Isotope research before Isotopy: George Hevesy's early radioactivity research in the Hungarian context

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SUMMARY: 1.—Introduction. 2.—Two types of radioactivity research. 3.—The reception of radioactivity in Hungary. 4.—Starting migration: Béla Szilárd. 5.—George Hevesy's early radioactivity research. 6.—Hevesy's connection with the Hungarian scientific community in the 1910s. 7.—The origin of the radioactive tracer method. 8.—Delocalized research work. 9.—Budapest: The first applications of isotopes as indicators and tracers. 10.—Epilogue and concluding remarks.

ABSTRACT: This paper presents a framework for the study of George Hevesy's research in the 1910s by distinguishing two styles of radioactivity research: the analytical (as practices in Manchester and Vienna in some extent) and the natural historical styles (as practiced in Hungary). Georg Hevesy's approach combined the two types. Indeed, by studying Hevesy's research in context, I show that the earliest applications of isotopes were born in parallel with the establishment of the isotope theory of matter.

PALABRAS CLAVE: Hungría, metodología de trazadores radioactivos, radioactividad, Béla Szilárd, George Hevesy, estilos de investigación científica.

KEY WORDS: Hungary, radioactive tracer methodology, radioactivity, Béla Szilárd, George Hevesy, styles of scientific research.

1. Introduction

George Hevesy was one of the earliest scientists to initiate and carry out research work applying radioactive isotopes. Frederick Soddy created the word «isotope» in February 18, 1913 [1]. About the same time in 1913, Hevesy

1. Soddy, Frederick. The radio-elements and the periodic law. Chemical News. 1913; 107: 97-99.

worked with Fritz Paneth in Vienna and habilitated at the Budapest University. The first stages of his research were performed as part of a broad cooperation embracing many European radioactivists. Hevesy's intensive networking provided him with the intellectual background to merge two contemporary types of radioactivity research.

The first part of this paper briefly characterizes these two types of research in order to provide a framework for the study of George Hevesy's research in the l910s. While he was conducting his isotope research as part of this broad international collaboration, he also belonged to the Hungarian chemistry community. The question to be answered here is whether the Hungarian context contributed to Hevesy's work, and if it did, how. A further problem is related to the first: how were the two types of radioactivity research connected in his isotope research studies?

2. Two types of radioactivity research

Thomas Kuhn distinguished between two kinds of science that both had long traditions. Classical physical sciences include astronomy, harmonics, mathematics, optics and statics, which were later joined by the analysis of motion. «They might be better described as a single field, mathematics» Kuhn said, although this tradition was «empirical rather than a priori» [2]. He called the other tradition Baconian science, which had emerged in the seventeenth century. Kuhn characterized this type with a new empirical mode that did not aim to demonstrate «what was already known» or to expand upon existing theories. «Their typical products were», as Kuhn said, «the vast natural or experimental histories in which the miscellaneous data was amassed» [3].

Kuhn's two traditions corresponded with two ancient disciplines, natural history on the one hand and natural philosophy on the other. The goal of natural philosophy was to provide demonstrative analytical knowledge, to deduce statements from first principles, as exemplified by Euclidean geometry. This was close to Kuhn's classical physical sciences. As Steven

2. Kuhn, Thomas. Mathematical versus experimental traditions in the development of physical science. In: Kuhn, Thomas, ed. The essential tension. Chicago: Chicago University Press; 1977, p. 37-38.
3. Kuhn, n. 2, p. 43.

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Shapin explained, by the seventeenth century the natural philosophical program intended to place phenomena into the causal structure of nature. Indeed, natural philosophy possibly constructed its propositions in terms of causes, structures and mechanisms. Natural history, which Shapin contrasted with natural philosophy, was a practice of registering or cataloging observed effects. «The register of natural historical facts was to contain several kinds: naturally occurring entities and effects whether or not they were produced in the ordinary course of nature (...) and those that might be produced by human labor» [4]. Natural history has traditionally been engaged in collecting objects, describing facts, and mapping their occurrence in nature. The various collections of plants, insects, animals, minerals and many other objects with related facts had to be ordered in accordance with various taxonomies and presented in museums, books or pictures [5].

Both the practices of the two types of knowledge and their connections with each other have changed throughout history [6]. Kuhn characterized modern science as the mathematization of Baconian fields, such as heat, electricity and magnetism. Although the bridge between the two types grew strong in the 19th century, some fields, including botany, zoology, some chemistry and others, maintained their natural historical character even in the 20th century.

Radioactivity emerged as a new possible field of research in the late 19th century. In a similar fashion to electricity or heat more than a century earlier, radioactivity could be studied in a Baconian or natural historical way and in an analytic, theoretical, natural philosophical way. The natural historical approach aimed to describe radioactive rays and substances, work out methods for the description, collection and systematization of facts,

4. Shapin, Steven. The scientific revolution. Chicago: The University of Chicago Press; 1996, p. 85.
5. This activity had been analyzed in detail by Michel Foucault. He explained the methods of natural history. Foucault, Michel. The order of things. London: Tavistock; 1970.
6. A very vivid and colorful picture of the changing practice of natural history can be found in a collective volume: Jardine, N.; Secord, J. A.; Spary. E. C., eds. Cultures of natural history. Cambridge: Cambridge University Press; 1996. Roger French showed the intimate relationship between logical and historical argumentation in natural philosophical disputes until the early modern times. French, Roger. William Harvey's Natural philosophy. Cambridge: Cambridge University Press; 1994. John Pickstone put natural history into the historical and epistemological context of knowledge. I am grateful to Xavier Roqué for drawing my attention to this book. Pickstone, John. Ways of knowing. A New history of science, technology and medicine. Chicago: Chicago University Press; 2000.

and map how globally widespread radioactivity was. The center of this activity was the Curie laboratory in Paris. Many important findings were made there, such as the isolation of new elements (polonium, radium and actinium), the examination of the emitted rays, the constant emission of heat, the development of measuring instruments and many others. When this laboratory's research activity is considered to lack any hypotheses concerning the structure of the atom, and to be incoherent and directionless, the natural historical character of its work is disregarded. Many other laboratories conducted the same kind of radioactivity research all over Europe [7].

