1

The Emergence of Radioecology

1.1 Glowing in the Dark

On the frieze of the south-west facing Grenelle side of the Eiffel Tower in Paris, sandwiched between the names of Broca and Coriolis, is the name Becquerel. Paul Broca was a gifted physician and anthropologist and Gaspard-Gustave de Coriolis a famous engineer and scientist. Antoine César Becquerel, however, was a physicist who, with his son Alexandre-Edmund, had made major contributions to the study of electricity. Alexandre-Edmund was himself to make major contributions to science, not only in relation to the study of electricity but also to the early development of photography; he was very interested in the curious phenomena of luminescence and phosphorescence as well. But it was Alexandre-Edmund's son, Antoine Henri Becquerel, who was to stumble upon a completely different phenomenon that was to change forever our understanding of the world around us, and that phenomenon was radioactivity. Just as fortunate, perhaps, was the fact that Henri did so only because of the very recent discovery of what we now know as ionising radiation by Wilhelm Conrad Röntgen, and that discovery had been very dramatic indeed.

Röntgen had been experimenting with the discharge of electricity in vacuum tubes that had metal plates sealed into their ends so that they could be connected to a battery or an induction coil. Such tubes, first produced by Johann Geissler in 1857, had been used by Julius Plücker in 1858 to study the nature of electricity, the flow of which caused a glowing light to emerge from the negative plate (the cathode) and then disappear into the positive plate (the anode). If the beams were energetic enough to hit the glass wall, it fluoresced. The emissions coming from the cathode, the 'cathode rays', could be deflected by a magnet, but not much else was known about them and virtually nothing was known about what caused the glass walls to fluoresce. And then late in the afternoon on 8 November 1895, Röntgen noted something very strange. He had been using a tube to which a thin aluminium window had been added in order to permit the cathode rays to 'escape', and a cardboard covering had been placed over that end of it to protect the aluminium window from damage. But in spite of this covering of cardboard, which was light-proof, he observed that a small screen painted with barium platinum cyanide, which had been placed close to the aluminium window end of the tube, was glowing. Röntgen further observed that this was not caused directly by the cathode rays but arose from an area of fluorescence on the side of the tube. Further careful study revealed that these rays, unlike the cathode rays, were not deflected by a magnet and thus, as they had been previously unknown, he simply termed them 'x'-rays. He also

quickly discovered that these new rays passed through various objects placed in their way, although they did 'expose' photographic plates. Indeed, in less than two weeks after his initial discovery he took the very first x-ray picture: it was of the bones within a hand, and the hand was that of his wife, Anna Bertha.

About 10 weeks after the wave of excitement that inevitably followed Röntgen's discovery, particularly the worldwide distribution of the sensational x-ray image of his wife's hand (Figure 1.1), Henri Becquerel, who had become the third member of his family to occupy the physics chair at the Muséum national d'Histoire naturelle, recalled his father's work on luminescence and phosphorescence and therefore wondered if materials such as uranium salts might also emit these penetrating x-rays when illuminated by bright sunlight. (In contrast to fluorescent materials, phosphorescent materials do not immediately re-emit the radiation that they absorb, such as light, but may do so several hours later, at a different wavelength.) His first experiments, using uranium salts and photographic paper, appeared to confirm his suspicions. But when he was preparing to repeat these experiments on 26 February 1896 Paris was cloudy and he put the unexposed photographic plate, with potassium uranyl disulphate placed on top of it, away in a drawer. When developing the plate on 1 March, he unexpectedly found very intense images that had obviously been caused irrespective of any previous exposure of the uranium salts to sunlight. Becquerel still

