Development of new low energy EM models to describe ionisation in the atmosphere

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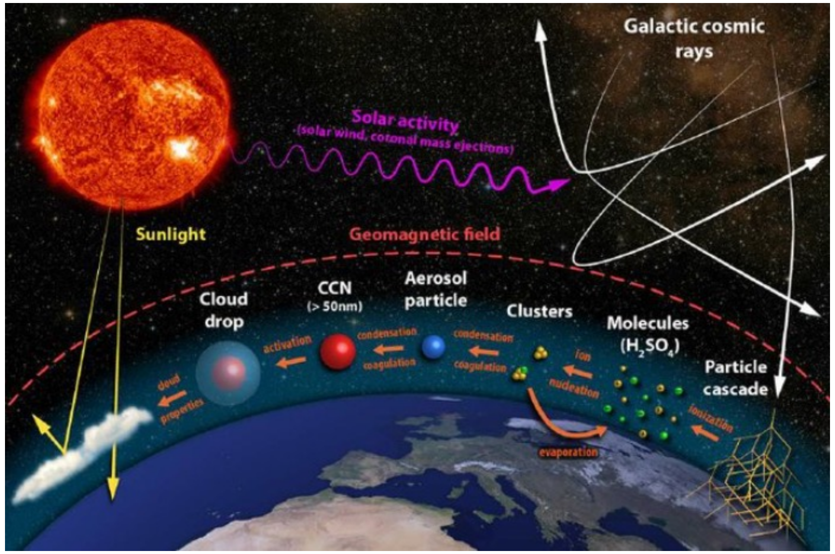
Motivation

What is the impact of cosmic rays and ions on atmospheric chemistry and climate evolution ?

precipitation
(Kniveton 2004)

Cloud cover
(Voiculescu et al. 2006)

ion-induced nucleation
(Svensmark et al. 2007)

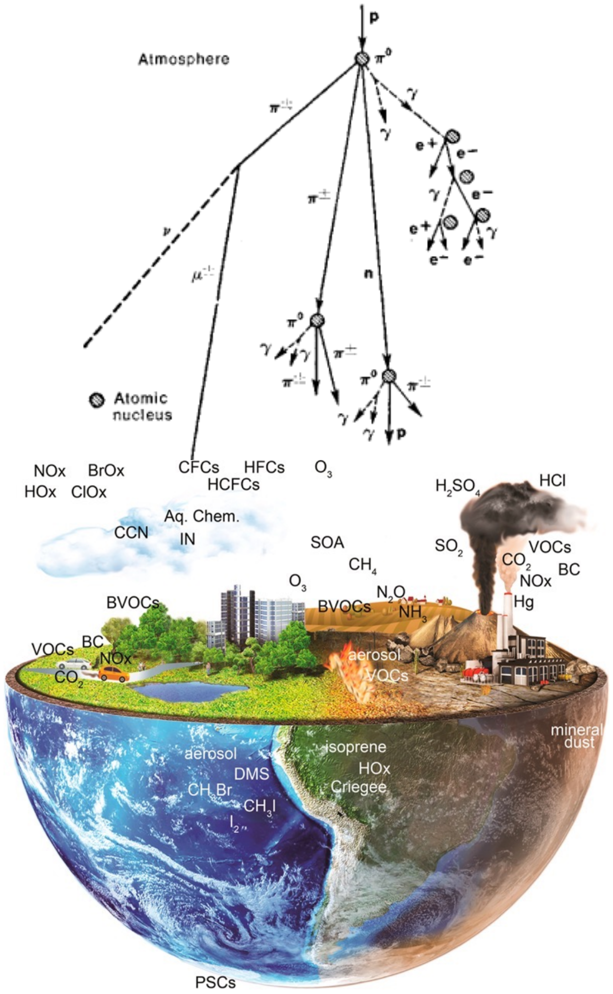


Ozone deplation
(Kniveton 2004)

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

- **The connection of cosmic rays with ions and the climate parameters remains a challenging topic.**

Motivation



▌Cosmic Rays impact with the atmosphere produce ions

▌**Even a small amount of ions can affect the atmospheric composition:**

- they trigger **faster chemical reactions** (up to 10 orders of magnitude higher)
- Partecipate in **catalytic processes**
- Inhomogeneous spatial distribution

State of the Art

Monte-Carlo codes: CR induced ionisation

Oulu CRAC:CRII (CORSIKA+FLUKA)
Usoskin et al., J. Atm. Solar-Terr. Phys, (2004).
Usoskin, Kovaltsov, J. Geophys. Res., (2006, 2010).

Bern model ATMOCOSMIC (GEANT-4)
Desorgher et al., Int. J. Mod. Phys. A, (2005)
Scherer et al. Space Sci. Rev. (2006).

AtRIS (GEANT-4)
Banjac et al., JGR Space Physics (2018)

RUSCOSMICS (Geant4)
Maurchev et al., Bull. Russ. Acad. Sci. Phys. (2019)

Output of the models: average production rate of ion pairs (ions cm-3 s-1)

BUT

- Condended history approach: it neglects local effects;
- Medium-high energy models: less accurate description of low-energy secondary radiation;
- No molecular description;

!

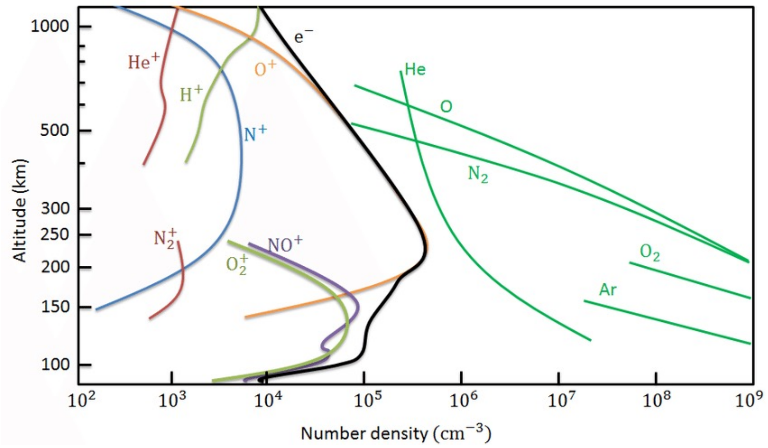
How many ions are produced? In what state? What spatial distribution?

