Development of EM models to describe ionisation in the atmosphere

Geant4 Collaboration Meeting 2024, Catania, Italy

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7-11/10/24

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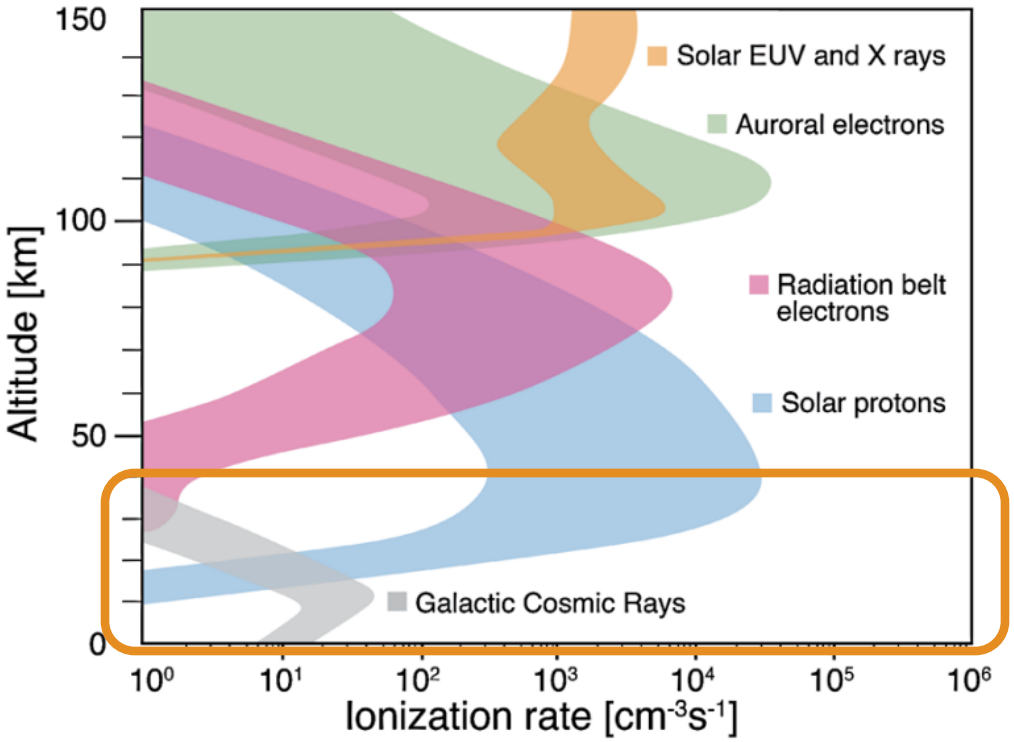
Motivation

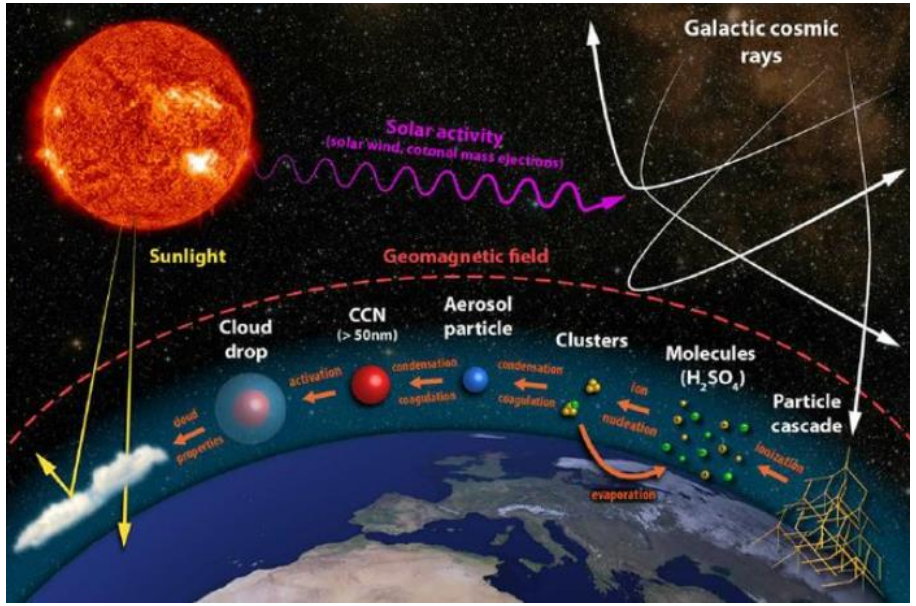
The impact of cosmic rays and ions on atmospheric chemistry and climate evolution is still poorly understood:

- ions affect reaction rates (**up to 10 orders of magnitude**) and types of reactions depending on the ionization state.



aerosol formation
(Shumilov et al. 1996
Mironova et al. 2005; Kazil et al. 2006)

precipitation
(Kniveton 2004)



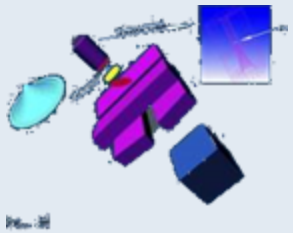
ion-induced nucleation
(Svensmark et al. 2007)

cloud cover
(Voiculescu et al. 2006)

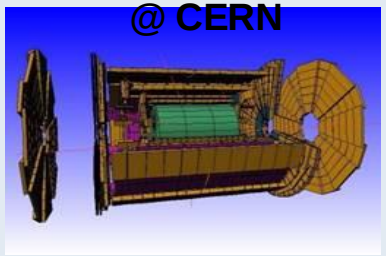
Monte Carlo for studying induced ionization

- Most of the simulation useful for the purpose are based on MC codes such as Geant4 (Oulu CRAC:CRII , ATMOCOSMIC, AtRIS, RUSCOSMICS, ..)

Medical linac

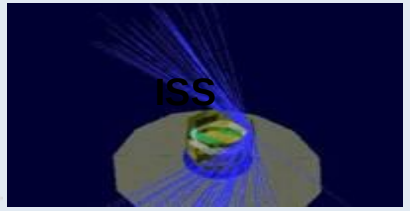


ATLAS, CMS, LHCb, ALICE @ CERN



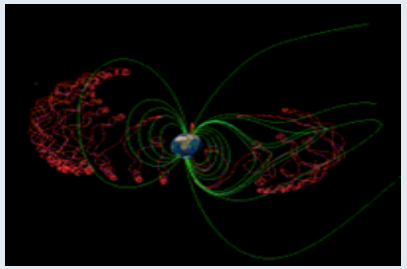
Geant4

GAIA

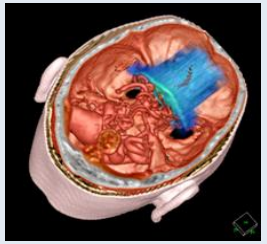


C++ toolkit for Monte Carlo simulation of hadronic and electromagnetic interaction of particle/radiation with matter

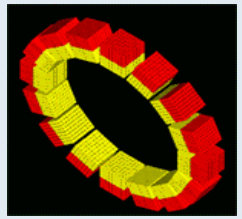
Earth magnetosphere



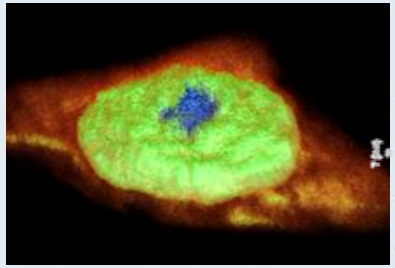
DICOM dosimetry

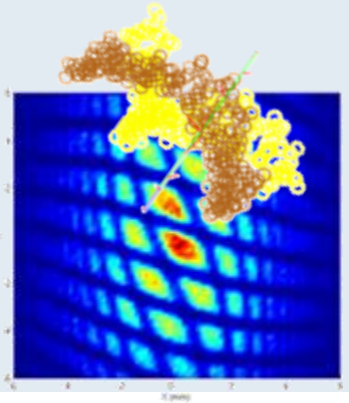


PET Scan (GATE)

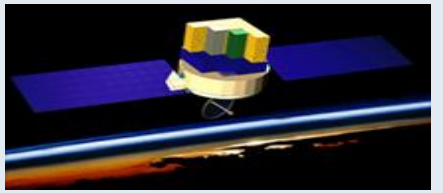


Geant4-DNA





GLAST/FERMI (NASA)



Monte Carlo for studying induced ionization

OUTPUT

Current State-of-the-Art approach:

- Condensed history
- Limited low-energy applicability
- No description of target molecular properties

- **Average number of ions pair $[cm^{-3}s^{-1}]$ (no ions description)**

A new approach with Geant4-DNA:

- Step-by-step modelling
- Description of target molecular properties
- Can simulate diffusion, dissociation, chemical reactions

- **Number of ions,**
- **Ionization states,**
- **Excitation states,**
- **Spatial distribution,**
- **Number of species produced**

