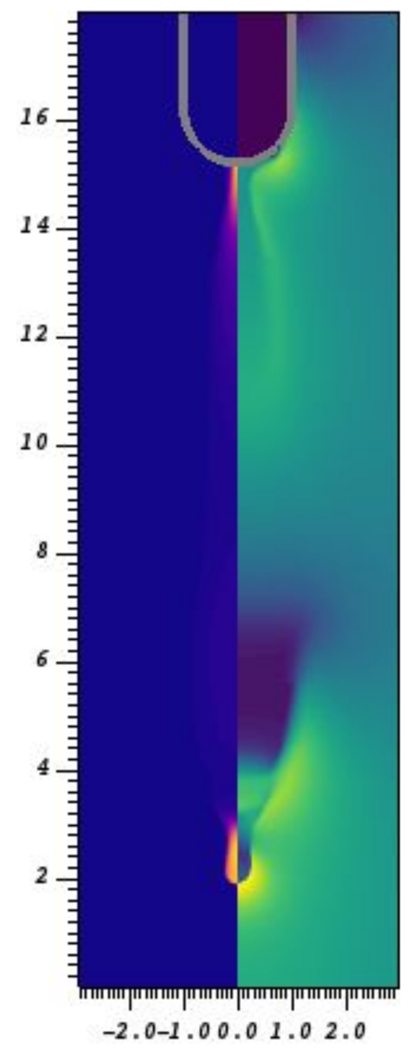
- Long time scale simulations in our code gives unphysical branching.

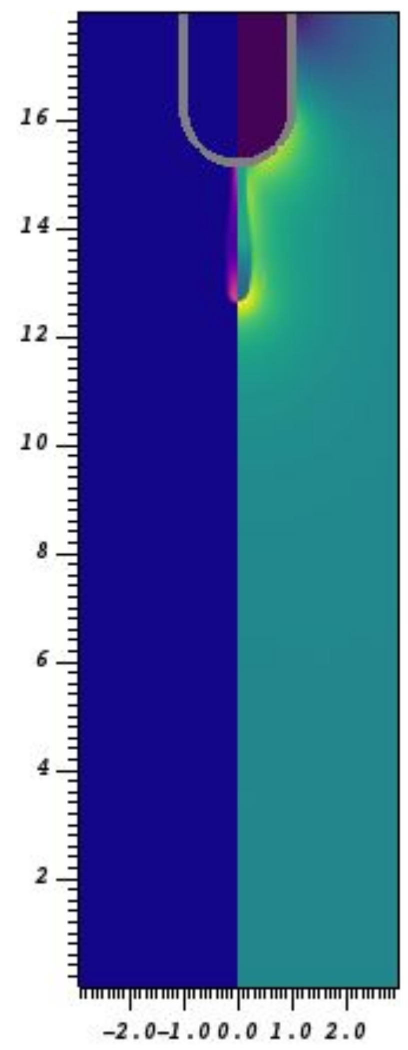
References

[1] Li, Yuan, et al. "Positive double-pulse streamers: how pulse-to-pulse delay influences initiation and propagation of subsequent discharges." *Plasma Sources Science and Technology* 27.12 (2018): 125003.

[2] Kossyi, I. A., et al. "Kinetic scheme of the non-equilibrium discharge in nitrogen-oxygen mixtures." *Plasma Sources Science and Technology* 1.3 (1992): 207.







1) This is an online poster. Make sure to kep it clean and simple, AND make the most important stuff stand out. Try not to fill it all with random stuff.
2) Highlight the research questions part (bold font, different color, large font). Add a line about the motivation of the topic (e.g., simulations of multiple pulses is so far not possible and we try to do it.)
3) Make a better figure of the simulation setup (maybe include/ shorten the initial description)
4) Remove the equations and just add the names (DD-LFA without ion motion), (Compressible equations for Gas D with a source term for the energy equation). Leave the equations for the heat transfer model.
5) Make a flow chart to explain the heating transfer process.
6) Move the pulses diagram nex/below the init condition figure
7) Add a fourth figure and highlight them all
8) The remaining space will have 2 figures- electron density and temperature
9) FInally highlight the conclusions and research questions (so that people can really notice it and give comments)
10) HIghlight the future questions--- chemistry related: using multiple data sets, one of which is super large, so we need to somehow determine which of the equations are important, etc
TO really determine the effect of heating, we need to go to long time scales, for that we have computational issues like branching, slow timesteps, etc

Fast Heating

Instantaneous transfer, due to chemical processes like ionization, attachment, electronic-excitation, detachment

Slow Heating

Slow transfer (Sslow), due to vibrational-translational energy relaxation. A fraction of the total energy goes into vibrationally exciting nitrogen molecules, which is then slowly released converted into heat at a characteristic time scale of Tau_slow.