

A comparative study of stopping power calculations implemented in Monte Carlo codes and compilations with experimental data



E. Vagena^{a,c}, E.G. Androulakaki^b, M. Kokkoris^{b,*}, N. Patronis^a, M.E. Stamati^a

^a Department of Physics, University of Ioannina, 45110 Ioannina, Greece

^b Department of Physics, National Technical University of Athens, Zografou Campus, 15780 Athens, Greece

^c Institute of Nuclear and Particle Physics, NCSR “Demokritos”, Aghia Paraskevi 153 10, Greece

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ABSTRACT

In the present work extensive stopping power calculations were performed, implementing general-purpose Monte Carlo codes (GEANT4, MCNP6.1, FLUKA, PHITS) and dedicated stopping power compilations (such as SRIM2013, PSTAR/ASTAR from NIST and ATIMA), having protons, alpha particles, ^{12}C , ^{16}O and ^{56}Fe ions as beam particles, in the energy range between ~ 1 and ~ 1000 MeV/u, impinging on a variety of thin or infinitely thick, pure, single-element targets, such as aluminum, iron, copper and silicon, which are typically used as shielding materials or components in complex devices, in order to investigate all the occurring differences and similarities in the obtained results. Water was the only compound target to be examined, since it is commonly used as a close substitute for biological tissue. In all the simulations the process of multiple scattering was ignored or disabled. The obtained results are presented in graphical form, along with all the available experimental data in the studied energy range and the occurring similarities and discrepancies are discussed and analyzed.

1. Introduction

The stopping power calculations, inherently implemented in all widely used, general purpose Monte Carlo codes, play a critical role in the determination of any expected reaction yield, when charged particles are present, either as beam particles or reaction products. Small changes in the stopping power values, and therefore in the corresponding particle energies, can lead to significant changes in the cross sections involved. This effect may be critical in a variety of problems, ranging from detector physics to dose calculations and—to the authors' opinion—despite the existence of pioneer, case-specific publications (e.g. [1–3]), requires further investigation over a broad energy range and for a variety of ion beam-target combinations, especially concerning the case of GEANT4, which is probably the most applied MC code for various detector and experimental setup studies, space and medical applications.

Thus, the aim of the present work is to examine the differences in the stopping power calculations between widely-used, general-purpose Monte Carlo codes, such as GEANT4 v.10.5.p01 [4], FLUKA v.2011.2x.6 [5], MCNP6.1 [6] and PHITS v.2.88 [7] and to compare the results (whenever possible) with all the available experimental datasets (retrieved from IAEA's repository [8], where the user can also obtain all

the corresponding references), as well as, against the widely used and partially benchmarked stopping power compilations, as implemented in the SRIM2013 [9], PSTAR/ASTAR (ICRU) codes [10] and ATIMA [11]. Since GEANT4 constitutes a unique case, due to the existence of several available options concerning the calculation of electromagnetic interactions, the study included not only the corresponding recommended option, but also a comparison among several of the available ones.

In general, in all Monte Carlo codes, protons, α -particles, ^{12}C , ^{16}O and ^{56}Fe ions were generated as beam particles in the energy range between 1 and 1000 MeV/u and were subsequently transported, impinging on a variety of thin (~ 1 μm) and/or infinitely thick, pure, single-element targets, such as aluminum, iron, copper and silicon, which are typically used as shielding materials or components in complex devices. Water was also examined, being a close substitute for biological tissue. The choice of this particular energy range was based on several considerations, such as (a) the fact that the vast majority of technological applications is indeed carried out for ion beam energies within the examined range, (b) the minimization of the non-electronic energy loss contribution and (c) the exclusion of ultra-high energy perturbations above 1 GeV/u.

The theory behind stopping power calculations in this particular energy regime, i.e. for high ion energies, has been beautifully analyzed

* Corresponding author.

E-mail address: kokkoris@central.ntua.gr (M. Kokkoris).

