Mean excitation energy determination for Monte Carlo simulations of boron carbide as degrader material for proton therapy

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1. Introduction

Proton therapy is a form of radiation therapy whose benefit in particular is the fact that protons release most of their energy at a specific depth (the so-called Bragg peak) to ensure maximum conformity of the deposited dose distribution to the tumour volume. State-of-the-art facilities use the so-called pencil beam scanning (PBS) technique \cite{1–3}, in which a pristine pencil beam of a defined energy is directed at a specific location of the target volume using steering magnets. For cyclotron-based facilities (which correspond to more than two-thirds of the proton therapy facilities worldwide, as of February 2020), the beam exits the accelerator with a fixed high energy, typically between 230 MeV and 250 MeV. To adjust the proton range to that required for a given treatment layer depth, the proton beam passes through an energy modulation system, which consists of material placed on the beam path either immediately before the patient or upstream of that soon after the cyclotron extraction. For upstream energy variations, such a system is called a degrader. Proton energies down to 70 MeV are routinely used in clinical treatments; the required degrader thicknesses for such an energy reduction are several centimetres, and consequently induce significant lateral scattering and momentum spread, which is then reduced using sets of collimators and a so-called energy selection system (ESS) \cite{4}.

Boron carbide is a material proposed as an alternative to graphite for use as an energy degrader in proton therapy facilities, and is favoured due to its mechanical robustness and promise to give lower lateral scattering for a given energy loss. However, the mean excitation energy of boron carbide has not yet been directly measured. Here we present a simple method to determine the mean excitation energy by comparison with the relative stopping power in a water phantom, and from a comparison between experimental data and simulations we derive a value for it of $83.1 \pm 2.8$ eV suitable for use in Monte-Carlo simulation. This is consistent with the existing ICRU estimate ($84.7$ eV with 10–15\% uncertainty) that is based on indirect Bragg additivity calculation, but it has a substantially smaller uncertainty. The method described can be readily applied to predict the ionisation loss of other boron carbide materials in which the atomic constituent ratio may vary, and allows this material to be reliably used as an alternative to graphite, diamond or beryllium.

Additionally, inelastic scattering and nuclear interaction processes cause a further reduction of the flux of particles, for example converting some of the incident protons into neutrons. The whole process introduces substantial beam losses, and the transmitted beam to the patient at lower energies is often less than 1\% of the extracted current at the cyclotron. The losses lead to significant requirements when shielding the cyclotron and ESS, may cause activation of material along those parts of the beamline and radiation damage to equipment, and pose strong limits in the possible current delivered to the patient; treatment times and consequently overall costs will be affected. As the interest in the field of radiation therapy is shifting towards using much higher dose rates for so-called FLASH irradiation \cite{5,6}, the ability to deliver larger beam currents to the patient may become a priority in the not too distant future. For this purpose, the choice of the degrader material is of the highest importance. Different materials result in different beam sizes and scattering angles, and especially materials with a low atomic number ($Z$) may offer greater transmission for a given exit energy from the degrader, because of the smaller lateral scattering for a given ionisation energy loss \cite{7,8}. Graphite is the most common degrader material as it combines a reasonably low atomic number ($Z = 6$) with good mechanical robustness, economical cost and reasonable density. Beryllium ($Z = 4$) has been used but is unpopular due to the potential for...
toxicity when manufacturing and handling degraders. Boron carbide (B₄C, Z = 5.6) is mechanically similar enough to graphite but its lower average Z is advantageous at lowering lateral scattering. In addition, several innovative designs have been proposed with the goal of limiting the beam size growth due to the degrader [9,10].

Standard beam dynamics codes that simulate particle transport (such as TRANSPORT/TURTLE [11] or OPAL [12]) are not aimed at precise estimation of beam losses and optimisation of degrader materials considering particle-matter interactions [13]. Usually such codes lack precise scattering cross sections (particularly inelastic ones). Monte-Carlo particle interaction codes – such as those based on GEANT4 [14–16] – can provide more precise predictions of degrader physics and other relevant phenomena when studying degraders [17,8]; however, they rely on the precise definition of the different characteristics of the materials under study, which are not always well-known and therefore can introduce significant uncertainties in any predictions [18]. Earlier studies of graphite and beryllium as degrader materials [17] have highlighted the importance of the mean excitation energy $I_Z$ in accurate predictions of the ionisation slowing, and matching of simulations with experimental data by tuning $I_Z$ is not always consistent with reference data such as those reported by the International Commission on Radiation Units and Measurements (ICRU) [19,20]. An incorrect use of $I_Z$ directly leads to incorrect definitions of other quantities such as the material thickness, that in turn influences the predicted lateral scattering; a precise determination of $I_Z$ matched to the Monte-Carlo model used is required to obtain reliable calculations of other simulated phenomena. On the other hand, a direct measurement of $I_Z$ is very challenging, as it requires precise knowledge of several correction factors, as recently highlighted in Seltzer et al. [20].