The analytical, classical physical scientific or natural philosophic approach was seeking a mechanism for spontaneous radiation originating from radioactive substances, and to explain the phenomena related to radioactivity. Adherents to this line of thought gathered around Ernst Rutherford in Manchester, and later in Cambridge [8]. Many theoretical breakthroughs, such as the transmutation of elements and the nucleus of the atom, originated from this laboratory. The difference between the intellectual features of the research centers in Paris and Manchester, which both employed a considerable number of visiting researchers from other countries, seems to be a matter of descriptive natural historical versus analytical natural philosophical approaches rather than the differing general philosophical commitments of their leaders [9].

3. The reception of radioactivity in Hungary

Chemistry in Hungary has traditionally been inorganic and analytical. This was a characteristically natural historical approach that sought to describe and map the country's natural resources. While in the late 18th and early 19th centuries, the analysis of minerals played an important role, later on

7. See J. L. Davis' detailed analysis of the Curie laboratory's work at the beginning of the 20th century. Davis, J. L. The research school of Marie Curie in the Paris faculty 1907-1914. Annals of Science. 1995; 52: 321-355.
8. About the early British researches see Jeff Hughes' article in this volume.
9. Malley, like Davis, has considered the Parisian laboratory less successful than the one in Manchester and attributed this assumed lack of success to Pierre Curie's positivism which she missed to analyze. Malley, Marjorie. The discovery of atomic transmutation: Scientific styles and philosophies in France and Britain. Isis. 1979; 70: 213-223.

in the 19th century and into the early 20th century, mineral water analysis became a central subject of chemical research. This can be explained by the country's richness in mineral water springs, and Lake Balaton, the largest lake in Central Europe, was also of balneological significance.

Radioactivity arrived early in Hungary, but not directly from Paris. While Henry Becquerel's paper announcing the discovery of the radiation of a uranium mineral appeared in February 1896, the first Hungarian review of it was published some months later. The review on «Láthatatlan sugarak» (invisible rays) was based on a German report of the original French paper. The reviewer was a young physicist, Károly Tangl, who twenty-five years later became a professor of experimental physics at Budapest University [10].

However, the research work on radioactivity started in chemistry not in physics. Béla Lengyel, professor of inorganic chemistry, was the first Hungarian to do research in the field. Lengyel published his first results on «the impact of some gases upon the photograph plate» in 1898, the same year that the term was coined by Marie Curie [11]. Lengyel was one of the highest authorities on chemistry, a professor and director of the II. Chemistry Institute (there were two) at the Budapest University, and a member of the Hungarian Academy of Sciences. His most important results included the production of high purity calcium and strontium, precise mineral water analyses and the usage of spectroscopy for analytical purposes.

Lengyel criticized Becquerel's method relying on black plots observed on photographic plates caused by radioactive radiation. The natural historical approach was seeking precise descriptions of radioactivity and to find new radiating substances. According to Lengyel, researchers found more radioactive substances, including calcium and other materials, than in fact existed. He attempted to prove that these elements do not radiate anything. He thought that the dark plot on the photographic plate (the indicator of radiation) was often due to certain reductive gases coming from the minerals they investigated, rather than from radioactivity. In 1900, he questioned whether polonium, radium and actinium were chemical elements in their own right or merely derivatives of other elements. He found radium and barium to be too similar to each other. It was impossible, he argued, for the two substances to only differ from each other in terms of their radiating

10. Tangl, Károly. Láthatatlan sugarak. Mathematikai és Physikai Lapok. 1896; 5: 188-189.
11. Lengyel, Béla. Néhány gáz hatása a fotográf lemezre. Mathematikai és Természettudományi Értesítő. 1898; 16: 365.

potential. Lengyel assumed that radioactivity was a feature some materials can receive during their formation. He produced some iron oxides and aluminium oxides that he measured to be radioactive, although the initial components were inactive [12].

He considered the subject important and recruited students to investigate it. This small school of radioactivity research produced some theses and papers that followed Lengyel's line of thinking. Methodological problems were discussed and measuring methods were worked out. Following on from natural historical traditions, they applied their methods to the search for radioactive resources in Hungary.

One of Lengyel's collaborators, Gyula Weszelszky, took over the research of this subject around 1912. With his students, he developed a special electrometer with useful additional devices for measuring the radioactivity of waters and gases. To this group, radioactivity appeared to be a new feature, which they had to search for in chemical substances. They were active in mapping the Hungarian minerals and mineral waters according to their radioactive components. One student, Irene Götz, the first woman to ever teach at a university in Hungary, spent some time in Paris studying methods used by Mme. Curie's in her Paris laboratory.

Under Weszelszky's direction, the research group was granted the status of an independent institution in 1915 and was given some rooms in the building of the II. Chemistry Institute in 1916. This was remarkable, because in the German academic system of the time, there was no place for such specialized institutes in Hungary [13]. The system was based on large institutes and chairs, teaching broad fields, such as chemistry or physics. However, in Germany the growing chemistry institutes focused on synthetic organic chemistry, while organic chemistry did not play an important role in the work of Budapest University's chemical institutes. Because none of the chairs were willing to adopt radioactivity, considered a brand new and perhaps suspicious subject, the university authorities sought a proper institutional solution. They decided to connect the small institute to an already existing service, the glassblowing institute, belonging to the II.

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12. Lengyel, Béla. A radioaktív baryumról. Mathematikai és Természettudományi Értesítő. 1900; 18: 121-127; Lengyel Béla. A radioaktív testektől. Magyar Chemiai Folyóirat. 1901; 7: 33.
13. On the institutional structure of chemistry Germany at that time see Johnson, Jeffrey. The Kaiser's chemists: Science and modernization in Imperial Germany. Chapel Hill: The University of North Carolina Press; 1990.

