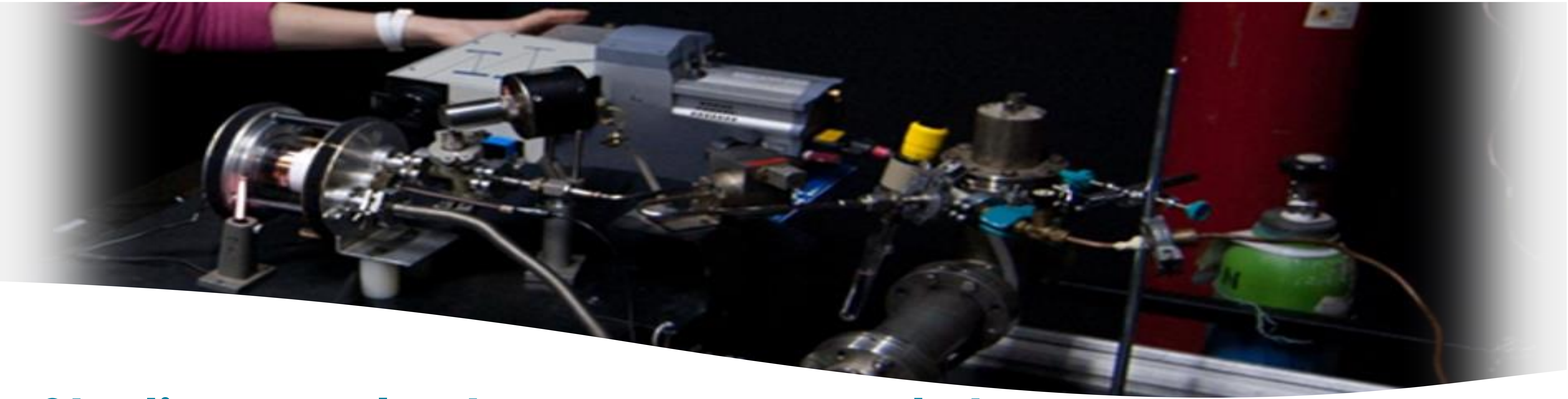

Studies on electron swarms and streamer discharges in environmentally friendly RPC gas mixtures under LHC-like conditions

Saša Dujko[1], Ilija Simonović[1], Danko Bošnjaković[1], Zoran Lj. Petrović[2] and Jaime de Urquijo[3]

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Science Fund
of the Republic of Serbia

XVII international Conference on Resistive Plate Chambers and
Related Detectors (RPC2024), September 9–13, 2024,
Santiago de Compostela, Spain

Why ultra-low GWP gases?

- **Resistive plate chambers**
- **Plasma processing technology**
- **Gaseous dielectrics in HV technology**
- **Refrigerants**

Environmental Impact!

- **EGWIn Project**
- **E**xploring ultra-low **G**lobal **W**arming potential gases for **In**sulation in high-voltage technology: Experiments and modelling

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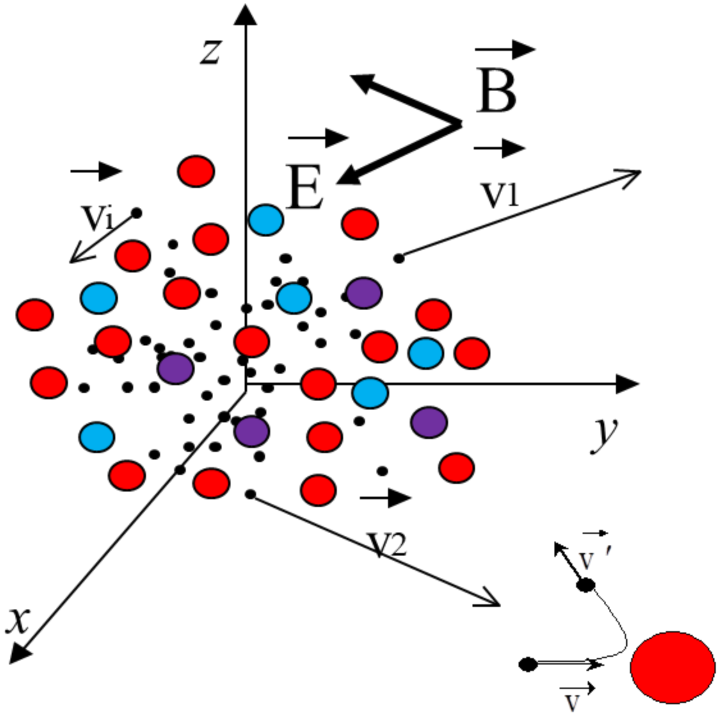
What is a swarm of charged-particles?

Swarm conditions ≡ Free diffusion plasma limit



🔵 🔴 🟣 Neutral gas atoms/molecules
• A swarm particle

Boltzmann equation:

$$\frac{\partial f}{\partial t} + \mathbf{c}\cdot\frac{\partial f}{\partial \mathbf{r}} + \frac{q}{m}\left(\mathbf{E}+\mathbf{c}\times\mathbf{B}\right)\cdot\frac{\partial f}{\partial \mathbf{c}} = -J\left(f, f_0\right)$$

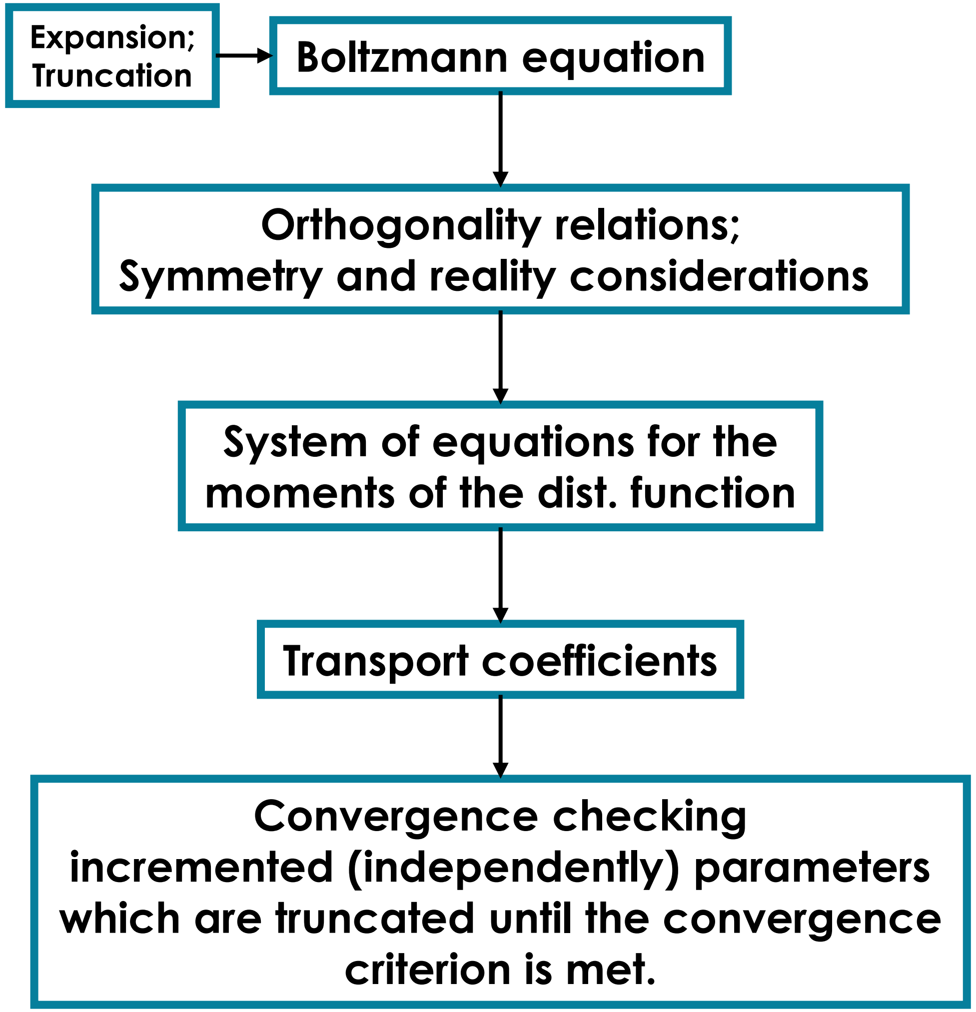
Ludwig Boltzmann (1844-1906)

Swarm conditions:

- ☐ Low density of charged particles:
  - Neglect interactions between charged particles
  - Neglect space charge effects
- ☐ **E** and **B** fields are spatially homogeneous and externally prescribed
- ☐ Small spatial gradients in number density
- ☐ Minimal boundary effects

1872 → 2022: 150th anniversery of the Boltzmann equation!

How do we solve the Boltzmann equation?



Expansion; Truncation

Boltzmann equation

Orthogonality relations; Symmetry and reality considerations

System of equations for the moments of the dist. function

Transport coefficients

Convergence checking incremented (independently) parameters which are truncated until the convergence criterion is met.

Dujko *et al.* 2010 *Phys. Rev. E* **81** 046403

$$\frac{\partial f}{\partial t}+\mathbf{c}\cdot\frac{\partial f}{\partial \mathbf{r}}+\frac{q}{m}\left(\mathbf{E}+\mathbf{c}\times\mathbf{B}\right)\cdot\frac{\partial f}{\partial \mathbf{c}}=-J\left(f,F_0\right)$$

- Resolving the angular dependence in velocity space:

$$f(\mathbf{r},\mathbf{c},t)=\sum_{l=0}^{\infty}\sum_{m=-l}^{l} f_m^{(l)}(\mathbf{r},c,t)Y_m^{[l]}(\hat{\mathbf{c}})$$

- Projecting out the space dependence:
  - Hydrodynamic regime:

$$f_m^{(l)}(\mathbf{r},c,t)=\sum_{s=0}^{\infty}\sum_{\lambda=0}^{\infty}\sum_{\mu=-\lambda}^{\lambda} f(lm\,|\,s\lambda\mu;c,t)G_\mu^{(s\lambda)}n(\mathbf{r},t)$$

  - Non-hydrodynamic regime:
    - finite difference
    - pseudo-spectral

- Resolving the speed dependence:

$$f(lm\,|\,s\lambda\mu;c,t)=\omega(\alpha,c)\sum_{\nu=0}^{\infty}F(\nu lm\,|\,s\lambda\mu;\alpha,t)R_{\nu l}(\alpha c)$$

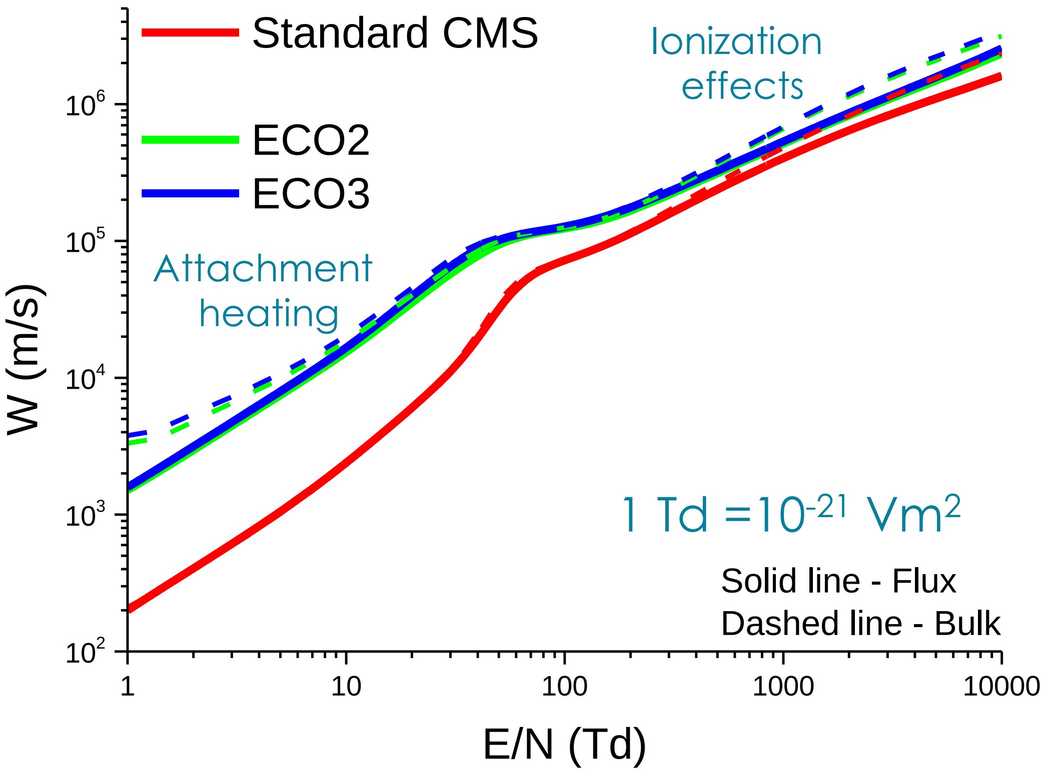
Transport coefficient duality

- **Two families of transport coefficients: Flux and Bulk**
  - Defined under hydrodynamic conditions!
  - Independent of the method of measurement!

$$\mathbf{\Gamma}(\mathbf{r},t)=\mathbf{W}^{(*)}n(\mathbf{r},t)-\mathbf{D}^{(*)}\cdot\nabla n(\mathbf{r},t)+\mathbf{Q}^{(*)}:\nabla\nabla n(\mathbf{r},t)-\dots$$

$$\frac{\partial n}{\partial t}+\mathbf{W}\cdot\nabla n-\mathbf{D}:\nabla\nabla n+\mathbf{Q}\vdots\nabla\nabla\nabla n-\dots=-R_a n$$

- **Swarm Experiments:**
  - Time-of-flight
  - Pulsed-Townsend
  - Steady-state Townsend
  - Arrival-time spectra, …
- **Fluid modelers must be aware of the origin of the transport coefficients they are using in their models!**



Standard CMS: R134a/i-C_4H_{10}/SF_6 95.2/4.5/0.3
ECO2: HFO1234ze/CO_2/ /i-C_4H_{10}/SF_6 35/60/4/1
ECO3: HFO1234ze/CO_2/ /i-C_4H_{10}/SF_6 29/65/5/1

RPC ECOGas@GIF++ Collaboration

Dujko *et al.* unpublished

Cross sections for electron scattering in $C_2H_2F_4$, C_3HF_5 and $C_3H_2F_4$

Cross-section set for electron scattering in $C_2H_2F_4$ (R134a)

Cross sections for electron scattering in $C_2H_2F_4$:
(1) Elastic momentum transfer, (2)-(12) Vibrational excitation, (13)-(14) Electronic Excitation, (15) 3-body attachment, (16) Dissociative attachment, (17) Ionization

Šašić *et al.* unpublished

Quantemol-N code calculations:

- Electronic excitation
- Ionization
- Dissociative electron attachment

Vibrational excitations:

Yamada et al. (1998) have calculated harmonic vibrational frequencies v1 – v18. The number of cross sections for vibrational excitations is reduced to 11.