We are currently investigating the potential of B₄C as a degrader material to be used in the Paul Scherrer Institute Centre for Proton Therapy (PSI) [21]. B₄C is a material in which the B to C atomic ratio can vary significantly [22], and it is important to carefully quantify that ratio, its homogeneity and the mean density in any given sample used. In the context of Monte-Carlo degrader studies, we are proposing a method to determine the value of $I_Z$ from experimental measurements of a given degrader or other material sample. With our study, we aim at answering the following questions:

- How can we determine the $I_Z$ parameter for a material, keeping the uncertainties under control?
- Can such a method be reliably and easily applied in a clinical facility, to help different institutes setting up their own simulation environments?

In this paper, we demonstrate a simple method that for the first time derives an estimate of $I_Z$ for B₄C from a comparison with experimental data. Such a value is suitable to use for Monte-Carlo predictions and design. The method described below – based on a relative measurement versus a water phantom – may be straightforwardly used for other material samples, and we have applied it here also to conventional graphite degrader materials.

2. Materials and methods

2.1. B₄C as a degrader material in the PSI proton therapy system

An optimal combination of low atomic number (to minimise multiple scattering angle) and sufficiently high density (to minimise the amount of material needed for the energy degradation) is the goal when choosing degrader material [7]. The present degrader material used at PSI is graphite, with density $\rho = 1.870 \pm 0.005$ g cm⁻³, and is used to degrade the energy for treatments in the range 70–230 MeV. But due to the losses introduced in the degrader and ESS sections, in the PSI proton therapy beamline the overall transmission efficiency at the lowest beam energy of 70 MeV – from the cyclotron extraction to the treatment nozzle – is only around 0.2%, comparable to other similar facilities. We are also investigating an alternative candidate material – boron carbide (B₄C); the lower Z together with the higher density ($\rho = 2.52$ g cm⁻³) should lead to lower emittance growth after the degrader in comparison to graphite, and thereby could enable treatments with proton energies below 70 MeV at an acceptable dose rate.

B₄C is the third-hardest material known (after diamond and cubic boron nitride), and as such is used in such applications as body and vehicle armour and to coat cutting tools. It can also absorb neutrons without forming long-lived radionuclides, and is widely used in nuclear reactor control rods, shielding materials and for neutron detection [23]; in particle physics it has also been investigated for use in muon experiments [24]. A first experiment with B₄C at PSI showed promising results, indicating a transmission increase of more than 30% using B₄C with respect to graphite, although with a simplified degrader geometry and not at the lowest 70 MeV energy [21]. A follow-up study is in progress that examines a wider range of energies and uses the PSI double-wedge degrader geometry.

The mean excitation energy of B₄C is presently known only from an estimate by Seltzer and Berger [19], who use a 13% correction to the value predicted by the Bragg additivity rule; $I_Z$ was estimated in that work to be 84.7 eV. This estimate and the correction is however based upon experimental values from other compounds – and particularly compounds not even containing boron – and has never been validated experimentally. Uncertainties on this value of $I_Z$ are conservatively estimated to be somewhere from 10% to 15%; this estimate is reported in the ICRU tables and in several simulation codes (including GEANT4).

Variation of the B:C ratio from 3.5:1 to 10.5:1 gives a variation of the additivity-derived $I_Z$ value of 0.7 eV. Several blocks of B₄C were manufactured using hot powder pressing by an industrial supplier; the manufacturer estimate of B:C atomic ratio was 78.3%:21.7%, slightly less than the nominal 4:1 ratio. Porosity was estimated by the supplier to be below 0.5% by volume; we independently assessed the quality of the samples (manufacturing defects, uniformity and density variations) by carrying out computed tomography (CT) scans. The observed Hansfield unit variation in the CT scans were at the level of the scanner noise – indicating good homogeneity – and there were no observed voids or other significant defects. We used these samples in tests in the PSI Gantry 2 treatment room, to estimate the mean excitation energy of the material from range measurements. The experimental setup for the measurements is explained in Section 2.2, whilst the method used for the estimation of $I_Z$ is explained in Section 2.4.