Chemistry Institute. The glassblowing institute's role was to assist all chairs in producing or repairing their glassware without having any research or teaching obligations. The organizational position of the radioactivity institute differed from any of the other institutions at the university. The Radiology Institute, or Radium Station (it was mentioned under different names), was a teaching and research institute with a special status of some independence from the disciplinary university institutes. In any case, Weszelszky, the leader, was never appointed as a full professor [14].

4. Starting migration: Béla Szilárd

A highly active young radioactivity expert, Béla Szilárd, despite working outside the chemistry community, preferred the descriptive style. He graduated from the faculty of pharmacy in 1904 and became an assistant at the Technical University in Budapest. Szilárd received a national stipend to go to Paris in 1907. He joined the Curie laboratory as a «travailleur libre» and stayed there until 1910, establishing contacts with its researchers, who included Albert Laborde, Pierre Curie's co-author in establishing the energy production of radium, and Ellen Gleditsch, who later played an important role in Norwegian science [15]. In 1912, Szilárd opened a small private company in Paris called Laboratoire de Produits Radioactifs. The firm produced special electrometers for radioactive measurements. Rather than return to Hungary during World War I, he moved to Spain to work in the *Instituto de Radioactividad* at the University of Madrid. In 1920, he returned to Paris. Five years later he received the Légion d'honor for his work on the measurement of radioactivity. Szilárd came to an untimely death in 1926 at the age of 42 [16].

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14. I gave a detailed and widely documented picture about the organization and the work of this institute in an earlier book titled (in Hungarian): Radioactivity and Chemical Theory of Atom: Crisis in the Views on Structure of Matter in Hungarian Chemistry. Gábor, Palló. Radioaktivitás és a kémiai atomelmélet: az anyagszerkezeti nézetek válsága a magyarországi kémiában. Budapest: Akadémiai Kiadó; 1992.
15. About Gleditsch career see Lykknes, Annette; Kragh, Helge; Kvittingen Lise. Ellen Gleditsch: Pioneer woman in Radiochemistry. Physics in Perspective. 2004; 6: 126–155.
16. Béla Szilárd's scientific biography has been addressed in Gábor, Palló. Szilárd Béla tudományos életrajza. Századok. 1981; 115: 770-798.

Szilárd followed the natural historical line in his research when writing on the radioactivity of mineral waters and in his extensive popularization work. His short book, titled Rádium és Radioaktivitás (Radium and Radioactivity), the first on radioactivity in the Hungarian language, focused on the description of rays, radioactive substances and their medical applications [17]. He carried out some experiments on special forensic photography, on the measurement of Roentgen rays, and on the determination of the appropriate doses for medical applications.

While at the Curie laboratory, he collected data and published a map of the spread of radioactive substances around the world [18]. He studied colloids containing radioactive substances, which he called «radiocolloids» [19]. Like other Hungarians, he constructed electrometers for specialised usage of radioactive measurements and performed experiments with them [20]. The company he established in Paris before the war was based on these devices. As an inventor, he produced a lightening rod with radioactive substances at the top [21]. The idea was that the radioactive rays had an ionizing effect upon the air, easing the discharge of the atmospheric electricity and guiding it into the rod. Another application with a similar effect was to remove the static electricity formed in textile factories by sliding the material across the machines.

In Spain he implemented certain innovations in his devices and started to map the country's radioactive substances [22]. He also discovered a new site of pitchblende occurrence [23].

His experiments and inventions received some publicity in the daily newspapers, which shows how Szilárd had established himself as a scientist in a third country. His results originated from the natural historical approach that he had learnt about in Hungary, one which was prevalent in Paris and seemed just as important in Spain. He tried to forge connections between

17. Szilárd, Béla. Rádium és radioaktivitás. Budapest: Mai H; 1905.
18. Szilárd, Béla. Tables de principaux minerais de radium et thorium. Le Radium. 1909; 6: 80.
19. Szilárd, Béla. Sur les composés colloïdaux des éléments radioactives. Comptes Rendus. 1907; 145: 463.
20. See Szilárd, Béla. Sur un appareil destiné á la mesure de la radioactivité. Le Radium. 1909; 6: 1-4.
21. Szilárd, Béla. Sur un paratonnerre au radium. Comptes Rendus. 1914; 158: 561.
22. Szilárd, Béla. Nuevo aparato para el reconcimento y determinación rápidos de los minerales de radio y de torio. Revista Minerales. 1918; 2: 665.
23. Szilárd, Béla. La pecblenda española. Revista Minerales. 1918; 2: 581.

the Hungarian and French radioactivity communities, but he hardly had any relations with Hungarian chemistry circles, not even Weszelszky's. At the international radioactivity conference organized in Brussels, 1910, both Weszelszky and Szilárd exhibited electrometers, but made no reference to each other. This is even stranger when we consider that Szilárd played a role in the organization of the conference.

5. George Hevesy's early radioactivity research

George de Hevesy came across the subject of radioactivity outside of the Hungarian scientific community. He was educated as a chemist, not in the Hungarian, but in the German and Swiss scientific community. Before he decided to become a chemist, he lived in Hungary. He received his first ideas about chemistry there. He was born in Budapest to an assimilated Jewish family, originally called Bischitz, receiving the «Magyarized» name of Hevesy in 1906. He graduated from the catholic Piarist high-school and started his chemistry studies at Budapest University. After the second semester, he continued his studies in Germany, and received his PhD in Zurich, in 1908, for his thesis on the electrolysis of alkali metals. He also published the content of his thesis in Hungarian [24].

Hevesy, however, did not return to work in Hungary after graduating, but eventually ended up in Manchester. In 1911, he started working at Ernst Rutherford's laboratory, the centre of the analytic, natural philosophical approach aimed at exploring the causes, mechanisms and consequences of radioactivity. At the time of Hevesy's arrival, the laboratory was working on a most important breakthrough, the detection of the nucleus and construction of the new structural model of the atom. The young Niels Bohr had just joined the lab, and became a lifetime friend of Hevesy's.

