Development of Monte Carlo Track Structure Codes

Larry Toburen
East Carolina University
Greenville, North Carolina

What is a charged particle "track"?

The definition of a charged particle "track" depends on the time in its history as well as the intended application. In general, we can say that a charged particle track is formed by all of the spatial products of ionization, both primary and secondary, and excitation formed as the charged particle and its secondaries slow and stop in the "stopping medium."

When ionizing radiation interacts with matter, by its basic definition, it produces free electrons and residual positive ions. Thus, one might think initially that all ionizing radiation should produce the same biologic damage. This, however, is not so. When what we traditionally call x-rays (energies from a few 10s of eV to a few hundred keV) interact with an absorber, whether it is a gas, liquid, or solid, they produce "fast" mono-energetic electrons having energies near that of the incident x-ray, i.e., photo-electrons with energies $E_{pe} = E_{x\text{-}ray} - \varphi$, where φ is the initial binding energy of the electron and is generally a few 10s of eV more or less; the binding energy is obviously more, and the difference between photon and electron energy greater, when sufficient energy is available to ionize inner shell electrons. When gamma-rays, traditionally defined as having energies from a few hundred keV to a few MeV, interact with matter, they can also produce photo-electrons, but as the energy increases above a few hundred keV, they produce free electrons with energies from a few eV to a few keV through particle-like collisions with absorber electrons; this is called Compton scattering. Thus gamma-rays of a defined energy produce free electrons with a broad distribution of energies. All fast electrons, regardless of how they are produced will subsequently undergo interactions with the bound electrons of the media and loose energy.

When heavy charged particles, such as alpha particles from radioactive decay or proton recoils from interactions of fusion neutrons interacting with hydrogenous material, interact with matter they produce secondary electrons with a wide spectrum of energies along their path. All fast moving heavy charged particles, whether resulting from radioactive decay or cosmic rays, or from high energy accelerator-produced ions, generate secondary electrons and ions along their path as they slow and stop in the absorbing medium. These interactions, resulting in ionization and excitation of the medium, form what we refer to as the track of the charged particle. Thus all ionizing particles from x-rays to gamma rays to electrons and heavy ions produce essentially the same products in the absorber. It must, therefore, be something about the *distribution* of these interactions in time, energy, and space that leads to the differences in damage that is observed for different types of radiation.

The investigation of the slowing and stopping of fast charged particles began with the discovery of the electron and the observation of its course in matter (Thomson, 1912, Bohr, 1913). Vast differences in the paths of electrons and alpha particles led to theoretical investigation leading to concepts of electronic energy loss (Bohr, 1948). Thus began the development of the most general model of track structure; that of describing the characteristics of a charged particle based on the energy it lost per unit path length as the particle interacted and slowed in the media it traversed. This description of the "stopping power" of a moving charged particle was used, and still is a primary area of charged particle

study, to describe the properties of charged particle interactions. A major advantage of this track description is that it can be very accurately determined by both experimental and theoretical means, with accuracies of better than 1% in modern studies. The disadvantage of this track description, as we shall see, is that it lacks detail, providing only an average of all interaction processes leading to the slowing of charged particles in matter, and particles with vast differences in velocity and charge can have the same stopping power, thus rendering its application highly unreliable in the estimation of relative chemical and biological responses. Still, it is a powerful concept and any more detailed model must provide an accurate representation of the stopping power when integrated over the parameters providing additional detail.

LET

It was not long after the development of stopping power that it was found that stopping power was not unique to a moving charged particle, and that particles of the same stopping power did not necessarily elicit the same chemical or biological response. Stopping power has no information other than average energy lost by the particle per unit path length; it is a particle-centered quantity that does not address the parameters of importance to predicting media response, i.e., how much of that energy is locally absorbed and by which physical processes will ultimately determine the physical, chemical, and biological response. Some of the energy imparted can be transported from the site of interest by photons or fast electrons emitted by the absorbing atoms, or there can be nuclear processes initiated that are not involved in the response of interest. In an attempt to overcome some of these limitations, the concept of linear energy transfer (LET) was developed. This conceptually is the media-centered analog to stopping power in that it addresses that fraction of the stopping power actually locally absorbed. In fact, one often considers the "energy restricted" LET_r where r is the designation of an energy threshold, or length threshold, above which one assumes the energy is transported away from the site of interest. This quantity has some advantage in chemistry and biology where the event of interest, e.g., DNA or chromosome damage, might occur in a very small volume and the energy losses above, say 100 eV, are transported by photons and secondary electrons (delta rays) to distances far from the target molecules. Still, in the end we are left simply with the average or mean energy loss per unit track length and this is hardly sufficient to define the damage processes actively involved in many areas of research.