Required input for chemistry models

Geant4-DNA for atmosphere

Goal

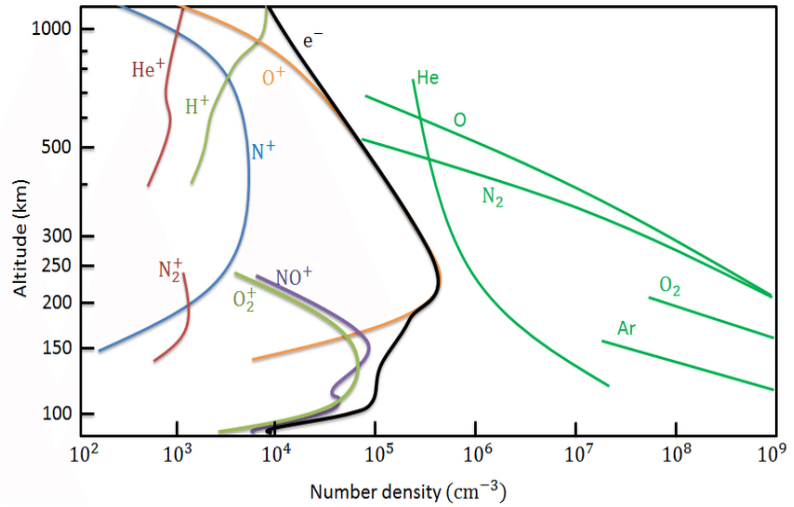
Accurately describe ions produced by CR interaction in the atmosphere

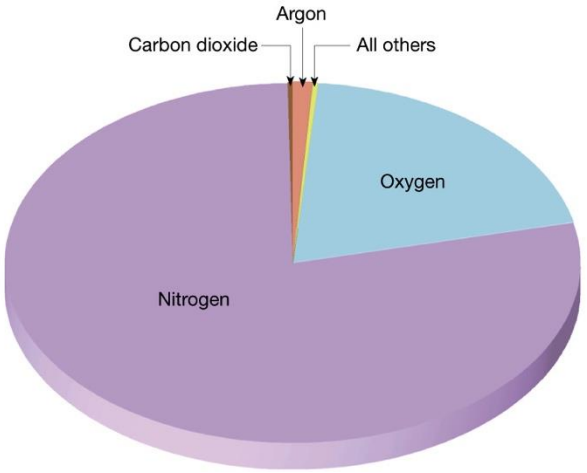
How?

Including in Geant4-DNA new models for particle-impact interactions with relevant molecules for climatology

Starting point

- Primary particles: electrons
- Target molecules: N_2, O_2





N_2: 78,084%
O_2: 20.946%
Ar: 0.934%
CO_2: 0.037%
…

N_2 and O_2 implementation in Geant4-DNA (10eV-10MeV)

[MODELS ARE DESCRIBED IN THE G4AtmXXXModel.cc .hh CLASSES]

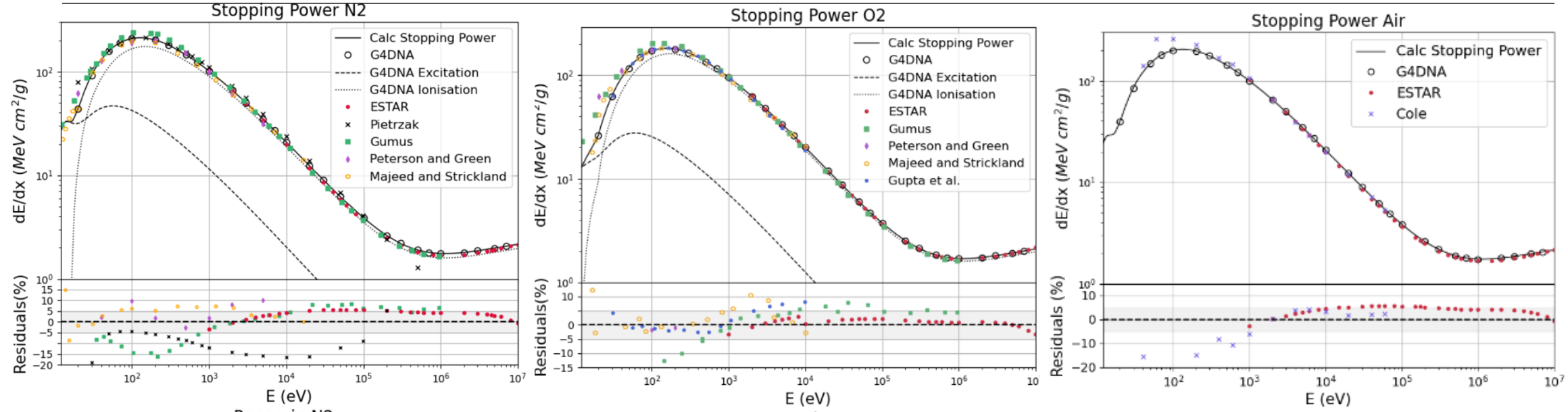
3 new model classes, 2 new model structure classes

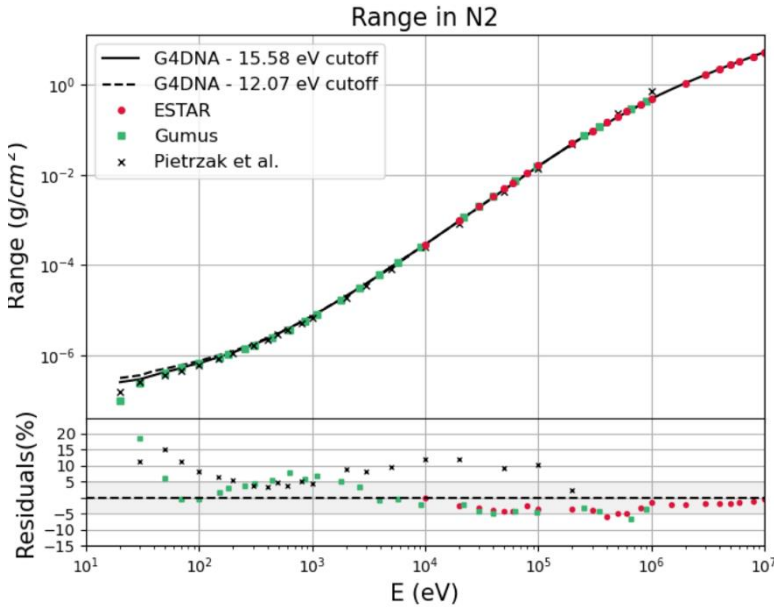
Designed to handle a mix of materials

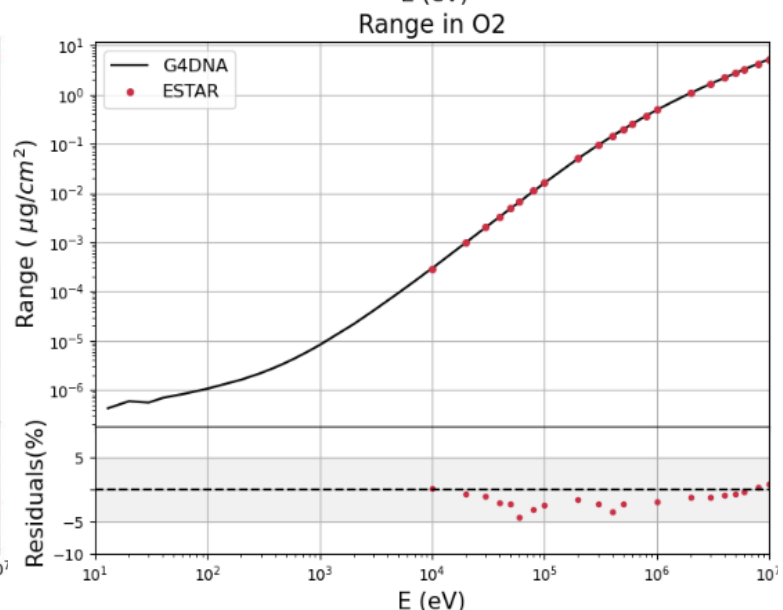
Interaction type	Cross section type	Model name	Implementation type
Impact ionisation	Total and differential (production energy of secondaries) - including partial cross sections for 6 subshells for O2, 5 for N2,	RBEB (except for k-shells ionisation which used the averaged RBEB)	Analytical
Elastic scattering	Total and differential (scattering angle)	SCAR (Screening Corrected Additivity Rule) with optimized free potential parameters	Data tables: sigmadiff_cumulated_elastic_e-_N2_atm.dat sigma_elastic_e-_N2_atm.dat
Electronic excitation	Total - 27 states for O2, 32 states for N2, Autoionization process is included	Formulas based on Porter et al, fitted cross section parameters	Data tables: sigma_excitation_e_N2_atm.dat

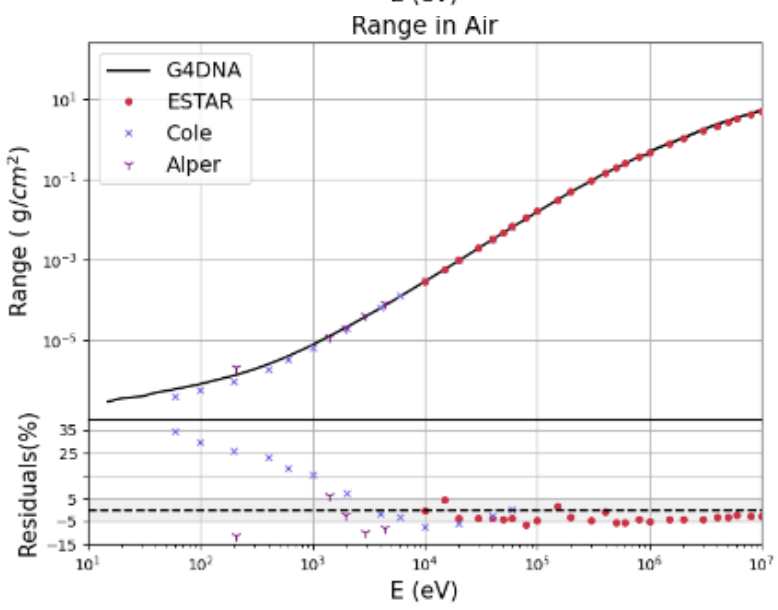
Nicolanti F, et al. . Calculation of electron interaction models in N2 and O2. In: Phys Med.
F . Nicolanti, et al. Geant4-DNA development for atmospheric applications: N2, O2 and CO2 models Implementation (Accepted by Phys Med. 2024)