Hevesy conducted research on two problems that were seemingly independent of each other. The first problem he received from Rutherford was to separate radium-D from lead, because Rutherford wanted to perform

24. About Hevesy's biography see Cockroft, J. D. George de Hevesy 1885-1966. Biographical Memoirs of the Fellows of the Royal Society. 1967; 13: 125-166. (This is based on Hevesy's memoirs.) Levi, Hilde. George de Hevesy: Life and Work. Copenhagen: Rhodos; 1985; Niesse, Siegfried. Georg von Hevesy: Wissenschaftler ohne Grenzen. Dresden: Forschungszentrum Rossendorf; 2005; and Gábor, Palló. Hevesy György. Budapest: Akadémiai Kiadó; 1998.

experiments with the former. This was a clear task for a chemist. Hevesy's other problem was related to the genetic order of radioactive elements. By that time it was experimentally established that radioactive elements, while disintegrating, produce new elements, some radioactive, others not. The radioactive ones produce other elements when they decay, forming series of disintegration, families of chemical elements. The question was how these elements related to each other, from which element a given element originates, and how the chemical properties of the new elements depended on the quality of the rays that produced them. To answer all these questions, the researchers had to do extensive and painstaking experimental work. By that time, it had been essentially accepted that they were indeed chemical elements that needed to be placed in the periodic table. To find their place, the chemical properties of the substances had to be determined.

Most of Hevesy's papers published in 1912 and 1913 were associated to solving the genetic problem concerning placement on the periodic table [25]. Although his papers discussed the diffusion and electrochemistry of radioactive substances, these papers cannot be considered merely reports on data gathering or descriptions given in a natural historical way. Hevesy sought to determine the valence of radioactive elements by these methods (he had been familiar with electrochemistry since his graduate studies), in order to collect data on the relationship between the radiation of the mother element and the valence of the daughter element. Through these papers, he contributed to the search for the displacement laws, which was conceived almost simultaneously by many researchers. According to his own and James Chadwick's account, A. S. Russell, who had worked with Soddy in Glasgow, visited Manchester and discussed the results found by another Glaswegian researcher, Alexander Fleck, with Hevesy. Fleck, after conducting systematic research for two years, obtained some basic data on the valence of radioactive substances. Frederick Soddy, the head of the

25. I think of articles such as Hevesy, George. On the solubility of actinium emanation in liquids and in charcoal. Journal of Physical Chemistry. 1912; 16: 429 (Physikalische Zeitschrift. 1911; 12: 1214; Mathematikai és Természettudományi Értesítő. 1912; 30: 125); The detection of actinium emanation in solutions of minerals. Journal of Physical Chemistry.1912; 16: 451; The electrochemistry of radioactive bodies. Philosophical Magazine. 1912; 23: 628 (Mathematikai és Természettudományi Értesítő. 1912; 30: 903); Über den Zusammenhang zwischen den chemischen Eingenschaften der Radioelemente und der Reihenfolge radioaktiver Umwandlungen. Physikalische. Zeitschift. 1912; 13: 672; Die Valenz der Radioelemente. Physikalische Zeitschrift. 1913; 14: 49 (Philosophical Magazine. 1913; 10: 390).

laboratory in Glasgow, suggested the research. Russell also published his experimental work on the subject. Before Christmas 1912, Hevesy stopped in Karlsruhe on his way back to Hungary to discuss these latest results with Kasimir Fajans, who had speculated on the same problem without carrying out any experiments of his own [26]. Fajans wrote his famous paper on the displacement laws right after Hevesy's visit, slightly earlier than Soddy.

Meanwhile, Hevesy had not neglected his other task, the separation of radium-D from lead. He tried many different kinds of analytical and physical chemistry methods with the help of Dr. Pring, a researcher in Rutherford's laboratory, but he failed. By late 1912, other researchers also found that some radioactive elements are inseparable. According to Soddy, the problem of chemical inseparability had already emerged in 1905 when Otto Hahn discovered radiothorium. McCoy, Rossi, Boltwood, Marckwald and some others reported on similar cases. As Soddy remembered in 1910, he already felt that the non-separable elements showed not just «close similarity but complete chemical identity» [27]. Fleck, in 1912, when investigating the chemical properties of radioelements, noticed that some elements could not be separated because of their nature, and not because of any lack of methodological ideas [28].

Nevertheless, Hevesy did not give up, although the assumption of non-separability even concerning radium-D and lead had appeared in an article written by Herschfinkel as early as 1910 [29]. In letters written en route to Budapest, he suggested the use of certain new methods to Dr. Pring. Hevesy reported to Rutherford on the progress of their work. In February 1912, he expressed his hope of producing «perfectly clean or at least highly concentrated radium-D» [30]. In March, he was still optimistic. But by the end of the year he had almost lost all hope.

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26. The story was told in an oral history interview with James Chadwick in the framework of the Sources for History of Quantum Physics (SHQP). The copies are available in several archives. For example, in the American Institute of Physics, Niels Bohr library, College Park, Main, USA.
27. Soddy, Frederick. The origins of the conceptions of isotopes. Nobel Lectures. Chemistry 1901-1921, p. 382.
28. Fleck, Lord Alexander. Early work in the radioactive elements. Proceedings of the Chemical Society. 1963; November, 330.
29. Herschfinkel, Heinrich. Sur le radio-plomb. Le Radium. 1910; 7: 198.
30. Hevesy to Rutherford, letter. February, 14, 1912. In: Hevesy Scientific Correspondence, (HSC) Niels Bohr Archive, Copenhagen. I am grateful to Dr. Finn Aaserud, director and Felicity Pors, archivist for their hospitality and the assistance they provided to my research.

Before arriving in Budapest, he stopped in Vienna to visit the Institute of Radium Research. The following February he gave an account of his impression to Rutherford:

> «The laboratory is a very pleasant place indeed, very quiet and agreeable. Professor Meyer is a very kind chief. No great discoveries are made but a lot of good, very accurate work is done. With the exception of yourself, everybody I spoke to about the Vienna laboratory had a poor opinion of the place, I must say this opinion is wrong, the work of those who sacrifice themselves by doing less interesting work with great accuracy for the benefit of us all should be held in very high esteem» [31].