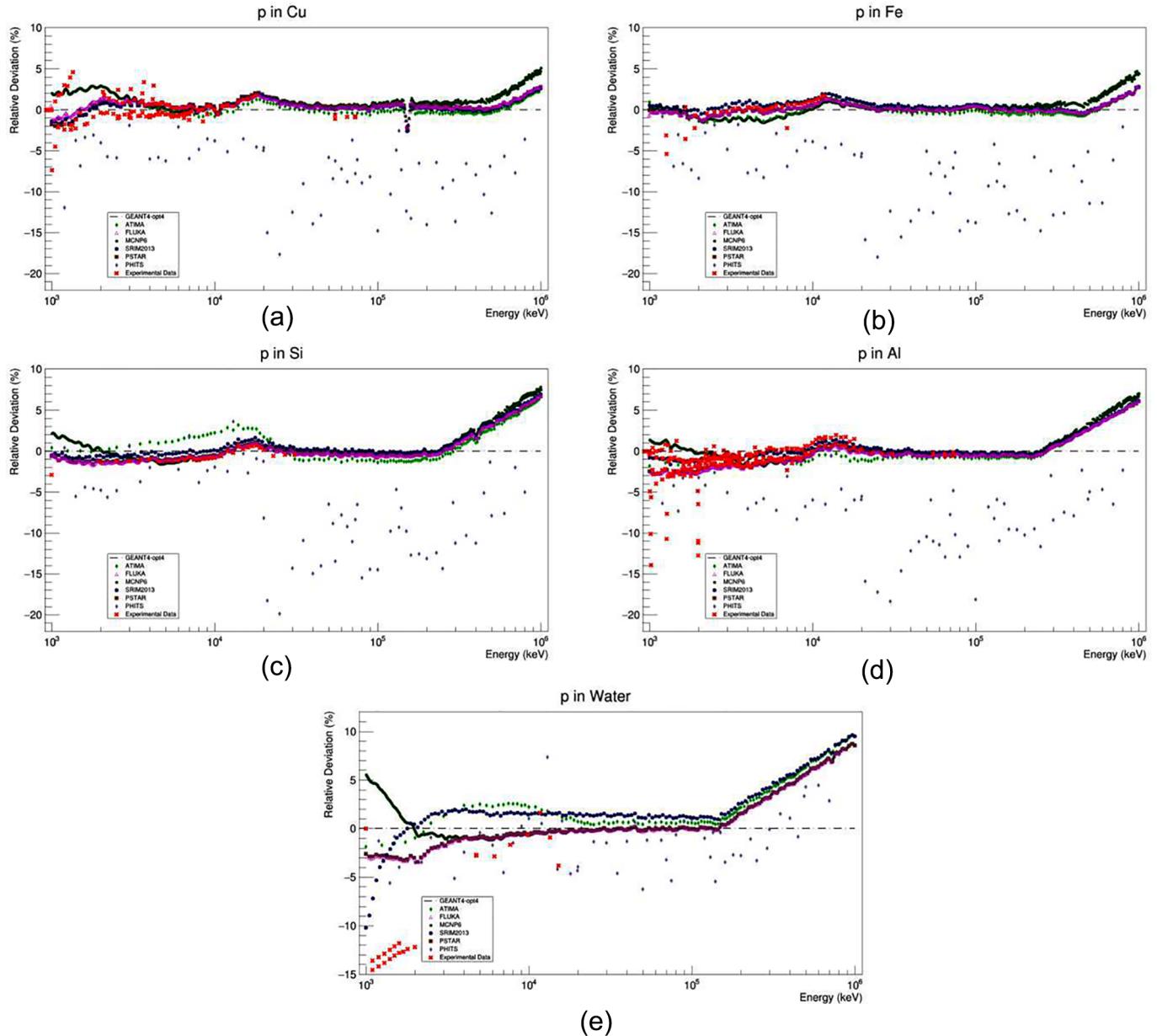


Fig. 1. a–e. Stopping power results, between 1 and 1000 MeV/u, concerning the following cases (implementing natural targets): (a) p in Cu, (b) p in Fe, (c) p in Si, (d) p in Al and (e) p in water. Energies are presented in units of keV, while the y-axis denotes the relative deviations from the GEANT4 predictions in %.

in detail in [12] and has been summarized in [3]. Briefly stated, it can generally be described by the relativistic Bethe theory, where the mass stopping power S/ρ (where $S = -dE/dx$ is the linear stopping power or stopping force) is a function of ρ , the density of the material, the atomic number, energy and velocity of the impinging ion, the atomic number and mass number of the target and is proportional to the stopping number L , which is given by:

$$L(\beta) = \ln \frac{2mv^2}{(1 - \beta^2)} - \ln I - \beta^2$$

where $\beta = v/c$, m corresponds to the mass of the electron, and I is the mean ionization potential. This equation is valid for high energies, but not so high as to render the density correction appreciable [3], while, in order to extend the validity to lower energies, shell, Barkas-Andersen and Bloch corrections should be considered. In the simulations these corrections are added mainly via the use of semi-empirical parameters, which vary according to each implemented code or compilation and projectile-target combination. It is important to note here that the

energy range covered in the present work is well beyond the stopping power maximum for protons and α -particles, while for heavier ions, in all projectile-target combinations the maximum occurs for energies below 1–2 MeV/u, thus there exists only a very small overlap with the region where electron screening may indeed become important. For methodological reasons the comparison of the results was carried out in three distinct energy ranges, namely, 1–10 MeV/u ('low energy', i.e. where relativistic corrections are minimal), 10–100 MeV/u ('medium energy') and 100–1000 MeV/u ('high energy', where, for heavy ions, having $A > 19$, the non-perturbational Lindhard–Sørensen theory has been reported to yield more accurate results [3,13]).

In all the simulations the process of multiple scattering was disabled, while, in certain cases, tables with stopping power values were generated for comparison, when available. The obtained results showed significant discrepancies for specific beam particle–target combinations in certain energy ranges. Especially at relatively low energies, deviations were observed from the SRIM2013, PSTAR and ASTAR predictions which have been thoroughly investigated and partially validated in the