Stopping power and Range validation

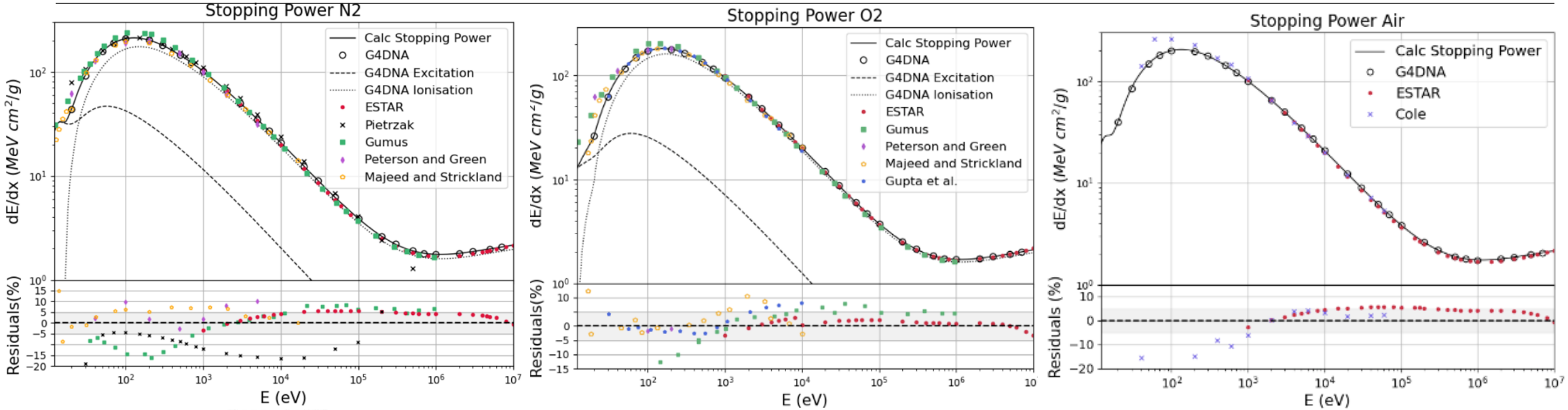


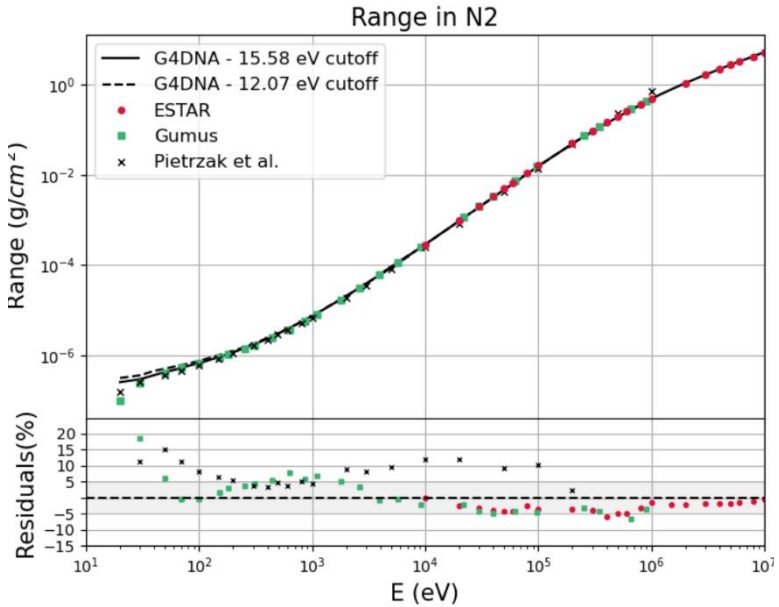


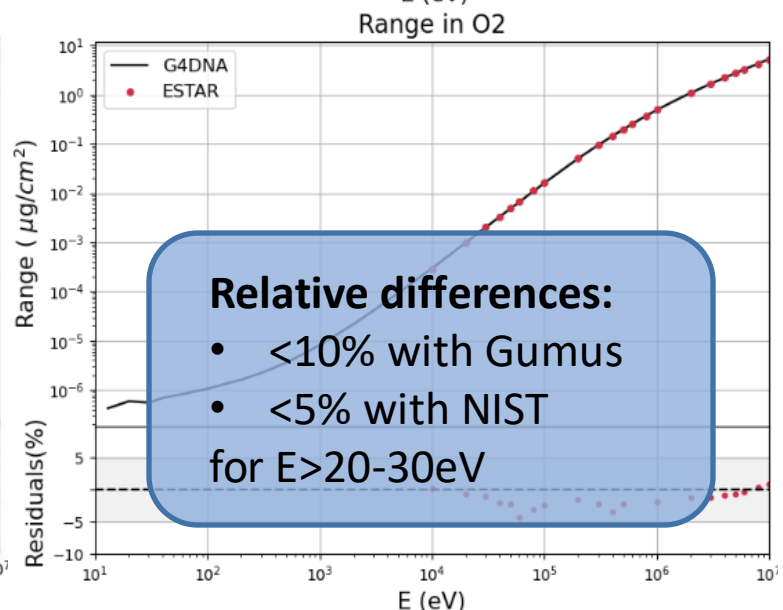


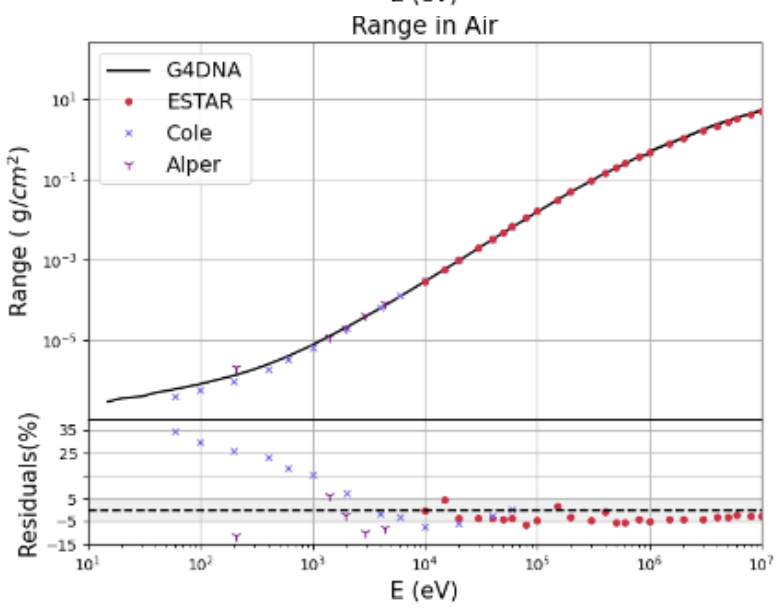


Stopping power and Range validation









Relative differences:

- <10% with Gumus
- <5% with NIST

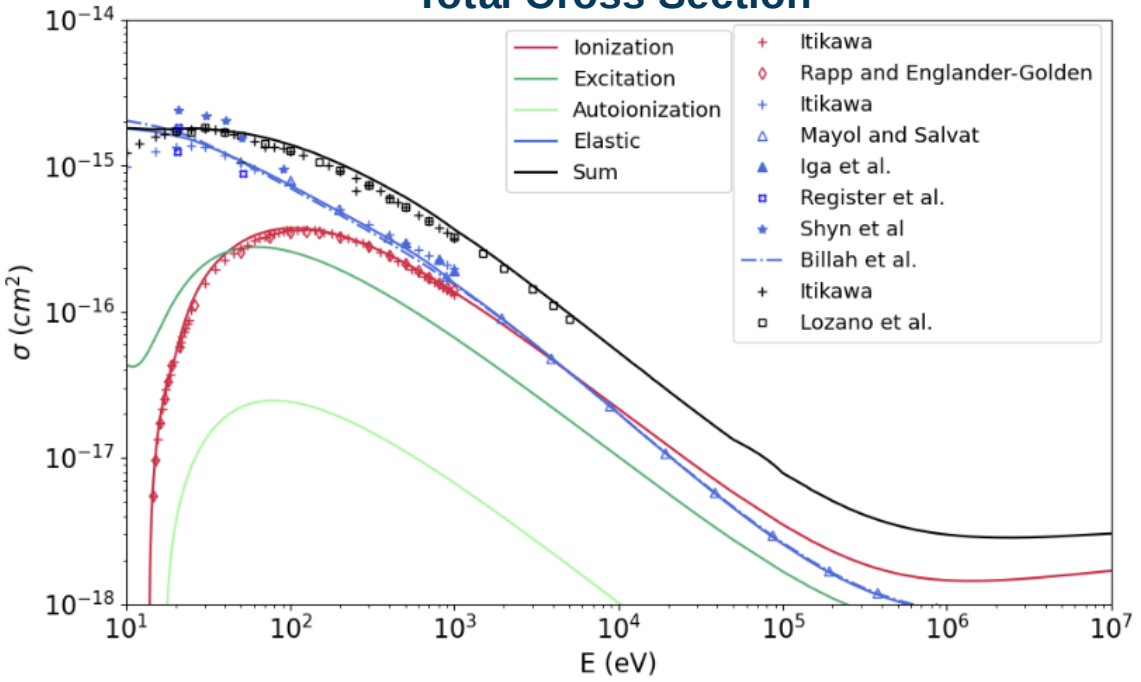
for E>20-30eV

CO_2 molecule

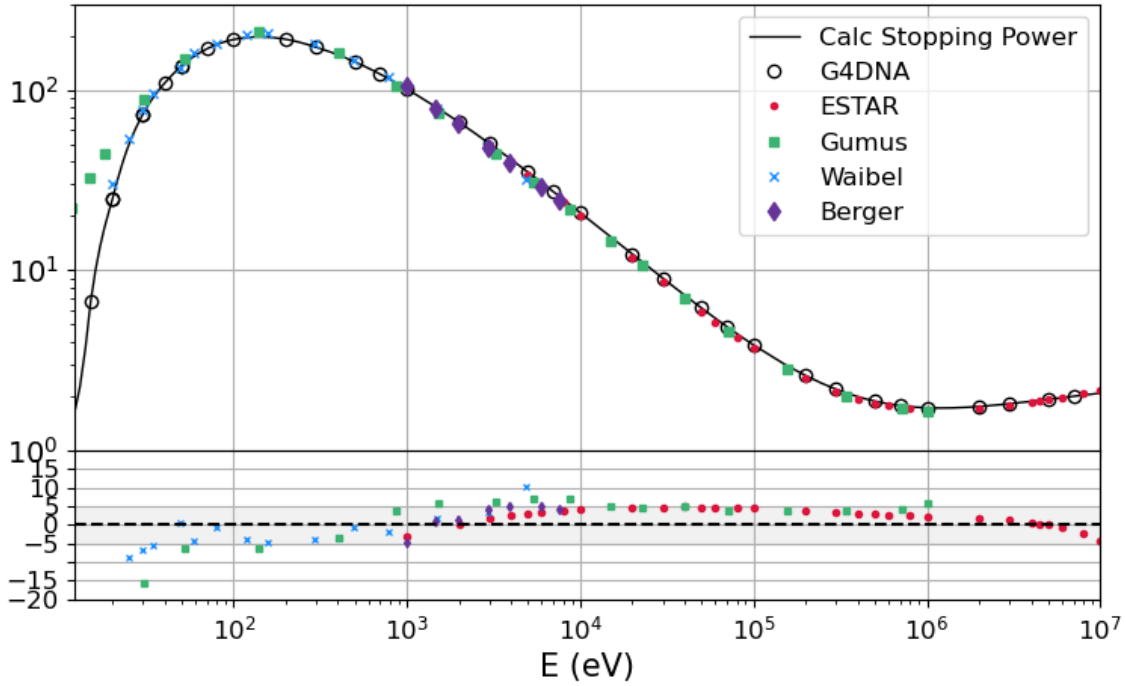
Recently added CO_2 molecule:
(Air composition fraction: 0.04%)

- Elastic: TCS and SDCS
- Ionization: TCS and SDCS
- Excitation: TCS for 15 excitation states (corrections applied to Porter's CS as described in [1], auto-ionization included)

Total Cross Section



Stopping power



New **data files:**

- sigmadiff_cumulated_elastic_e-_CO2_atm.dat
- sigma_elastic_e-_CO2_atm.dat
- sigma_excitation_e_CO2_atm.dat

Cosmic Rays simulation

To study the impact of primary cosmics on atmospheric ionisation a two-steps simulation was developed:

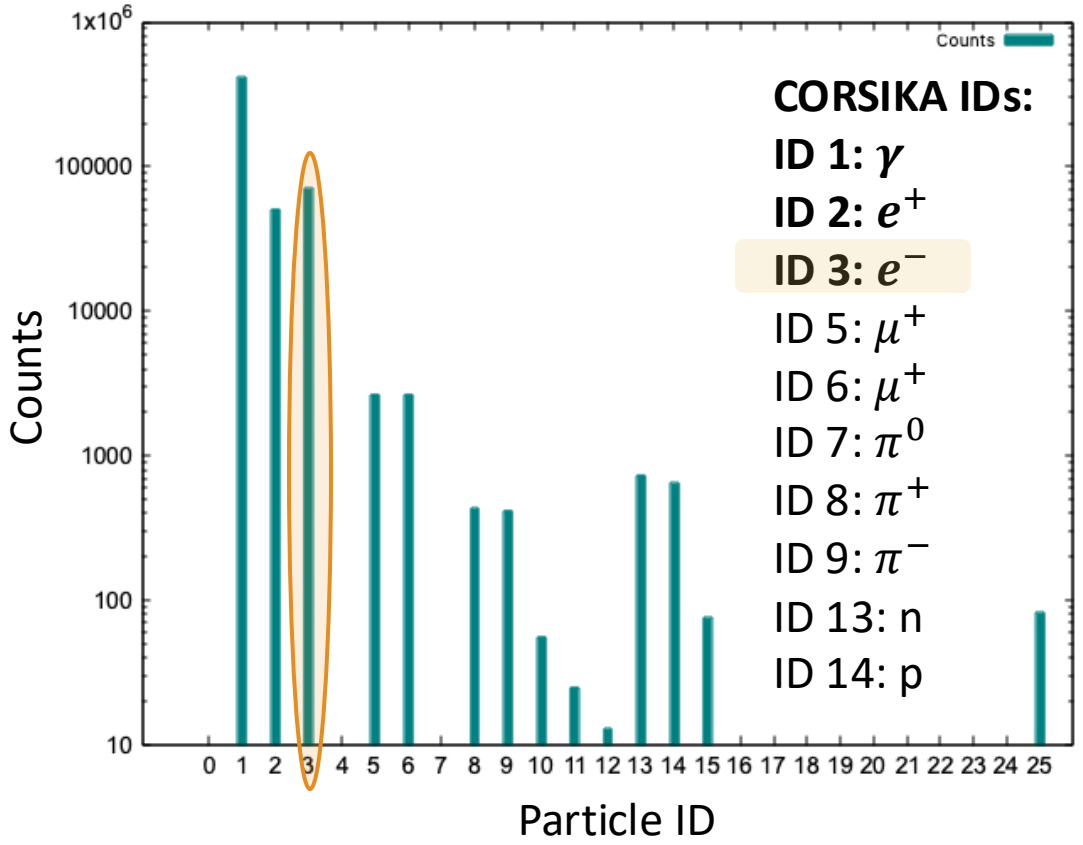
CORSIKA code:

- for simulating CR spectra at 10 km altitude (primary protons energy: 10E14 - 10E16 eV)

A Geant4 simulation:

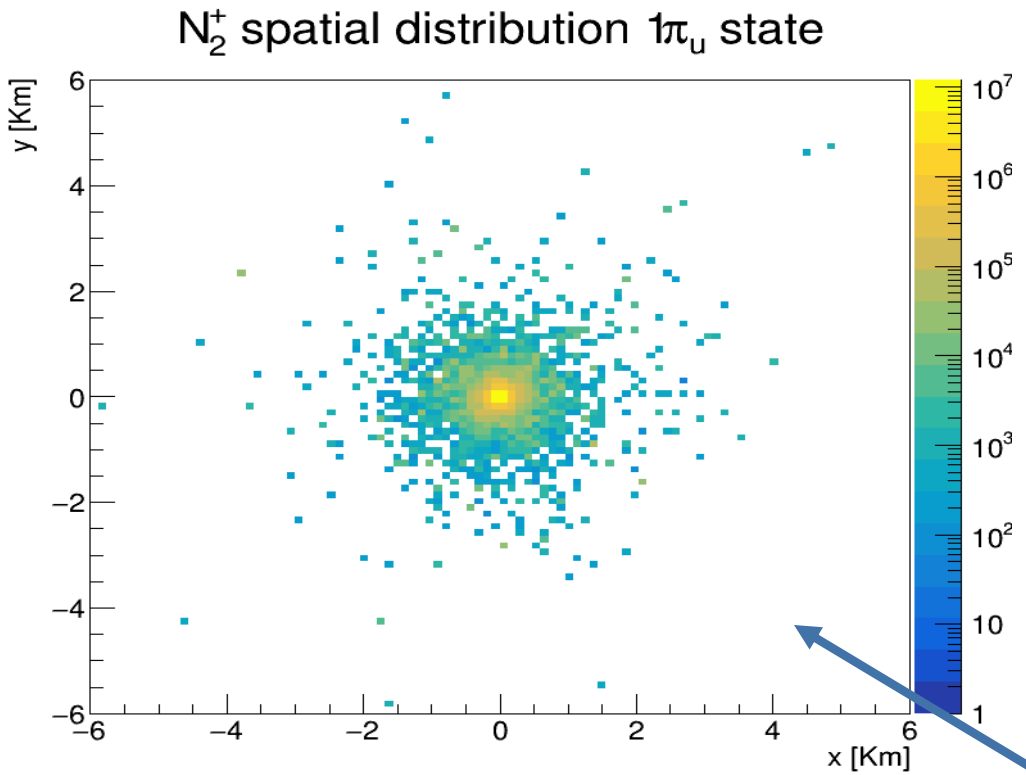
- **Primaries generator:**
  - A custom thread-safe primary generator was implemented for importing CORSIKA output
- **Combined Physics:**
  - emstandard_opt4
  - G4AtmDNAPhysics for $e-$ (E<10MeV)
- uniform **magnetic and electric field** are incorporated into the simulation