A view from Radiation Chemistry

To overcome the limitations inherent to both stopping power and LET, many different approaches have been developed, each incorporating more detailed descriptions of the interactions along a charged particle track and each averaging the interactions at a different level of organization in the description. The chemists were among the first to recognize that different chemical reactions might be initiated in different regions along the track. Because energy deposition is a stochastic (i.e., random) process, it results in the formation of clusters of interactions of different sizes along with corresponding regions of no interactions along the track. Since many chemical reactions require an energy threshold for reactivity, these clusters might lead to different products. Clusters of different sizes were defined including spurs, blobs, and short secondary electron tracks to illustrate the stochastic process of energy deposition – but always the integrated quantities were required to equate to the stopping power (Mozumder and Magee, 1966 a-c). These authors noted that the best models of cluster size and frequency also included the requirement that there be random displacements of these track entities from the track's central path. Their study of the dependence of chemical reactions based on the density of ionization in a charged particle track was successful in providing much improvement to understanding radiation chemistry and led to a very successful model of charged particle tracks developed by Chatterjee beginning in the 1970s (Chatterjee et al., 1973, Chatterjee and Schaefer, 1976).

Track structure in radiobiology

In the study of radiation biology, the field of Microdosimetry developed in a similar manner to radiation chemistry to consider energies deposited in volumes representing the sensitive parts of a cell. Usually these models included an average over a volume representative of a cell nucleus as the important size parameter in radiation biology. This field coupled theories of mammalian cell response with the frequency spectra of energy deposition in volumes of "tissue-equivalent" gas ionization-detectors to enhance the detailed information of the energy deposition process along charged particle tracks. The charged particle beam was characterized either by the mean energy deposited in the volume coupled with the frequency distribution of energy depositions, or the radial distribution of ionization along the path; the latter determined by wall-less proportional counters located on or near the particle path to determine the spectrum of energy deposition events characteristic of the charged particle of interest. To couple the measured frequency of energy deposition events to biologic response required some assumed model of biologic action resulting from the energy deposited. Such models as the dual radiation action DRA (Rossi and Zaider, 1996) or the hit size effectiveness HSE (Varma et al., 1994) were advanced and provide some insight into the radiation damage process. Perhaps the most widely successful initial use of track models was pioneered by Katz (Butts and Katz, 1967) who applied physical theory to describe the radial distribution of energy around fast charged particles along with a multi-hit target theory to describe the relative effectiveness of different heavy charged particles in producing various biologic endpoints.

Event-by-event track structure development

So far the "track theories" discussed have all contained a good deal of averaging of the initial events along the track from the early development of stopping power theory to the radial distribution models of Chatterjee and Katz [it should be noted that all these mathematical techniques continue to be developed and applied to this day providing critical insights although the initial developers have passed on]. To many investigators it seemed appropriate, even in the early days, that an event-by-event record of all the interactions along a charged particle track was needed if one was to truly understand the pathways leading from energy deposition to cell response. This desire, combined with the introduction of a new generation of computers, the development of high-tech laboratory technology, and advances in cell biology, called for a dedicated effort to develop the models and data needed to make major advances in our understanding of radiation response. Thus in 1965 the US Atomic Energy Commission (later evolving to become the US Department of energy (DOE)) funded a new initiative of both experimental data development and computer modeling to make detailed track structure models available to radiation biologists. This new program brought together a number of the scientists who had introduced the evolving methodology of track structure as well as new laboratories involved in experimental physics and radiobiology. The new program included those innovative scientists who introduced the first track structure, such as Katz and Chatterjee; proponents of the microdosimetry methodology such as Rossi, Roesch, Braby, Varma and Bond; and a new set of experimentalists to study cross sections for photon and electron interactions, (Inokuti and group at ANL and Birkhoff and Arakawa and their groups at ORNL), heavy ion interactions (Toburen and Group at PNNL, Tobias and group at LBNL); computer modelers and atomic theorists, Wilson (PNNL), Inokuti (ANL), Ritchie, Turner (ORNL), and a number of others at universities and national laboratories.