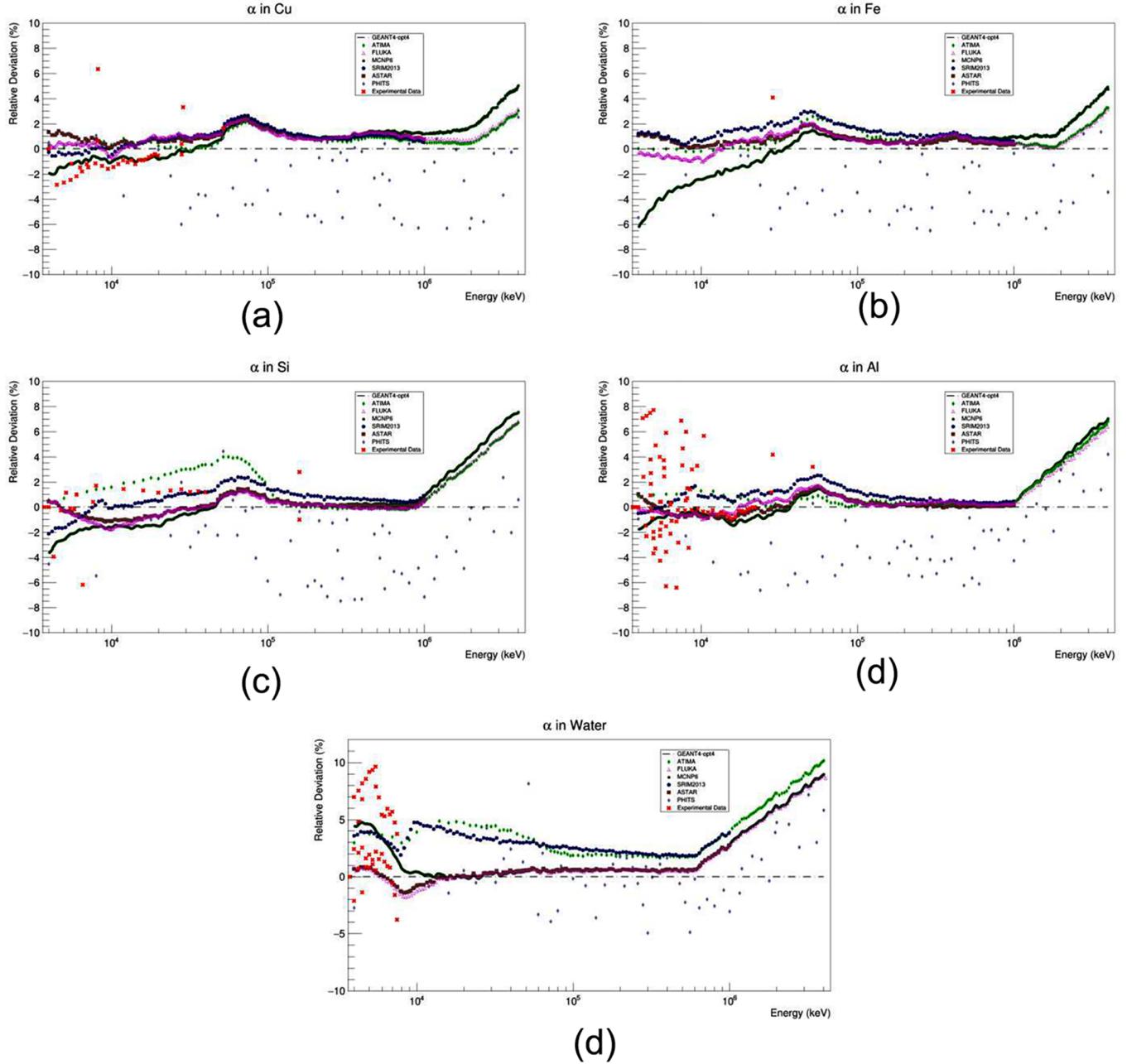


Fig. 2. a–e. Stopping power results, between 1 and 1000 MeV/u, concerning the following cases (implementing natural targets): (a) α -particles in Cu, (b) α -particles in Fe, (c) α -particles in Si, (d) α -particles in Al and (e) α -particles in water. Energies are presented in units of keV, while the y-axis denotes the relative deviations from the GEANT4 predictions in %.

past [3]. The final values are presented in graphical form and the observed similarities and discrepancies are discussed and analyzed. Nevertheless, since experimental data have not yet been fully benchmarked over a broad energy range, and for a large variety of ion beam–target combinations, the final assessment of the obtained results relies on the user. It should be stressed here that, in compliance with the FLUKA user license, no comparisons of extracted specific internal physics models are performed with data or with other codes or models (only tabular values are presented) and any validation of the Monte Carlo codes used is beyond the scope of this work.

It should also be noted that the present work is a detailed technical report aiming at complementing and extending the critical overview of stopping power programs published in 2013 [3], with respect to the most widely used materials for space applications, including all relevant experimental datasets (present in the updated IAEA repository as of 9/

2019), with particular emphasis put on the general-purpose, widely used Monte-Carlo codes and in particular GEANT4, which have been updated several times during the last six years. This is the reason for the exclusion of the purely theoretical, pioneer and widely used PASS [14] and CasP [15] codes from the present study, along with certain other specific codes included in [3]. Moreover, a comprehensive report, extending the present study to lower projectile energy values, in order to thoroughly investigate the area of the stopping power maximum and the nuclear stopping regime as well, including all the available experimental points, stopping power codes and compilations will be the subject of a future work.

2. Calculation details

There is a large variety concerning the adopted approaches for the

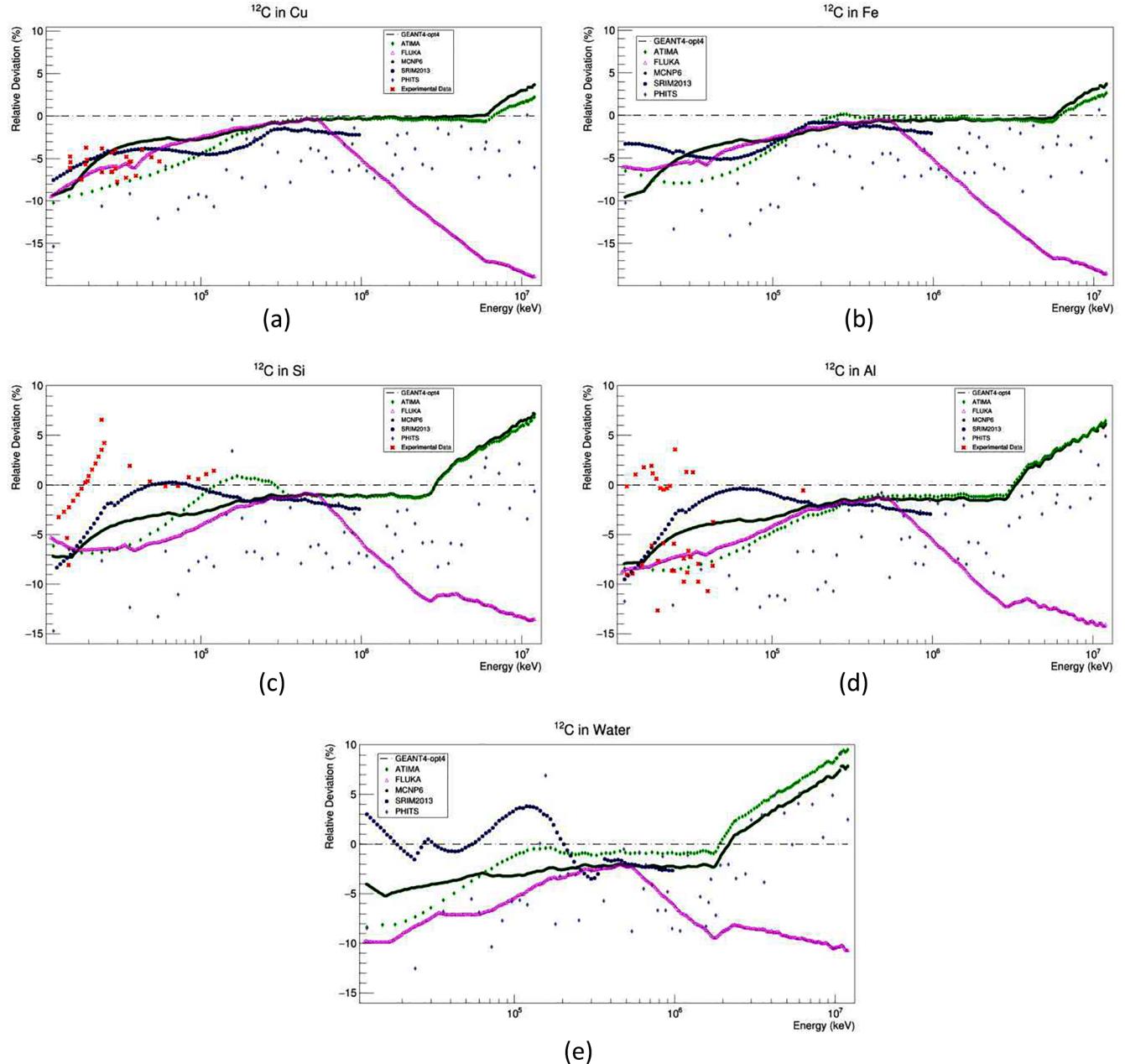


Fig. 3. a–e. Stopping power results, between 1 and 1000 MeV/u, concerning the following cases (implementing natural targets): (a) ^{12}C ions in Cu, (b) ^{12}C ions in Fe, (c) ^{12}C ions in Si, (d) ^{12}C ions in Al and (e) ^{12}C ions in water. Energies are presented in units of keV while the y-axis denotes the relative deviations from the GEANT4 predictions in %.