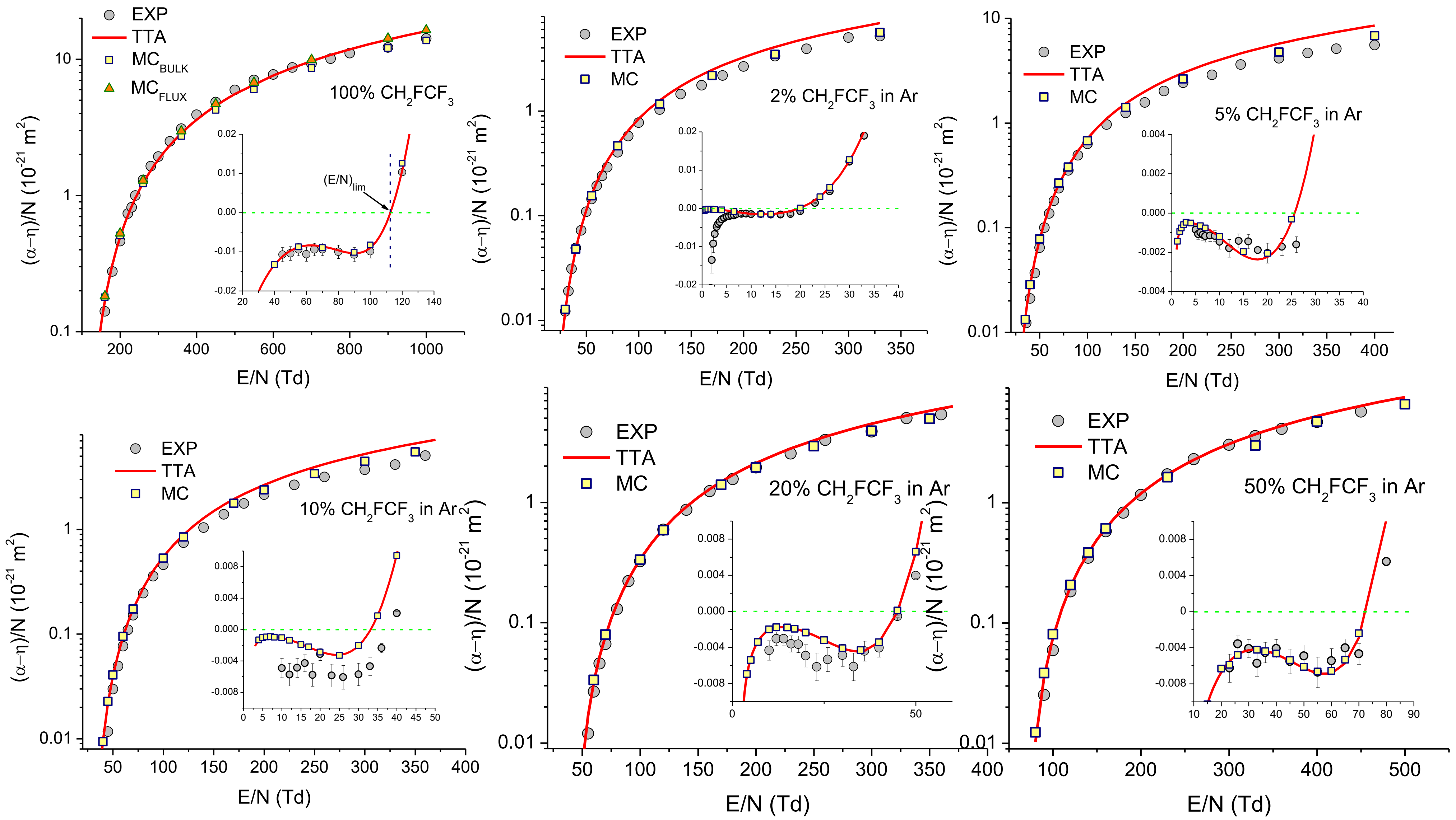
T. Yamada, T.H. Lay and J.W. Bozzelli, *J. Phys. Chem. A* 1998, **102** 7286-7293

Cross-section for 3-body attachment:

Initially developed by Biagi (2010). In the present work it is modified to fit the effective ionization coefficient measured by Basile *et al.* (1999).

G. Basile, I. Gallimberti, S. Stangherlin, T.H. Teich, *in Proceedings of the XX International Conference on Phenomena in Ionized Gases, edited by M. Vaselli,* Vol. 2, 1991, p. 361

Effective ionization coefficient in Ar-$C_2H_2F_4$ mixtures

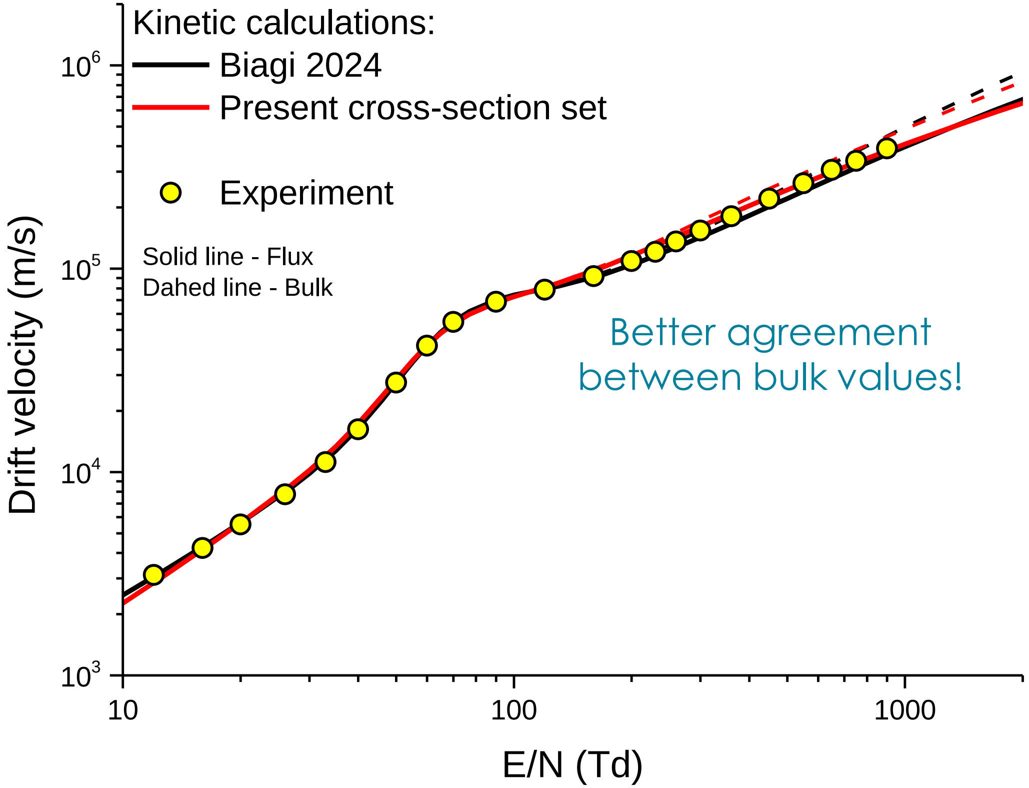


- Very good agreement is reached between calculated and measured data in pure $C_2H_2F_4$ and its mixtures with Ar.

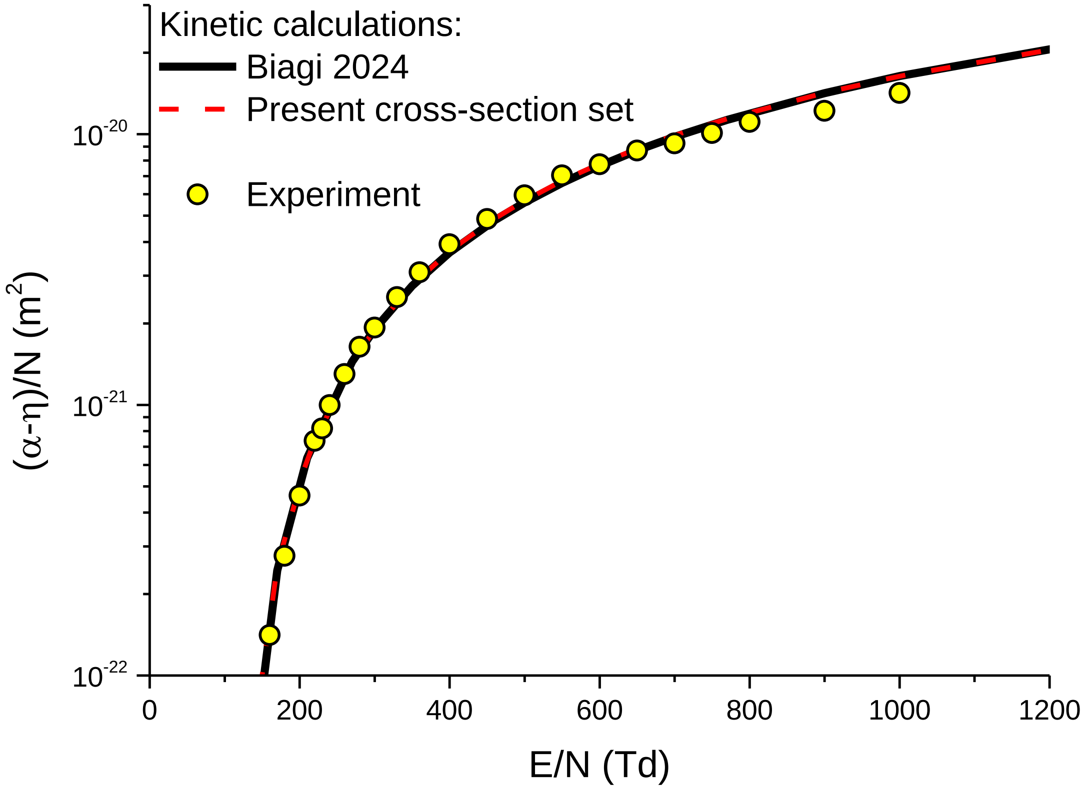
- In most cases differences are about 10% indicating that the inelastic losses are determined with sufficient accuracy over the wide range of the applied E/Ns

- Critical electric field of 112.5 Td for pure $C_2H_2F_4$ agrees very well with the value determined by Basile et al. (1991).

Present cross-section set vs. Biagi 2024



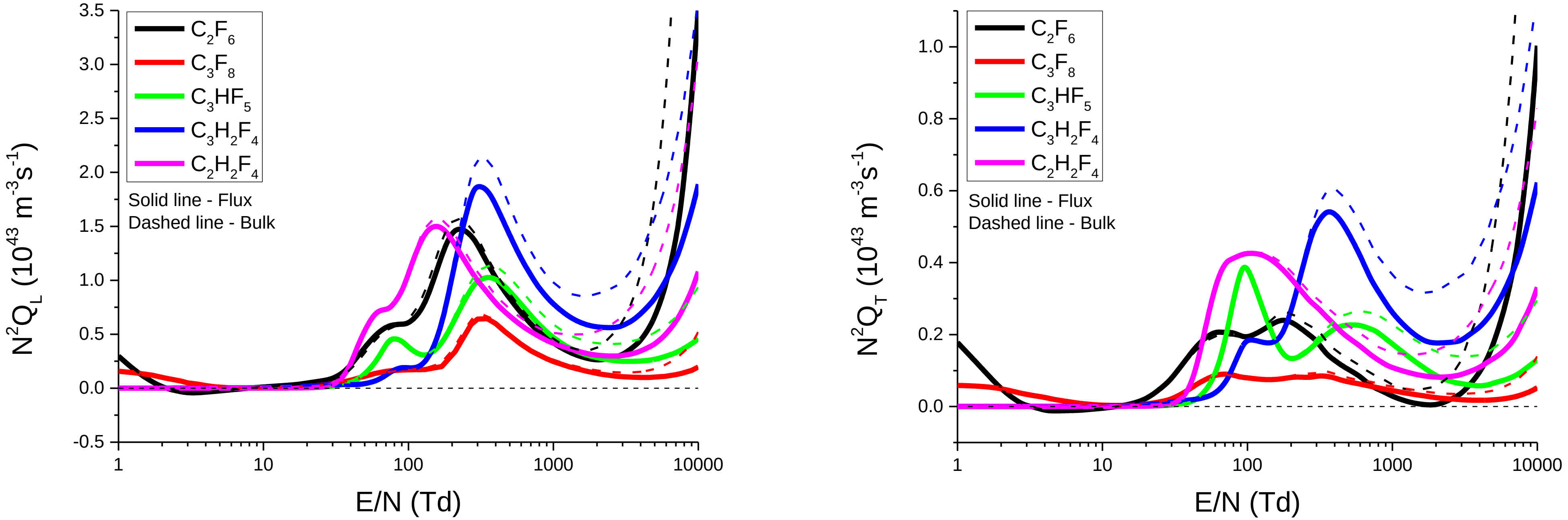
Better agreement between bulk values!



- Good agreement between the swarm data obtained using the two cross-section sets.
- **Example of non-uniqueness:** two completly different cross-section sets provide good agreement between measurements and kinetic calculations.

Biagi 2024: Marnik Metting van Rijn *et al.* (2024) *J. Phys. D: Appl. Phys.* **57** (2024) 355202

Why third-order transport coefficients?



- Required in swarm analysis for converting transport data measured in various experiments into hydrodynamic transport coefficients. They can be negative!
- Necessary for describing deviations of spatial density profile from an ideal Gaussian.
- Since they are very sensitive with respect to the energy dependence of cross sections - their measurement and calculation would improve the accuracy of cross section fitting procedure (reducing the non-uniqueness!).

Dujko *et al*. unpublished

Is C_3HF_5 a good candidate for replacing $C_2H_2F_4$ in RPCs?

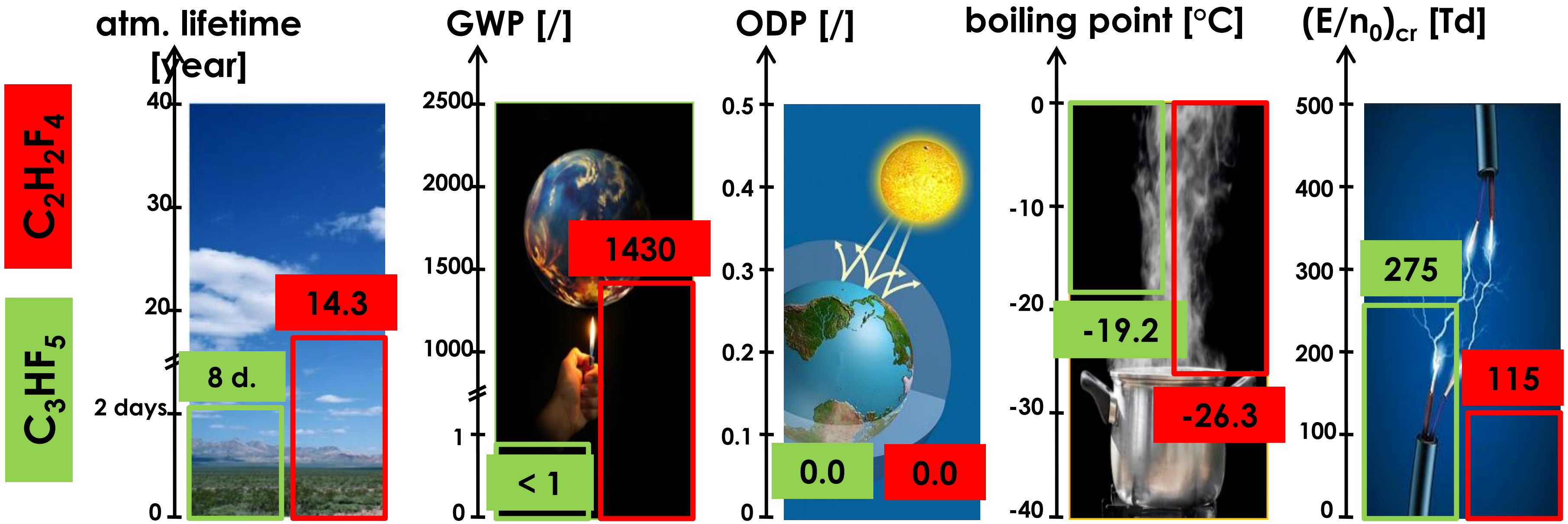
Pentafluoropropene C_3HF_5:

- Also known as HFO1225ye(Z) or R1225ye(Z)
- Considered as (1) medical propellant, (2) possible component of an alternative refrigerant blend, (3) plasma processing gas, and (4) gaseous dielectrics. **So far it has not be considered in RPCs!**

Pentafluoropropene C_3HF_5 vs R134a

+ Low-toxicity, Non-flammable, Good chemical stability, Good thermal stability.