2.2. Range scanner measurements at PSI Gantry 2

We used an in-house built water phantom – the so-called Range Scanner (RS) shown in Fig. 1 – to measure the Bragg peak profile as a function of depth in water. The scanner box is made of Plexiglass and allows continuous movement of an ionisation chamber through the contained water over a range of 450 mm and with a resolution of ±0.025 mm, using a motor controlled by an in-house-programmed LabView interface. Several chamber holders may be mounted inside the scanner that allow for measurements using a variety of ionisation chambers; in our experiment we mounted as measurement chamber a large, plane-parallel ionisation chamber (80 mm sensitive area diameter), the same as used for the range commissioning and quality assurance (QA) tests for treatments at our centre.

The LabView interface allows the user to define the step resolution for the measurement, allowing the combination of regions of coarser resolution (for example, in the buildup region) with others of finer resolution (for example, around the Bragg peak); this gives flexibility and efficiency in taking the data. The normalization of the dose

1 Ceratec Technical Ceramics, www.ceratec.nl
measured along the peak is done using a reference chamber. At PSI Gantry 2 we use the dose monitor in the gantry nozzle as a reference chamber; in the LabView interface it is possible to define a preset number of monitor units (MU) for the reference chamber, and the measurement system will accumulate the integrated charge (IC) collected in the measurement chamber while the reference chamber reaches that preset. The charge collected as a function of position of the measurement chamber in the RS is used to produce the Bragg peak profile. The fall-off of the curve is then fitted with a cubic spline interpolation and the range is extracted as the 80% point from the peak height (to make this better visible to the reader, we normalised all plots in the paper to the peak height). In our present measurement, we used the same settings for preset and step resolution as in our yearly QA tests, to ensure good quality of our measurements.

The measurements were performed in the Gantry 2 treatment room; therefore the beam characteristics are the same as described in Pedroni et al. [25], with typical spot sizes (σ) at the isocentre of around 2.3 mm at 230 MeV. The nominal beam energy is defined by range measurements performed with the RS and matched to the ICRU 49 tables [26]. The energy spread (around 0.7%) is estimated from the energy selection system acceptance and from matching to measured Bragg peak profiles.

### 2.3. GATE/GEANT4 simulations

GEANT4 is a toolkit for Monte-Carlo simulation of particle interaction with matter. Its use case mainly originated in high-energy physics, but its models have been expanded to cover a broad range of particle types and energies, including applications in space radiation, cosmic ray modelling, nuclear physics, heavy-ion and radiation physics, and medical applications. For the simulations described in this paper we used the GATE application for GEANT4 [27,28]. Although initially conceived for imaging applications, GATE has been developed to allow radiotherapy applications, including proton and hadron therapy [29]. We use the term ‘GEANT4’ for brevity, meaning our simulations developed using GATE and GEANT4; we used GATE version 8.1.p01 and GEANT4 version 10.04 patch 2.

Results obtained using GEANT4 simulations are dependent on the physics models employed and by tuning parameters such as range cuts and energy limits. We followed the recommendations of the GATE collaboration - summarised in Grevillot et al. [29] and updated in Fuchs et al. [30] and Resch et al. [31] for scattering validation – to fix the parameters and the physics processes used. GEANT4 provides different electromagnetic transport methods with varying precision; we used G4EmStandardPhysics_option4 (also called EM2) as it provides the greatest precision [32] for the low-energy regime. As noted for example by Sabin et al. [33], a particular mean excitation energy I₂ must be matched to the correct physics model in GEANT4 to give a reliable energy loss estimate. We used the settings of Grevillot et al. [29] for the Bethe-Bloch energy loss, which can be used down to 2 MeV; below that a parameterisation based on ICRU 49 is implemented. This list uses the highest precision low-energy models (G4KleinNishinaModel, G4LowEPComptonModel, G4PenelopeGammaConversion and G4GoudsmitSaundersonModel). For proton multiple scattering simulations, it has been shown that the models available in the GEANT4 physics lists show different degrees of agreement with experimental data; Fuchs et al. [30] have shown that the WentzelVI model for multiple Coulomb scattering (MCS) results in a better agreement with data published in the literature; the WentzelVI model has been the default MCS model used in the EMZ option since version 10.02 of GEANT4, and is used also by us in the present work. For nuclear models, we also followed the recent recommendation of Resch et al. [31] and used the QGSP_BIC_HP physics list for our simulations; this list contains elastic scattering cross sections that have been shown to provide a better agreement with experimental data than other models (for example QBB) for the energy range of interest of our simulations (between 60 MeV and 250 MeV). However, the difference between the two physics lists is mainly in the predicted transverse dose profiles, and – since we are integrating the dose over a large area transverse to the beam direction – we don’t expect a large impact from the choice of the elastic scattering model. The additional option ‘HP’ used with this physics list ensures that high-precision neutron models and cross sections are used in the simulation.