The Viennese laboratory was engaged in research that involved data gathering and precise descriptions in a natural historical way.

Concerning the problem of the separation of radium-D from lead, Hevesy told Rutherford in another letter that,

> «Stephan Meyer felt very unhappy about their unsuccessful attempts to separate radium-D from lead; they now have very large quantities of radiolead, and Prof. Meyer tried to induce me to take up the problem again. I must say I do not feel very confident now, though it is possible to concentrate it up to a certain, unfortunately very narrow, limit. He said the same to Prof. Meyer and other people have been disappointed as well» [32].

On this visit, he met Fritz Paneth, a young physical chemist, who remained his closest friend throughout his life. They decided to cooperate and try once more to separate radium-D from lead, but again without any success. Hevesy seemed to have gradually come to the conclusion of inseparability. In May 1912, he gave a talk at a meeting of the Bunsengesellschaft in Germany saying that radioactive methods, including radioactive indicators, could be applied to electrochemical measurements. The idea of radioactive indicators probably appeared for the first time in this talk [33].

31. Hevesy to Rutherford, letter. 28. February 28, 1913. HSC.
32. Hevesy to Rutherford, letter, December 7, 1912. HSC.
33. Siegfried Niesse found this Hevesy statement and considered it the first sign of radioactive indicators. Niesse, n. 24, p. 33.

6. Hevesy's connection with the Hungarian scientific community in the 1910s

In December 1912, Hevesy arrived back in Budapest. He planned to cooperate with Paneth in Vienna and to establish himself as part of the Hungarian chemistry community. Hevesy published most of his 1912 papers in the form of Hungarian translations, making himself known in Hungarian scientific circles. His specific goal was to go through a habilitation process and become a «privat dozent» at the University of Budapest. Although his scientific approach significantly differed from natural historical tradition, his measurements concerning some radioactive substances, such as uranium, actinium emanation and radium-D, as well as the new physical chemistry data he published on these substances could be considered important contributions to the description of these materials. In addition, the measurement of actinium emanation content was related to mineral water analyses as the radioactive components of mineral water could be determined through the emanation they emitted. In this way, they were not so alien to the traditional Hungarian natural historical approach.

Hevesy started his habilitation process in January 1912, when he was working in Manchester. Budapest University appointed two chemistry professors to be responsible for the matter. Béla Lengyel, as a major authority, was an obvious choice despite his old age. He died in 1913. The second person, Gustav Buchböck, was a physical chemist, but was not yet a full professor.

In the evaluation process, Hevesy received highly positive reviews from both professors. Buchböck seemed enthusiastic. On January 24, Hevesy passed his habilitation exams, and on January 28 he successfully gave his privat dozent lecture [34]. His «Habiltationsschrift» was published under the title of «The features of the electron and the constitution of the atom» [35]. In this, Hevesy applied the structural approach, rather than the usual Hungarian natural historic approach. He wrote about the recent views on the structure of the matter, including quantum theoretical views and some references to the theory of relativity in relation to the mass of

34. The documents can be found in the Archives of the Eötvös Loránd University, BTK DH 963/1911-12.
35. Hevesy, György. Az elektron tulajdonságai és az atom konstitucziója. Magyar Chemiai Folyóirat. 1913; 19: 53-59; 69-74; 88-94.

the electron. Hevesy explained the Rutherford model (Bohr published his famous triad some months later). When writing about radioactive substances, Hevesy remarked that «on the one hand some elements with equal atomic weights can show different chemical properties, on the other we know of several cases in which elements with different atomic weights have entirely identical chemical properties» [36]. This sentence should be understood in relation to his and others' failures in some separation experiments with radioelements, and almost to be a definition of the isotopy that had not been crimed at that time.

In early 1913, Hevesy spent most of his time in Budapest. He gave lectures at various scientific meetings and earned his new degree, although his mentality and approach were totally different from the traditional way of thinking of Hungarian chemists, not to mention his unusually wide international network and information. Apart from a review of Weszelszky's book, there is no sign that he had any contact with Weszelszky's radioactivity laboratory or with its staff. He knew the laboratory existed but he did not cooperate with it [37]. Also, apart from a reference to Szilárd's publication on radiocolloids, Hevesy probably had no knowledge of Béla Szilárd's activity.

7. The origin of the radioactive tracer method

While based in Budapest, Hevesy commuted between Vienna and Budapest, to continue his research with Paneth. The Radium Research Institute had perfect devices, good personnel and was rich in radioactive substances as Joachimstahl, one of the most important pitchblende resources belonging to the Austro-Hungarian Monarchy. In 1913, Hevesy published papers on the valence of radioelements, based on their diffusion and electrochemical properties, and another six publications with Paneth as a co-author. These could be considered natural historical descriptions but also further articulations of the genetic sequences of radioelements, and a search for displacement laws. They continued the investigations on the separation of radium-D from lead with the usual failure but now they made a big leap forward.

36. Hevesy, n. 35, p. 94.
37. Hevesy's book review is: Vegyészeti Lapok. 1918; 13: 70.

As Hevesy mentioned in his talk given to the meeting of the Bunsen Society in 1912, Hevesy and Paneth used radioactivity as an indicator. Their reasoning was clear and almost obvious. If the active and inactive substances have the same chemical properties, they cannot be distinguished from each other in any other way than their radioactivity. Consequently, if radium-D is added to a radioactive element in the inactive lead, the radiation of radium-D will detect the presence of the lead.

In February 1913, Hevesy described the birth and motifs of the tracer method:

> «I felt somewhat annoyed by the unkind criticism by several chemists of the usefulness of radioactivity and radioactive methods for chemical purposes and decided to demonstrate how wrong they are by determining the solubility of PbS in water by mixing it with a known amount of RaD and measuring how much RaD+RaE went in solution. PbS is a most important body for the analytical chemist yet no reliable data has been obtained for the solubility of PbS in water, the latter being exceedingly small. Having great amounts of RaD at our disposal (corresponding to several curies of emanation) we managed to determine the solubility by this very simple method» [38].