- Boiling point (-19.2 °C at 0.1 MPa), Difficult to directly apply in gas insulated HV equipment (must be mixed with buffer gases), **RPCs: Too high operating voltages, More prone to streamer formation, More expensive**



Cross sections for electron scattering in C_3HF_5

Elements of swarm analysis

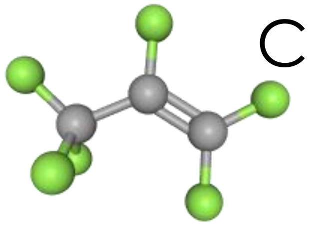
We use individual cross sections for electron scattering in C_2F_6, C_3F_6, and C_3F_8 to construct the initial set.

C_3F_6 and C_3HF_5 have drift velocities that are quite similar. This applies to the effective ionization rate coefficient above the critical electric field as well.

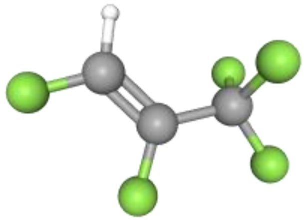
Cross sections for ionization and dissociative attachment are calculated using Quantemol-N code.

Pulsed-Townsend measurements of effective ionization coefficient, drift velocity, and longitudinal diffusion coefficient were used as a set of reference data.

The three-body attachment cross section was developed manually using measurements of the pressure-dependent effective ionization coefficient.

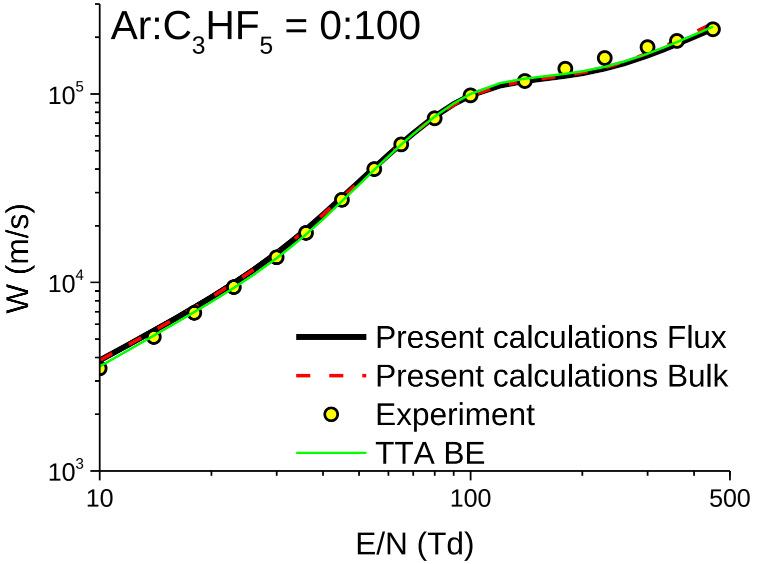


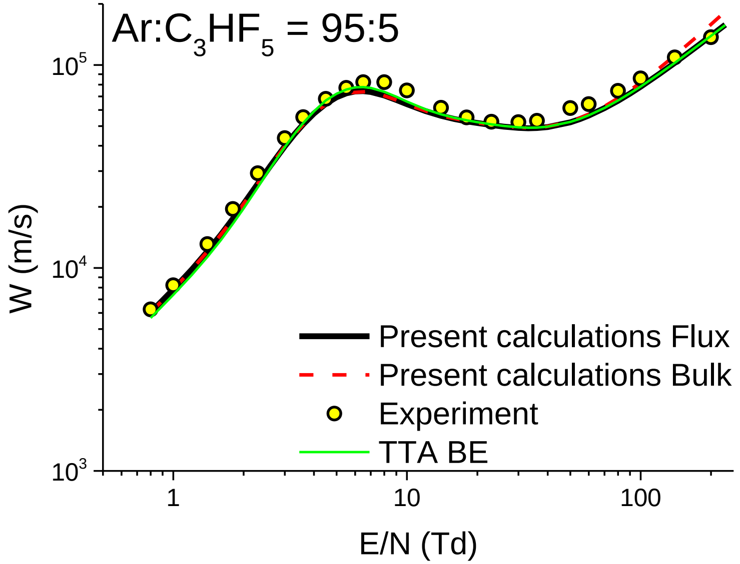
C_3F_6

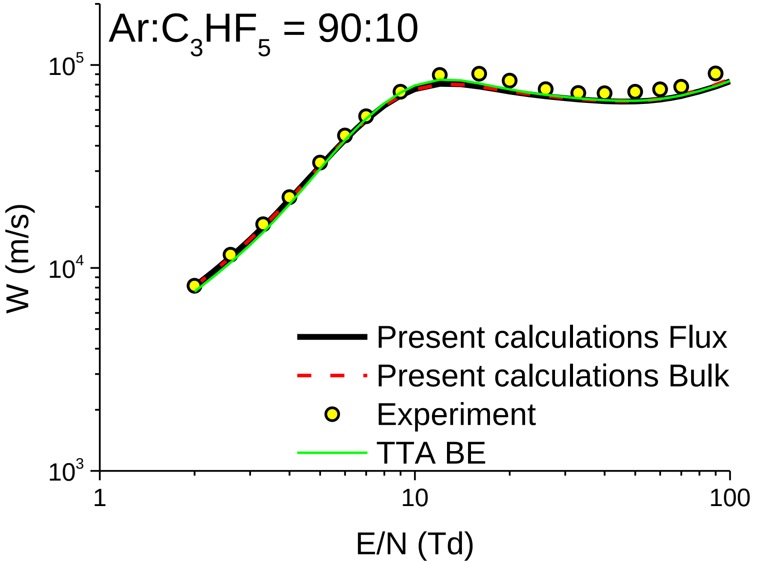


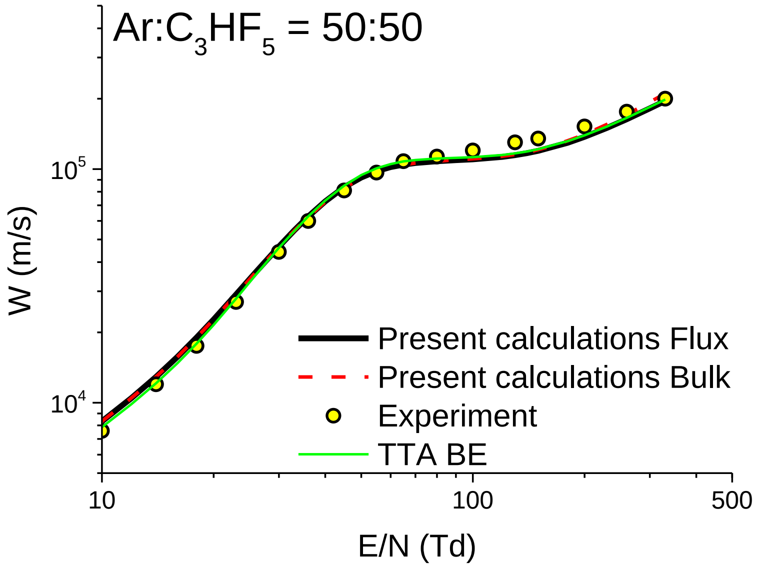
C_3HF_5

Drift velocity in Ar-C_3HF_5 mixtures





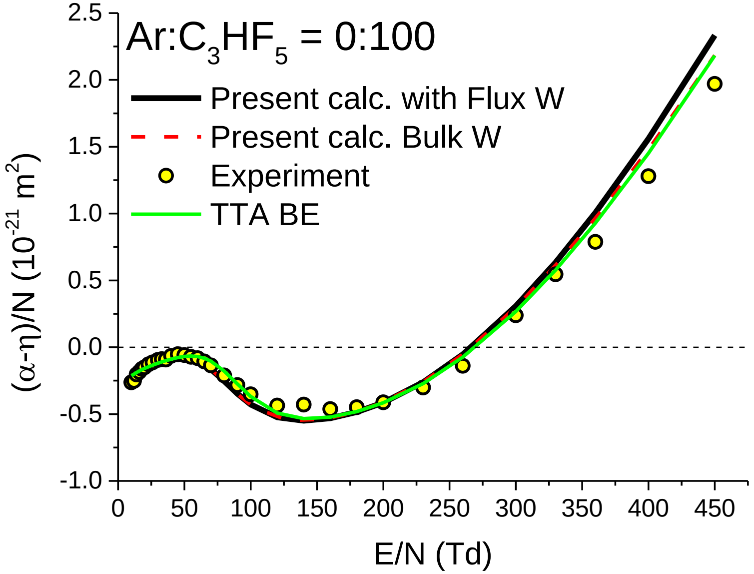


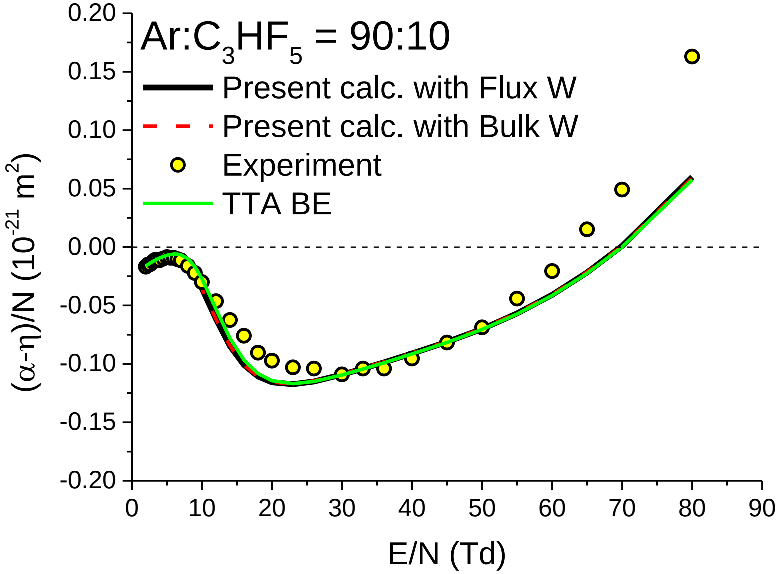


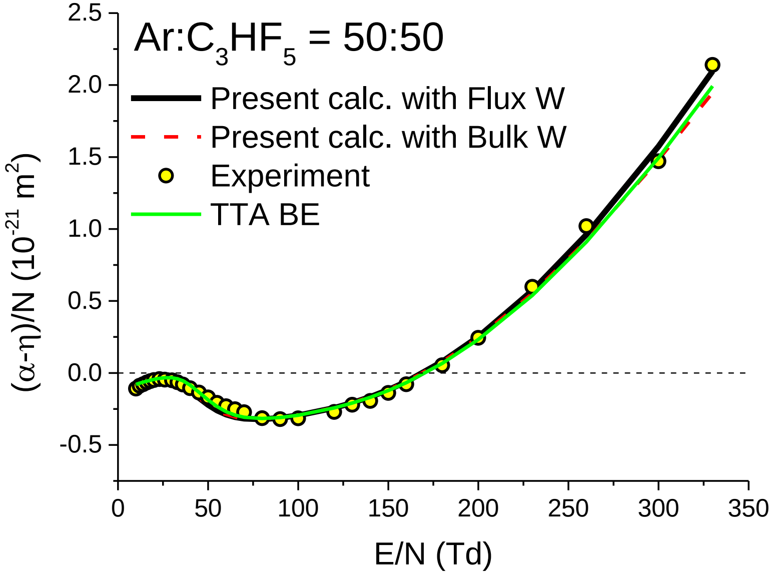
We observe the following:

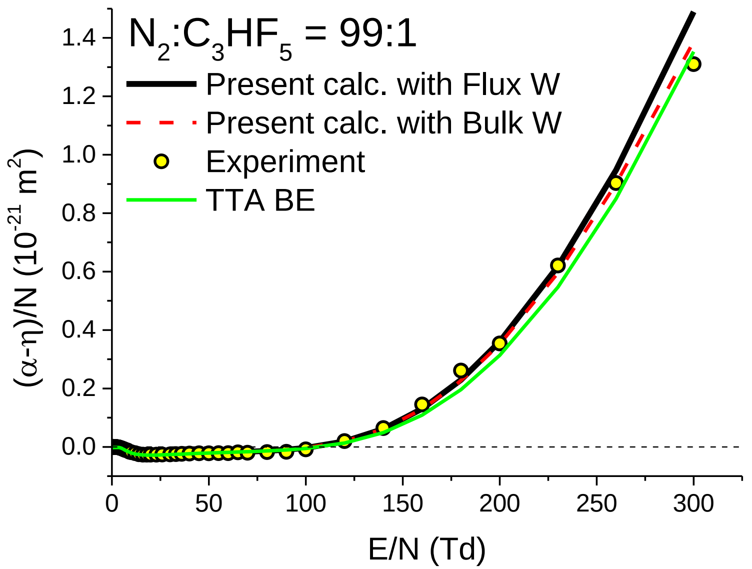
- We have reached the optimal fit with the present data.
- Cross sections for electron scattering in Ar were taken from Hayashi’s database.
- **What is NDC?** Negative differential conductivity (NDC) is a decrease of the drift velocity with increasing E/N.
- Good agreement between measured and calculated drift velocity in the presence of NDC is a good indicator of momentum balance in our cross-section set.