GEANT4 simulations can also be affected by several other parameters; in particular, the production threshold for secondary particles (electrons/positrons and gammas) can be defined as a distance (or range cut-off) that is internally converted to an energy for individual materials. Production thresholds are defined for a given geometrical region. We fixed the production threshold at 1 mm for the region surrounding the samples and the water phantom, and to a lower value for the target materials and the water phantom where a better precision is needed. The impact of this lower production threshold was verified using simulations without degrader samples, by checking that the 80% proton range in water does not change as the threshold is lowered. A lower production threshold of course increases computing time, and therefore a higher production threshold is to be preferred for computational efficiency. We observed a 0.02 mm change in predicted proton range when varying the production threshold from 0.01 to 0.001 mm, for an 80 MeV simulated beam onto a water phantom; reducing the production threshold comes at a great additional expense in computing time (from 128 primaries/s.

Fig. 1. Experimental setup used for the estimation of I₂ for BαC: a) the PSI range scanner phantom; b) the samples mounted in front of the range scanner (the films indicate the beam size of the 230 MeV beam used in the experiment).
for 0.01 mm production cut to 1.8 primaries/s for 0.001 mm production cut). We chose an intermediate 0.005 mm production cut for our simulations, to minimise the impact of the production cut on the resulting range but at the same time to keep the computing time reasonably low. We chose an intermediate 0.005 mm production cut for our simulations. Due to irregularities in the shapes (presence of grooves etc.), checking the consistency of the volume between all samples proved difficult. So, to estimate conservatively the impact of possible density variations on the measurements, we compared ranges measured with the beam impacting on a different position on a sample and by using two samples, and considered the resulting difference as a density uncertainty.

The Bragg curves and resultant proton range predicted from simulations depend on a variety of parameters [18], and uncertainties in variables or changes to the physical models used to determine the uncertainty of the range. A variation of the used value of the $I_z$ for water, or the choice of physics list and production cuts, gives a systematic shift in the predicted range. However, when calculating $\Delta R$ these systematic shifts are expected to cancel out. The same is true when considering the uncertainty on the incident proton energy; we consider this briefly in Section 3.1, but again the effect of (small) variations of the incident energy upon $\Delta R$ is also small. In contrast, uncertainties such as the precision with which the thickness of the degrader sample is known, the density variation within that sample, and statistical uncertainties in the Monte-Carlo simulation itself do not cancel in $\Delta R$ and therefore contribute to the final uncertainty.

To validate our method and to investigate its limitations we applied it also to a graphite sample, here a spare degrader as used in the current PSI degrader, the results are reported in Section 3.4.

3. Results

3.1. Benchmarking of GATE/GEANT4 simulations of the experimental setup

The experimental setup corresponds to the setup used during commissioning and yearly QA measurements of the clinical beam line of PSI Gantry 2; therefore, we benchmarked our simulations against the last yearly QA measurements, performed only three months before our experiment. Fig. 2 shows a comparison between four simulations of the PSI Gantry 2 beam in the water phantom and how they compare with the measured Bragg curves; we observe a good match in the peak width when using the known energy spread of 0.7%, whilst the range predicted by the simulation is slightly underestimated with respect to the data; the mismatch is corrected by using a mean beam energy in simulation that is 0.35 MeV lower than the nominal beam energy expected at the entrance to the experiment (note that proton energy loss in the intervening air is already taken account of). We repeated the Bragg curve measurements for a 230 MeV beam incident on the same day of the experiment – without the degrader samples – and found a excellent match between those two curves; the measured difference in the fitted 80% range