By 9 April, 1913, Hevesy was ready to provide the general conclusion in writing to Rutherford:

> «RaD being non separable from lead it is clear that we can use RaD as an indicator of lead and investigate the behavior of lead in minute concentrations, for instance the solubility of insoluble lead salts. We prepared a strong Lead sulphide + Radium D sulphide and used this precipitation to determine the solubility of PbS by measuring the ß activity of the saturated and evaporated solution» [39].

Two weeks after writing this letter (April 24, 1913), Hevesy and Paneth published their results as the texts of two talks given at a session of the Viennese Academy of Sciences. The texts were published in the Academy's journal. The first paper «On the experiments for separating radium-D from lead» detailed all the unsuccessful methods they had used, including distillation, adsorption, diffusion, and dialysis, about twenty different

38. RaE was a bismuth isotope, insignificant in our point of view. Hevesy to Rutherford, letter. February 28, 1913. HSC.
39. Hevesy to Rutherford, letter. April 9, 1913. HSC.

methods altogether. However, as the authors concluded, they «could not indicate any increase in the concentration of RaD» [40]. The next report in the journal describes the practical application of non-separability under the title of «On radioelements as indicators in analytical chemistry» [41]. This is the starting point of the practical usage of isotopes before isotopy had even been defined and coined [42]. The term «isotopes» appeared for the first time in December 1913, in a letter Soddy sent to Nature [43].

Hevesy wrote to Rutherford about this as follows:

> «The idea that some of the common elements might be mixtures of non-separable elements of different atomic weight in constant proportions (...) was advocated a long time ago by Soddy. The whole idea of the 'non-separability' of some elements originates from Soddy. Soddy's view was that the fact that RaD and lead have yet to be separated is not due to the lack of skill of the experimenters but to the principle of the 'non-separability' of those and other elements. I must confess I hoped for a long time that it might be possible to separate those bodies and would not accept Soddy's view on this matter» [44].

8. Delocalized research work

While collaborating with Paneth, Hevesy commuted between Budapest and Vienna, spending more time in the Austrian laboratory than his home in Budapest. In late March 1913, he left Vienna, rested in Budapest, then in

40. Paneth, F.; Hevesy, G. Über Versuche zur Trennung des radium D von Blei. Mitteilungen des Instituts für Radiumforschung. In der Sitzung am 24. apr. 1913. Monatsschrift Für Chemie und verwandte anderer Wissenschaften. Wienna. 1913; 34: 1393.
41. Paneth, F.; Hevesy, G. Über Radioelemente als Indikatoren in der analytischen Chemie. Mitteilungen des Instituts für Radiumforschung. In der Sitzung am 24. apr. 1913. Monatsschrift Für Chemie und verwandte anderer Wissenschaften. Wienna. 1913; 34: 1401.
42. These two papers were published in a journal known locally only. Therefore, another article, appeared somewhat later, is considered the first radioactive indicator paper: Hevesy, G.; Paneth, F. Die Löslichkeit des Bleisulfids und Bleichromats (RaD). Zeitschrift für anorganische Chemie. 1913; 82: 223. This work was also the starting point of a long series of researches that lead to Hevesy's chemistry Nobel Prize in 1943. I have written about the more than twenty years long repeated nominations in Pallo, Gabor. Scientific recency: George de Hevesy's Nobel Prize. In: Elisabeth Crawford, ed. Historical studies in the Nobel Archives: The prizes in science and medicine. Tokyo: Universal Academy Press; 2002, p. 65-78.
43. Soddy, Frederic. Intra-atomic charge. Nature. 1913; 92: 399-400.
44. Hevesy to Rutherford April 25, 1913. HSC.

Meran, a resort in South Tyrol. There he worked on some manuscripts and meditated on theoretical problems related to the structure of matter. He wrote a number of letters to his friends and colleagues all over Europe.

In May, after a short stop in Germany, he returned to England, basically to Manchester, to work on some problems related to the disintegration series and a partly new subject, the separation of chemically non-separable elements by diffusion, based on the difference between their atomic weights. He stayed in England until the end of the summer. In September 1913, he participated in an important conference in Vienna, and then went to Budapest and played an active role in Hungarian scientific life. At the end of the year he rested in various resorts, including Meran and Tatra Lomnic. He spent the first month of 1914 in Budapest and Vienna, and returned to Britain in summer with short stopovers in Vienna, Prague, Berlin and Scheveningen, Holland. In Britain, besides Manchester, he spent some time in London, in Wales, and in Oxford with Moseley to learn the Roentgen diffraction method for use in investigating the chemistry of rare earth elements.

He was always on the move and did not belong to any one local community, making him a delocalized researcher. Wherever he went, he met colleagues to discuss theirs and others' latest results, and made new friends. He had extensive correspondence with many people, particularly Bohr, Rutherford, Paneth and Fajans, speaking about almost all important names in his field, which proves Hevesy's widespread networking activity. He did not seem to belong to any national community, but rather to the whole of European science.

9. Budapest: The first applications of isotopes as indicators and tracers

In the 1910s, despite his activity being so delocalized, Hevesy wanted to establish himself as a Hungarian scientist. His habilitation, his Hungarian publications and his participation in the local scientific life served this goal. In Manchester in 1912, he cooperated with a young Hungarian researcher, László Putnoky, on some problems with uranium's disintegration series. As Hevesy wrote to Fajans, he was highly satisfied with Putnoky's work [45]. Putnoky returned to Hungary in 1913 and soon became a professor of

45. Hevesy to Fajans, April 15, 1913. HSC.

inorganic chemistry at the Budapest Technical University. He was unjustly fired from his post as a result of the political screening carried out after World War II [46].