stopping power calculations among the existing codes and the corresponding theoretical literature is rich, therefore each studied case presented unique characteristics. More specifically:

- (a) As mentioned above, to model the transport of particles through matter, GEANT4, v.10.5.p01, incorporates several models concerning the description of the electromagnetic interactions (EM) [4]. GEANT4 results indicate that for protons the most suitable option is the G4EmStandardPhysics_option4 (Opt4) one, whereas for heavier ions all the available EM models seem to converge. For the Opt4 selection the step limit function was modified to (0.02, 0.01 μm), as it was suggested by a previous study [16] in order to improve the calculations at relatively low energies (between 1 and 10 MeV/u), while the particle production cut was kept at 1 mm. Since stopping power tables were not generated, extensive simulations were carried out varying the energy step by no more than

5% of the corresponding energy value, implementing both thin ($\sim 1 \mu\text{m}$) and infinitely thick (excluding the effect of multiple scattering – that is, energy and lateral straggling) targets divided into ultra-thin, 1 μm slices. The statistical error was kept as low as 0.1% in all cases.

- (b) In FLUKA, electronic stopping powers are computed starting from the Bethe–Bloch formalism. Several corrections to the standard formulation have been implemented in the recent years, which, according to the code's documentation, follow, with modifications, extensions and refinements, the functional forms presented by ICRU, complemented by Ziegler compilations at the lowest energies. In the present work the default values for the average ionization potentials (I-values) and density effect parameters of pure elements were adopted, as implemented in the code, without any user intervention, while the results were obtained exclusively in a tabular form.

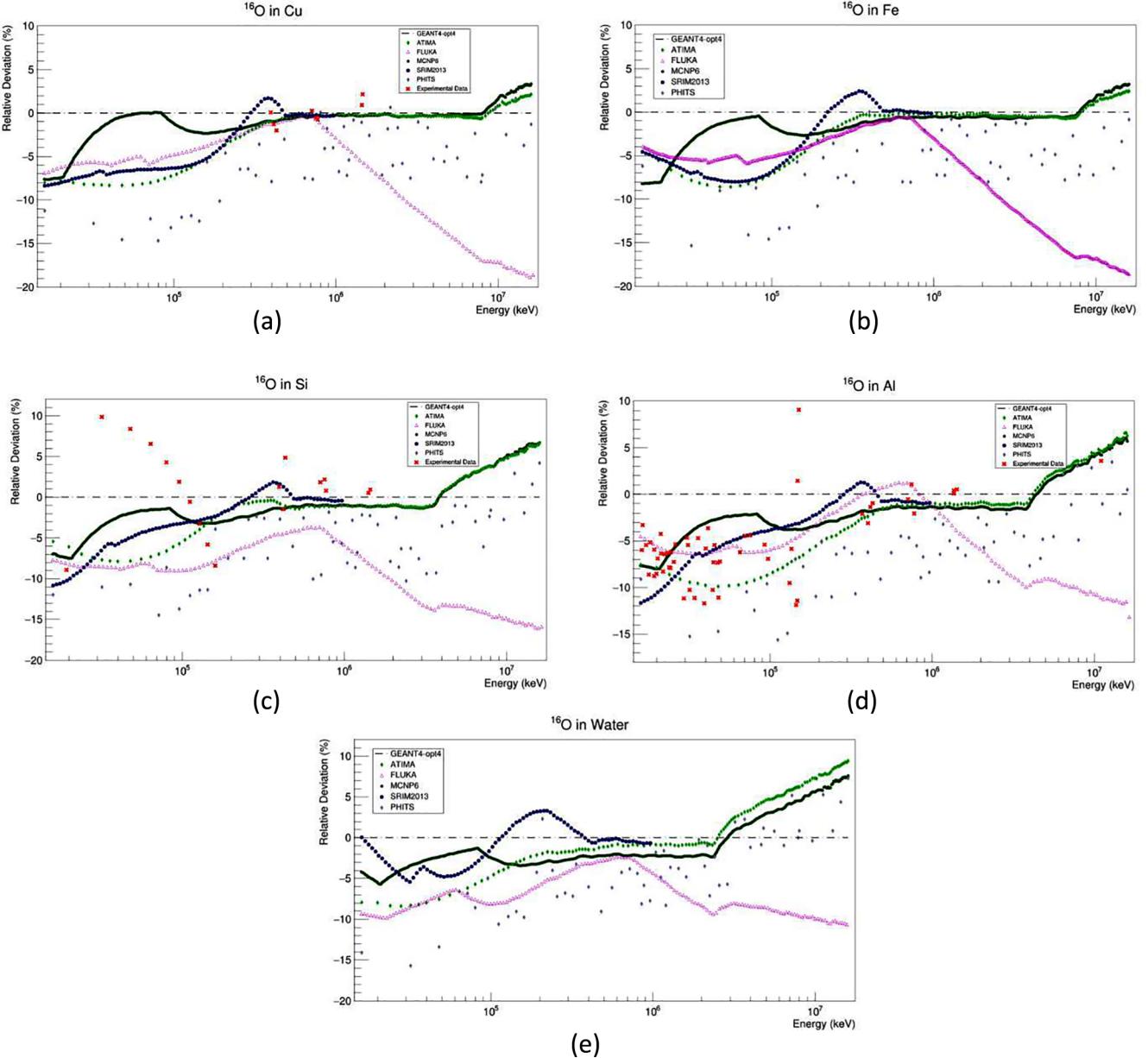


Fig. 4. a–e. Stopping power results, between 1 and 1000 MeV/u, concerning the following cases (implementing natural targets): (a) ^{16}O ions in Cu, (b) ^{16}O ions in Fe, (c) ^{16}O ions in Si, (d) ^{16}O ions in Al and (e) ^{16}O ions in water. Energies are presented in units of keV, while the y-axis denotes the relative deviations from the GEANT4 predictions in %.