Geant-DNA for atmosphere

GOAL

Accurately **describe ions produced by CR** interaction in the atmosphere:

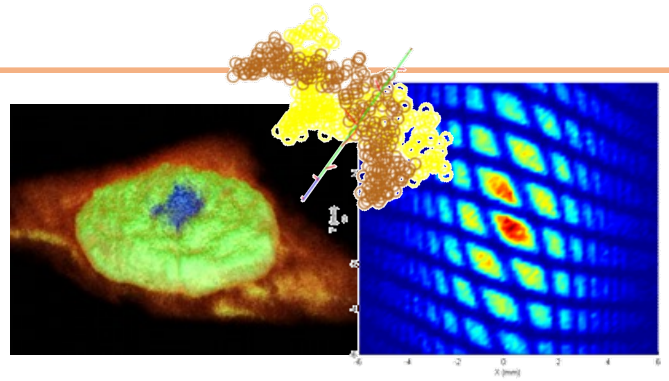
- Ionic density
- State of ionization
- spatial distribution of ions
- chemical species produced



HOW

By **including in** **Geant4-DNA** **new models** for particle-impact interactions with relevant molecules for climatology

- Track Structure approach
- physics models reach low energy (10 eV)
- Description of target molecular properties:



STARTING POINT

e- impact interaction models on N2 and O2

- **Ionisation, elastic scattering, electronic excitation**
- **energy range 10eV – 10 MeV**

N2 and O2 cross-sections implemented in Geant4-DNA

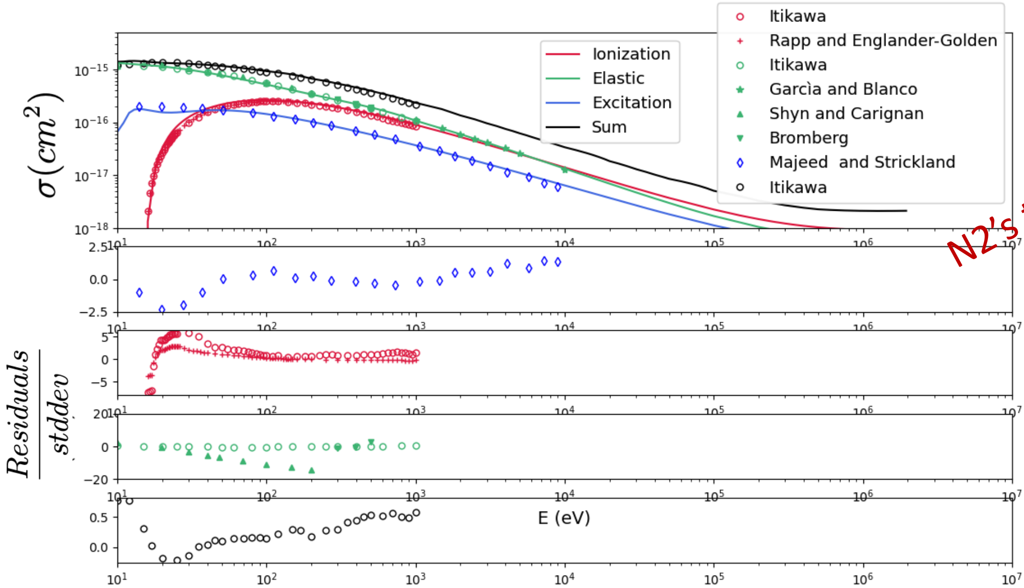
Cross section models have been reviewed;

Selected cross section models:

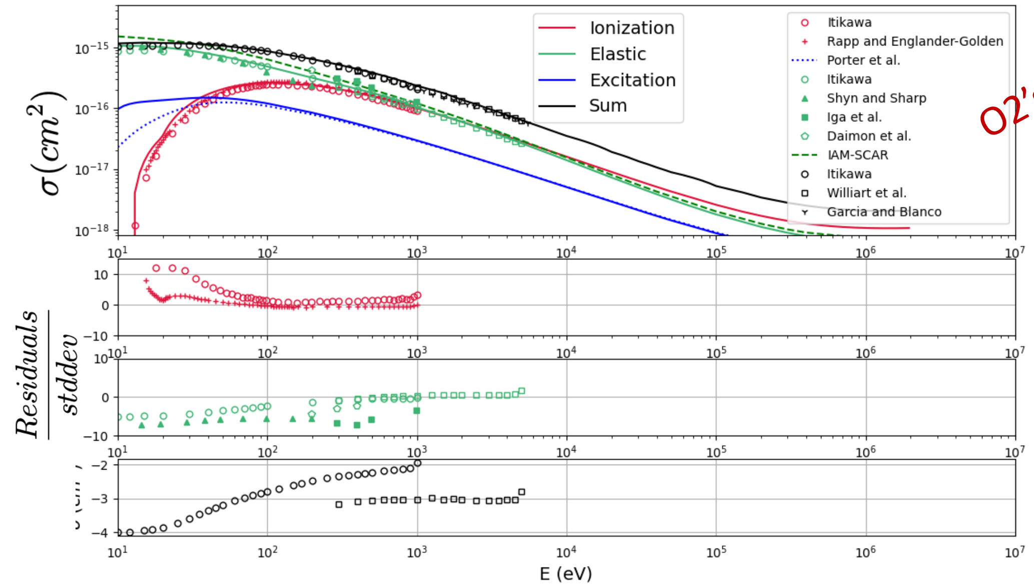
- **Impact ionisation**: Relativistic Binary Encounter Bethe (RBEB) model
- **Elastic scattering**: Screening Corrected Additivity Rule(SCAR) model
- **Electronic excitation**: Porter's formulas