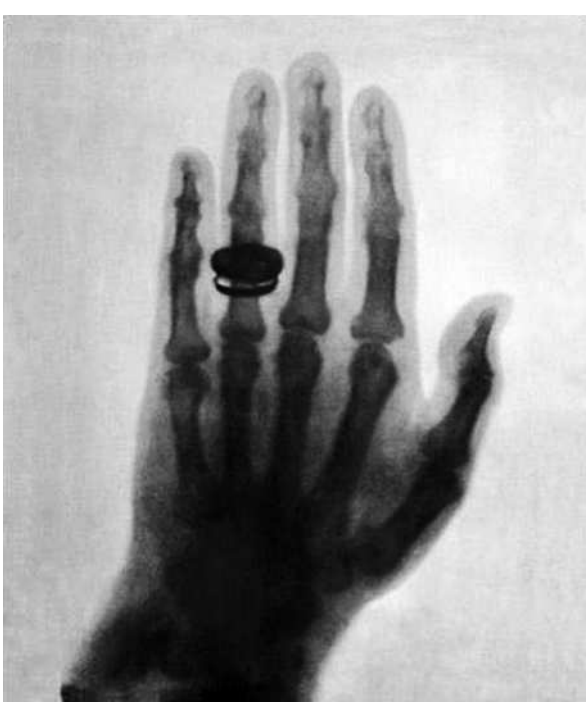

Figure 1.1 The first x-ray image ever taken, of the hand of Anna Bertha, the wife of Wilhelm Conrad Röntgen, in 1895.

thought that the effect might have been the result of some strange form of luminescence but, following various careful experiments, he was able to demonstrate that the 'rays' causing the exposure of his photographic plates also ionised gases, and that their intensity in a sample could be measured using a crude gold-leaf electroscope.

Becquerel's discoveries certainly lacked the dramatic impact of those of Röntgen, and were indeed subsequently regarded as being simply serendipitous, although perhaps it would be more accurate to describe them as being the result of deserved good fortune. The results were nevertheless eventually pursued with great interest by a young Polish student, Marie Skłodowska, and her new husband Pierre Curie, who were both at university in Paris. They showed that the element thorium also possessed the property that Becquerel had demonstrated with uranium, and it was the Curies who in 1898 coined the term 'radio-actif' in order to describe this phenomenon. The Curies went on to discover polonium and radium and to explore the radioactive properties of these elements. They also demonstrated the immense amount of heat generated in radioactive decay and hence the large amount of energy involved in the process. And they obtained the first information on the rates of radioactive decay.

But what of the magnetically deflected cathode rays? It had fallen to J. J. Thomson in Cambridge to elucidate their nature because in 1897 he described how he had set out to determine the ratio of charge to mass of the particles that constituted these rays and, as a result, demonstrated that such 'electrons', as he called them, were lighter than hydrogen. Fortunately, a young New Zealand physicist, Ernest Rutherford, was working under Thomson at the Cavendish Laboratory at that time and by 1899 he had discovered that there were in fact two types of rays being emitted by these uranium salts: one of them could be stopped by thick paper, whereas the other could not only pass straight through such paper but also expose photographic plates. He named them alpha and beta rays. Then, in 1900, it occurred to Becquerel that he could also measure the charge-to-mass ratio of the 'beta' particles by using the same method that J. J. Thomson had used to study cathode rays and indeed found them to be the same. He therefore suggested that the beta rays were, in fact, electrons. And to these two 'rays' the French chemist Paul Villard was, in 1900, to add a third and more powerful and penetrating type of 'ray' that he had discovered emanating from radium. These rays were recognised by Rutherford as being of a fundamentally different kind from the previously named rays and thus, in 1903, he named Villard's rays gamma rays. Rutherford also noted that gamma rays were not easily deflected by a magnetic field, another property making them different from alpha and beta rays. Furthermore, in 1902 and then working at McGill University in Canada with Frederick Soddy, who was later to coin the term 'isotope', Rutherford developed a theory of atomic disintegration demonstrating, as indeed Marie Curie had previously suggested, that radioactivity was an atomic phenomenon. Rutherford and Soddy had remarkably shown that radioactivity involved the spontaneous disintegration of atoms into other types of atoms, and that one element could therefore change into another.