Now looking back to the 1960s and the early 70s, it is hard to realize just how little was known of the details of the interaction process for the stopping of charged particles in matter. To develop a detailed track model one needed first to know the cross sections for ionization of tissue by electrons, but also similar data were needed for other charged particles of interest to the Department of Energy (DOE), such as the heavy charged particles produced as recoils from neutron interactions in tissue from fission reactor

operation. One of the goals of the program was to understand the relationship between radiation damage from electromagnetic radiation (X-, and γ-rays) and high LET radiation (neutrons and alpha particles).

Early work with water vapor

To be able to develop the 3-dimensional event-by-event description of any particle's track structure, the inelastic interaction cross sections for ions and electrons needed to be differential in the ejected/secondary electron (from ionization) initial energy and the emission angle into which it was ejected. The available data bases had essentially none of these doubly differential ionization cross sections, for electrons or heavy charged particles. Ideally, cross sections were needed for all the material within a mammalian cell. Unfortunately, technology did not exist to make single collision cross section measurements in a solid or liquid – the range of a low-energy electron was too small for the electron to emerge from even the thinnest sample conceivable without undergoing secondary scattering. To make measurements (and modeling) feasible the assumption was originally made that water vapor was a reasonable surrogate for tissue (tissue is considered about 85% water) and the first measurements and models used this assumption. It was also assumed that excitation of the target material was a negligible contribution to the total energy loss; a 10% contribution to energy loss was a common assumption based on limited information.

One of the problems with documenting the progress near the beginning of the development of event-by-event Monte Carlo modeling was the lack of publications by the scientists in main-stream publications. Most of the information exchange among researchers occurred during the annual DOE contractors meetings, the Radiation Research Society meetings and the periodic meetings of the International Congress of Radiation Research and Microdosimetry Symposia. Only these latter two meeting series have proceedings published, and these volumes had limited distribution making old volumes difficult to find. Often the detailed documentation of progress waited years to make it to retrievable first-line journal publications.

As the Department of Energy's program was getting underway a similar effort was underway at the Institut für Strahlenschutz der GSF München in Neuherberg, Germany (now the Helmholtz Zentrum München, Institut für Strahlenschutz) also focusing on the basic processes in the production of radiation damage in tissue. It was there that H.G. Paretzke had developed one of the first event-by-event computer models for electron transport using Monte Carlo techniques to follow the slowing down of fast electrons in simulated water (Paretzke, 1974). In Paretzke's model the cross sections for ionization and excitation were calculated using Bethe-Born theory (Inokuti, 1971) and experimental data for the optical oscillator strengths for water. Bethe-Born theory provided the differential cross sections for energy loss that Paretzke was able to classify in terms of the electron energy levels of the water molecule (Paretzke, 1988). To obtain doubly differential inelastic electron scattering cross sections, i.e., energy and angular differential cross sections, he relied on recent measurements to model the angular distributions of primary and secondary electrons produced in water by fast electron collisions (Opal et al., 1971, 1972).

In 1973, Paretzke spent several months working with Walter Wilson at the Pacific Northwest National Laboratory (PNNL) where experimental work was underway to measure the energy and angular dependent electron emission cross sections for interactions of protons with atomic and molecular gas targets. The goal was to merge the electron transport code of Paretzke with an electron source term derived from experimental data for proton interactions. This resulted in one of the first event-by-event heavy-ion track structure codes and was first documented in a presentation at the 4th Symposium on Miocrodosimetry in 1973 (Paretzke, 1974). One of the weaknesses of this code that was pointed out by the radiation-biology research community was that this code was based on atomic and molecular cross

sections, whereas the body is a condensed phase material. How relevant can an atomic model of tissue be to real tissue? It was generally accepted that liquid water was a good surrogate for tissue, tissue being about 85% water, but could one really use atomic and molecular cross sections to represent a condensed system? At that time, and even up to the present time, the codes must generally rely on either measured cross sections for gas targets, or on untested theoretical results for the condensed phase; the decision in the early 70s was that the GSF model was to use theory tested by experimental findings rather than the theory based on liquid water. This promoted considerable discussion among researchers of the time.

Coincident with the work of Wilson at PNNL and Paretzke and the GSF group, there were experimental studies of liquid water and theoretical work to calculate liquid water cross sections underway at Oak Ridge National Laboratory (ORNL). A milestone publication occurred in 1974 when Heller et al. (1974) presented optical oscillator strengths from reflectance data on liquid water. This provided the basic data to evaluate the dielectric response functions needed in the theory of energy loss in liquid water (Hamm et al. 1975; Ritchie et al. 1991; Dingfelder et al. 2008). Ritchie, Hamm and coworkers at ORNL worked to refine the dielectric response theory and applied the cross sections derived to the Monte Carlo study of electrons and ion transport in liquid water. Although experimental differential cross sections for liquid water could not be obtained for direct validation of the Monte Carlo (MC) input data, MC model results compared with the time dependent yield of products of irradiation of water gave a secondary test and these G-values measured by time-resolved chemistry were in excellent agreement with the new MC results.