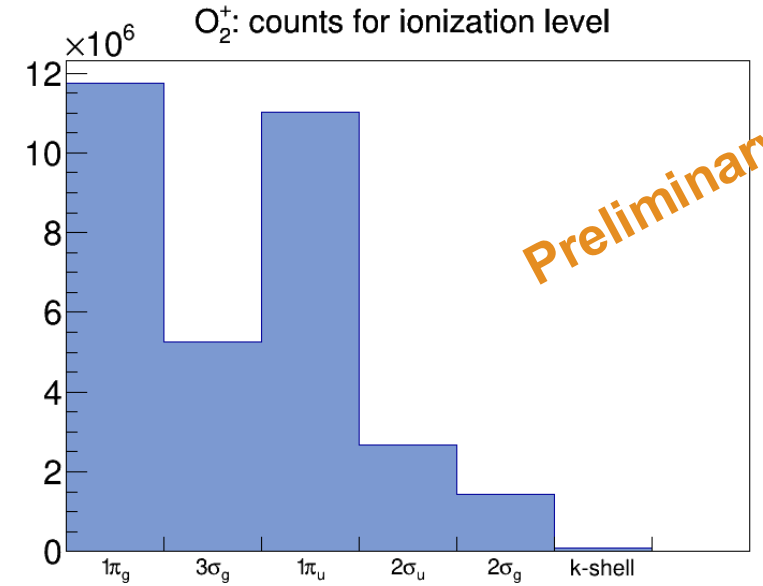
Average particles composition at 10Km from 100 10^{14} eV proton:



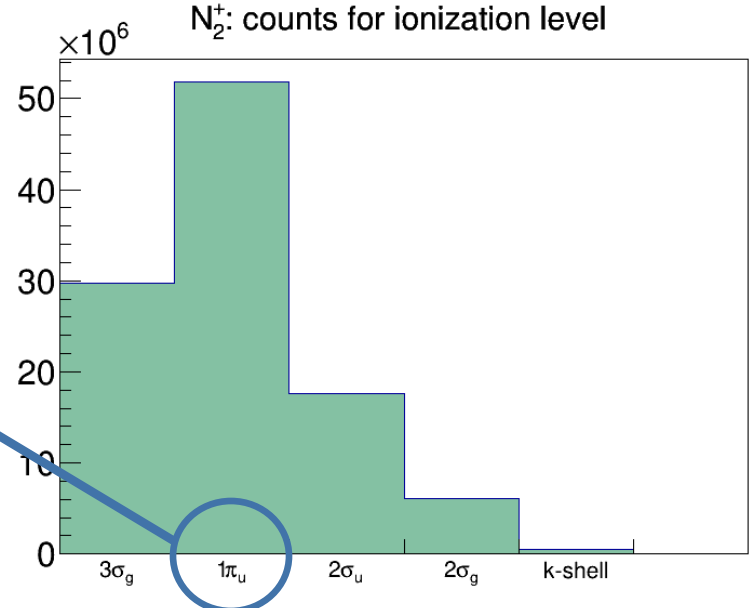
Cosmic Rays simulation

What an output from one 10^{14} eV proton looks like:





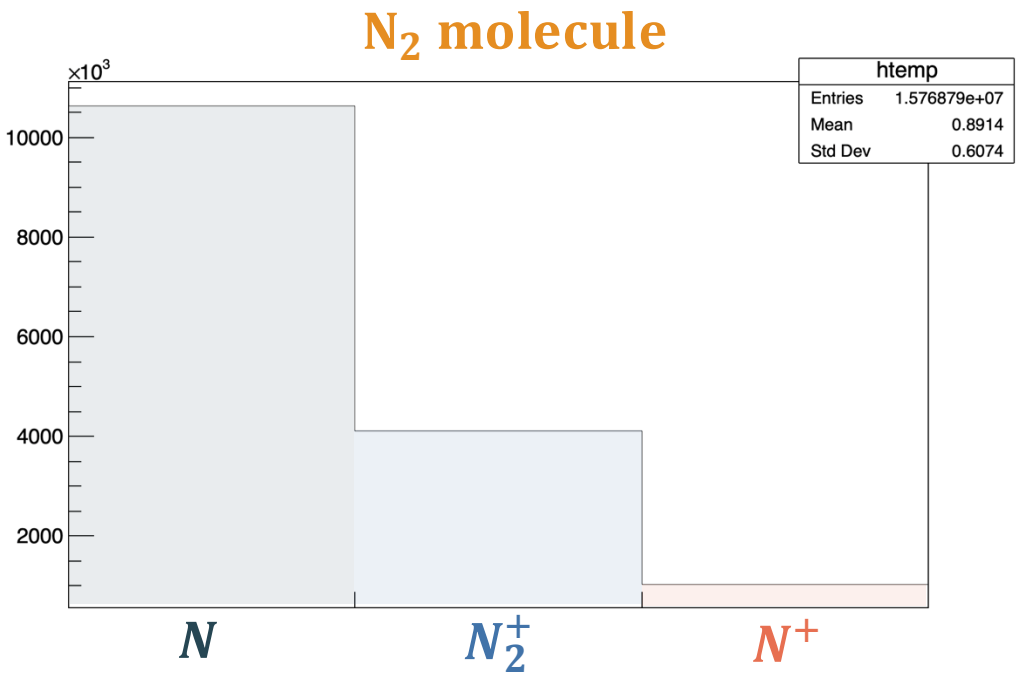
Preliminary



Resources requested from CINECA GALILEO100 for running at HIGH STATISTICS!

Physico-chemical stage

- After physical interaction molecules can **dissociate or relax**
- The physico-chemical stage has been implemented for N_2 and O_2 (work in progress for $\boldsymbol{CO_2}$)



N2	Process		Type	Channels	Branching ratios (%)
	Ionization	N_2^+	-- Dissociative decay	N_2^+ N+N^+	80 20
	Excitation	$vib, A^3, B^3, W^3, B'^3, a'^1, w^1, E^3, a''$	Non-dissociative deexc.	$N_2 + \Delta E$	100
		$a^1, C^3, b^1, b'^1, c_4'^1$	Dissociative deexc. Non-dissociative deexc.	N + N $N_2 + \Delta E$	12, 50, 95, 84, 10 (respectively) (100-dissociative)
		Rydberg, 17.3 eV and 15.8eV peak	Dissociative deexc.	N+N	100

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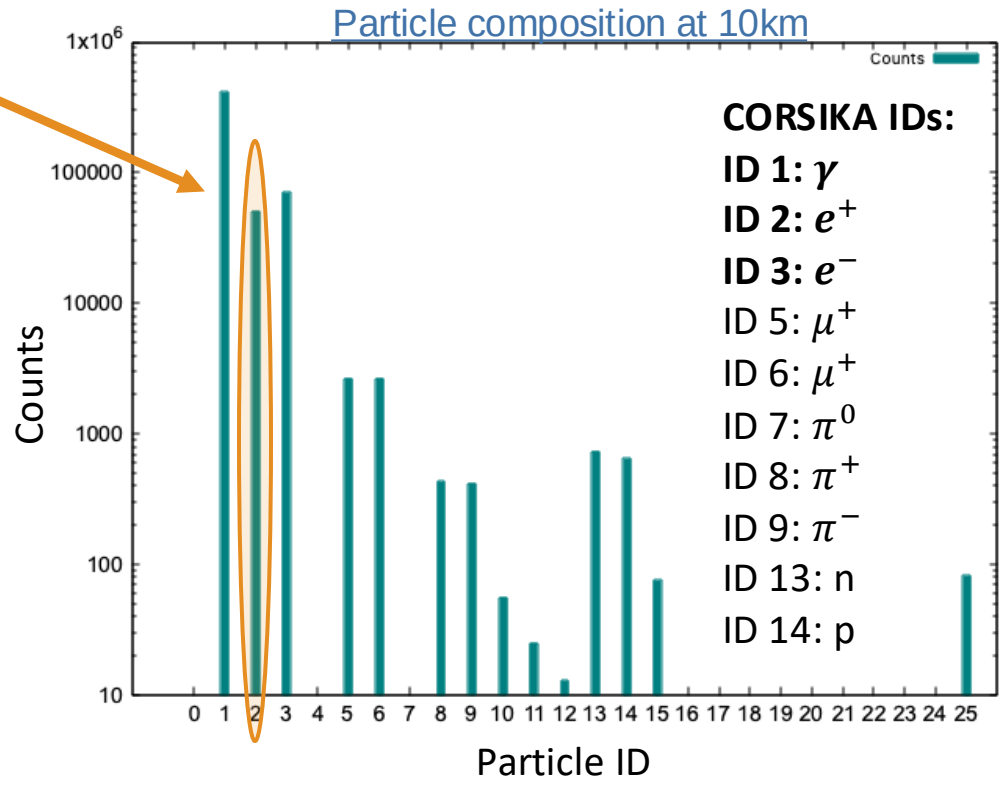
Extending physical models to positron impact