Between September 1913 and March 1914, then during World War I and after, Hevesy lived mostly in Hungary. Besides giving courses at Budapest University, he built up a research laboratory that, as he reported to Bohr, «means a lot of trouble and work, especially here in Budapest, where everything takes such a long time and I am obliged to do everything on my own» [47]. He had two «research students», one working on diffusion problems of salts in water, the other on melted salts. He seemed to have adapted to the Hungarian situation.

However, Hevesy's mentality, his networking activity and his structural approach, did not fit in with the radioactivity research that followed the natural historical, analytical tradition in Hungary. Because Hevesy was not connected with Weszelszky's group that was working with a group of students at the II. Chemical Institute, he set up his laboratory at the newly opened III. Chemical Institute. In 1911, the university had decided to divide the I. Chemistry Institute into two parts, and to organize the III. Chemical Institute from a part of the I. Chemical Institute. The III. Institute was opened in 1914. The I. Institute specialized mostly in analytical chemistry, while the III. Institute specialized in teaching chemistry to students of medicine and pharmaceutics. Gustav Buchböck was appointed as a full professor and the director of the III. Institute. He was one of the professors responsible for the evaluation of Hevesy's habilitation process. Buchböck, Hevesy's friend and an expert on physical chemistry, had fresh eyes, and much interest in and appreciation of Hevesy's work. Therefore, without making any official noise, Hevesy was able to open his laboratory, and work with students without concerning himself with the research going on at the only official radioactivity institute in Hungary.

One of Hevesy's research students, Erzsébet Róna, published an investigation of the uranium series, which could be considered a continuation of the Hevesy-Putnoky article [48]. Róna acknowledged Hevesy's important

46. Hevesy, Georg; Putnoky, Laszlo. Über die Diffusion des Urans. Physikalische Zeitschrift. 1913; 14: 63.
47. Hevesy to Bohr, letter. October 27, 1913. HSC.
48. Erzsébet, Róna. Az urán átalakulásairól. Mathematikai és Természettudományi Értesítő. 1914; 32: 350-386.

advice in her research but she was the only author. Her next publication was co-authored with Hevesy. This was a study on the speed of the solution of lead and bismuth ions in nitric acid. They showed that an exchange process could be detected by the radioactive indicator method between active lead chloride and inactive lead nitrate, and between the solid and liquid phases of the lead salts. They used ThB, a radioactive lead isotope, as an indicator [49]. This work was the first application in Hungary of the radioactive tracer method in exchange processes, which was the main focus of Hevesy's interest at that time.

In World War I, Hevesy served in the Austro-Hungarian Army, but even then he was able to continue publishing. He became interested in colloid chemistry and in particular the colloids of radioactive substances, much like Béla Szilárd, who also published a substantial study on the subject [50]. After the war, like all Hungarians, Hevesy went through a turbulent period. The Austro-Hungarian Monarchy collapsed, revolutions and counter revolutions broke out, a communist regime ruled the country for a while in 1919, anti-Semitism was on the rise, the economy could hardly work, and the Romanian army occupied a large part of the country. Hevesy was first appointed as an extraordinary professor in 1918, later, in 1919, to full professor and head of the II. Institute of Physics, which was a transitional position before establishing a physical chemistry chair.

His publications were written from somewhere new. Although he was still on friendly terms with Buchböck, he did not return to the III. Chemical Institute. Instead, he worked outside the university, at the Budapest veterinary school. His change of site was probably the reason why Erzsébet Róna also wrote her publication from this school in 1917. She acknowledged Hevesy's help in her research into a typical Hevesy topic: the measurement of the diffusion constant of radium emanation in various solvents, such as ethanol, benzene and toluene [51]. This was the last study they made together. After that, they went their separate ways.

49. Hevesy, György; Rona, E. Die Lösungsgeschwindigkeit molekularer schichten (Thorium B). Zeitschrift für physikalische Chemie. 1915; 89: 294 and 303. Hevesy selected this paper for publication in his collected works: Hevesy, George. Adventures in radioisotopes research. Collected Papers of Georges Hevesy. London: Pergamon Press; 1962, p. 89-96.
50. Hevesy referred to Szilárd's article quoted in note 19.
51. Róna, Erzsébet. A rádium-emanáció diffúzióállandója és atomátmérője. Magyar Chemiai Folyóirat. 1917; 23: 156-160 (Zeitschrift für physikalische Chemie. 1917; 92: 213).

At the veterinary school, Hevesy continued his studies of exchange processes with another collaborator, László Zechmeister, an excellent organic chemist. They extended Hevesy and Róna's earlier investigations of the exchange processes of inorganic compounds to organic compounds, to cases in which changes of valence take place. They mixed radioactive tetravalent lead acetate (Pb/CH3 COO/4) with inactive bivalent lead acetate (Pb/CH3 COO/2) and measured how the activity of the former gradually diminished to half of the original value, proving that the active isotopes exchanged with the inactive ones. They also proved that an exchange process between the atoms of the same molecules could only be experienced if the molecules are in dissociated state [52]. This article extended the isotope indicator method to the organic world.

The explanation that Hevesy was working in a new laboratory in Budapest probably lies in the appointment of his friend, Gyula Gróh, to head of the veterinary school's chemistry department. The school had a laboratory that might have seemed appropriate to Hevesy for radioactivity research. An important two-part article was published under the name of Gróh and Hevesy on the self-diffusion of lead. A mixture of lead and ThB was placed at the bottom of a vertical tube. At the top, lead was placed at three times greater a height than that of the mixture. The whole column was heated to 300 C° and left at this temperature for three days. Then it was slowly cooled down and cut into pieces. The activities of the pieces were measured. From this, the speed and other features of self-diffusion could be calculated. In another investigation, lead was left at 280 C° for a year, and the activity of RaD was measured along the column. In this way, they could observe how high the lead atoms rose by spontaneous diffusion from the bottom. These measurements can be considered a further step in the application of the indicator method, as isotopes were used as real tracers here [53].