(c) PHITS generally uses the code ATIMA, developed at GSI (also implemented in LISE+ + [17]), which calculates the slowing down of protons and heavy ions in matter for energies ranging from 1 keV/u to 450 GeV/u. Above 30 MeV/u the stopping power is obtained from the theory by Lindhard and Soerensen (LS) [13], as implemented in the code, including the shell corrections, a Barkas term and the Fermi-density effect. Below 10 MeV/u an older version of Ziegler's SRIM is adopted. In the intermediate energy range the code interpolates between the two. PHITS stopping power tables were not generated, and extensive simulations were carried out, as in the case of GEANT4, implementing both thin ($\sim 1 \mu\text{m}$) and infinitely thick targets divided into ultra-thin, $1 \mu\text{m}$ slices but with a larger energy step. The statistical error was again kept as low as 0.1%. In all cases, default values of the corresponding parameters were used, as implemented in the code, without any further user interventions. Additionally, tables were generated using ATIMA v.1.2 [17] and were also included in the graphs, as an independent

check for possible deviations.

- MCNP6.1 is based on a combination of stopping power results obtained from the rather old SPAR code [18], along with data tables from Janni, J. F. (1982) [19,20], using a weighting (interpolating) technique; namely at low energies SPAR prevails, while, progressively, Janni results become predominant. Results can be obtained in a tabular mode and are identical with those obtained following microscopic calculations.
- SRIM2013 stopping power values were also generated from tables, while for water, the composition, density and correction factors used were adopted from the incorporated compound dictionary. The same applied to the PSTAR and ASTAR (ICRU) codes. The calculations were performed up to the maximum beam energy allowed by the corresponding compilations.

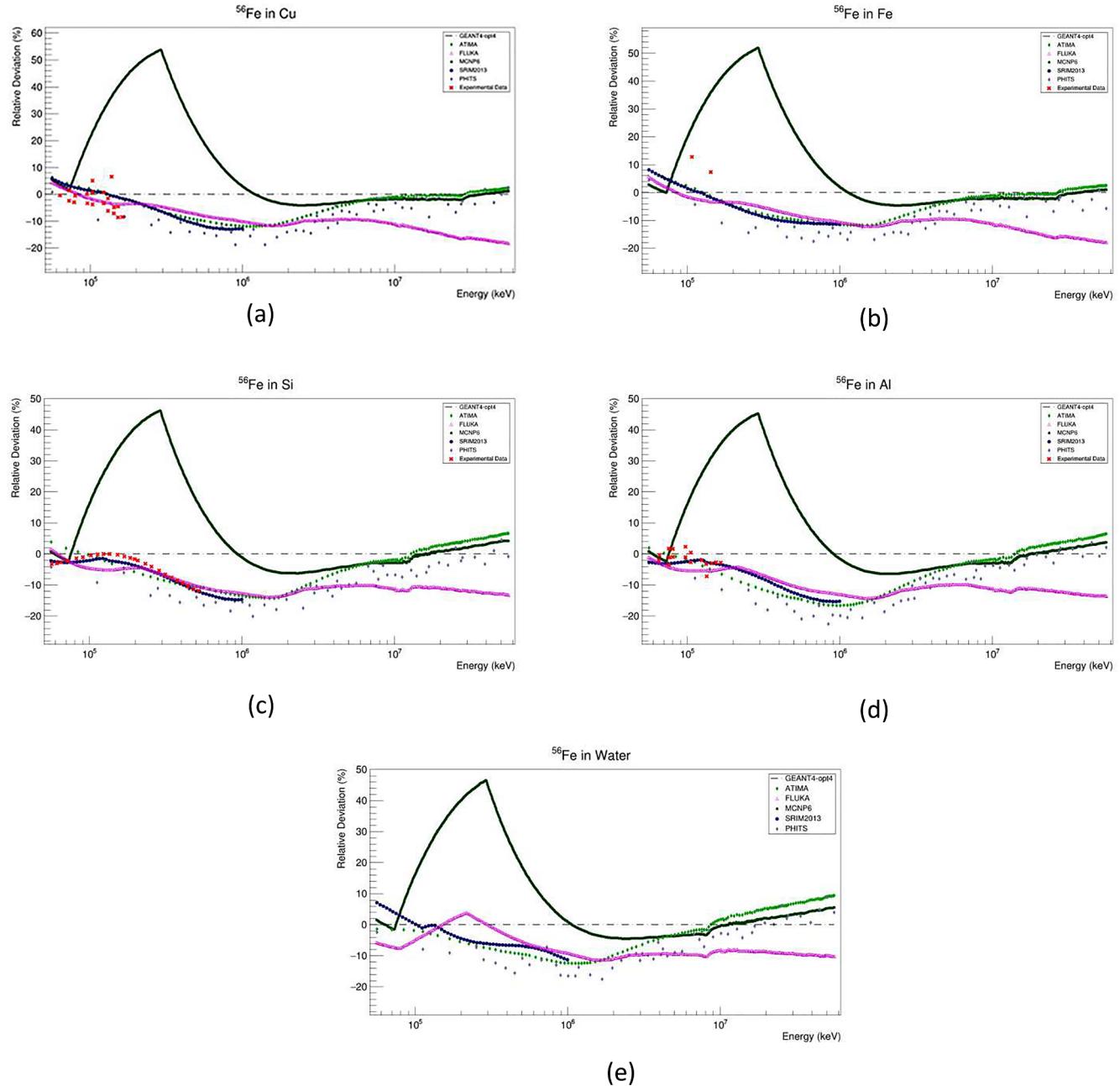


Fig. 5. a–e. Stopping power results, between 1 and 1000 MeV/u, concerning the following cases (implementing natural targets): (a) ^{56}Fe ions in Cu, (b) ^{56}Fe ions in Fe, (c) ^{56}Fe ions in Si, (d) ^{56}Fe ions in Al and (e) ^{56}Fe ions in water. Energies are presented in units of keV, while the y-axis denotes the relative deviations from the GEANT4 predictions in %.