Goal: have a rough first estimate using the same cross-section models modified for e^+ impact (repulsive columbian interactions, no exchange effects)

Processes:

- **Ionisation:** RBEB (Relativistic Binary Encounter Bethe) with the Wannier-type threshold law [1] (RBEB-W)
- **Elastic:** (SCAR) Screening Corrected Additivity Rule
- **Excitation:**
  - Experimental data for e^+ impact on N_2, O_2 scarce or absent
  - Treating the excitation of N2 and O2 by e+/e- in the same way [2]

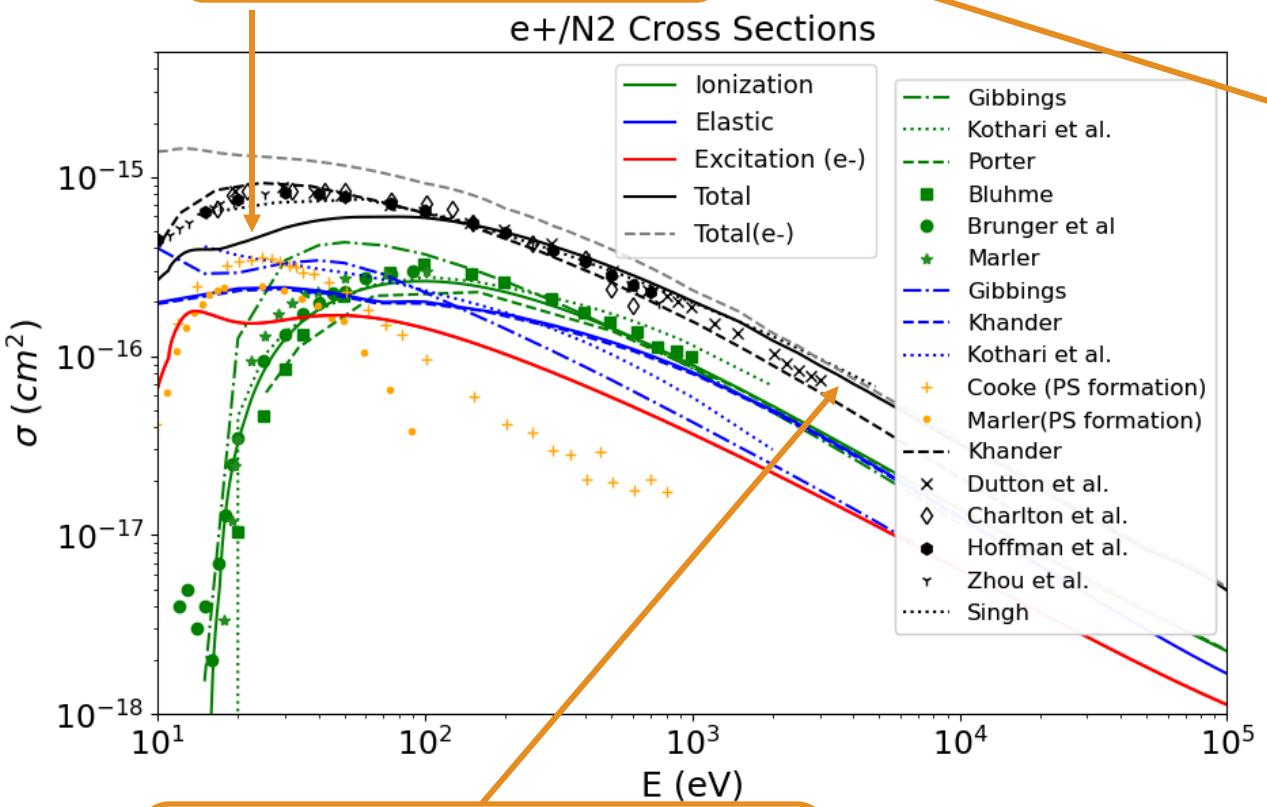


[1]Franz,M.; Wiciak-Pawłowska, K.; Franz, J. Binary-Encounter Model for Direct Ionization of Molecules by Positron-Impact. Atoms 2021, 9, 99.

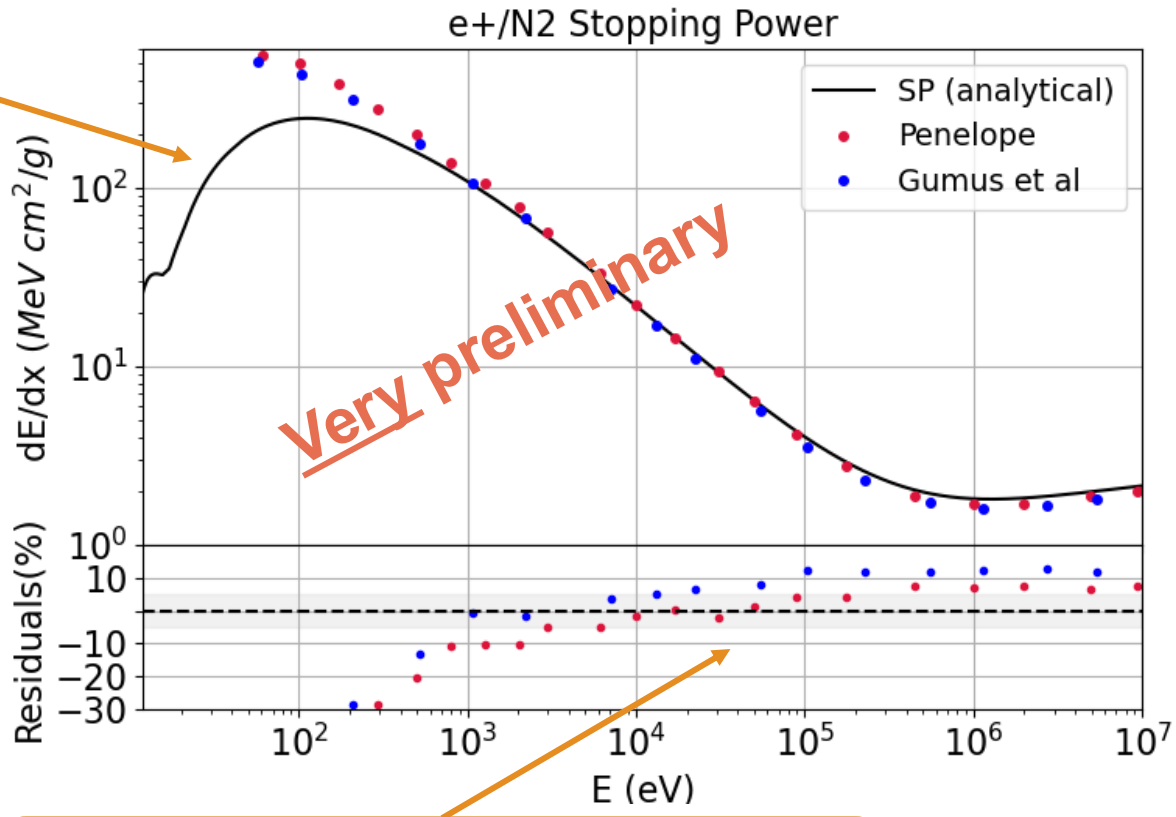
[2] Champion C, Le Loirec C. Positron follow-up in liquid water: I. A new Monte Carlo track-structure code. Phys Med Biol. 2006 Apr 7;51(7):1707-23.

Extending physical models to positron impact

Missing contribution from Positronium formation



Overestimated total cross section
Up to 20% compared with Dutton's data at 3keV



Overestimated stopping power:
Up to 8% compared with Penelope program
Up to 12% compared to Gumus model

Next steps

- Extending physical models to **positron** impact
    - Fitting the free parameters of RBEQ and Porter's formula to the total cross section obtained by application of the optical theorem
- **Running** simulation at high (with the help of Cineca GALILEO100)
    - The simulation output will be used as input for chemistry models

Perspectives:

- Including **chemical stage** (experimental G-values required)

Thank you for your attention!

[GOAL: **Studying the impact of ionisation on atmospheric chemistry**]

Geant4 models:

- Not cares what happens to the medium after the physical interactions

Geant4-DNA approach:

- Allows to obtain the number of ions produced, the ionisation states, chemical species, ..