52. Hevesy, G; Zechmeister L. Über den intermolekularen Platzwechsel gleichartiger Atome. Berichte der Deutsche chemische Gesellschaft. 1920; 53: 410. Über den Verlauf des Umwandlungsvorgangs nomerer Ionen. Zetschrift für Elektrochemie. 1920; 26: 151. Egynemű atom intermolekuláris kicserélődéséről. Magyar Chemiai Folyóirat. 1920; 26: 58-64. Also published in Hevesy's Collected Papers. Adventures in Radioisotopes Research. Collected Papers of Georges Hevesy. London: Pergamon Press; 1962, p. 103-109.
53. Hevesy, G.; Gróh J. Die Selbstdiffusionsgeschiwindigkeit des geschmolzenen Bleis. Annalen der Physik. 1920; 63: 85; Hevesy, G.; Gróh J. Die Selbstdiffusionsgeschwindigkeit in festem Blei. Annalen der Physik. 1921; 65: 216. This was also published in Hevesy's collected papers Col-

This was Hevesy's last work in Hungary. The experiments at the veterinary school lasted for five years until 1920, when Hevesy left Hungary for good.

10. Epilogue and concluding remarks

Hevesy's group soon disappeared from Hungarian scientific life. After the communist revolution, in 1920 Hevesy was expelled from Budapest University and even lost his title of privat dozent and «venia legendi», the right to teach. The veterinary school proved instrumental at the time, because Gróh was able to secure a laboratory for his outcast friend. In 1920, Niels Bohr offered Hevesy shelter and a research opportunity in Copenhagen. This was Hevesy's first escape for political, mostly anti-Semitic reasons, followed by two others later in his life [54].

Hevesy's collaborators met a similar fate. Before meeting Hevesy, Róna studied in Karlsruhe with Fajans, who became his friend. Her interest and activity in modern chemistry were not Hungarian products. Soon after the political turmoil, she also left Hungary, worked in Vienna for some years, then in Sweden and Norway with Ellen Gleditsch, who was related to Béla Szilárd at the Curie laboratory in Paris. Róna ended up in the USA [55]. She never really left the field of radioactivity, or nuclear science, in which she had made a good start as Hevesy's contributor in Budapest.

László Zechmeister was appointed professor of chemistry at the newly established University of Pécs, Southern Hungary, in 1922. As a former student of Willstätter in Germany, he did not return to the field of radiochemistry, but instead continued his master's line related to the chemistry of plant pigments, using chromatography, of which he became a major exponent.

lected Papers: Adventures in Radioisotopes Research. Collected Papers of Georges Hevesy. London: Pergamon Press; 1962, p. 110-113. Somewhat later Hevesy repeated this investigation with a Russian contributor: Hevesy, G. Obrutsheva, A. Seldiffusion in solid metals. Nature. 1925; 115: 674.

54. I have written about the details of Hevesy's emigration in Pallo, Gabor. Why did George von Hevesy leave Hungary? Periodica Polytechnica. 1986; 30: 97-115.

55. Rona, Elisabeth. How It came about: radioactivity, nuclear physics, atomic energy. Oak Ridge, Tenn.: Oak Ridge Associated University Press; 1978; Rentetzi, Maria. Gender, Politics, and radioactivity research in Interwar Vienna: The case of the Institute for Radium Research. Isis. 2004; 95: 359-393.

He left Hungary in 1941 by invitation of Linus Pauling at Caltech, where he stayed until his death in 1972.

Only Gyula Gróh stayed in Hungary. He became a renowned professor for generations of Hungarian chemists until he was dismissed from Budapest University in 1950, where he had succeeded Buchböck in 1936. His textbook-writing activity was highly appreciated. Gróh maintained his relationship with Hevesy throughout his life. They corresponded and sometimes met inside and outside Hungary, even in Copenhagen, where Gróh was invited by Hevesy to give a talk. Gróh nominated Hevesy for the Nobel Prize in 1939 [56]. After Hevesy left, Gróh did not immediately give up his research on self-diffusion. He, and one of his students later, published on solubility problems investigated by radioactive indicators. He solved lead in mercury and the amalgam was in contact with lead nitrate containing ThB, a radioactive lead isotope, and measured the growth of the radioactivity of the amalgam [57]. Gróh's student, László Auber, completed this investigation in 1928 [58]. At this point, the continuity of Hevesy's influence on the isotope research in Hungary came to an end.

Nevertheless, radiochemistry lived on at the Radium Station. The traditional, descriptive, natural historical data gathering was able to continue under Weszelszky's guidance until his retirement in 1937, when the new head, Lajos Imre, modernized the Institute's research program. Gradually, the institute absorbed the new results in the field from international journals, after having missed the opportunity to learn from colleagues experimenting in the adjacent building at the university, and lecturing in the same scientific institutions.

The traditional natural historical approach only geographically met the modern structural views in the field of radiochemical research in Budapest. In a letter to Fajans, Hevesy complained that the leading Hungarian chemists, including some professors and Weszelszky, considered him a «charlatan» for his modern chemical views [59]. The local practice proved too stabile, closed and independent to easily absorb a research style imported from a wide region of Europe by a young delocalized researcher. Most of the open,

56. Móra, László. Gyula Gróh élete és munkássága. Budapest: Technika Alapítvány; 1996.
57. Gróh, Gyula. Az ólom és a bizmut oldódási sebességéről és abszolút elektrolytos oldási tenziójáról. Mathematikai és Természettudományi Értesítő. 1927; 44: 544-554.
58. Auber László. Az ólom oldódássebességéről. Magyar Chemiai Folyóirat. 1928; 34-62 and 77.
59. Hevesy to Fajans, October 4, 1920. HSC.

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receptive minds that collaborated with Hevesy left Hungary and became parts of the wider scientific community. However, the natural historical approach proved fruitful to Hevesy's research, mainly in working out the method of radioactive indicators, the starting point of isotope applications. The Viennese Radium Institute represented the same data gathering approach, but its devices, personnel and mentality offered an incomparably better atmosphere and opportunity to Hevesy than its parallel in Budapest. That was how the descriptive, natural historical style that was so characteristic of Hungarian chemistry at that time met the British structural approach in Hevesy's delocalized early research into isotopes. ∎