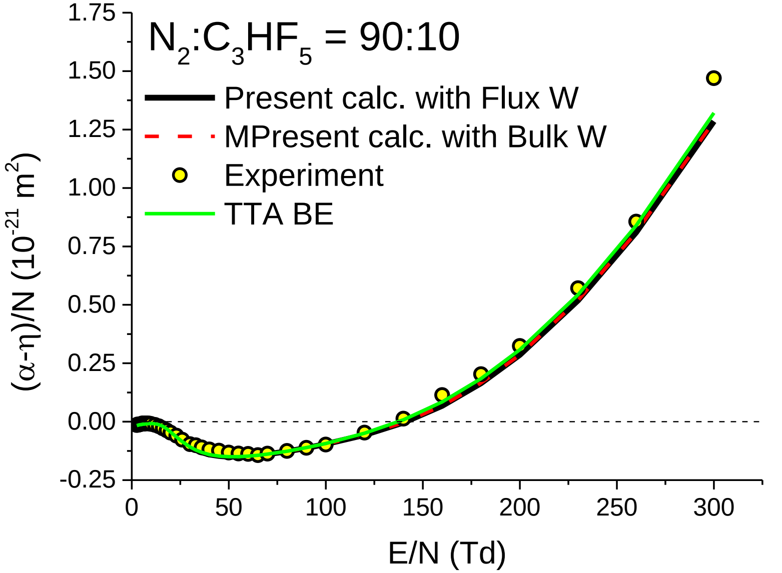
Effective ionization coefficient in Ar-C_3HF_5 and N_2-C_3HF_5 mixtures

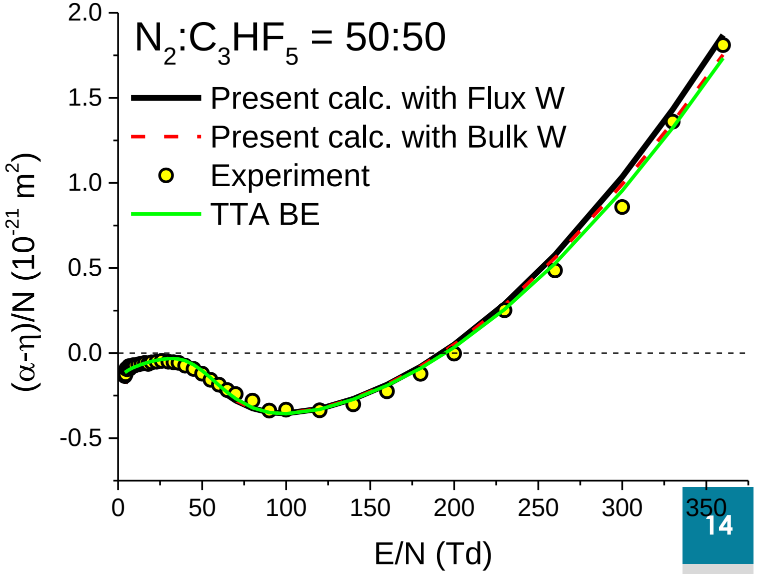




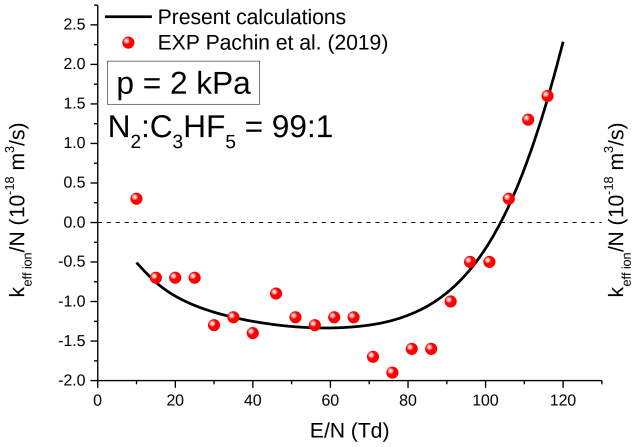


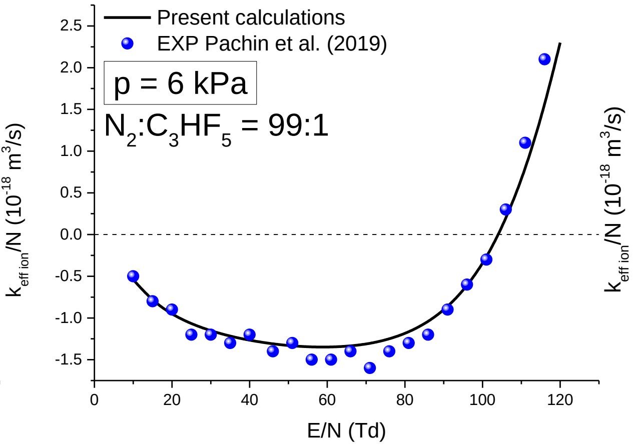


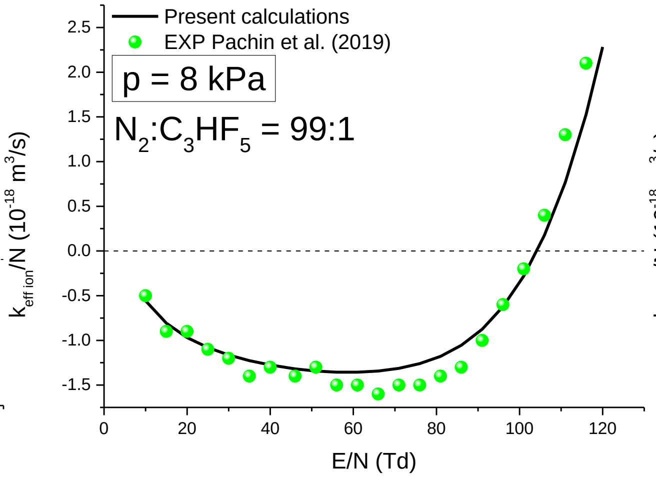


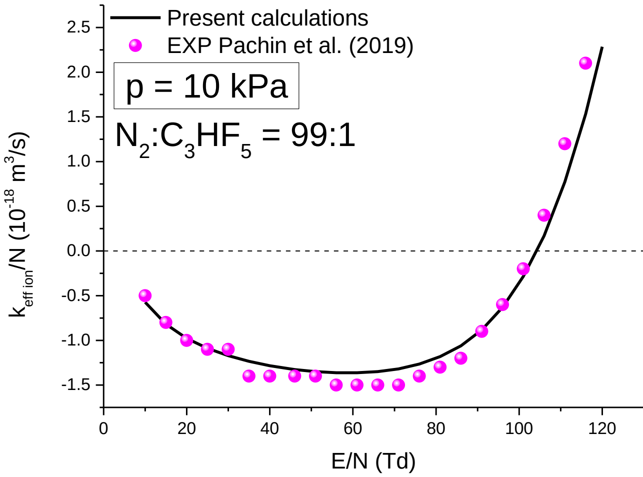


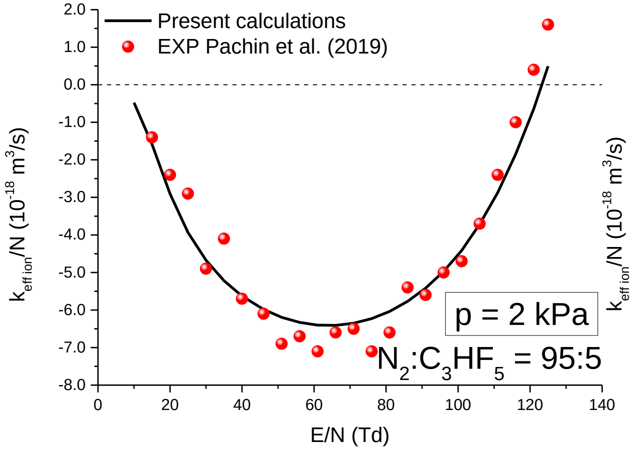
Effective ionization coefficient in N_2-C_3HF_5 mixtures: Our data vs. HV ETH Zurich experimental data

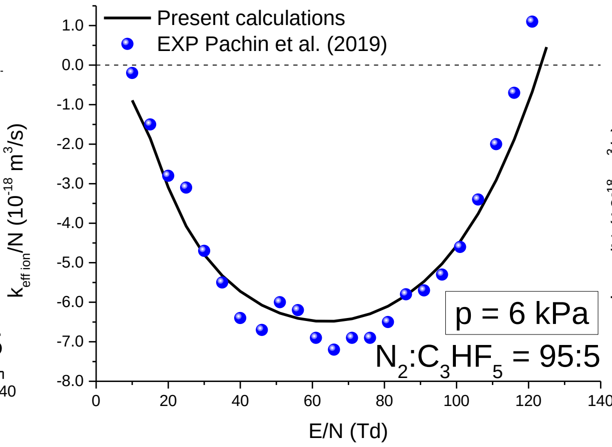


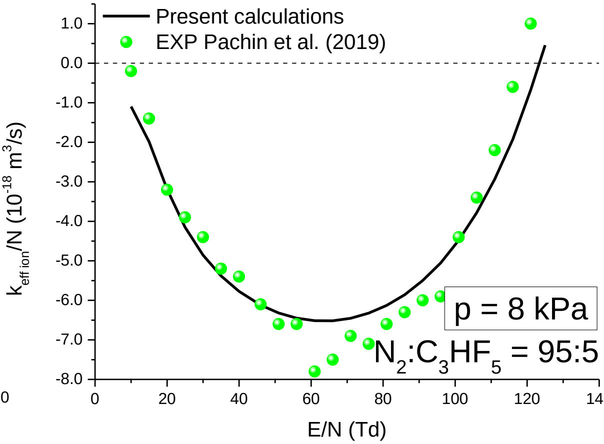


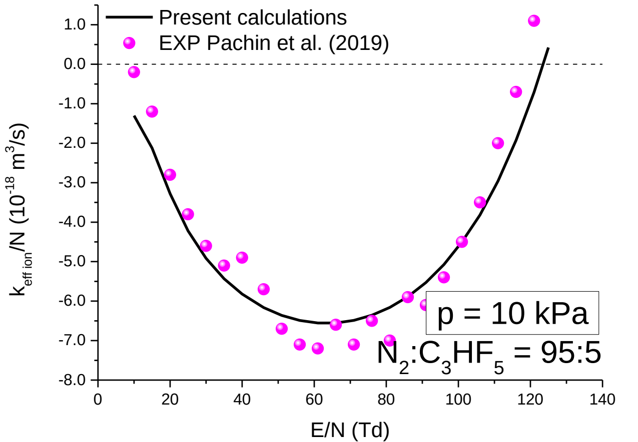






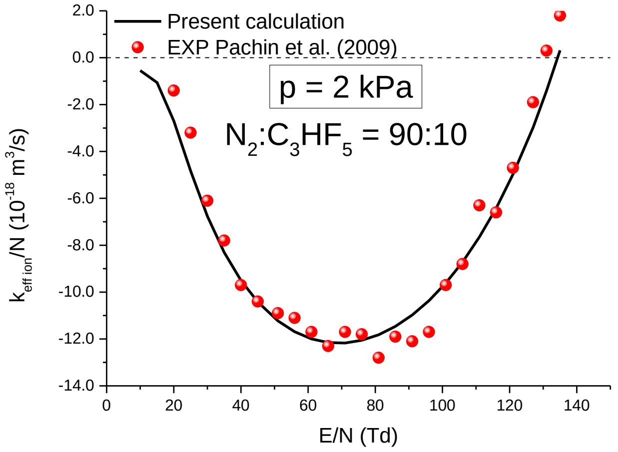


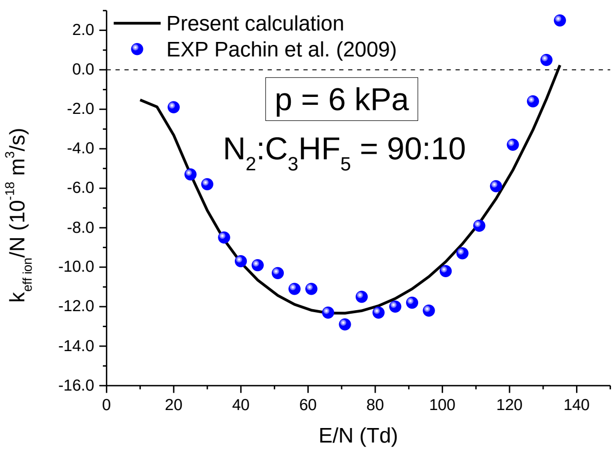


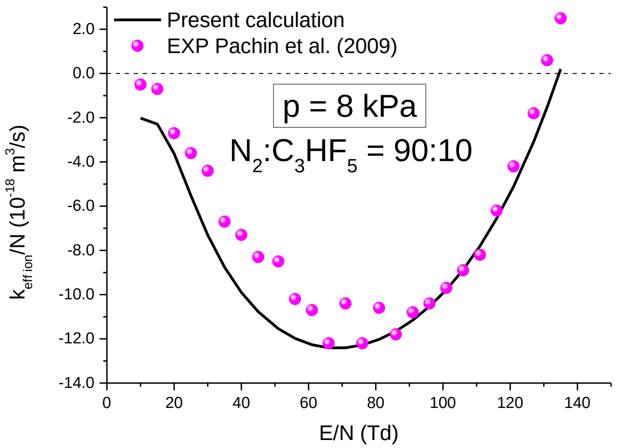


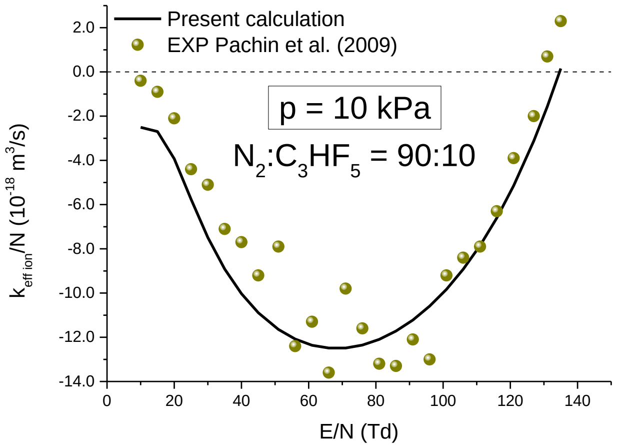
Pachin *et al.* (2019) *J. Phys. D: Appl. Phys.* **52** 235204

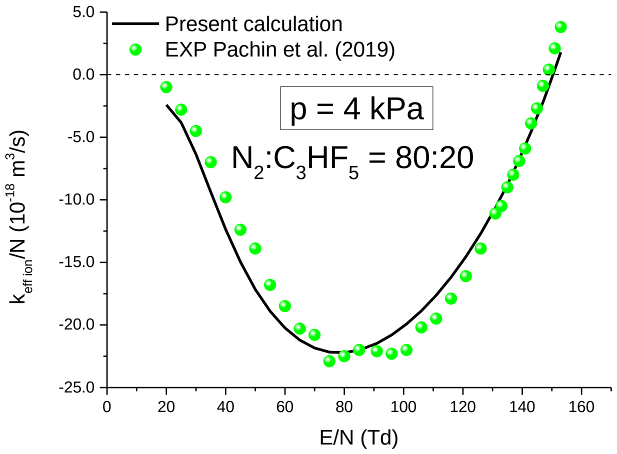
Effective ionization coefficient in N_2-C_3HF_5 mixtures: Our data vs. HV ETH Zurich experimental data

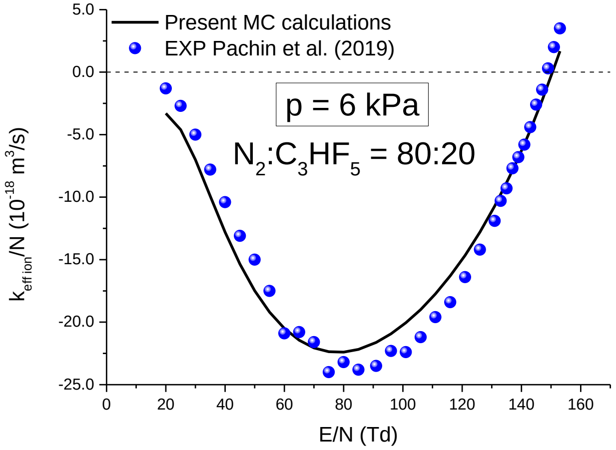


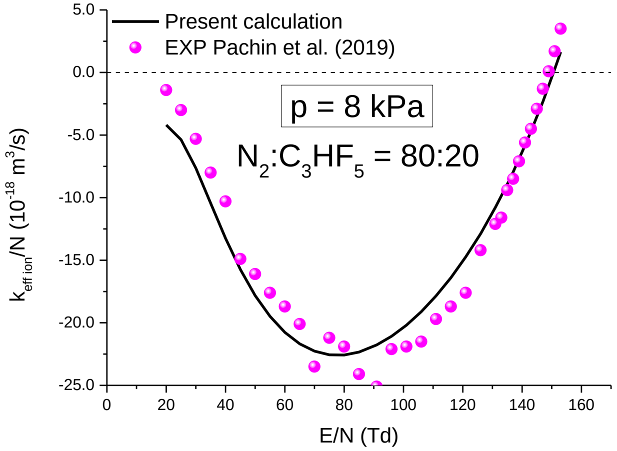


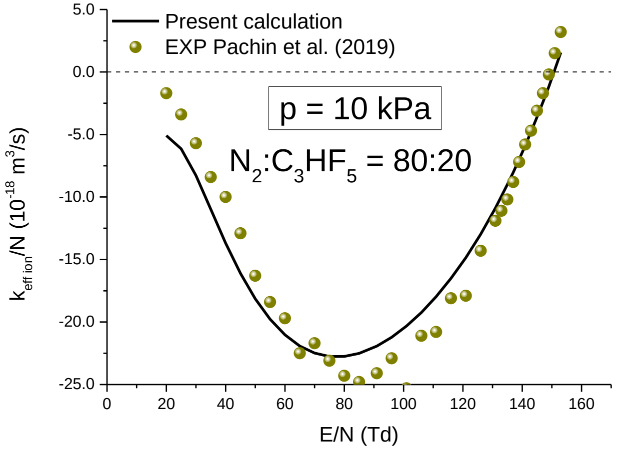












Pachin *et al.* (2019) *J. Phys. D: Appl. Phys.* **52** 235204

Is $C_3H_2F_4$ a good candidate for replacing $C_2H_2F_4$ in RPCs?