Over the years many different groups have been actively involved in the development of Monte Carlo-based event-by-event track structure simulation codes. A comprehensive study of these codes in radiation research can be found in a review article by Nikjoo et al. (2006). As the applications have increased in radiation biology and confidence has been gained in the dielectric-response model, this model seems to be a dominant force in radiation biology. Two of the most common codes, PARTRAC developed at GSF Neuherberg (essentially an upgrade of the earlier MOCA series including liquid cross sections) and NOREC (an upgrade of the original OREC code) developed at ORNL are described in detail by Dingfelder et al. (2008). Also see the article here in THREE for a more detailed review of the field of Monte Carlo code development (Dingfelder, 2014).

As noted above, the DOE program as well as complementary work in Europe continued in all aspects of data development, microdosimetry and track structure, including further developments on analytic as well as Monte Carlo track structure models. The radial distribution model of Katz received broad application, including its application to risk assessment by NASA (NASA Risk Model 2013). The core-penumbra model of Chatterjee has also been widely used in radiobiology. One of Chatterjee's most exciting results was that his model was successful in enabling molecular biologists to determine the appropriate structure for the DNA in a chromosome configuration (Rydberg et al., 1988) by track structure analysis of molecular fragments measured following irradiation of cells. The successful applications of track structure in molecular biology are also clearly observed in the identification of DNA strand-break frequencies and locations in mammalian genes (Friedland et al., 2001). There are currently many examples of the importance and successes of various track structure codes throughout the world dealing with everything from x-rays to heavy ions, from molecular radiation biology to nuclear medicine; some of this is reviewed elsewhere in THREE (Plante, 2010).

The future of Monte Carlo codes

Has the development of mathematical descriptions of charged particle track structure reached its logical conclusion? Here I believe the answer is a resounding – no. There are many issues that are not fully resolved, both in the physics of track development and in the subsequent understanding of biological response. In particular, we do not know how to deal with the complexity of heavy ions that carry bound

electrons. Although considerable data exist on the physics of charge transfer and ionization by partially dressed ions, these concepts are only beginning to be incorporated into viable codes (Dingfelder et al. 2005). In addition, the very high density of ionization near track-ends and multiple ionization processes initiated by heavy ions are not generally considered in track codes. Many of these continuing challenges were discussed by Toburen in a recent publication (Toburen, 2011). There are still questions about the reliability of elastic and inelastic cross sections derived from dielectric response theory. For instance, the distribution of measured low-energy electron emission from amorphous solid water differs for Monte Carlo simulation of electron transport in liquid water by as much as a factor of 5; the theory predicts 5 times the low energy electrons observed at about 5 eV (Toburen et al., 2010).

The ever growing assembly of track structure codes is having considerable success in many fields, particularly radiation biology and medical physics. Which code should be used depends on many factors. Perhaps the most important factor is the degree of averaging over particle and target parameters that can be tolerated. Even in medical applications where stopping power (or LET) may still provide an adequate description of the energy deposition process for dosimetry of light particles in large organs, they will undoubtedly require a highly complex event-by-event code for stopping proton and heavier ions in a small organ where the dose might be delivered by a small number of particles undergoing considerable straggling. Certainly understanding the detailed findings in molecular radiation biology will require ever increasing accuracy in the point-by-point slowing and stopping of charged particles in a well-constructed target matrix.

References

Bohr N, 1913. On the theory of the decrease of the velocity of moving electrified particles on passing through matter, Phil. Mag. **25**: 10-30, 1913)

Bohr N, 1948. The penetration of charged particles through matter. Det Kgl Danske Videnskabernes Selskab Mathematiskfysiske Meddelelser XVIII **8**:1–144.

Butts JJ, Katz R. 1967. Theory of RBE for heavy-ion bombardment of dry enzymes and viruses. Radiation Research **30**:855–871.

Chatterjee A, Maccabee HD, Tobias CA. 1973. Radial cutoff LET and radial cutoff dose calculations for heavy charged particles in water. Radiation Research **51**: 479–494.