They **have been optimized for N2,O2 based on comparison with experimental data.**

Calculation of electron interaction models in N2 and O2 / F. Nicolanti, B. Caccia, A. Cartoni, D. Emfietzoglou, R. Faccini, S. Incerti, I. Kyriakou, M. Satta, H. N. Tran, C. Mancini-Terracciano. Physica Medica



N2's total cross section



O2's total cross section

N2 and O2 cross-sections implemented in Geant4-DNA

[MODELS ARE DESCRIBED IN THE G4AtmXXXModel.cc .hh 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, and 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	Formulas based on Porter et al, fitted cross section parameters	Data tables: sigma_excitation_e_N2_atm.dat

[1] Kim, Y. (2000), Extension of the Binary-Encounter-Dipole Model to Relativistic Incident Electrons, Physical Review A (Atomic, Molecular and Optical Physics)

[2] Francesc Salvat; Aleksander Jablonski; Cedric J. Powell (2005). elsepa—Dirac partial-wave calculation of elastic scattering of electrons and positrons by atoms, positive ions and molecules. , 165(2), 157–190.

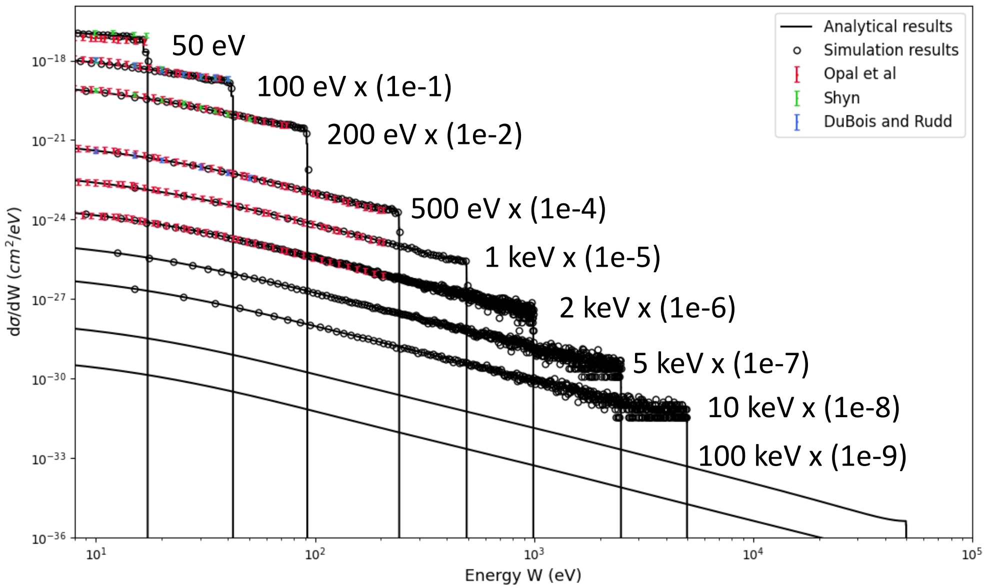
[3] H. Porter, C. Jackman, and A. Green, Efficiencies for production of atomic nitrogen and oxygen by relativistic proton impact in air, J. Chem. Phys. 515 65 (1) (1976) 154–167

Validating implementation

- ✓Differential cross sections
- ✓Stopping power
- ✓Range

Differential Cross Sections (N2)

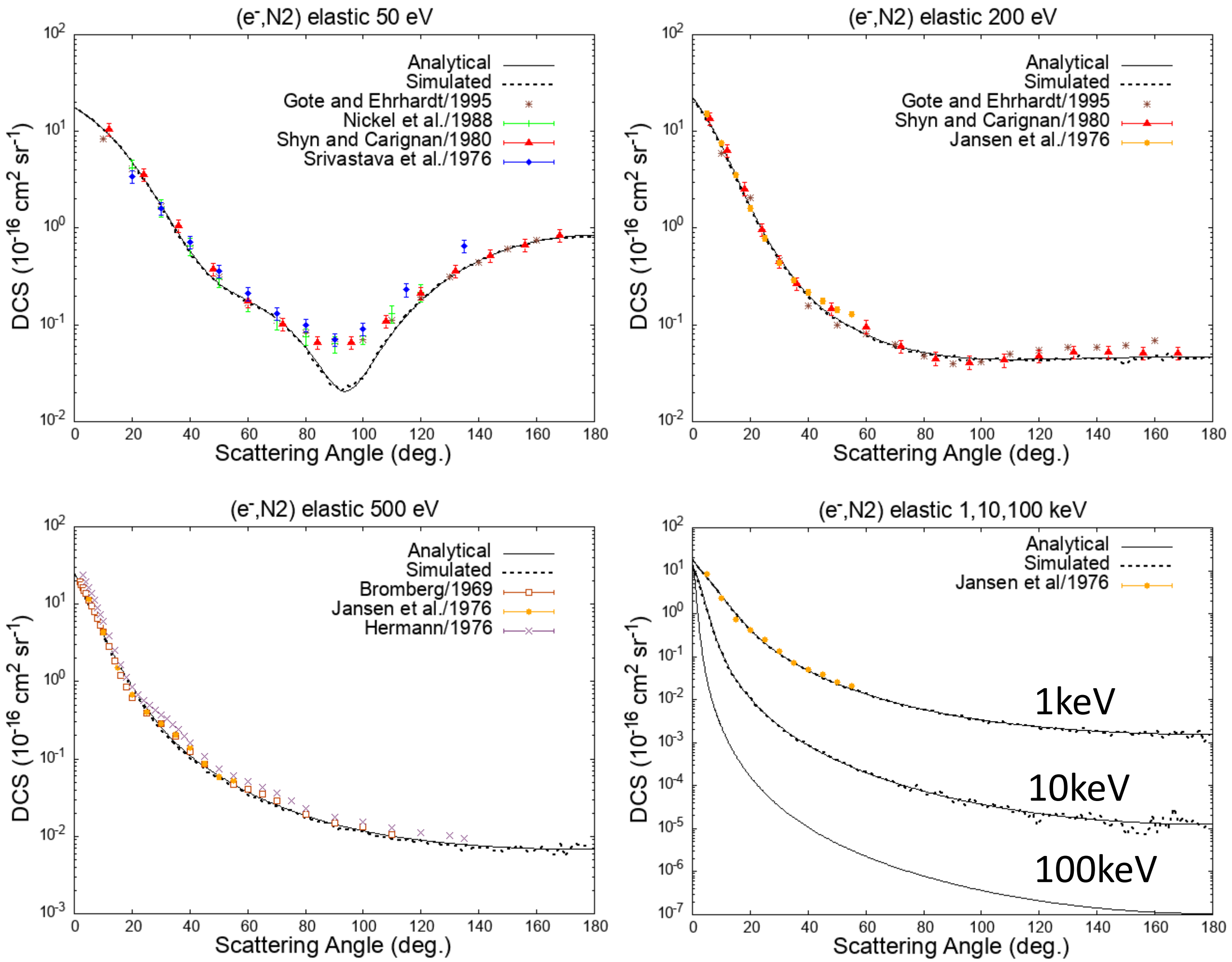
Ionisation: secondary electrons distribution