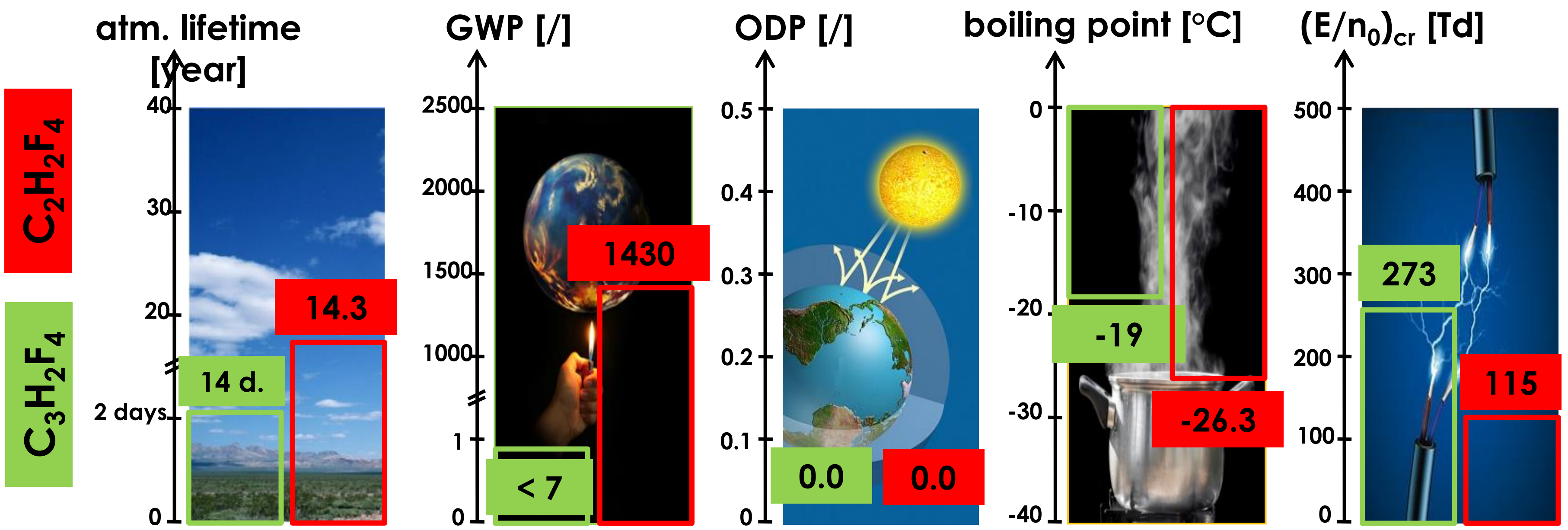
Tetrafluoropropene $C_3H_2F_4$:

- Also known as HFO1234ze(E)
- Applications: (1) used as a refrigerant gas as a replacement of hydrofluorocarbon R134a ($C_2H_2F_4$) (2) plasma processing gas, (3) gaseous dielectrics, (4) used in RPC detectors as a replacement of R134a!

Pentafluoropropene $C_3H_2F_4$ vs **R134a**

+ Low-toxicity, Non-flammable, Good chemical stability, Good thermal stability.

- Boiling point (-19 °C at 0.1 MPa), Difficult to directly apply in gas insulated HV equipment (must be mixed with buffer gases), **RPCs: Cannot be used as a replacement of R134a (must be mixed with R134a, CO_2 or He)**



Cross sections for electron scattering in $C_3H_2F_4$

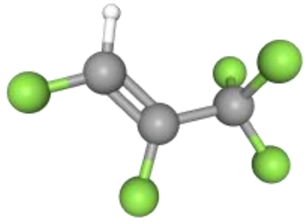
Elements of swarm analysis

The initial cross-section set is constructed using individual cross sections for electron scattering in C_2F_6, C_3F_6, and C_3F_8, and a set of cross sections for C_3HF_5.

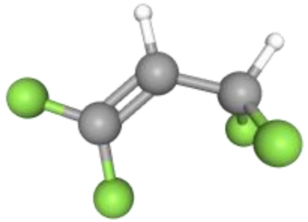
Cross sections for ionization and dissociative attachment are calculated using Quantemol-N code.

Pulsed-Townsend measurements of effective ionization coefficient, drift velocity, and longitudinal diffusion coefficient were used as a set of reference data.

The three-body attachment cross section was developed manually using measurements of the pressure-dependent effective ionization coefficient.



C_3HF_5



$C_3H_2F_4$

Drift velocity and effective ionization coefficient for electrons in pure $C_3H_2F_4$

Very good agreement between experimental results and calculations.

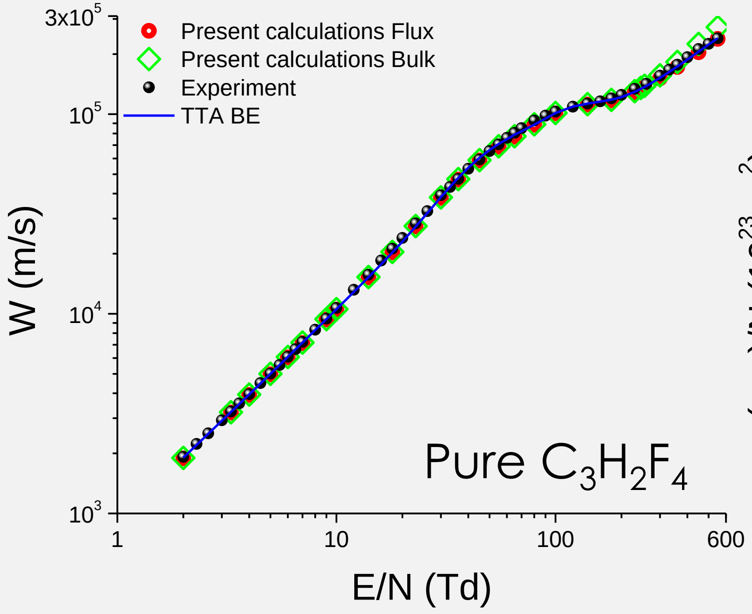
Drift velocity: within 5% for E/N<300 Td, and 12% for higher values of E/N.

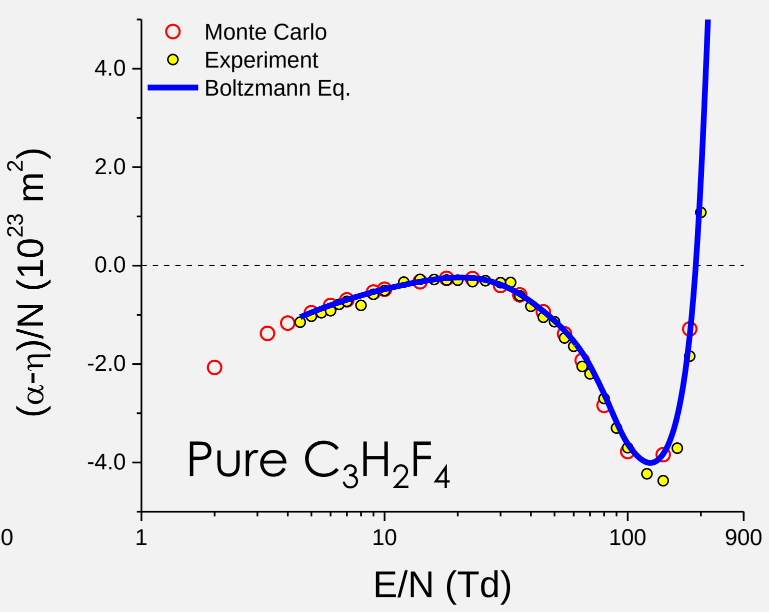
Effective ionization coefficient: within 10% with the exception around critical electric field.

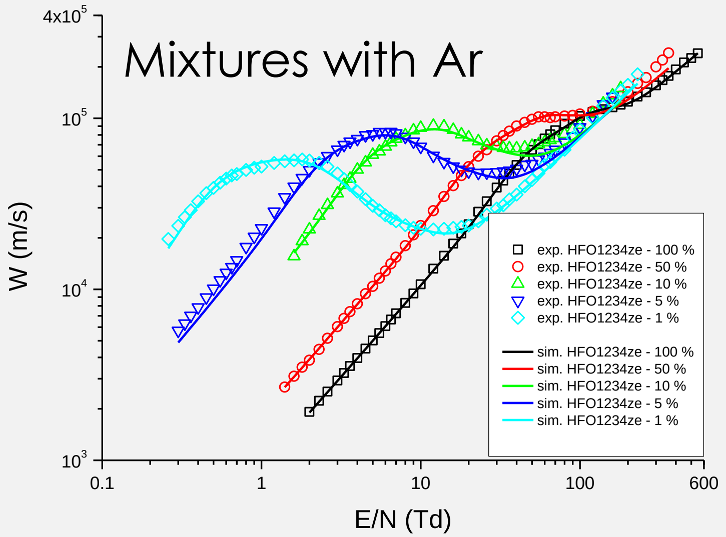
In pure C3H2F4 there is no NDC in the profile of drift velocity.

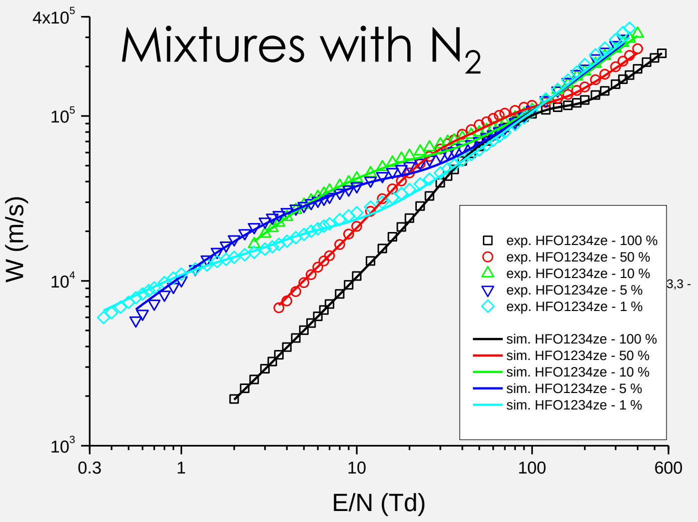
Critical electric field is pressure-dependent (at 1 bar pressure 273 Td).

Mixtures with argon: Good agreement between measured and calculated drift velocity in the presence of NDC





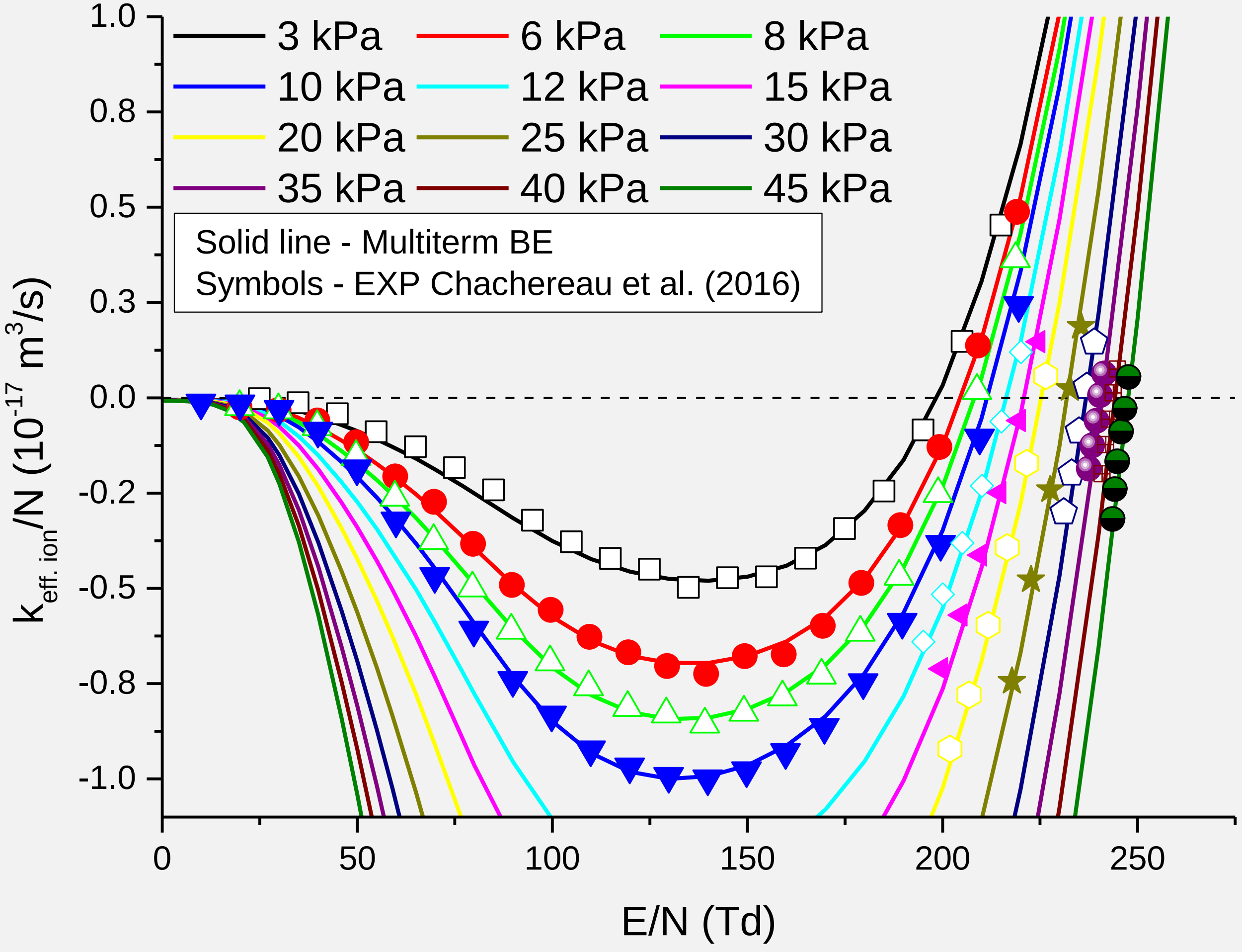




Mirić *et al.* unpublished

Effective ionization coefficient in pure $C_3H_2F_4$

- Excellent agreement between our kinetic calculations and measurements under pulsed-Townsend conditions.
- Scaling factor for the cross section of a 3-body attachment is linear function of the gas pressure (no detachment!).
- Gas pressure has no impact on drift and diffusion. This suggests that the 3-body attachment has small implicit effects on the distribution function.
- Critical electric field increases with increasing gas pressure. At 1 bar pressure, the Ecr is approximately 275 Td.