Chatterjee A, Schaefer HJ. 1976. Microdosimetric structure of heavy ion tracks in tissue. Radiation and Environmental Biophysics **13**: 215–227.

Dingfelder M, Toburen LH, Parertzke HG. 2005. An effective charge scaling model for ionization of partially dressed helium ions with liquid water. In The Monte Carlo Method: Versatility Unbounded in a Dynamic World, Available on CD-ROM American Nuclear Society, LaGrange Park, IL.

Dingfelder M, Ritchie RH, Turner JE, Friedland W, Paretzke HG, and Hamm RN. 2008. Comparisons of calculations with PARTRAC and NOREC: Transport of electrons in liquid water. Radiat. Res. **169**: 584-594.

Friedland W, Li WB, Jacob P, and Paretzke HG. 2001. Simulation of Exon Deletion Mutations Induced by Low-LET Radiation at the HPRT Locus. Radiat. Res, **155:**703–715.

Hamm RN, Wright HA, Ritchie RH, Turner JE, and Turner TP. 1975. Monte Carlo calculation of transport of electrons through liquid water. In *Proceedings of the 5th Symposium on Microdosimetry* (J. Booz, H. G. Ebert and B. G. R. Smith, Eds.) Commission of the European Communities, Luxembourg:1037-1050.

Heller Jr. JM, Hamm RN, Birkhoff RD, and Painter LR. 1974. Collective oscillations in liquid water. J. Chem. Phys. **60**:3483-3486.

Inokuti M. 1971. Inelastic collisions of fast charged particles. Rev. Mod. Phys. **43**:297-347.

Mozumder A, Magee J. 1966a. Models of tracks of ionizing radiation for reaction mechanisms. Radiation Research **28**:203–211.

Mozumder A, Magee J. 1966b. A simplified approach to diffusion controlled radical reactions in the tracks of ionizing radiations. Radiation Research **28**:215–231.

Mozumder A, Magee J. 1966c. Theory of radiation chemistry. VII. Structure and reactions in low LET tracks. Journal of Chemistry and Physics **45**:3332–3341.

Nikjoo H, Uehara S, Emfietzoglou D, Cucinotta FA. 2006. Track structure codes in radiation research. Radiat. Meas. **41**:1052-1074

Opal CB, Beaty EC, Peterson WK. 1971. Tables of energy and angular distributions of electrons ejected from simple gases by electron impact, Joint Institute for Laboratory Astrophysics (JILA) Report No. 108, University of Colorado, Boulder.

Opal CB, Beaty EC, Peterson WK. 1972. Tables of secondary electron production cross sections. At. Data Nucl. Data Tables **4**:209-253.

Paretzke HG. 1974. Comparison of track structure calculations with experimental systems. In Proceedings of the Fourth Symposium on Microdosimetry, Verbania-Pallanza, Italy (J Booz, H.G. Ebert, R Eickel, and W. Walker, Eds.) pp. 141-165. Commission of the European Communities. Luxembourg, 1974. [EUR 5122d-e-f.]

Paretzke HG. 1988. Simulation von Elektronenspuren im Energiebereich 0.01-10 keV in Wasserdampf. GSF-Berich 24/88. Gesellshaft für Strahlen-und Umweltforschung, München.

Ritchie RH, Hamm RN, Turner JE, Wright HA, and Bolch WE. 1991. Radiation interactions and energy transport in the condensed phase. In *Physical and Chemical Mechanisms in Molecular Radiation Biology* (W. A. Glass and M. N. Varma, Eds.), Plenum Press, New York:99-133.

Rossi H, Zaider M. 1996. Microdosimetry and its applications. Springer, New York.

Rydberg B, Holley RH, Saira Main I, Chatterjee A. 1988. Chromatin Conformation in Living Cells: Support for a Zig-Zag Model of the 30 nm Chromatin Fiber. J. Mol. Biol. **284**:71-84.

Thomson JJ. 1912. Ionization by moving charged particles. Phil. Mag. **23**:449-457.

Toburen LH, McLawhorn SL, McLawhorn RA, Carnes KD, Dingfelder M, Shinpaugh JL. 2010. Electron emission from amorphous solid water induced by passage of energetic protons and fluorine ions. Radiat. Res. **174**:107-118.

Toburen LH. 2011. Challenges in Monte Carlo track structure modeling. Int. J. Radiat. Biol. **88**: 2-9.

Varma MN, Wuu CS, Zaider M. 1994. Hit size effectiveness in relation to the microdosimetric site size. Radiat. Prot. Dos. **52**: 339-345.