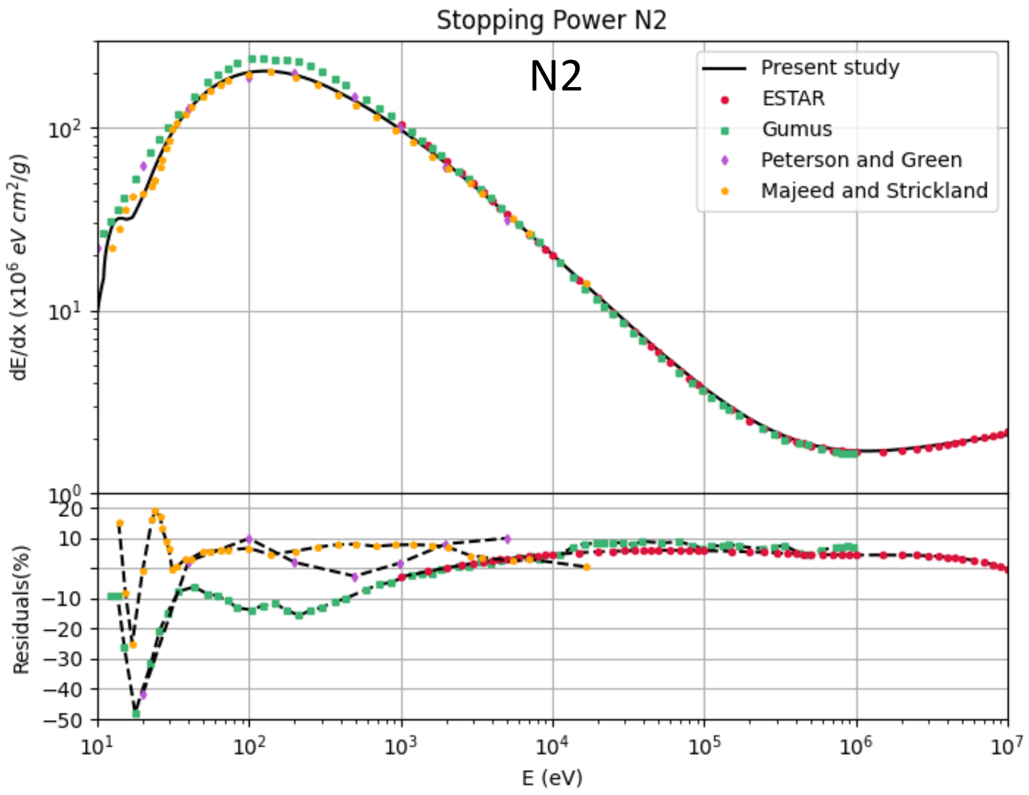
- Primaries and secondary electrons distribution have been:
  - compared to analytical calculations
  - validated through experimental data

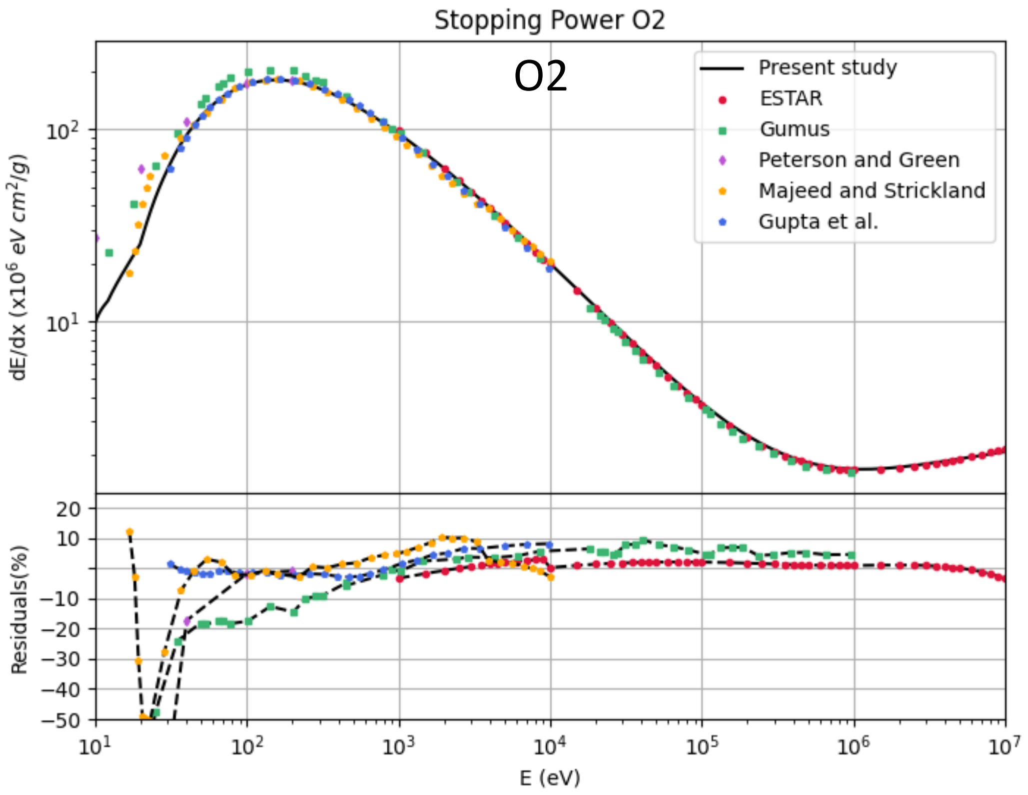
- General **good agreement** is observed