And so by the turn of the twentieth century the basics of 'radioactivity' had, literally, come to light. Röntgen was awarded the first Nobel Prize in physics in 1901, and the 1903 prize was shared by Henri Becquerel and the Curies. Sadly, Pierre Curie was killed in

a carriage accident in 1906, but Henri Becquerel continued to study various aspects of uranium salts and died, famous, at the age of 55. Marie Curie went on to receive another Nobel Prize, in chemistry, in 1911 and died of aplastic anaemia in 1934 at the age of 66. Rutherford received the Nobel Prize for chemistry in 1908, and Frederick Soddy was awarded the same prize in 1921. Rutherford then went on to become director of the Cavendish Laboratory at Cambridge University in 1919, although his greatest scientific achievements were yet to come. He died as Lord Rutherford of Nelson and was interred near Sir Isaac Newton's tomb in Westminster Abbey. And having been awarded the Nobel Prize in 1906 for demonstrating that cathode rays, the electrons, were particles, J. J. Thomson was to see his son, George, receive the same prize in 1937 for demonstrating (equally correctly) that the electrons behaved like waves. But Röntgen, whose discovery of x-rays was immediately taken up and applied worldwide, with incalculable ultimate benefit to human health, and having declined to patent any of his discoveries, died a bankrupt in 1923 at the age of 77, having donated his Nobel Prize money to the University of Würzburg. A truly remarkable man.

The events over this short period of time were to spark a whole new era of science whose origins related in one way or another to the discovery of radioactivity. Much of the basic mathematics pertaining to the various aspects of what had been observed was quickly set out over the subsequent two decades, and by the 1920s the basic features of this new and exciting area of science were already the subject of extensive reviews [1–3]. As to be expected, after the initial investigations into the nature of the phenomenon of radioactivity, much scientific effort was expended on attempts to unravel the structure of the atom, which resulted first in the discovery of the proton by Rutherford in 1917 and then of the neutron by James Chadwick in 1932. Numerous experimental investigations were made, and the sheer genius of Albert Einstein with his theories of special and general relativity, together with the theoretical contributions of Erwin Schrödinger, George Gamow, Werner Heisenberg, Paul Dirac, and others, all contributed to unravelling the fundamental – as was then thought – structure and properties of matter and the energy associated with it.

Not many studies were made of the phenomenon of radioactivity in an environmental context, however, and it was generally believed that ionisation observed in the air was caused by radiation arising from radioactive elements in the ground, or by the radioactive gases they produced and emanated from it. As early as 1901 Hans Geitel and Julius Elster had extracted radioactive materials from the air and the following year C. T. R. Wilson extracted them from rain. Geitel and Elster examined air in caves and showed that radioactive gases emanated from the ground, resulting in higher concentrations within underground chambers. Measurements of ionisation rates above the ground also showed a decrease with increasing height, which researchers assumed partly resulted from the absorption of the ionising radiation by the air. This view was soon to change, however, because in 1909 Theodor Wulf developed an electrometer that measured the rate of ion production inside a hermetically sealed container, and he later used it to show that the levels of radiation were higher at the top of the Eiffel Tower than at its base. Perhaps not surprisingly, his results were not immediately accepted; but with a series of ascents in a balloon, starting on 17 April 1912, Victor Hess carried three 'improved' electrometers to

an altitude of 5,300 metres in a balloon flight on 7 August 1912 and found that the ionisation rate at such an altitude was almost an order of magnitude greater than at ground level. Hess also ruled out the obvious possibility that the source of this radiation was the Sun by making a balloon ascent during a near-total solar eclipse, achieving much the same results. It was Robert Millikan, however, who in 1922 extended observations up into the stratosphere and further demonstrated the extent to which such cosmic rays, as he termed them, penetrated material on Earth. Both Millikan (in 1923) and Wilson (in 1927) were awarded Nobel Prizes in physics, although for Wilson this was largely in recognition of his development of the 'cloud chamber' that became a vital tool for the detection of sub-atomic particles. Victor Hess belatedly received the Nobel Prize in physics in 1936, jointly with Carl David Anderson, for his contribution to the discovery and nature of cosmic radiation.