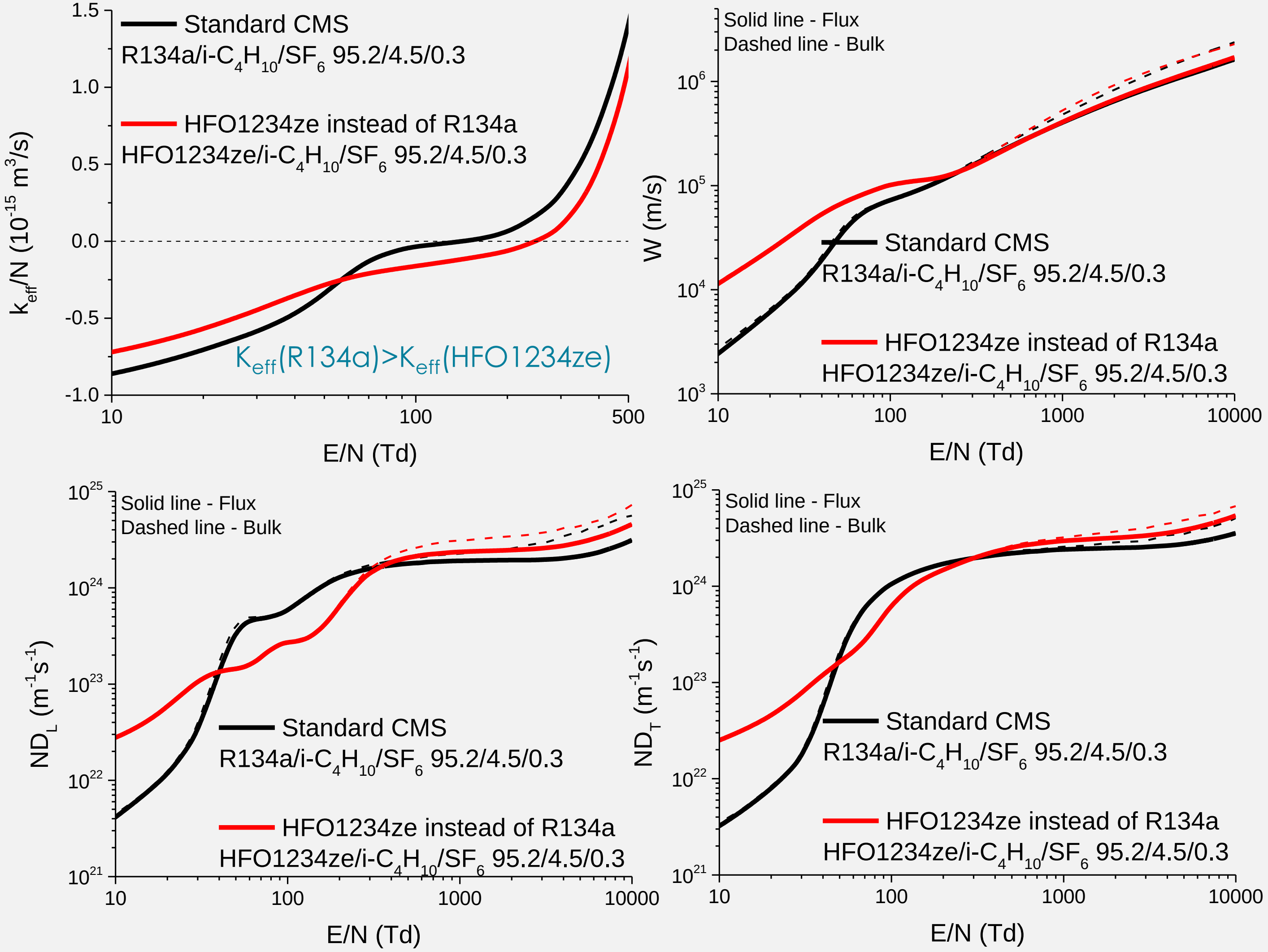
Chachereau *et al.* (2016) Plasma Sources Sci. Technol. **25** 045005

Dujko *et al.* unpublished

Electron transport in eco-friendly RPC gas mixtures

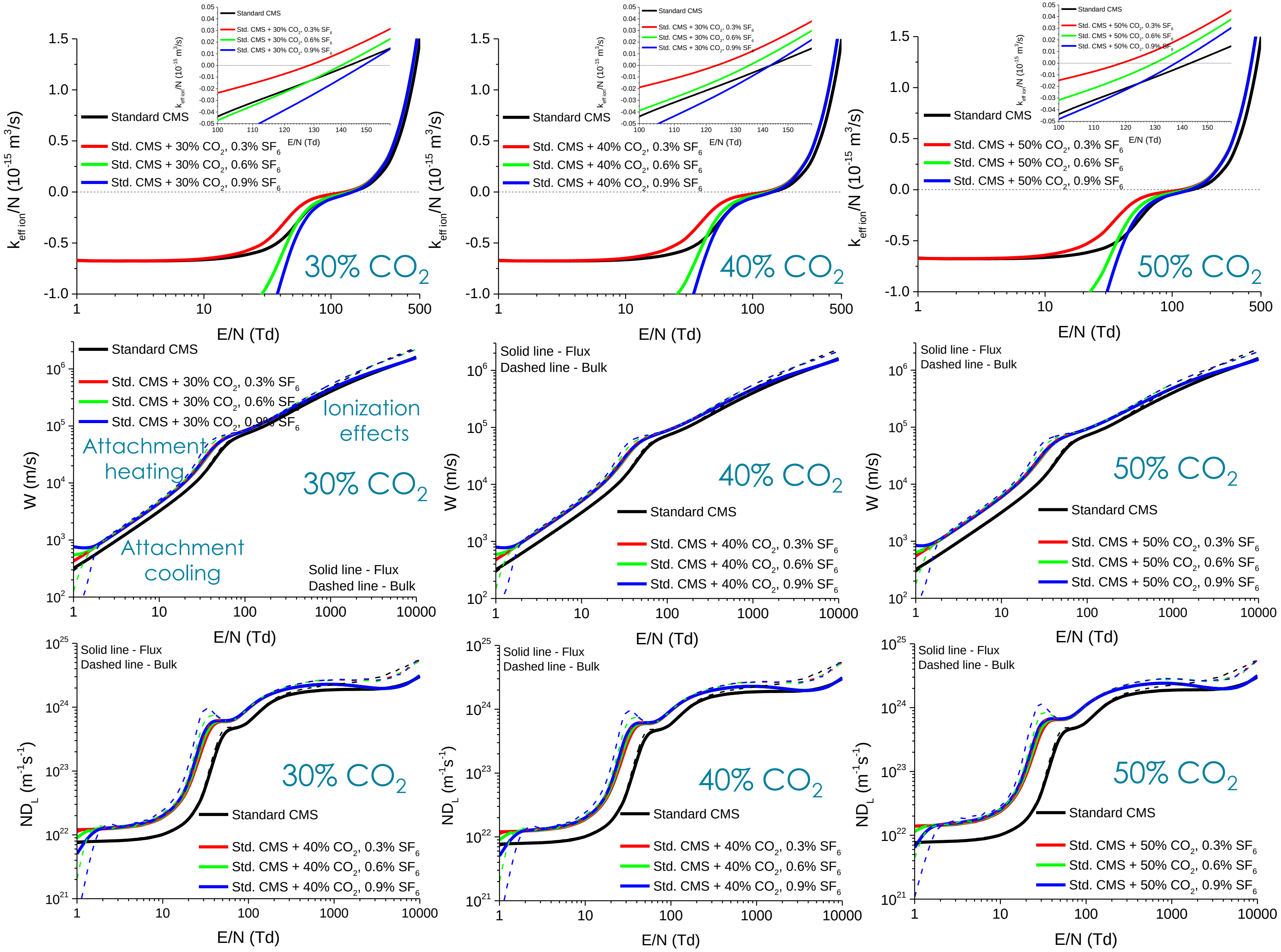
Considering the very low GWP factor for HFO1234ze, why not just replace R134a with HFO1234ze?

- Critical electric field of the standard CMS mixture is 149 Td.
- When $C_2H_2F_4$ is entirey replaced by $C_3H_2F_4$, the critical electric field is increased to 250 Td.
- Over the entire range of E/N drift velocity is higher when R134a is replaced by HFO1234ze.
- For approximatelly 30<E/N<350 Td ND_L (R134a) > ND_L (HFO1234ze).
- For approximatelly 50<E/N<300 Td ND_T (R134a) > ND_T (HFO1234ze)
- **Too high operating voltages to be compatible with the high voltage systems and readout electronics employed in the LHC experiments!**
- **HFO1234ze must be mixed with CO_2 or He!**



Dujko *et al.* unpublished

Electron transport in standard CMS mixture with the addition of CO_2



Rigoletti *et al.* (2023) *Nucl. Instrum. Meth. Phys. Res. A* **1049** (2023) 168088

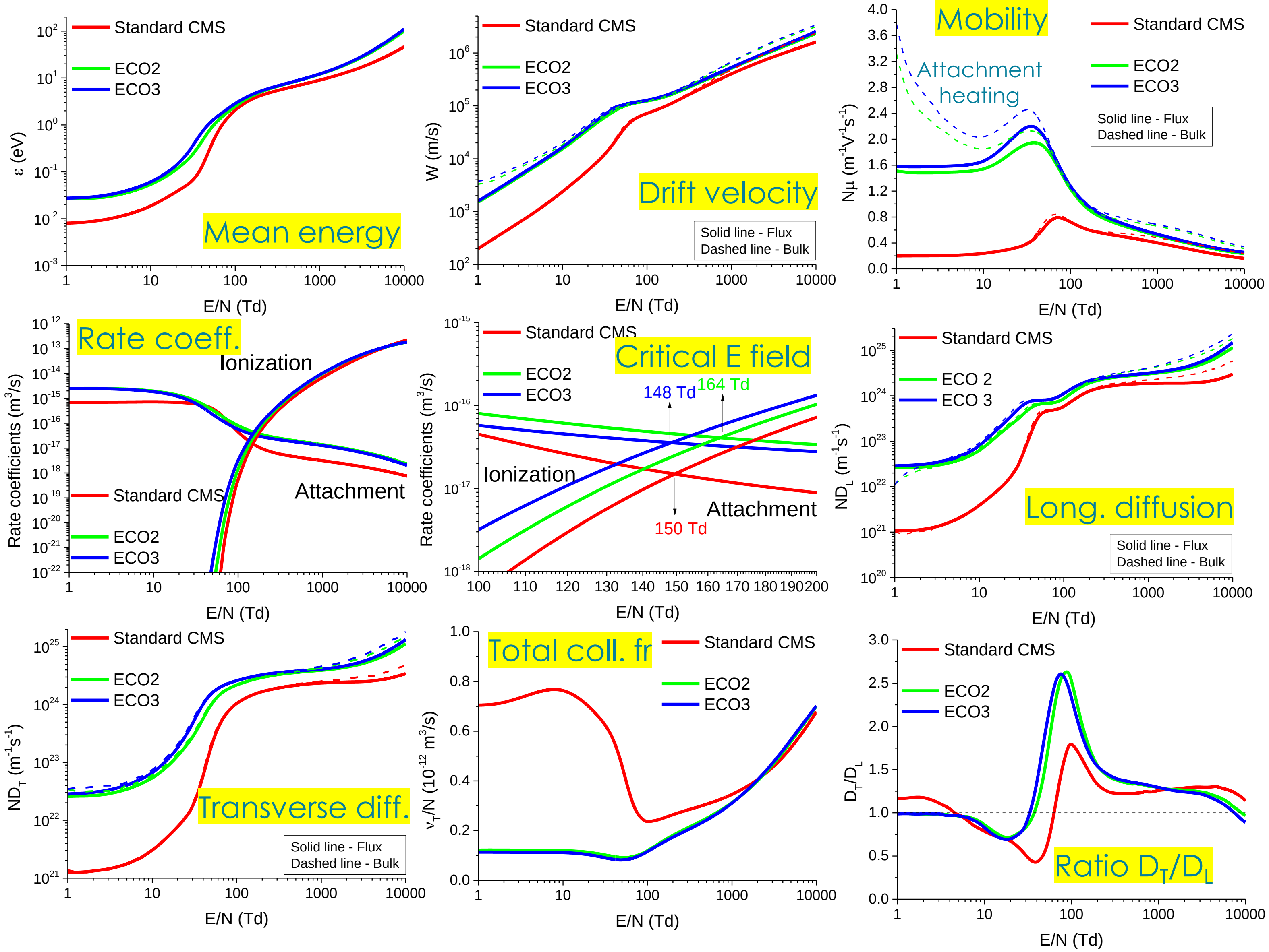
- 30%CO_2: Ecr(SF_6 0.3) < Ecr(SF_6 0.6) <Ecr(Std) < Ecr(SF_6 0.9)
- 40%CO_2: Ecr(SF_6 0.3) < Ecr(SF_6 0.6) <Ecr(Std) = Ecr(SF_6 0.9)
- 50%CO_2: Ecr(SF_6 0.3) < Ecr(SF_6 0.6) <Ecr(SF_6 0.9) < Ecr(Std)
- Bulk W and ND_L are enhanced with increasing fraction of SF_6.

↓

- Each 10% increase in CO_2 decreases the critical electric field by approximately 5 Td. If we assume the STP and a 2 mm gap, this suggests a shift of the working point by approximately 250 V.
- More CO_2 in the mixture leads to more charge release and a higher probability for the occurrence of streamers.