Elastic scattering: scattering angle distribution



Stopping Power in N2 and O2





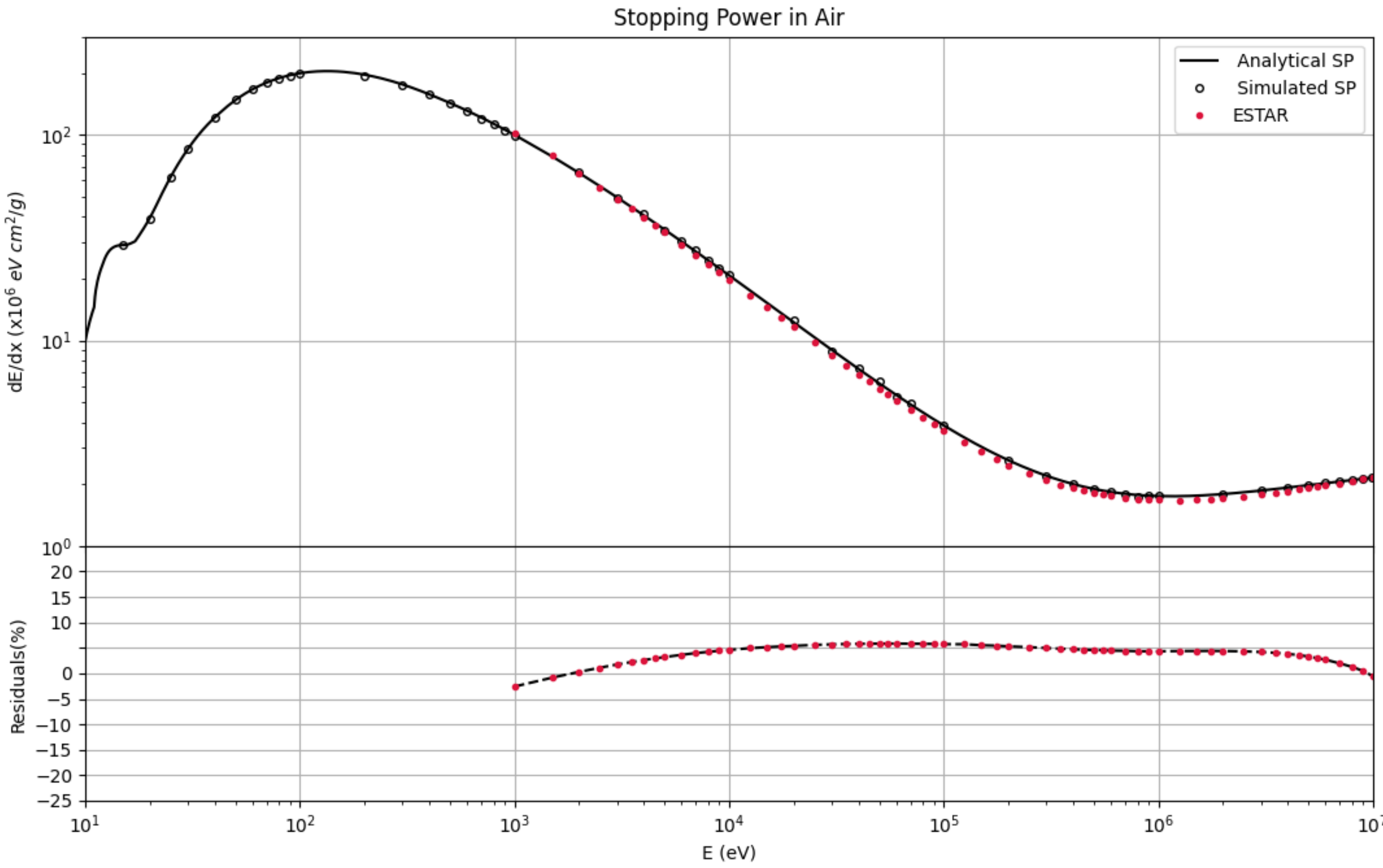
Relative differences:
- <10% with Gumus model and Peterson
- <5% with NIST