One aspect of radioactivity that was soon appreciated was its capacity to enable the age of rocks to be assessed. Acting on a suggestion made by Rutherford, Bertram Boltwood in the USA established that the final decay product of uranium was lead and, noting that the lead : uranium ratio was greater in older than in younger rocks, was in 1907 the first person to measure their age by the decay of uranium to lead. Another aspect that was seen to be of great value was the fact that extremely small (invisible) quantities of radioisotopes could be detected by suitable instruments, and thus could be used to trace the location and movement of elements in biological systems. George de Hevesy, a Hungarian physicist, was the first to use such a technique, using a radionuclide of lead (^{212}Pb). He carried out his first, but unpublished, experiment apparently when he was studying in Manchester under Ernest Rutherford between 1910 and 1913. He was convinced that his landlady was habitually recycling the leftovers of his meal, serving them up to him again the following day in a brown 'hash'. He proved his point by first spiking his leftovers with the nuclide and then demonstrating, with a gold-leaf electroscope, how 'radioactive' the following day's hash was compared with all of the other food on his plate, much to his landlady's astonishment! But, more professionally, he later used the same nuclide to follow its absorption and subsequent translocation throughout the stems and leaves of broad bean plants, publishing his results in 1923. He was awarded the Nobel Prize in chemistry in 1943 for his key role in the development and use of radioactive tracers in biological systems.

One other important consequence of the discovery of radiation was that the x-ray photographs taken by Röntgen of Anna Bertha's hand were immediately seen as having immense practical benefits, not only for the practice of medicine but also for biological sciences as a whole. Indeed, within a matter of weeks, Josef Maria Eder, a director of an institute for graphic processes in Vienna, and Eduard Valenta, a photochemist, produced *Versuche über Photographie mittelst der Röntgen'schen Strahlen* (*Experiments in Photography by Means of X-rays*), which was a portfolio of 15 positive and negative images including skeletal forms of animals (Figures 1.2 and 1.3), human limbs, and an assortment of various materials such as metal, wood, and glass.

Equally remarkable was the fact that the dangers of exposure to radiation were also quickly realised. With considerable foresight, Ivan Romanovich Tarkhanov, a retired Russian physiologist at St Petersburg University with a particular interest in electrophysiology had, early in 1896 and thus again within a few weeks of Röntgen's discovery, exposed frogs and insects

Figure 1.2 X-ray image of a snake taken by J. M. Eder and E. Valenta in Vienna in 1896. Public domain.

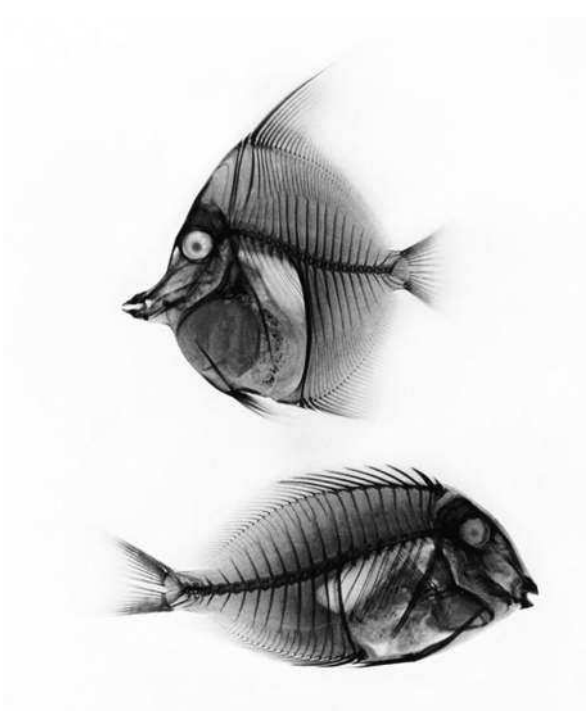

Figure 1.3 X-ray images (positive) of fish taken by J. M. Eder and E. Valenta in Vienna in 1896. Public domain.

to these newly discovered rays and concluded that not only did they 'expose' photographic plates but they could also 'affect the living function' of the organisms exposed to them. These experiments led him to believe that the impairment of reflexes that he had observed in animals after x-ray exposure resulted from changes in metabolism and blood circulation. Sadly, his considerable achievements in this field seem never to have been recognised.