3. Results and discussion

The obtained results in the case of protons as impinging ions are presented in Fig. 1a–e. All values are depicted in the form of relative deviations (RD in %) from the GEANT4 (G4) predictions, namely, the y-axis corresponds to $\text{RD} = 100 [\text{S(i)}-\text{S(G4)}]/\text{S(G4)}$, where S(i) denotes the stopping power obtained from each MC code or compilation, while the corresponding S(G4) values were obtained using the more accurate TSpline3 fitting curve model for the interpolations (<https://root.cern.ch/doc/master/classTSpline3.html>). Experimental data points for all targets have also been added in the graphs, when available. As reported in [3] there has been a considerable trend towards convergence of the corresponding stopping power values between all the MC codes over the last decade and, as a result, differences in the high energy region (100–1000 MeV/u) did not exceed 8% for low (Al, Si), 5% for high-Z

(Fe, Cu) targets, and 8% for water, (with microscopic calculations implementing PHITS being the only exception, yielding differences up to 10–20% in specific ion beam–target combinations). The medium energy region (10–100 MeV/u) was the area of the best convergence among all MC codes and compilations yielding differences less than 5% among them, again with the exception of PHITS. In the low energy region (1–10 MeV/u), for high- and low-Z targets the agreement was generally within 5%, with the PHITS microscopic calculations converging within 10%, while for the specific case of water, MCNP6 seemed to overestimate the average prediction of GEANT4 by 5–6%, while SRIM2013 underestimated accordingly by ~5–10%. The experimental data agree with these MC simulations within 5% for the atomic targets, except for Al below 2 MeV. In the case of liquid water, Fig. 1e shows an underestimation of the stopping power in the data at $E < 2$ MeV. This fact has already been noted and analyzed before in literature [21].

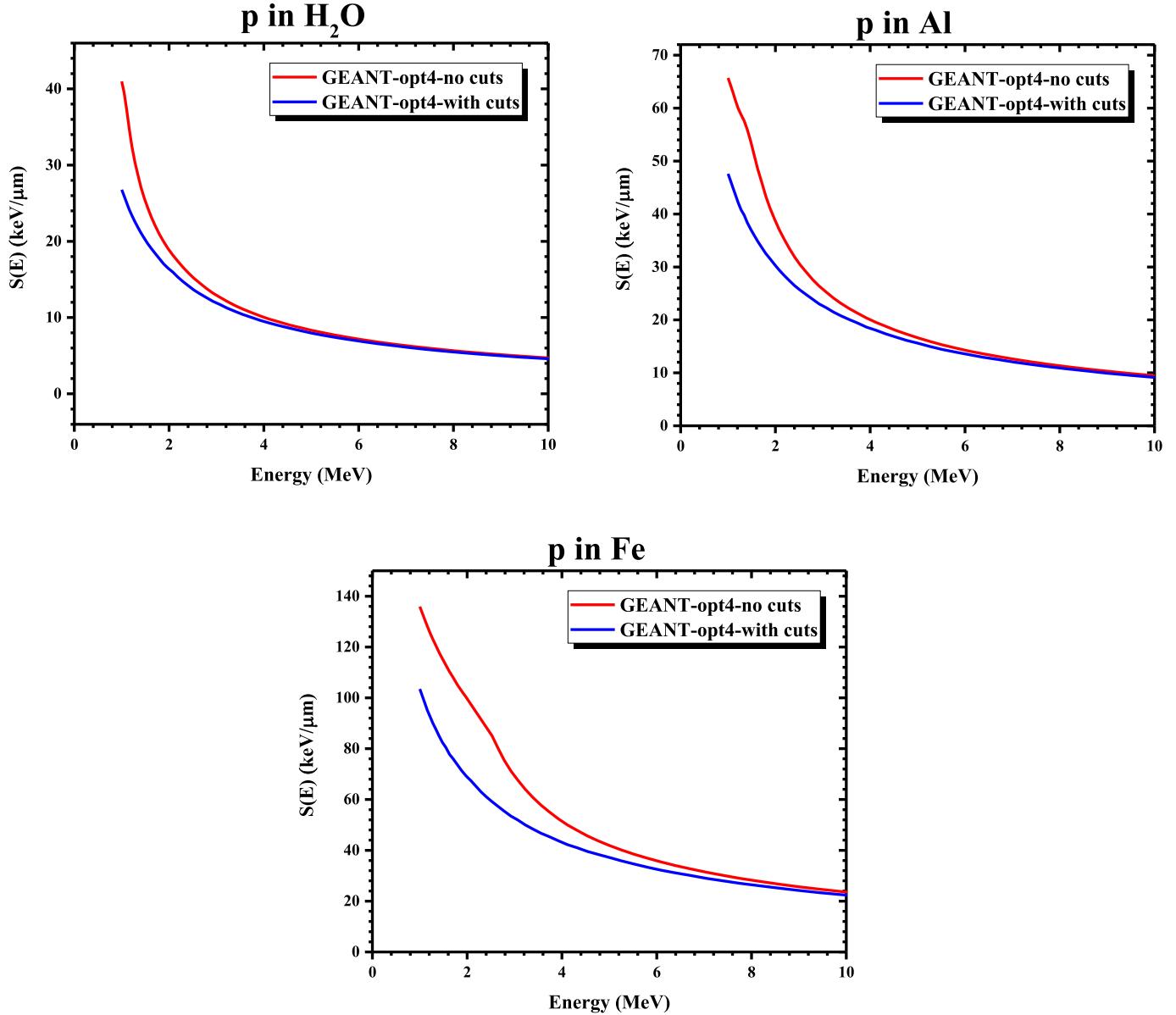


Fig. 6. a–c. Comparison between GEANT4 electromagnetic physics list models, namely, Opt4 with default values for the cuts and Opt4 with the cuts recommended in [14], in the case of: (a) protons in water, (b) protons in aluminum, (c) protons in iron.

The quite satisfactory agreement between the stopping power values continued in the case of α -particles as impinging ions, as demonstrated in Fig. 2a–e. Once again, differences in the high energy region (100–1000 MeV/u) did not exceed 5–8% for low-Z (Al, Si) and high-Z (Fe, Cu) targets, and 10% for water, (with microscopic calculations using GEANT4 and PHITS yielding systematically lower values in this case). In the medium energy region (10–100 MeV/u) the differences were less than 4% (with the exception of PHITS yielding systematically lower values up to 8%), while in the low energy region (1–10 MeV/u), for high-Z targets the agreement was better than 5–6% and for low-Z ones around 4%. For the specific case of water the variation was within 5%. Concerning the experimental points in the low-energy region, for the case of α -particles impinging on iron, only one experimental point could be retrieved at 28.8 MeV and considerable scattering of the available values was generally observed for aluminum and water. This variation was not so pronounced in the cases of copper and silicon.