for E>20eV

Relative differences:
- <10% with Gumus, Gupta, and Peterson
- <3.5% with NIST

for E>30eV

Stopping Power in Air



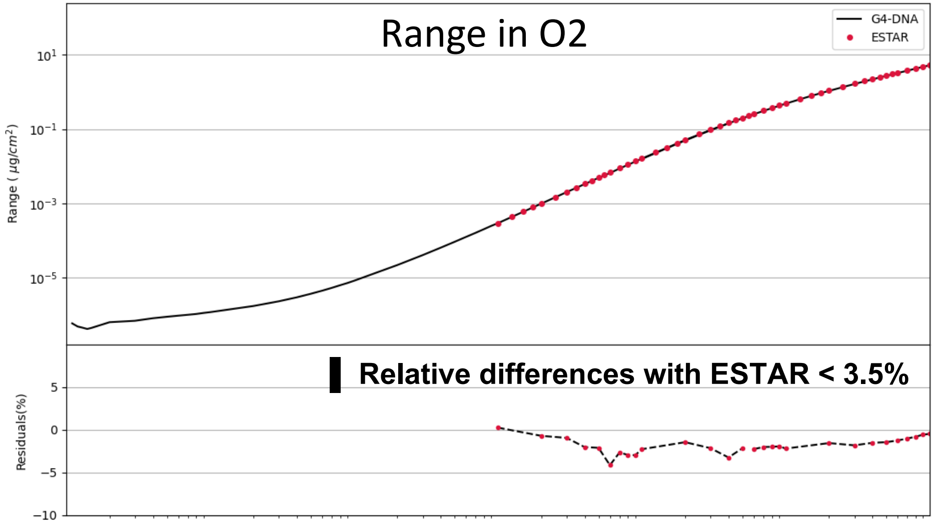
- Air composition (NEAR SEA LEVEL):
  - 76% N2
  - 24% O2

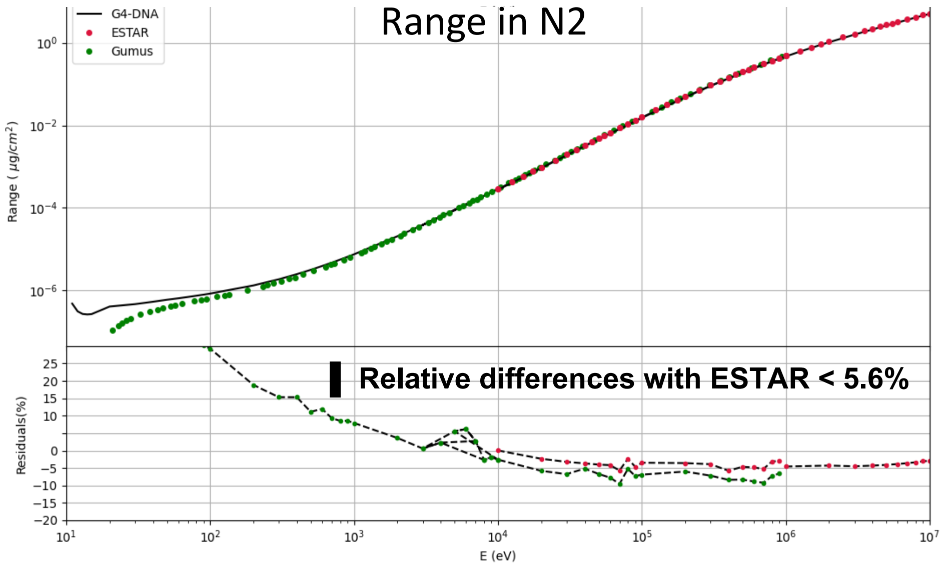
  Density (g/cm3): 1.205E-03

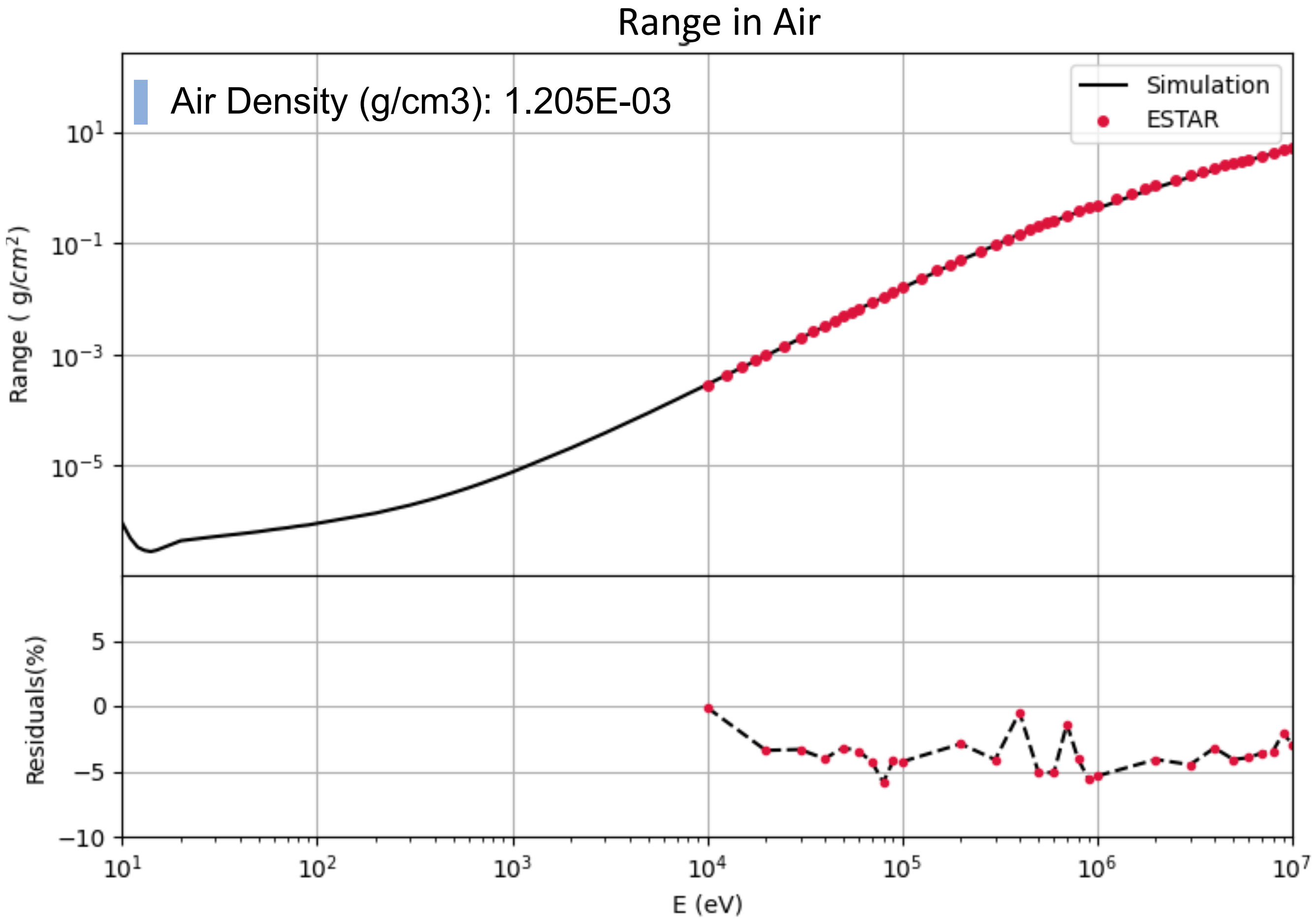
- **Relative differences:**
  < 5.4% with NIST