One extraordinary pioneer at that time was Mihran Kassabian, an Armenian-American radiologist who became director of the Röntgen Ray Laboratory at Philadelphia General Hospital in 1902. His clinical work included the acquisition of fluoroscopic images and he first noted reddened areas of skin on his hands in 1900, which he initially thought was due to the use of chemicals. Two years later he sustained a serious radiation burn to his left hand and six years later two fingers were amputated. He kept a journal and took photographs of his hands as his tissue injuries (skin cancer) progressed (Figure 1.4). Within another two years he had further cancer-related surgery, this time to remove muscle from his chest, and died shortly afterwards on 14 July 1910.

Another pioneer was William Herbert Rollins, an obscure New England dentist who, more significantly, made experiments in 1901 and 1902 that not only demonstrated that this new 'x-light' could cause skin burns and even sterilisation, as had been noted by others, but also that it could kill adult animals (guinea pigs) and the foetuses of their pregnant females. Just as importantly, he also showed how partial shielding and other steps could reduce damage to animal tissues. Indeed he was, in effect, the founding father of what is now known as radiological protection. It is clear that Rollins was frustrated by the slow response of others to his findings, and hence with the lack of measures to do something about them.

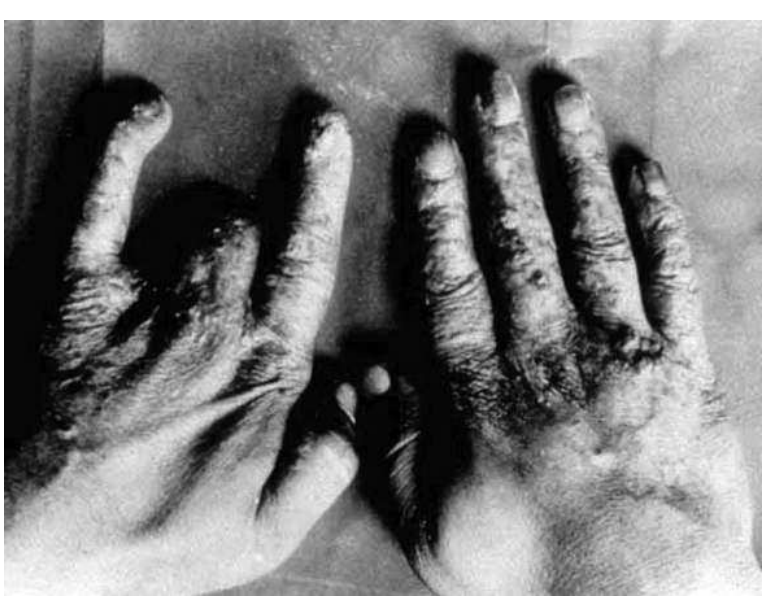

Figure 1.4 The hands of the pioneer Mihran Kassabian, 1909.

The dangers associated with exposure to radiation did, however, become increasingly apparent over the first decade of the twentieth century, and a number of national committees started to appear prior to the outbreak of World War I to address them. Yet it took another decade after the ending of that war for some form of international approach to be developed. At the First International Congress of Radiology in London in 1925 various ideas were discussed, and at the Second Congress in 1928 two bodies were established: the International X-Ray Unit Committee, with the primary objective of defining a unit for the measurement of radiation as applied in medicine, and the International X-Ray and Radium Protection Committee. Essentially, the role of the former was to define the quantities and units and that of the latter to advise on how they should be used for the purposes of radiological protection. In 1950 their names were changed to the International Committee for Radiological Units (ICRU) and the International Commission on Radiological Protection (ICRP), respectively. The ICRP issued its first report in 1928 that, together with its initial recommendations, related to the medical applications of radiation. However, the critical ICRP reports that were to become a driving force behind the necessity to acquire data on the behaviour and fate of radionuclides in an environmental context were not to emerge until after a very momentous event in history that was to herald the dawn of the atomic age.