Dujko *et al.* unpublished

Electron transport in standard CMS, ECO2 and ECO3 mixtures



Standard CMS: R134a/i-C_4H_{10}/SF_6 95.2/4.5/0.3
ECO2: HFO1234ze/CO_2/ /i-C_4H_{10}/SF_6 35/60/4/1
ECO3: HFO1234ze/CO_2/ /i-C_4H_{10}/SF_6 29/65/5/1

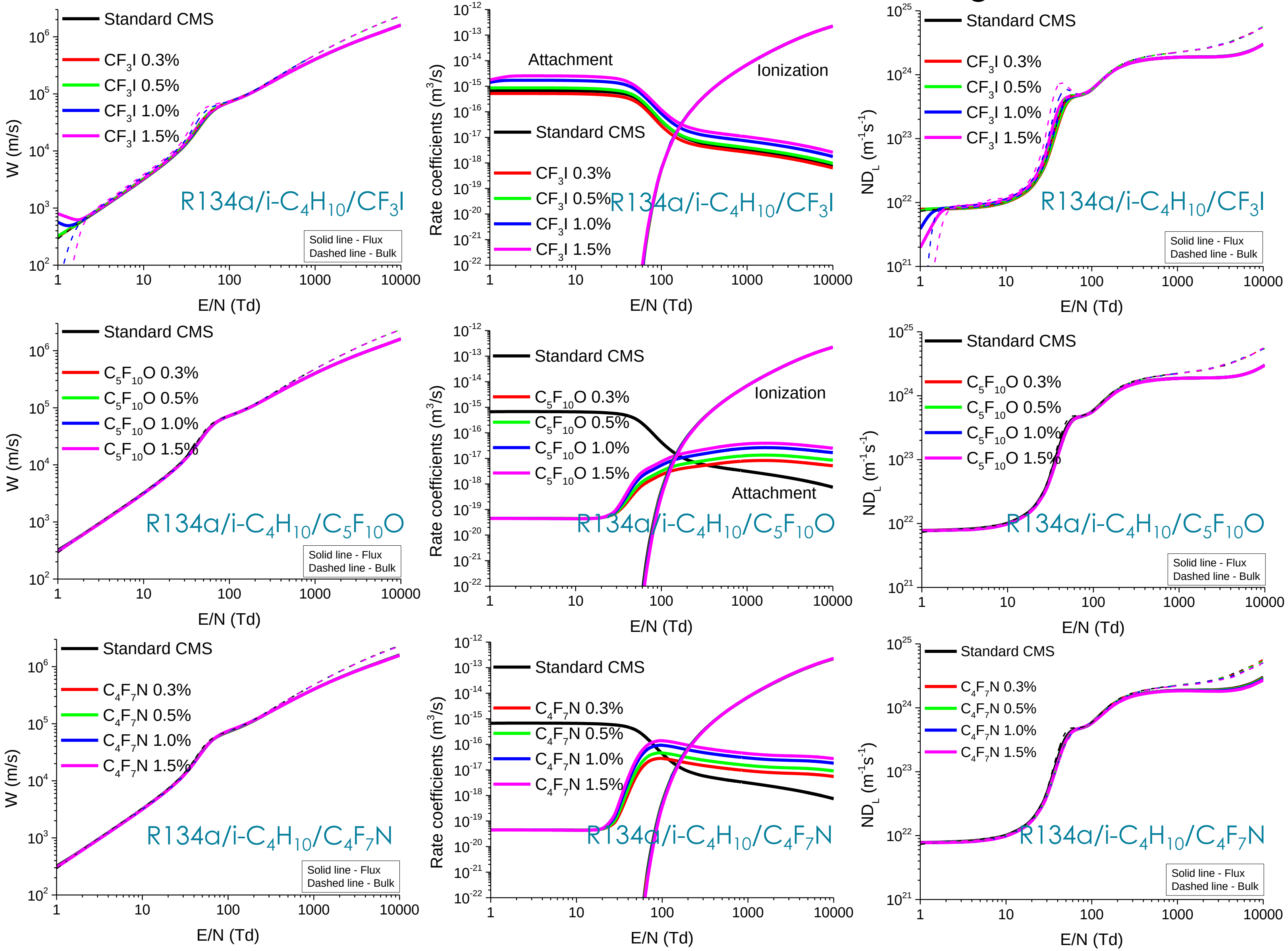
Abbrescia *et al.* (2024) *Eur. Phys. J. C* (2024) 84:300

- Attchment heating and explicit effects of ionization are evident
- Drift and diffusion are enhanced in ECO2 and ECO3 mixtures
- Surprisingly, the critical electric field for the ECO3 mixture is slightly lower than that of the standard CMS
- Anisotropic nature of the diffusion tensor is more pronounced for ECO2 and ECO3 mixtures

- Faster transition from an avalanche into a streamer
- Stronger field enhancment, more liberated charge, larger streamer velocity, stronger signals in RPCs

Dujko *et al.* unpublished

Possible alternatives to SF_6



Critical electric fields

Standard CMS mixture, Ecr = 149 Td

CF_3I 0.3%, Ecr = 142 Td
CF_3I 0.5%, Ecr = 150 Td
CF_3I 1.0%, Ecr = 162 Td
CF_3I 1.5%, Ecr = 171 Td

Similar to std cms

$C_5F_{10}O$ 0.3%, Ecr = 121 Td
$C_5F_{10}O$ 0.5%, Ecr = 128 Td
$C_5F_{10}O$ 1.0%, Ecr = 141 Td
$C_5F_{10}O$ 1.5%, Ecr = 152 Td

Similar to std cms

C_4F_7N 0.3%, Ecr = 159 Td
C_4F_7N 0.5%, Ecr = 173 Td
C_4F_7N 1.0%, Ecr = 197 Td
C_4F_7N 1.5%, Ecr = 215 Td

Higher working point!

Gas	Pro	Cons
CF_3I	Low GWP <1, High Ecr, 437 Td	Toxic
$C_5F_{10}O$	Low GWP <1, High Ecr, 765 Td	Boiling point 27°C
C_4F_7N	High Ecr, 972 Td	High GWP ~2200, Toxic

Dujko *et al*. unpublished

Fluid modelling of streamer discharges in eco-friendly RPC gas mixtures

Classical fluid model

- Advection diffusion reaction equation for the time evolution of the number density of electrons:
  - $\frac{\partial n_e}{\partial t} + \nabla(n_e \boldsymbol{W} - \boldsymbol{D}\nabla n_e) = n_e(\alpha - \eta)|\boldsymbol{W}| + S_{ph}$
- Reaction equations for the time evolution of the number densities of ions:
  - $\frac{\partial n_p}{\partial t} = n_e \alpha |\boldsymbol{W}| + S_{ph}$ $\frac{\partial n_n}{\partial t} = n_e \eta |\boldsymbol{W}|$
- Local field approximation
- Total electric field:
  - $\mathbf{E} = \boldsymbol{E}_{applied} - \nabla\Phi_{space\_charge}$ $\Delta\Phi_{space\_charge} = -q_e \frac{n_p - n_e - n_n}{\varepsilon_0}$
- Photoionization model is implemented for N_2-O_2 mixture using the Zheleznyak model.

Dujko *et al.* (2013) J. Phys. D: Appl. Phys. **46** 475202
Simonović *et al.* (2024) *Plasma Sources Sci. Technol.* **33** 085012 (19pp)

Numerical solution in the AMReX library

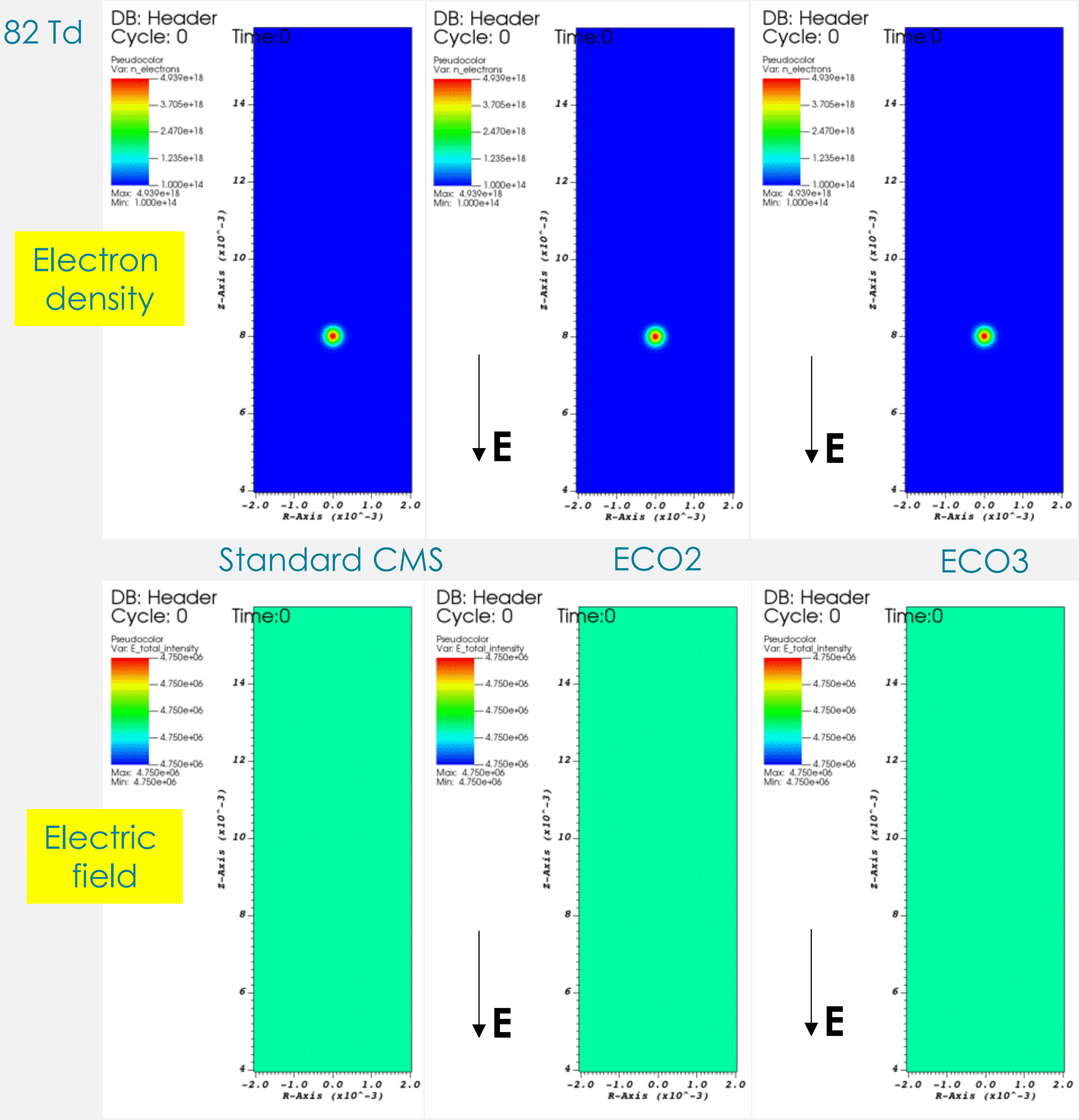
- Spatial discretization: Finite volume method
- Scalar variables are defined at cell centers, while vector variables are defined at cell faces
- Flux limiting schemes are employed to interpolate electron density from the cell centers to the cell faces to calculate the flux of electrons
- Time integration: 2nd order Runge-Kutta
- Time step restriction criteria: the CFL condition, the dielectric relaxation time and the time step restriction due to rates of nonconservative processes

- AMRex - An open-source C++ library for massively parallel, block-structured adaptive mesh refinement (AMR) applications
- AMReX includes inbuilt geometric multigrid solvers for the Poisson equation and the Helmholtz equations
- Allows both MPI and OpenMP parallelization, as well as parallelization on graphic processing units
- Adaptive mesh refinement is applied to correctly describe streamer dynamics at the streamer front

Simonović *et al.* (2024) *Plasma Sources Sci. Technol.* **33** 085012 (19pp)

Two-headed streamers in RPC’s mixtures

- Positive streamer starts slower than the negative streamer, but later (after a few ns, depending on E/N) it quickly makes up for this.
- Positive streamer is narrower and therefore its field enhancement is larger than in the negative streamer.
- Negative streamers have a larger radius. Streamer radius is a complicated function of time.
- In eco-friendly mixtures there is a faster transition from an avalanche into a streamer, a larger amount of charge is released, with a more intense field amplification at the front of the streamer. These properties become more obvious with increasing externally applied electric fields.