![Fig. 2. Benchmarking of GATE/GEANT4 simulation setup, showing the variation of relative dose (for the data, obtained from the integrated charge measurement) with water-equivalent range (WER). The beam input parameters (energy and energy spread) were validated against Bragg peak measurements in a water phantom. The nominal beam energy of the beam in the measurement is 230 MeV, based on comparison of the measured range with the ICRU 49 [26] range/energy table. The range/energy points in the measurement corresponded to an equivalent 229.65 MeV energy in the GEANT4 simulation.](image-url)
between data and simulations is 0.02 mm, similar to the RS resolution.
This shows that variation in measured range is small. We investigated
whether the range mismatch with the simulations arises from the scat-
tering model used, and/or from the chosen value of the $I_Z$ for water. As
shown in Fig. 3a, the scattering model does not impact the range
measured in our experimental conditions. However, the choice of the
mean excitation energy of water used in the simulation does. The
nominal beam energy is defined by matching the measured ranges with
the ICRU 49 report [26], which uses an $I_Z$ of 75 eV for water; GEANT4
uses 78 eV, so the discrepancy in range is due to this difference in $I_Z$, as
shown in Fig. 3b. However, since the method we use to determine the $I_Z$
of the B$_4$C sample is based on the differential measurement $\Delta R$, the
additional uncertainty caused by the range mismatch will cancel out
almost completely. For this reason, we decided to match the Bragg
curves between measurements and simulations by adjusting the energy
of the input beam in the GEANT4 simulation.

3.2. Range measurements with and without degrader samples at PSI
Gantry 2

Fig. 4 shows an example of the measured curves with the range
scanner—both with and without the B$_4$C degrader sample—demon-
strating the shift in the Bragg peak when the B$_4$C sample is placed in the beam; measurements with either of our B$_4$C samples gave almost the same range, and the difference was used in the
density uncertainty estimate. We optimised the number of points
collected along the Bragg curve for efficiency, using a coarser resolution
in the plateau region and a finer resolution around the peak. Each point
corresponds to a fixed number of monitor units ($100,000, about 9.2 \times
10^8$ protons), from which the dose is obtained using the dose monitor in
the PSI Gantry 2 nozzle as a normalization signal. The measured curves
are smooth, except for a few points where instabilities in the delivered
current from the accelerator occurred (for example, in the plateau of the
B$_4$C data in Fig. 4). These instabilities however occurred only in a few
points collected on the plateau regions of the measured curves; therefore
they do not affect the analysis of the falloff of the measured curves.

Fig. 5 shows how the variation of the $I_Z$ of B$_4$C affects the position of
the Bragg peak in the water phantom. We carried out simulations with
the B$_4$C sample for fourteen different values of $I_Z$, using a finer
resolution of 1 eV between 80 and 90 eV and a coarser resolution of 10
eV between 70–80 and 90–110 eV; a smaller range was used for the
comparative test with graphite since we have a fairly precise reference
value to compare to. We fitted the full distal edge of the curves with a
cubic spline interpolation, and extracted the range as the 80% point
from the fitted curve. The same fit was performed with the reference
simulation that had no material sample.

3.3. Determination of the mean excitation energy of B$_4$C

Fig. 6 shows the $\Delta R$ variations as function of $I_Z$ in the simulation,
overlaid onto the measured value; the bands shown indicate the
experimental and simulation uncertainties. As mentioned in Section 2.4,
using $\Delta R$ removes most sources of systematic uncertainties. For the
experimental data, the uncertainties left are the precision of the water
scanner measurement, which is $\pm 0.025$ mm, and the density uncer-
tainty, which is $\pm 0.1$ mm; when added in quadrature, the density

Fig. 3. Deposited dose as function of water equivalent range (WER), to investigate the energy mismatch observed between data and simulation. The scattering model (left) does not have a significant impact on the resulting range, while the choice of the mean excitation energy of water (right) does. The nominal beam energy at PSI Gantry 2 is based on ICRU 49 [26] range/energy table, which uses $I_Z = 75$ eV for water. The energy mismatch between data and simulations is due to this definition.