Same overestimation as observed for N2

Range

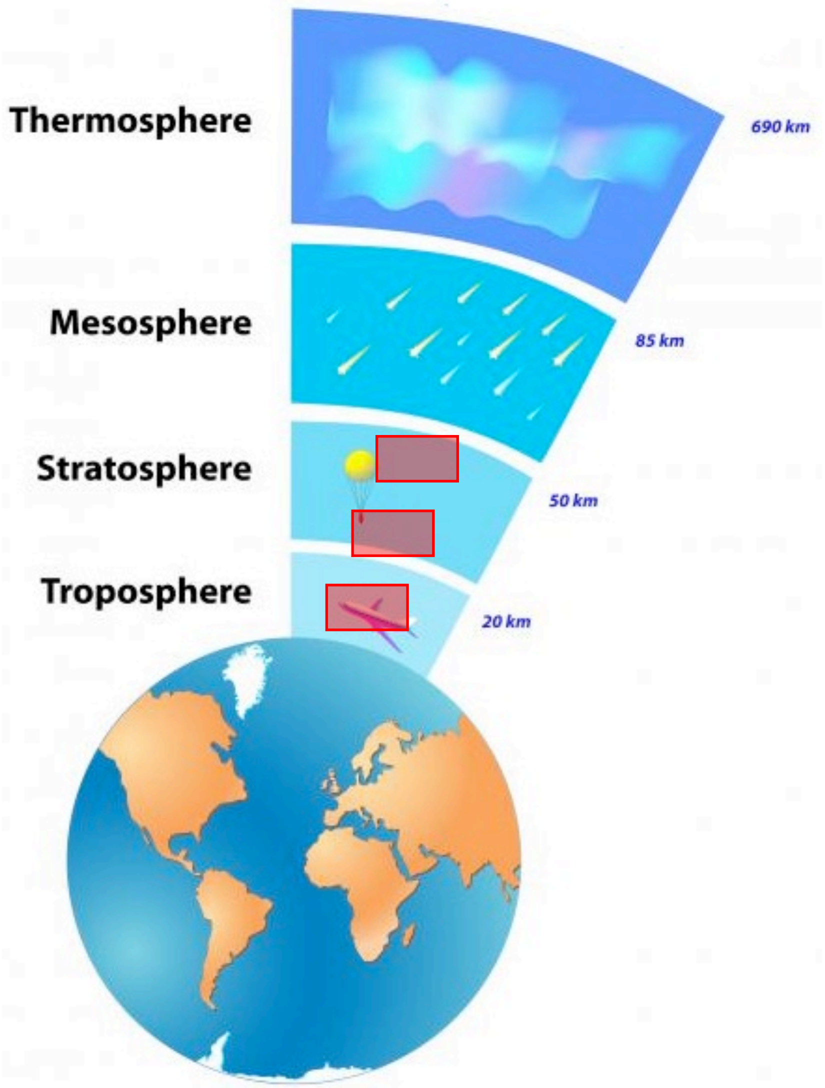






Relative differences with ESTAR database: < 5.5%

Further work



- Import cosmic rays spectra at different altitudes (5-25km) in Geant4
- Simulate primaries interactions using Geant4 models
- Use of the new electron-impact G4-DNA models in small volumes
- Development of a **Geant4-DNA example** to track electrons in the atmosphere and get the **ionic density**
- Experimental validation

- Development of the **physical-chemical stage** to simulate dissociation, diffusion, recombination, and chemical reactions

Conclusions

N2 and O2 gas cross-section have successfully been implemented in Geant4-DNA. This will allow to simulate interactions with a simplified **air volume** in the atmosphere

They will be available in a next Geant4 release togheter with an updated version of the spower example ***/examples/extended/medical/dna/spower***

Applications:

- Potential impact on the physics of the atmosphere
- Open the way for new space-related studies regarding chemistry and exobiology
- Can be applied to any kind of Geant4-DNA simulation (micro and nano dosimetry, discharge fenomena, ..).

Contact: francesca.nicolanti@uniroma1.it

Thank you for your attention!

N2 and O2 cross-sections implemented in Geant4-DNA

[**AUTOIONISATION PROCESS INCLUDED IN THE EXCITATION MODEL:**

The auto-ionisation process is part of the excitation model in both MC codes, where a probability of 50% auto-ionisation is assumed if the excitation energy of a Rydberg state is greater than the ionisation threshold for the molecule under study. The secondary electrons produced by auto-ionisation are emitted isotropically and their kinetic energy is calculated from the given excitation energy minus the ionisation threshold.