The situation slightly worsens as the Z of the beam ion increases. More specifically, as depicted in Fig. 3a–e, in the case of ^{12}C ions, which are particularly important for space and medical physics applications,

although differences in the high energy region (100–1000 MeV/u) did not exceed 7–10% for all the targets (with the exception of FLUKA which yielded systematically lower values by 6–19%, with the deviations increasing with energy) and in the medium energy region (10–100 MeV/u) the differences were again less than 5–8% (with GEANT4 systematically overestimating the results – except for the case of water – and PHITS underestimating them by up to 15%), in the low energy one (1–10 MeV/u) the simulated results deviated considerably. In particular, GEANT4 generally yielded higher values than the majority of the other codes, with copper being the most striking example, where the corresponding deviations reached values up to 8–9%. In the case of water (particularly important for hadron therapy applications), in the absence of experimental data, the overall maximum spread between the simulated values reached \sim 10–13% in the low-energy part. Experimental data could not be retrieved for the case of iron as well, while, for low-Z targets (Al, Si) the bulk of the experimental data were also quite scattered and agreed only within 15%. GEANT4 generally tended to overestimate the majority of the experimental values by \sim 5–10% (as e.g. in copper), while PHITS underestimated them

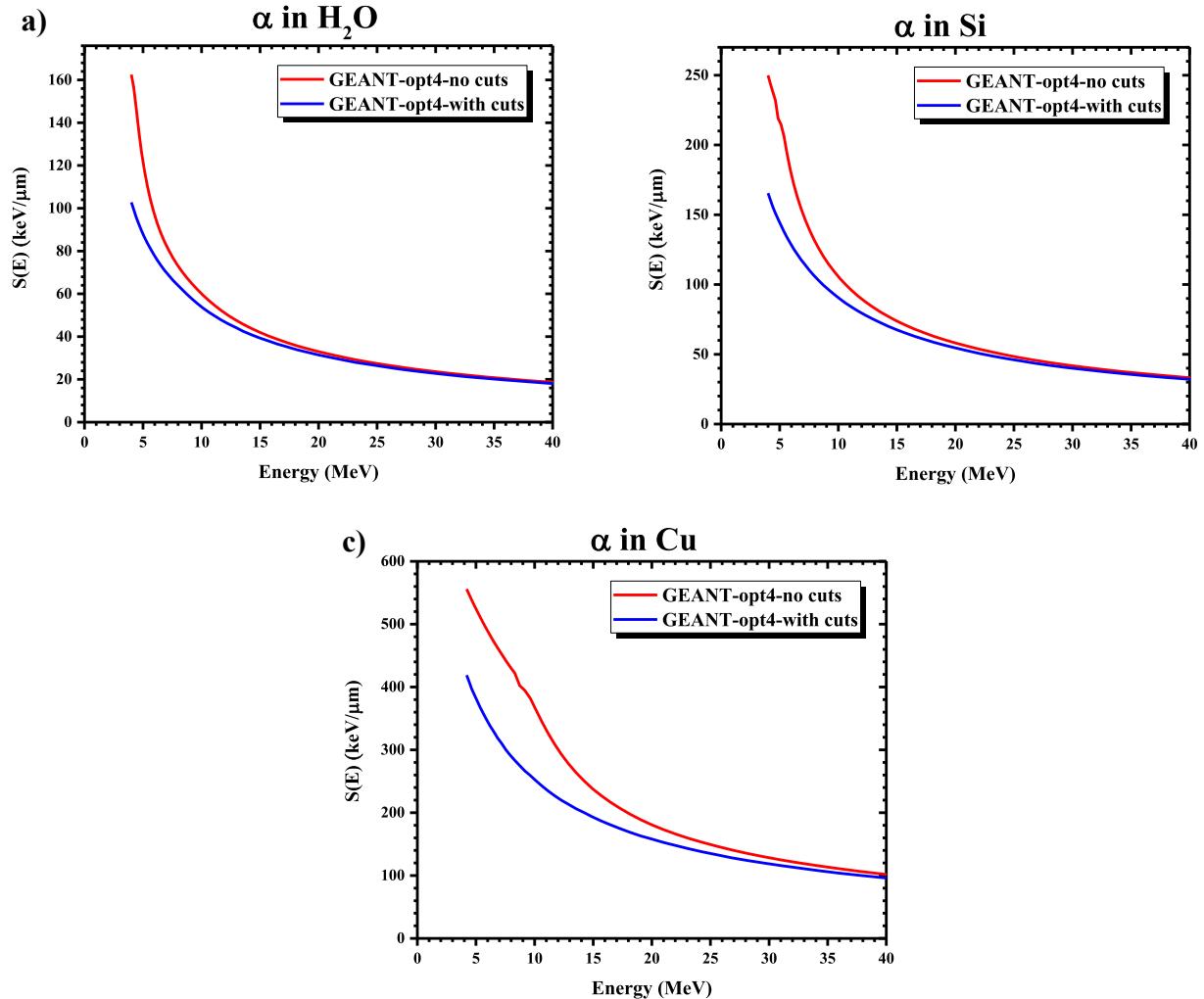


Fig. 7. a–c. Comparison between GEANT4 electromagnetic physics list models, namely Opt4 with default values for the cuts and Opt4 with the cuts recommended in [14], in the case of: (a) α -particles in water, (b) α -particles in silicon. (c) α -particles in copper.

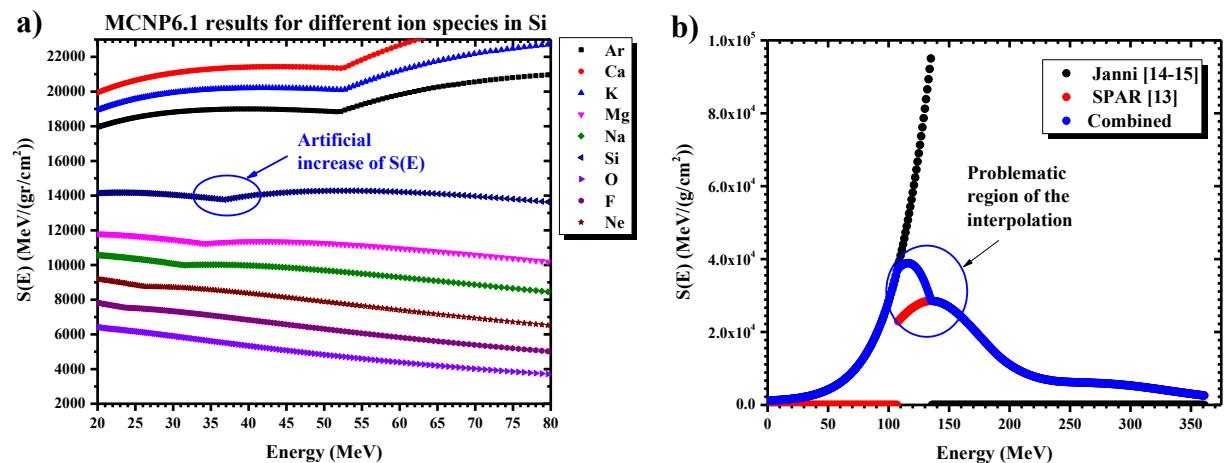


Fig. 8. a–b. (a) Stopping power value trends in MCNP6.1 for heavy ions with $Z > 8$ (b) An example of the actual interpolation between the SPAR [16] and Janni tables [17,18] in the case of ^{56}Fe ions impinging on Al.

accordingly by 5–10%.