Fig. 4. Water scanner water-equivalent range (WER) measurements without (blue) and with (red) the B$_4$C degrader sample. The shift in the Bragg peak is clearly visible, from which the change in range $\Delta R$ can be determined. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
In simulations, we repeated the simulations for a representative set of samples, for the statistical uncertainty in the simulations, as well as for the uncertainty in the range extracted from the fit of the falloff. To estimate the uncertainties due to thickness and density variations, we repeated the simulations for a representative $I_Z$ value with a 1% variation in the density and a 0.002 cm variation of the thickness, and obtained the resulting change in range. As mentioned in Section 2.4, from mass measurements we observe a maximum 1% mass variation across all samples; since we could not estimate precisely volume variations for the different samples, we have directly translated this variation into a 1% density uncertainty. Combining the thickness and density uncertainties gives an overall uncertainty in the WER of ±0.1 mm, while the interpolation and the statistical uncertainty – when obtaining the range from a fit to the Bragg peak – gives an uncertainty in WER of ±0.05 mm; since they are not correlated, we sum them in quadrature to give the overall uncertainty in WER.

To extract $I_Z$ from a comparison between data and simulations, we then performed a polynomial fit of degree 3 through simulated ranges. The uncertainty in the estimated range is obtained as the 68% (1σ) confidence interval in the polynomial fit. By comparing the measurements to the simulations, we obtained a value for the $I_Z$ of our B$_4$C samples to be 83.1 ± 2.8 eV.

To test the consistency of our result, we also estimated $\Delta R$ for an incident beam of 150 MeV and 83 eV mean excitation energy (it is worth reminding that $\Delta R$ is independent of the incident beam energy). The result was compatible with the one obtained in the 230 MeV simulation within uncertainties, further validating our simulation setup.

### 3.4. Methodological validation: determination of the mean excitation energy of graphite

Fig. 7 shows the application of the same method described above to a graphite sample, here a spare degrader for the PSI PROSCAN facility with a measured bulk density of 1.870 ± 0.005 g cm$^{-3}$. The figure shows the $\Delta R$ changes as $I_Z$ is varied in simulation. We used again a 1% uncertainty in the density of the graphite as for B$_4$C. We obtain an $I_Z$ for our graphite sample of 81 ± 5 eV. The relative error obtained is larger because of the larger uncertainties in the fit (lower number of points) as well as the lower density of graphite; this gives a smaller variation of $\Delta R$ with $I_Z$ with respect to B$_4$C. The value obtained for $I_Z$ is in good agreement with the accepted reference value: the latest ICRU estimate gives 81.0 ± 1.8 eV [20]), thus supporting the validity of our method.

### 4. Discussion

We have developed a simple method to determine the $I_Z$ value of a given material sample such that the $I_Z$ may be reliably used in Monte-Carlo simulations; this method overcomes the limitations of simulations that must today employ the Bragg additivity rule to estimate $I_Z$. Using our method, we estimate the $I_Z$ of B$_4$C to be 83.1 ± 2.8 eV; this uncertainty is larger than the uncertainty arising from variation B:C atomic ratio. Our method may readily be applied in any clinical proton treatment facility, since it utilises detectors that are already commonly used for beam commissioning and QA, and it can be modelled using a simple simulation setup.

Our measurement is the first experimental determination of the $I_Z$ of B$_4$C which was previously based only on the calculations of Seltzer and...
With Monte-Carlo simulation codes becoming more precise, faster and easier to set up, their use for proton therapy system design will only increase; this is already shown in the context of magnet design [38] and degrader design [8,17,21,13]. Our present work contributes to this by more carefully defining material properties for reliable simulation than has been previously described. With the present interest in high dose rates in the context of FLASH irradiation [5,39], optimisation of beamline design to achieve the best beam transmission will substantially increase in importance. The method reported here to obtain an improved determination in the $I_Z$ of $B_4C$ contributes to this, by allowing candidate degrader materials and configurations to be examined more reliably in proton therapy system design; this will be a first and essential step in designing future innovative degrader systems.

**Declarations of Interest**

EO received funding from the European Union’s Horizon 2020 research and innovation programme under the Marie Skłodowska–Curie Grant agreement No. 675265, OMA – Optimization of Medical Accelerators. HO gratefully acknowledges support from UKRI/STPC grant ST/P002056/1 (Cockcroft Institute). The other authors have no conflicts of interest to disclose.

**Acknowledgements**

We would like to thank A. Muelhaupt from the PSI accelerator department, for providing the weight and size estimates of the different samples used in the density estimation. SP would like to thank Z. Chowduri from the Centre for Proton Therapy at PSI, for her help with the range measurements. Finally, thanks to T. Boehlen, T. Lomax and D. C. Weber, from the Centre for Proton Therapy at PSI, for commenting the manuscript.

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