✓ We have developed and implemented electron impact model with N_2, O_2, CO_2 molecules, including dissociation and relaxation after the physical interactions

✓ We have written a Geant4-code to simulate CR propagations in air

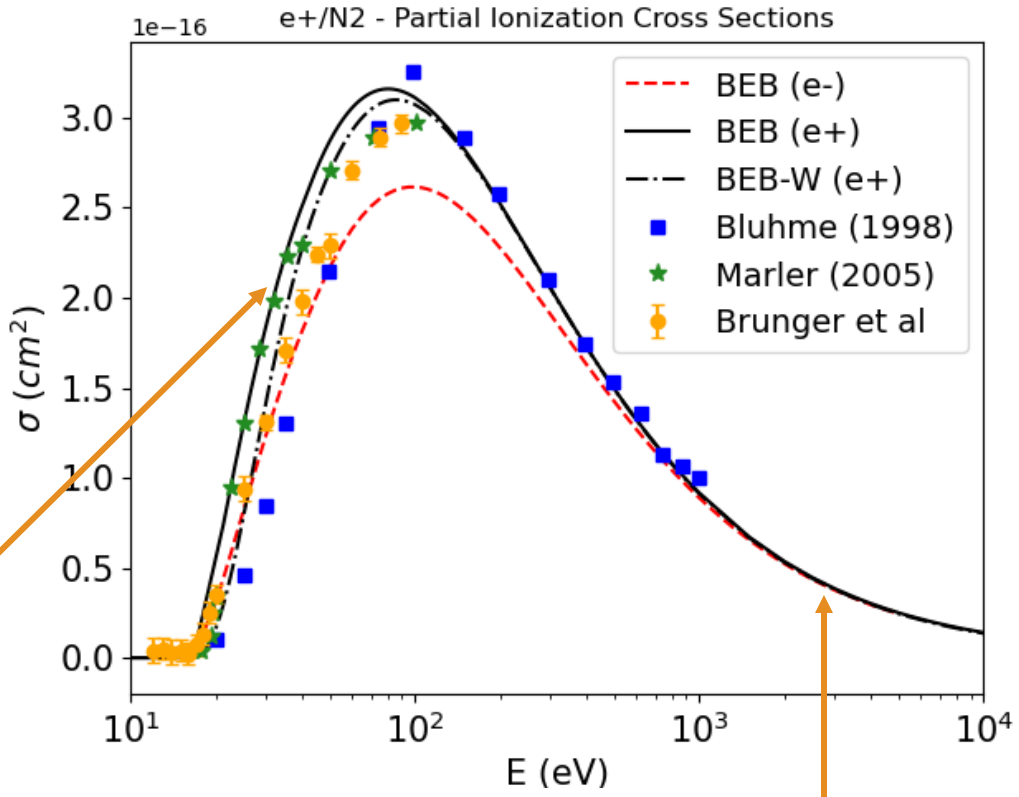
✓ **READY to RUN** with high statistic… !

Francesca Nicolanti, 09/10/24
Contact: francesca.nicolanti@uniroma1.it

Extending physical models to positron impact

Processes:

- **Ionisation:** BEB-W model [1]
- **Elastic Scattering:** SCAR
- **Electronic excitation:** ?



Wannier law near threshold
$\sigma(E) \propto (E - B)^{-\alpha}$.
(several parametrizations are possible)

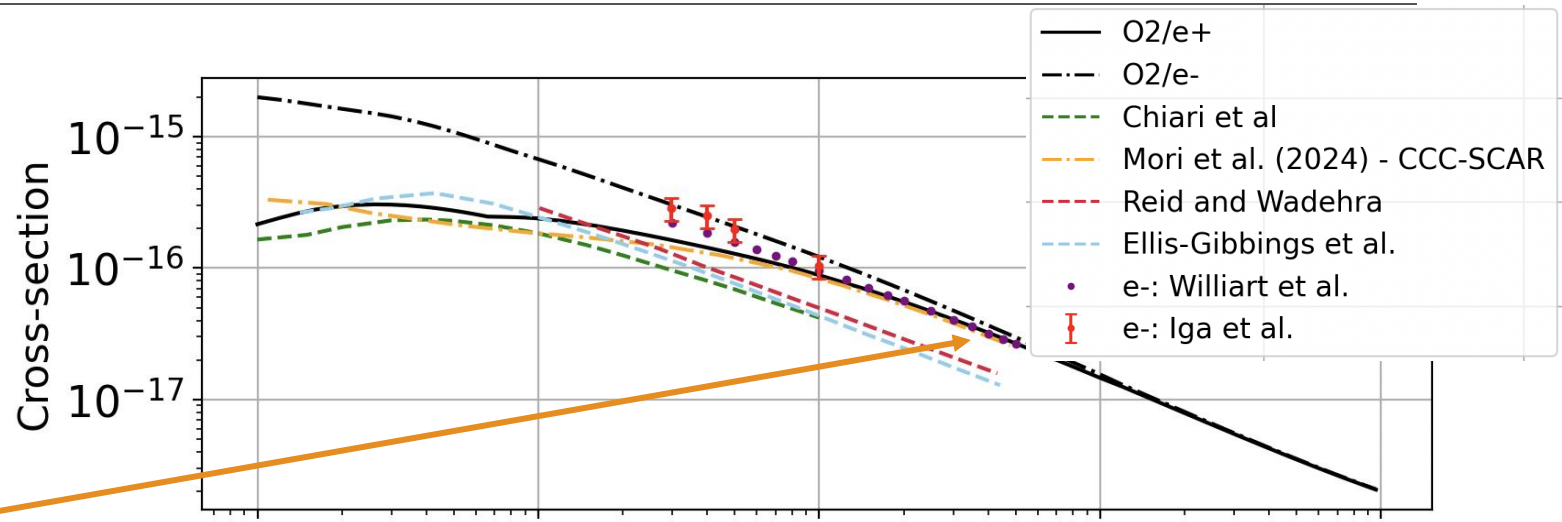
e+/e- cross section merge for ~ E>1keV

[1] Fedus, K.; Karwasz, G.P. Binary-encounter dipole model for positron-impact direct ionization. *Phys. Rev. A* **2019**, *100*, 062702.

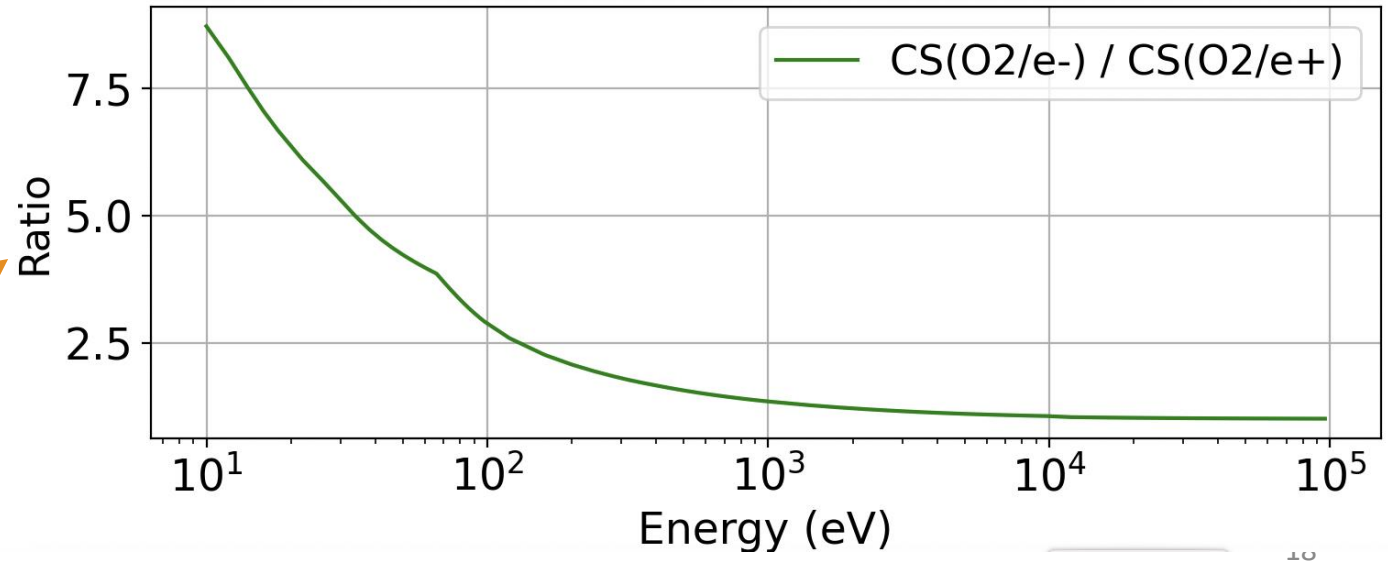
Extending physical models to positron impact

Processes:

- **Ionisation:** BEB-W model
- **Elastic Scattering:** SCAR
- **Electronic excitation:** ?



e+/e- cross section merge for ~ E>1keV



From ICRU 77 report, at E = 10 keV:

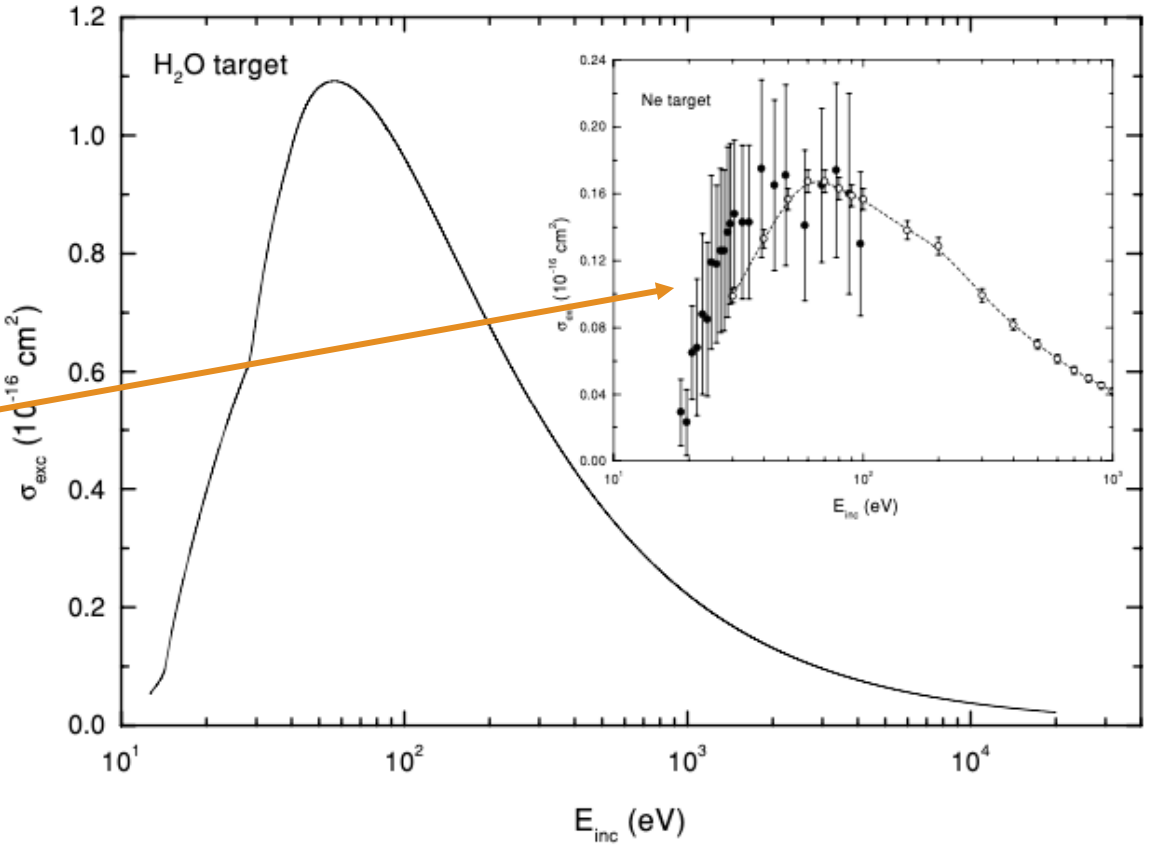
- Au : R =1.06
- Al : R = 1.05
- C: R = 1.02

For O2 at 10 keV: R = 1.025

Extending physical models to positron impact

- **Processes:**
  - **Ionisation:** BEB-W model
  - **Elastic Scattering:** SCAR
  - **Electronic excitation:** ?

CS of Ne excitation by positron and electron impact show very small differences



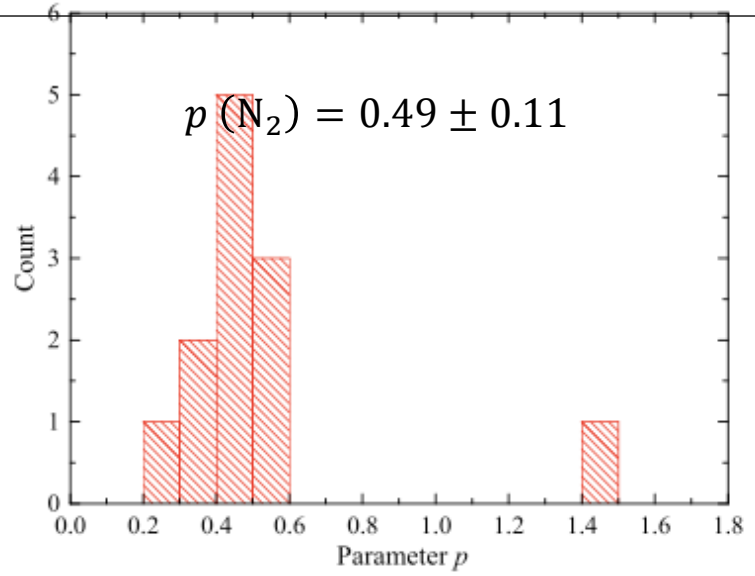
?

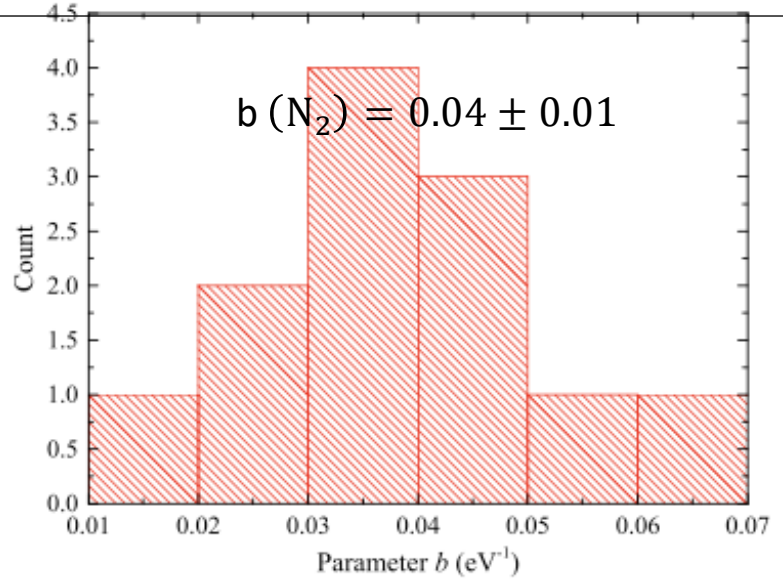
Treating the excitation of N2 and O2 by e+/e- in the same way may be an option [1]

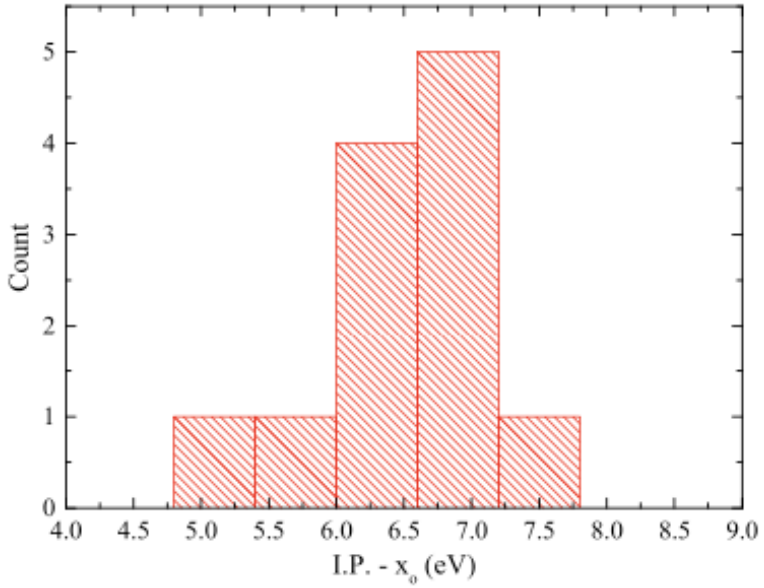
[1] Champion C, Le Loirec C. Positron follow-up in liquid water: I. A new Monte Carlo track-structure code. Phys Med Biol. 2006 Apr 7;51(7):1707-23. doi: 10.1088/0031-9155/51/7/005.

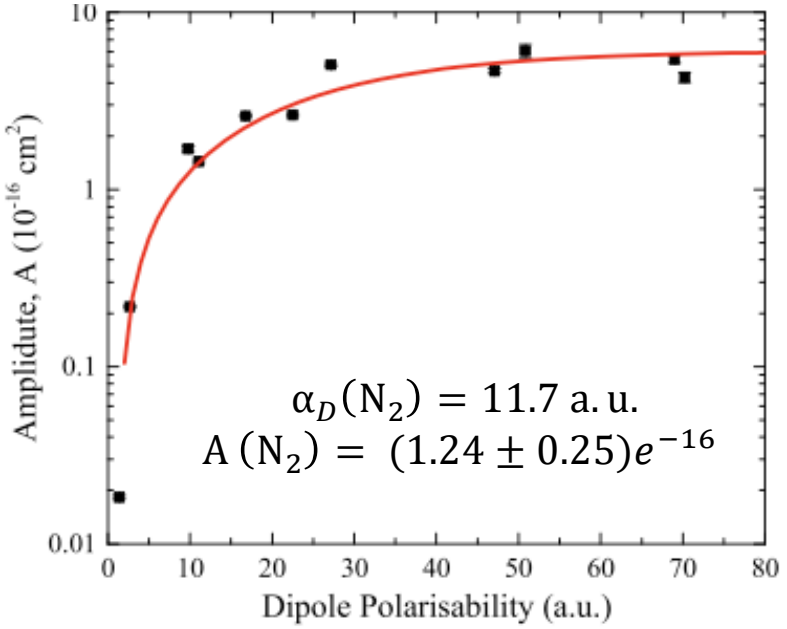
Extending physical models to positron impact:

Target
He
Ne
Ar
Kr
Xe
H_2
Water (H_2O)
Formic acid (CH_2O_2)
Pyrimidine ($C_4H_4N_2$)
THF (C_4H_8O)
3h-THF ($C_4H_8O_2$)
Uracil ($C_4H_4N_2O_2$)









Extending physical models to positron impact:

Regularities in positronium formation for atoms and molecules

J R Machacek[1], F Blanco[2], G Garcia[3], S J Buckman[1,4] and J P Sullivan[1]

- Semiempirical formula fitted to exp data

$$f(x) = Ax^p e^{-bx},$$

- Fitted parameters:

```
Parametri ottimali e errori associati:
x0 = 11.0729 ± 0.5541
A  = 1.2388e-16 ± 2.5049e-17
p  = 0.4931 ± 0.1140
b  = 0.0426 ± 0.0060
```

Positronium impact cross section $e^+ - N_2$