Indeed, it is important to bear in mind that the subject of radioecology did not arise out of mere academic curiosity but was a direct consequence of the development of nuclear weapons and, subsequently, of nuclear power, although it has in more recent years broadened in scope and its boundaries are still somewhat ill defined. So in order to understand why certain chemical elements have been studied more than others, to appreciate the controlled or uncontrolled nature of the experiments in relation to them that have been performed, and to recognise why only certain types of environments have been investigated rather than others, it is necessary to review very briefly the evolution of radioecology against the background of events that helped to end World War II. Perhaps even of greater relevance in many respects, it is also necessary to review it further against the rapidly changing world of science and politics that pervaded the Cold War that was to replace it. So let us begin with the former: the development of nuclear weapons leading up to, and eventually ending, the war in 1945.

1.2 An Explosive Entry into the Atomic Age

Many detailed accounts have been written of this crucial turning point in history, primarily in relation to the combined USA, UK, and Canadian programmes that resulted in the development and then deployment of the two bombs that ended World War II [4–6]. A reasonable starting point for this sequence of events is a grey morning in London on Tuesday 12 September 1933 when Leó Szilárd, a Hungarian physicist of Jewish descent who had fled to the UK from Nazi Germany, while waiting for the traffic lights to change opposite the British Museum, had a great idea. The idea related to the feasibility of a nuclear chain reaction sustained by neutrons, the critical mass necessary to sustain such a chain reaction, and the enormous explosion that would thus ensue. Having considered these ideas

further, on 28 June 1934 he lodged a patent for the idea (British patent 630726) under the innocuous title of '*Improvements in or relating to the transformation of chemical elements'* and, after its acceptance in 1936, he assigned the patent to the British Admiralty so that it could be covered by the UK's Official Secrets Act.

By coincidence Irène (the daughter of Marie and Pierre Curie) and her husband Frédéric Joliot-Curie had, in 1934, discovered that artificial radioactivity could be induced in stable elements by bombarding them with alpha particles, for which they were awarded the Nobel Prize for chemistry in 1935. Enrico Fermi had reported similar results when bombarding uranium with neutrons. Then in December 1938 Otto Hahn and Fritz Strassmann, working in Berlin, sent a manuscript for publication reporting that they had detected the element barium after bombarding uranium with neutrons. The paper was published on 6 January 1939. Lise Meitner, an Austrian physicist who had fled Nazi Germany and was exiled in Sweden, together with her nephew Otto Robert Frisch, who was working with Niels Bohr in Denmark, quickly and correctly interpreted these results as being due to the splitting of the uranium atom and considered the physics that lay behind such a reaction. They named the process *fission* because of its biological similarity to the splitting of one cell into two. Frisch also experimentally confirmed the Berlin studies, and two papers were rushed to press on Friday 13 January 1939, being published on 11 and 18 February. The implications of their insight sent a shock wave throughout the scientific community. News of Meitner's and Frisch's interpretations moved fast, and Frisch's experiment was even replicated on 25 January 1939 at Columbia University, New York. Otto Hahn was awarded the Nobel Prize in chemistry for his work in 1944, but the all-important contributions of Meitner and Frisch in explaining what had been observed were shamefully never recognised by the Nobel Committee.