E/N = 182 Td

Electron density

Electric field

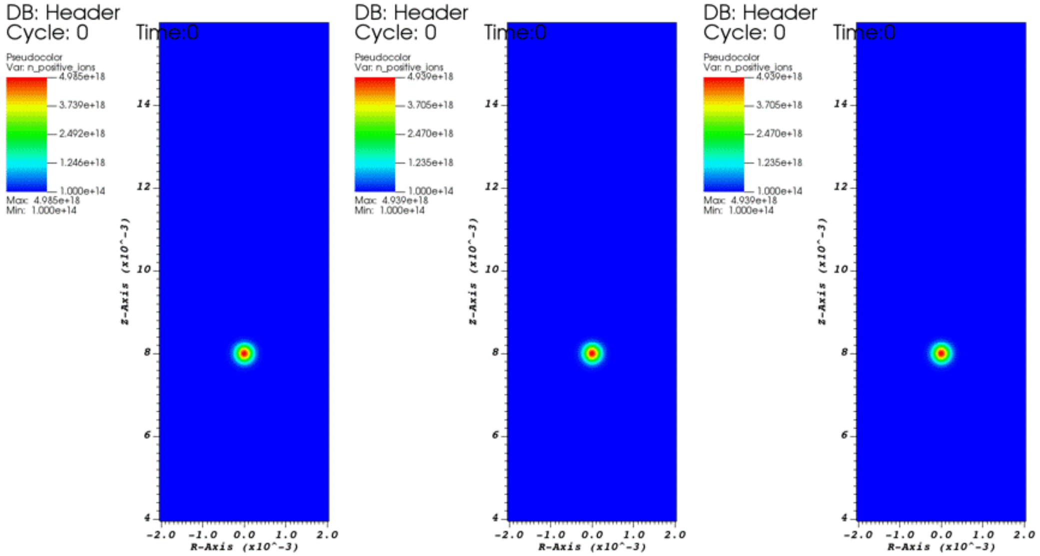
Standard CMS

ECO2

ECO3

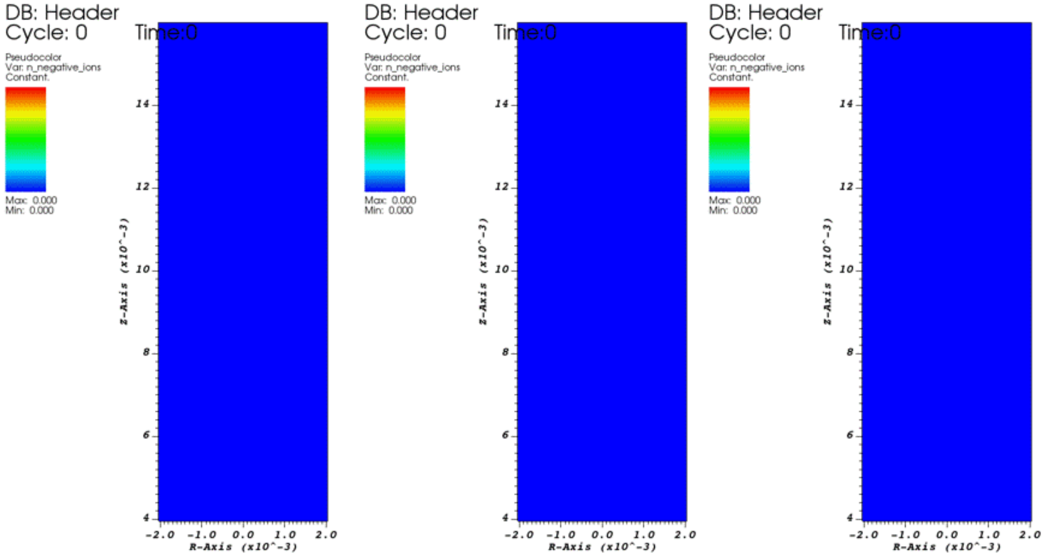
Dujko *et al.* unpublished

Positive ions

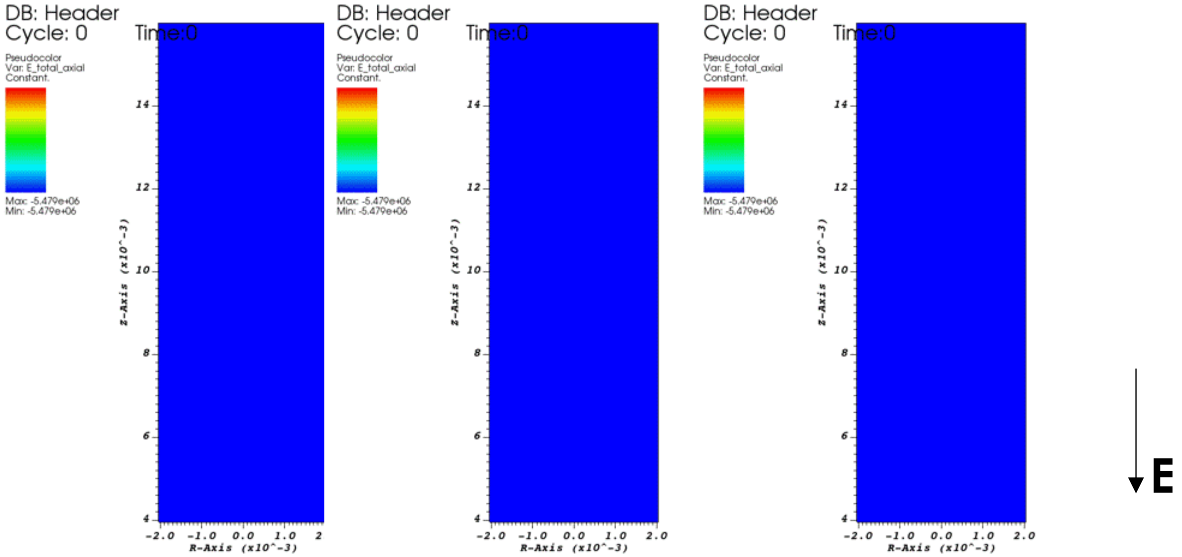


Negative ions

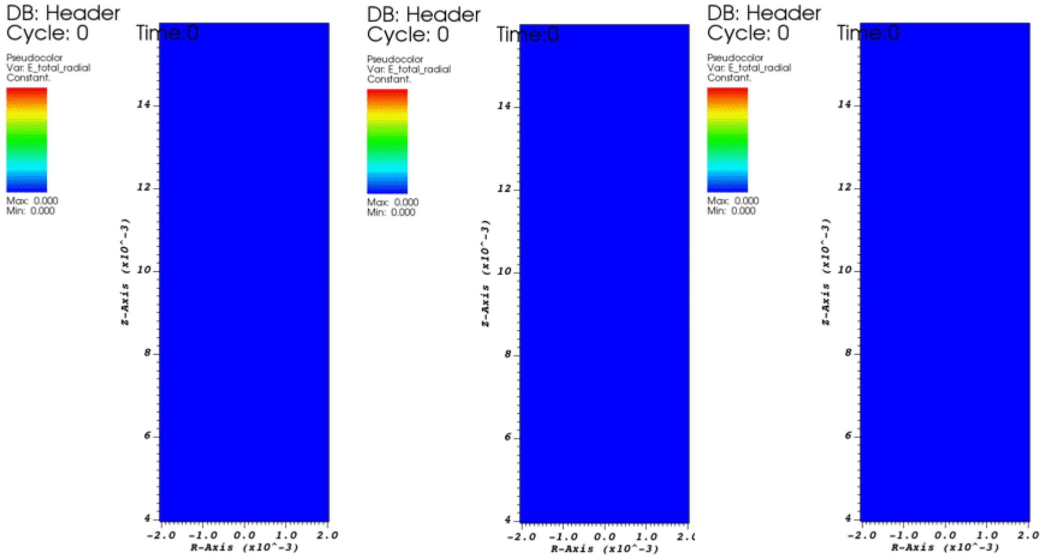
Dujko *et al.* unpublished



Axial electric field



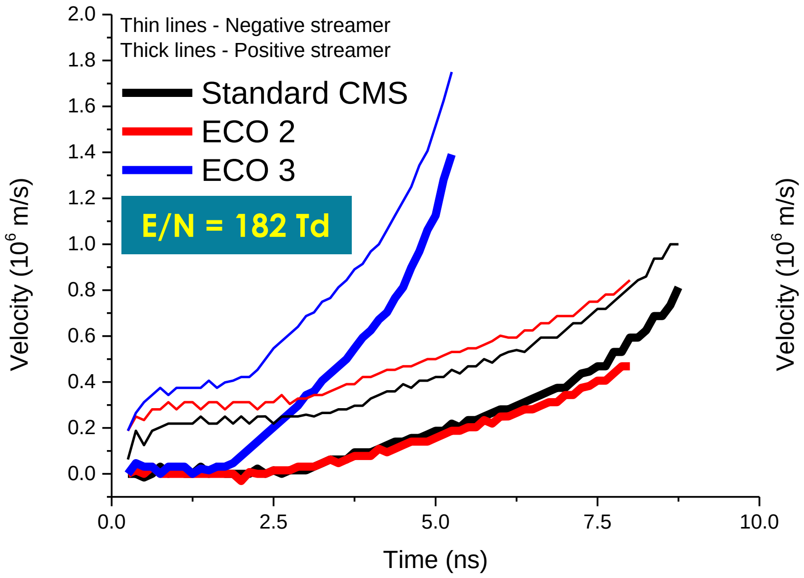
Radial electric field

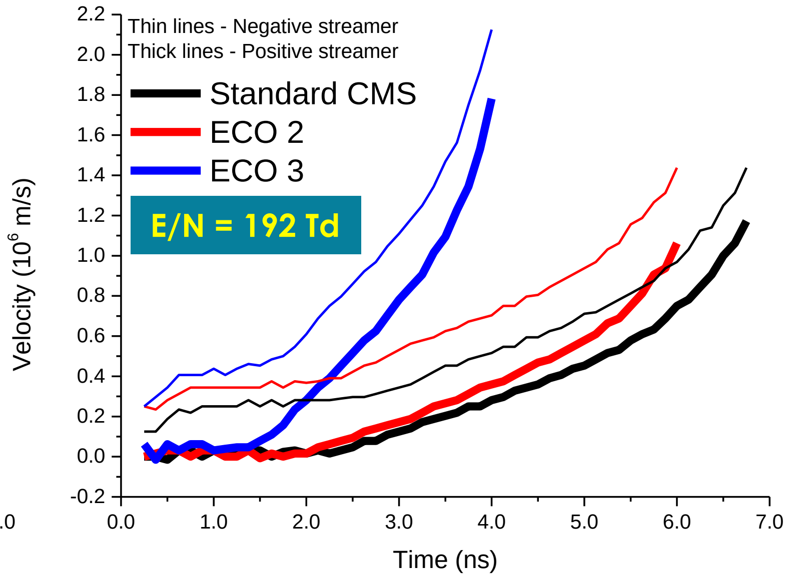


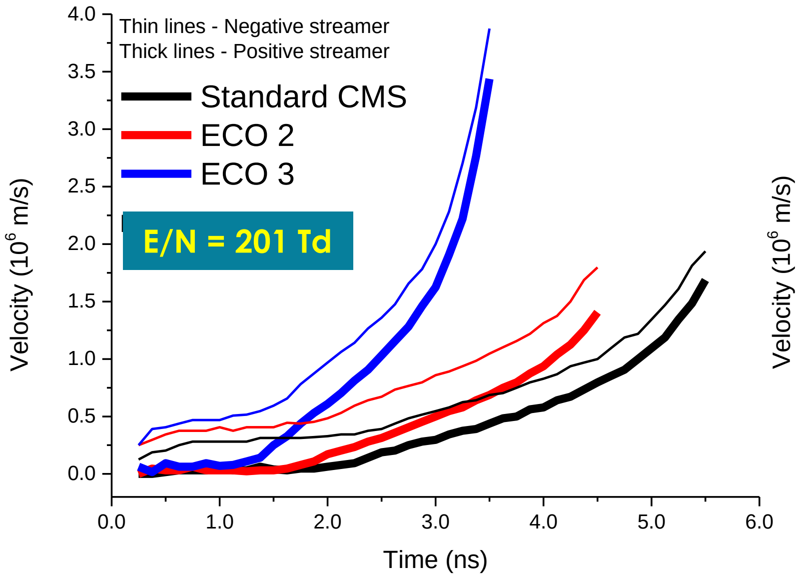
Standard CMS ECO2 ECO3

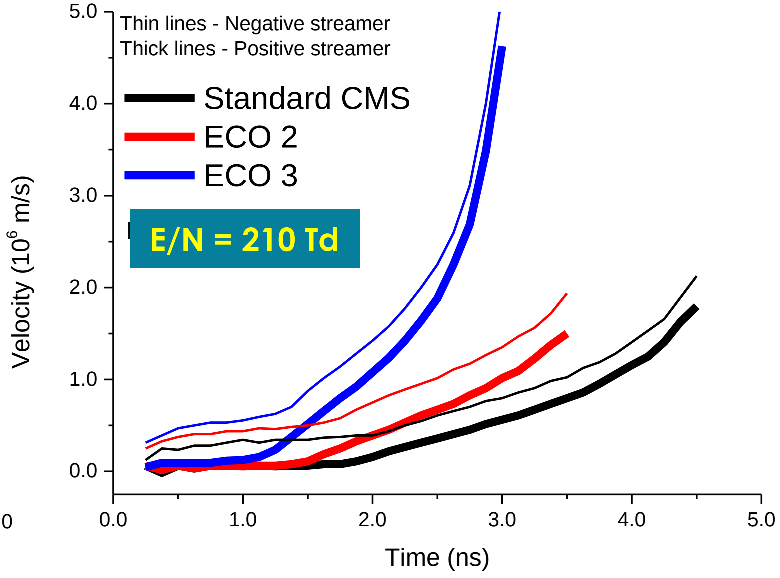
Standard CMS ECO2 ECO3

Streamer velocity in the standard CMS, ECO2 and ECO3 mixtures









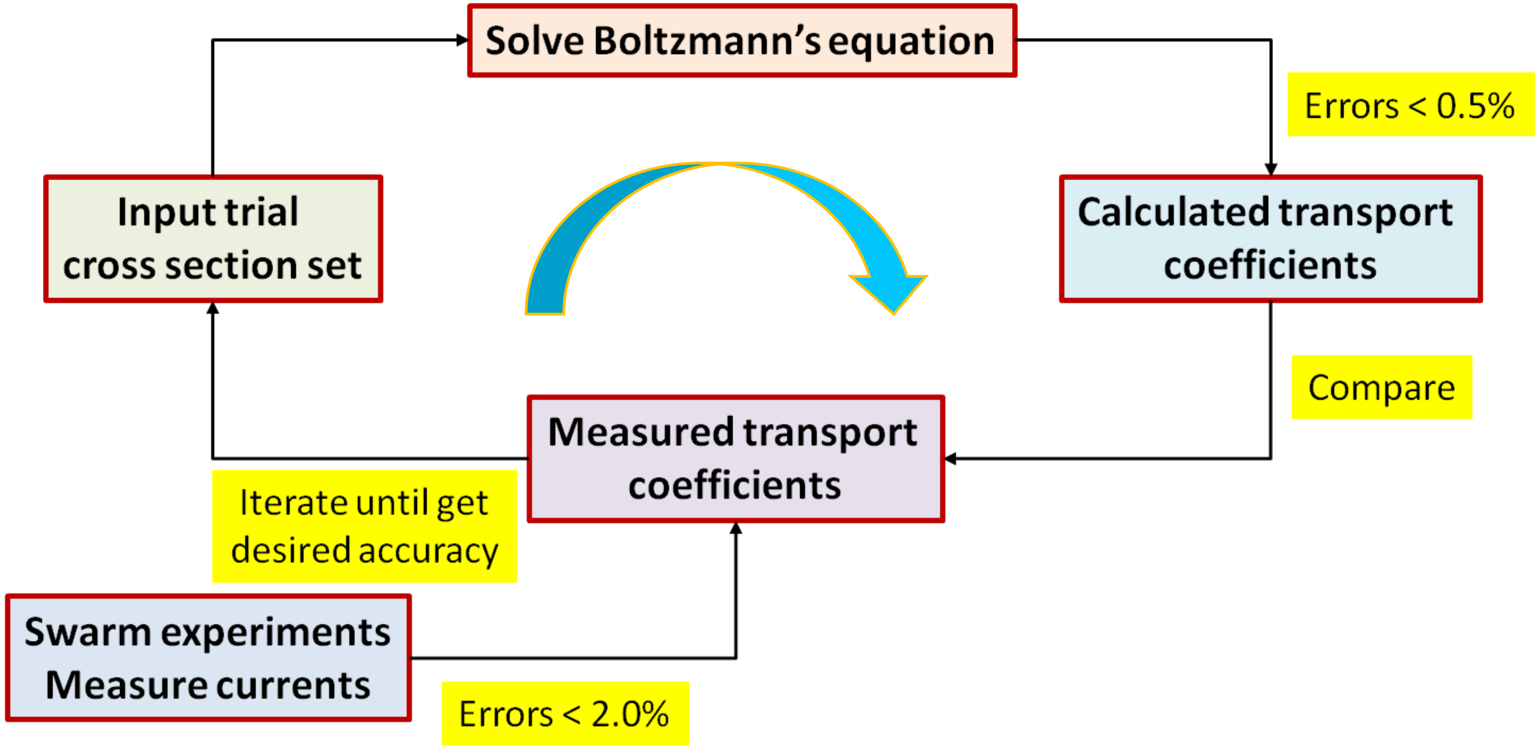
- Streamers accelerate after transitioning from an avalanche into a two-headed streamer
- Streamer velocity is higher than the electron drift velocity
- Negative streamers are faster than positive streamers
- Streamers in ECO3 mixture are the fastests
- Only for the lowest E/N of 182 Td, positive streamers in ECO2 mixture are slightly slower than those in the standard CMS mixture

Dujko *et al*. unpublished

Concluding remarks

- We have extended and generalized our multi-term theory for solving the BE and our MC code to study the transport of electrons in eco-friendly RPC gas mixtures.
- We have developed complete and consistent sets of cross sections for electron scattering in $C_2H_2F_4$, C_3HF_5, $C_3H_2F_4$, CF_3I, $C_5F_{10}O$, and C_4F_7N.
- Knowledge of cross sections and transport coefficients for electrons is of key importance in future modelling studies of RPCs and experimental measurements of time resolution, efficiency, charge spectra, etc.
- Concepts of attachment heating, attachment cooling, and the implicit and explicit effects of non-conservative collisions on drift and diffusion play a very important role in understanding transport coefficient duality.
- We have implemented the classical fluid model within the AMReX software environment. The numerical integrity of the code is verified in several benchmark calculations, with and without photoionization.
- On the time scale of a few ns, negative streamers are faster in the standard CMS, ECO2 and ECO3 mixtures.
- The field enhancement at the streamer front is stronger for positive streamers, while the streamer radius is larger for negative streamers.
- Transition from an avalanche into a streamer occurs faster for the eco-friendly gas mixtures.

Additional slide: What can swarms bring to the modelling of gaseous particle detectors?



Advantages:

- Completeness
- Absolute cross sections
- Direct applicability to model plasmas and particle detectors

Disadvantages:

- Non-uniqnuess
- Limited resolution
- Complexity and indirect nature of procedure

What has been done?

- Normalized sets: NO, N_2O, HBr, CF_4, …
- New sets: $C_2H_2F_4$, $C_3H_2F_4$, C_3HF_5, …

- **Kinetic models:** overcome/assess currently used approximations such as the TTA, effective field approximation, …
- **Fluid models:** provision of accurate swarm data, correct implementation of swarm data, information on non-local effects (temporal and spatial).