In the case of ^{16}O ions the same trend was verified, as shown in Fig. 4a–e. Deviations in the high energy region (100–1000 MeV/u) reached 7–9% for low (Al, Si) and high-Z (Fe, Cu) targets, and ~10% for water, (with FLUKA yielding systematically lower values by ~10–20%).

In the medium energy region (10–100 MeV/u) the differences were less than ~5–10% in all cases, with GEANT4 yielding systematically slightly higher values, while in the low energy region (1–10 MeV/u), for high-Z targets the agreement was within 14–15%, with PHITS yielding systematically lower values, for low-Z within 15–16% and for the specific

case of water again within 15%. Concerning the experimental points, in the cases of copper, silicon and aluminum the available values in the medium energy region agreed quite well with all the MC results. However, for the latter two targets, considerable scattering was observed in the low-energy part. No data points could be retrieved in the cases of iron and water.

The case of ^{56}Fe ions, important for cosmic-ray studies, was by far the most complicated one, especially due to the limited amount of experimental data related to all the projectile-target combinations studied in the present work, as retrieved from the IAEA repository. Excluding the weird artifact that appears in MCNP6.1 stopping power tables and microscopic calculations, which will be analyzed in a following paragraph, for low-Z targets, as shown in Fig. 5a–e, deviations among the simulation results in the high energy region were as high as $\sim 15\%$, in the middle one $\sim 20\text{--}25\%$ (with GEANT4 values being systematically higher) and in the low energy region around $\sim 10\text{--}15\%$. For high-Z targets the situation was similar, namely deviations were up to $\sim 20\%$ in the high energy region, slightly less than 20% in the middle and $\sim 10\text{--}12\%$ in the low energy one. For the specific case of water, the deviations reached $\sim 10\text{--}12\%$ in the high energy region, $\sim 15\text{--}20\%$ in the middle and $\sim 10\text{--}15\%$ in the low energy one. Moreover, in all cases, FLUKA results at high energies underestimated the bulk of the simulated data by $\sim 20\text{--}25\%$, depending on the case. Concerning the experimental points in the low-energy part, the agreement with the simulated results was quite satisfactory for copper and aluminum, mediocre for iron, while for silicon, the only existing dataset seemed to be in excellent agreement with SRIM2013 and ATIMA, in quite satisfactory with FLUKA and PHITS, and in mediocre with GEANT4. Once again, no experimental data could be retrieved for water.

It should be noted here that for heavy ions (e.g. ^{12}C , ^{16}O and ^{56}Fe) all the EM options in GEANT4 yielded practically the same results, while the modification in the cuts suggested in [16] did not affect the obtained values. This is the reason why default values for Opt4 are presented in the corresponding graphs. On the other hand, the reported modification is very important in the case of protons and α -particles, as shown in Fig. 6a–c for protons impinging in water, aluminum and iron and in Fig. 7a–c for α -particles impinging in water, silicon and copper respectively. GEANT4 users should be very careful when implementing default values for the cuts in stopping power calculations, since for protons differences ranged from $\sim 32\%$ for high-Z targets, to 52% for low-Z ones and $\sim 40\%$ for water, for energies around 1 MeV/u (and less than 10 MeV/u, since above this value both approaches converged), while for α -particles and for the same energy range, differences ranged again from $\sim 32\%$ for high-Z targets, to 52% for low-Z ones, but exceeded $\sim 60\%$ in the case of water.

Another intriguing result concerned the behavior of MCNP6.1 calculations in the case of heavy ion beams, which was mentioned above. As demonstrated in Fig. 8a, for different beam ion species having $Z > 8$, there appears an artificial increase in the stopping power values, a seeming discontinuity in the normal decreasing trend of $S(E)$ with energy (above 1–2 MeV/u), whose position varies with the beam energy and the Z (and consequently the mass) of the selected ion. This obvious discrepancy could be attributed to the partial overlap of the Janni [19–20] and SPAR [18] tables at low energies ($\sim 1\text{--}3$ MeV/u) which, for heavy ions, as shown in Fig. 8b, can lead to strange artifacts in the combined interpolated values.

4. Conclusions

In the present work a comparative study of stopping power calculations between widely used MC codes and compilations with experimental data has been presented in the energy range of 1–1000 MeV/u. Several projectile-target combinations revealed a good to quite satisfactory agreement between the codes in the cases of protons, α -particles and ^{12}C ions, but with increasing discrepancies for higher-Z ones,

such as ^{16}O and –mainly– ^{56}Fe , for which further experimental investigation is required, along with a proper subsequent tuning of the corresponding model parameters.

It is important to stress here, that since there do not exist benchmarked experimental stopping power datasets over a considerably broad energy range and for a large variety of ion beam–target combinations, the final assessment of the obtained results relies on the users, who should constantly exploit the new stopping power data compilation/repository developed under the auspices of IAEA for updates, especially for heavy ion projectiles and high energies [22]. It is also evident from the present report, that despite the tremendous progress achieved so far, a coordinated effort, both on the experimental and on the theoretical level is still required before a complete convergence of MC codes in the problem of stopping power values is accomplished.

CRediT authorship contribution statement

E. Vagena: Methodology, Software, Writing - review & editing. **E.G. Androulakaki:** Methodology, Software, Writing - review & editing. **M. Kokkoris:** Conceptualization, Methodology, Writing - original draft, Writing - review & editing. **N. Patronis:** Conceptualization, Methodology, Writing - review & editing. **M.E. Stamati:** Writing - review & editing, Visualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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