Just prior to these exciting developments, Arthur Jeffrey Dempster at the University of Chicago had in 1935 used a new technique called mass spectrometry to discover that uranium consisted principally of ^{238}U plus a very small fraction of ^{235}U, the latter eventually being shown to be about 0.7% of the total. Considering the physics that lay behind the conclusions of Meitner and Frisch in their joint paper of 11 February 1939, their colleague Niels Bohr quickly realised that it was the recently identified ^{235}U that was responsible for the fission and, together with the American John Wheeler, published his conclusions the following September, together with an extensive theoretical treatment on the mechanism of nuclear fission. In the meantime, Frisch and a colleague in the UK, Rudolf Peierls, had worked out a process by which an atomic device could be generated by simply using ^{235}U and conventional explosives that would not only create an enormous initial blast but also a great deal of consequent nuclear 'fallout'. Thus the concepts of fission, chain reactions, critical mass, and their explosive consequences had all come together on the eve of the outbreak of World War II.

There was no time to waste and thus research to construct a nuclear bomb quickly began in the UK under the code name of the Tube Alloys Project but, after the attack by the Japanese on Pearl Harbour in December 1941, the USA also began actively to pursue the same objective. This was to develop into the famous Manhattan Project, set up in August 1942 under General Leslie Groves with a scientific team led by Robert

Oppenheimer. The UK programme was well ahead of that of the USA, but the two were combined at the Quebec Conference of August 1943. Uranium was the key material and the initial challenge was to separate the lighter ^{235}U from ^{238}U, clearly a difficult task because the two would need to be separated by physical rather than chemical means.

But another element was also of relevance: plutonium. The discovery of plutonium was no mere accident. The first transuranic element (beyond uranium in the periodic table) had already been created by Edwin McMillan and Philip Abelson in 1940 at the Berkeley Radiation Laboratory, University of California, which was a facility that had been founded by Ernest Lawrence as early as 1931. At first not named as such, this element was neptunium (specifically, ^{239}Np). McMillan's work was then further extended by a team led by Glenn Seaborg, who in 1942 succeeded in producing the next element, plutonium (specifically ^{239}Pu) in sufficient quantities to be seen and weighed. They also realised that this specific nuclide was fissile and thus could also be used to produce a bomb; furthermore, being created from ^{238}U, the two nuclides could easily be separated chemically.

There were, however, considerable differences and difficulties in triggering a ^{239}Pu bomb compared with the relatively straightforward means of creating and sustaining a critical mass of ^{235}U, a problem solved by the mathematician John von Neumann. There was also the issue of creating ^{239}Pu in sufficient quantities. This required a controlled and sustained fission reaction that could be achieved only by building what was then called an 'atomic pile' – a nuclear reactor. In addition to the need for uranium as the fuel, there was also a requirement for the right material to act as a moderator to control the behaviour of neutrons in the atomic pile. Two possibilities were identified: 'pure' graphite and 'heavy' water. The former was pursued in earnest, and the first atomic pile (CP-1) was constructed at the University of Chicago using graphite as a moderator. It went critical on 2 December 1942. A multi-stage chemical process was then developed to produce larger quantities of plutonium, and this was further developed at the Clinton Engineer Works at Oak Ridge, Tennessee. The final, large-scale, production plant was established at Hanford, Washington State.

Meanwhile a group of scientists in France had considered the alternative possibility: the use of heavy water (deuterium oxide), a form of water in which the hydrogen atom has an extra neutron. Such hydrogen atoms occur naturally and deuterium had been identified in 1931 by Harold Urey; but only about 1 in 6,400 atoms are in this form and thus in water there is one molecule of heavy water per 3,200 molecules. Fortunately it was discovered that the two forms could be separated by electrolysis, and heavy water was already being produced at the Norsk Hydro plant (which provided a cheap source of electricity) at Vemork in Norway, simply as a by-product of the isolation of hydrogen for the production of ammonia as a fertiliser. Early in 1940 a group of scientists in Paris therefore asked the French Minister of Armaments to obtain as much heavy water as possible from Vemork. The French then discovered that Germany had already offered to purchase their entire stock, suggesting that the Germans were also pursuing the same route to an atomic bomb, and duly informed the Norwegians. Just prior to the German invasion of Norway on 9 April 1940, the Deuxième Bureau (the French military intelligence) removed 185 kg of heavy water from the plant in Vemork, the plant's managing director having agreed to 'lend' the heavy water