N2 and O2 cross-sections implemented in Geant4-DNA

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

- **Designed to handle a mix of materials different from liquid water**
- «G4DNAModelInterface» used to manage the physical processes and models of mixed material
- **Introduced for the first time in Geant4 10.4 for the use of DNA precusors material's cross-sections: THF, TMP, PU and PY** (*M. Bug et al, Rad. Phys and Chem. 130, 459-479 (2017)*)

«G4AtmPhysics» class

```
auto modElas = new G4AtmElasticModel();

auto air_e_elas = new G4DNAModelInterface("Air_e-_elas");
air_e_elas->RegisterModel(modElas);

auto e_DNAElasticProcess =
  new G4DNAElastic("Air_e-_elasticScattering");
e_DNAElasticProcess->SetEmModel(air_e_elas);

helper->RegisterProcess(e_DNAElasticProcess, part);
```

«G4AtmElasticModel.cc» class

```
if (fpG4_MOLNITRO != nullptr) {
  auto index = fpG4_MOLNITRO->GetIndex();
  AddCrossSectionData(index, p, "dna/sigma_elastic_e_N2_elsepa",
            "/dna/sigmadiff_cumulated_elastic_e_N2_elsepa", scaleFactor);
  SetLowELimit(index, p, 10. * eV);
  SetHighELimit(index, p, 10. * MeV);
}
if (fpG4_MOLOXYGEN != nullptr) {
  auto index = fpG4_MOLOXYGEN->GetIndex();
  AddCrossSectionData(index, p, "dna/sigma_elastic_e_O2_elsepa",
            "/dna/sigmadiff_cumulated_elastic_e_O2_elsepa", scaleFactor);
  SetLowELimit(index, p, 10 * eV);
  SetHighELimit(index, p, 10 * MeV);
}
if (!G4DNAMaterialManager::Instance()->IsLocked()) {
  // Load data
  LoadCrossSectionData(p);
  G4DNAMaterialManager::Instance()->SetMasterDataModel(DNAModelType::fDNAElastics, this);
  fpModelData = this;
```

N2 and O2 cross-sections implemented in Geant4-DNA

The cross sections **have been optimized for N2,O2:**

- Potential models and calculation parameters for elastic scattering:
  - based on discrete RMSE between the experimental data and calculated DCSs
- Free Porter's model parameters for excitation:
  - through fitting experimental data

Calculation of electron interaction models in N2 and O2 / F. Nicolanti, B. Caccia, A. Cartoni, D. Emfietzoglou, R. Faccini, S. Incerti, I. Kyriakou, M. Satta, H. N. Tran, C. Mancini-Terracciano. Physica Medica

SDCS

Ionisation: Relativistic Binary Encounter Bethe (RBEB)

$$\frac{d\sigma_{ion,MO}}{dW} = \frac{\pi a_0^2 \alpha^4 N}{(\beta_t^2+\beta_u^2+\beta_b^2)2b'}\Big\{\Big[ln\Big(\frac{\beta_t^2}{1-\beta_t^2}\Big) - \beta_t^2 - ln(2b')\Big]\Big[\frac{1}{(w+1)^3}+\frac{1}{(t-w)^3}\Big] - \frac{1}{t+1}\Big(\frac{1}{w+1}+\frac{1}{t-w}\Big)\Big[\frac{1+2t'}{(1+t'/2)^2}\Big] + \frac{1}{(w+1)^2} + \frac{1}{(t-w)^2} + \frac{b'^2}{(1+t'/2)^2}\Big\}$$

Depend only on three parameters:

B: Binding Energy
U: Average Kinetic Energy
N: Electron Occupation Number

Elastic: Screening Corrected Additivity Rule (SCAR)

- Independent Atom Approximation
- Dirac partial-wave approach
- Optical potential model:

$$V(r) = V_{st}(r) + V_{ex}(r) + \boldsymbol{V_{cp}}(\boldsymbol{r}) + iW_{abs}(r)$$

Depend on two **free** parameters

Electronic Excitation: Porter's formulas

Depend on four **free** parameters

$$\sigma_j^{exc,allowed} = \frac{q_0 A\phi(2W_j/m\beta^2c^2)}{(m\beta^2c^2/2W_j)\,W_j^2}\Big\{\ln\Big[4\Big(\frac{m\beta^2c^2}{2W_j}\Big)\frac{C_j}{(1-\beta^2)}+\mathrm{e}\Big]-\beta^2\Big\}$$

$$\sigma_j^{exc,forbidden} = \frac{q_0 A\phi(2W_j/m\beta^2c^2)}{(m\beta^2c^2/2W_j)\,W_j^2} \qquad \phi(2W_j/m\beta^2c^2) = \frac{\big[1-(2W_j/m\beta^2c^2)^{\alpha}\big]^{\beta}}{(m\beta^2c^2/2W_j)^{\Omega-1}}$$

ELSEPA interaction potential

Optical potential model:

$$V(r) = V_{st}(r) + V_{ex}(r) + \boldsymbol{V_{cp}}(\boldsymbol{r}) - iW_{abs}(r)$$

Electrostatic potential $V_{st}(\mathrm{r})$

- Potential model:

  Nuclear charge: Fermi distribution;

  Electron density: Dirac–Fock distribution.

Exchange potential $V_{ex}(r)$

Potential model:

Furness–McCarthy potential.

Correlation-polarization potential $\mathbf{V_{st}}(\mathbf{r})$

Influence at small scattering angles and E < 500eV

- Potential model:
  - Buckingham potential + LDA correlation (Perdew and Zunger)
- Free parameters:
  - static polarizability α_d = *1.562E-24(O2), 1.710E-24(N2) [cm^3]*
  - cut-off parameter $b_{pol}^2 = \max[(E - 20\text{ eV})/\text{eV},\, 1]$.

Inelastic absorption potential $-\mathbf{i}\mathbf{W}_{abs}(\mathrm{r})$:

Influence at intermediate and large scattering angles

- Potential model:
  - LDA potential (Salvat)
- Free parameters:
  - lowest excitation energy ϵ_1 = 0.98 eV(O2), 7.63 (N2)
  - absorption strength $A_{abs} = 2$

N.B. Empirical parameters are **validated for noble gases** and mercury