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NMB Innovators Series

NMB Innovators Series Professor Heinz Hubert Coenen, PhD

With this series of editorials, NMB honors the scientists in the radiopharmaceutical sciences domain that have dedicated their career to our field and made substantial contributions.

1. Introduction

The editors are pleased to present the second "NMB – Innovators" editorial featuring with Professor Heinz H. Coenen, PhD, Emeritus Professor at the University of Cologne (UoC). In July 2024 he celebrated his 75th birthday after more than 55 years of devoted activity in chemical sciences.

Throughout his academic career, Heinz H. Coenen (known to most as Heinz) achieved numerous ground-breaking advances in radiopharmaceutical chemistry. His chemical genealogy can be traced back to the institute of Otto Hahn in Berlin, following a long line of prolific pioneers of nuclear chemistry. His work benefited from both his rigorous knowledge of radiochemistry, acquired at the UoC, and the excellent research environment and interdisciplinary collaboration possibilities at the Forschungszentrum Jülich (FZJ). These conditions enabled him to apply radiochemistry as a tool for solving natural scientific, pharmacological and medical problems (Fig. 1).

Of his achievements, particularly noteworthy is his development of several radiotracers for SPECT and PET diagnostics. He was a pioneer in the development of novel radiolabelling methods and preclinical evaluation of promising tracers, as well as translation to the large-scale automated production of radiopharmaceuticals for clinical use. These innovations facilitated significantly to the breakthroughs necessary for widespread clinical application of PET. Besides these scientific advances, he also played a key role via professional organizations and committees in establishing the legal framework necessary for regulatory approval of PET radiopharmaceuticals for routine clinical practice. Early on, he became aware of stringent purity needs of radiopharmaceuticals, and the importance of regulatory and legislative requirements, compelling him to take actively part in task groups and committees to address these needs [1,2].

Heinz led a globally recognized research group and authored about 400 publications spanning nuclear chemistry, radiopharmaceutical chemistry and molecular imaging. His scientific impact is evident in numerous high-profile publications, as well as the supervision of over 60 PhD students - many of whom now lead successful radiopharmaceutical research groups. Teaching was particularly close to his heart; as professor at the UoC he successfully advocated for the preservation of nuclear chemistry as a field of study; making it one of only three institutions in Germany offering comprehensive instruction in this specialty. Besides his work at the UoC, he helped establish at the ETH Zurich a European postgraduate training program for radiopharmaceutical chemistry and radiopharmacy. He taught at this institution for more than 20 years, and lectured at the Pharmaceutical Faculty of the JWG-University, Frankfurt for 5 years.

His commitment to the field of radiopharmaceutical science extended to numerous professional organizations. Heinz co-founded the Working Group Radiochemistry and Radiopharmacy, which provides the networking of central European laboratories. He was a long-time board member of the International Radiopharmaceutical Chemistry Society, which evolved to the Society of Radiopharmaceutical Sciences, and served as its 4th president. He also organized numerous workshops, meetings, and conferences in his field, including the 17th International Symposium on Radiopharmaceutical Sciences (2007) and (with Syed M. Qaim) the 6th International Conference on Nuclear and Radiochemistry (2004), both in Aachen, Germany.

2. Start of his career and his path to radiopharmaceutical chemistry

Born and raised in a farming family, Heinz developed an early interest in the life sciences. He attended the University of Cologne, where he focused on chemistry, complemented by coursework in biochemistry and genetics. As a student working at FZJ, he became intrigued at the potential of radiotracers for studying biochemical processes in vivo. Like many of the pioneers of radiopharmaceutical chemistry at the time, he studied hot-atom chemistry and completed Diplom research under Prof. Gerhard Stöcklin on the reactions of hot ^{38}Cl recoil atoms with arenes [3]. His subsequent doctoral thesis research was also at the FZJ under the supervision of Gerhard Stöcklin with a research focus on reactions of nucleogenic bromine cations with arenes, and the practically carrier-free labelling of organic molecules with bromine-76 and bromine-77 [4]. The stringent requirements of hot-atom-chemistry proved to be valuable in later applications of nuclear techniques in biomedical research, in particular for the analysis of practically massless substances of highly active, short-lived compounds, as well as measurement of nuclear data and radionuclide production, targeting and labelling techniques.

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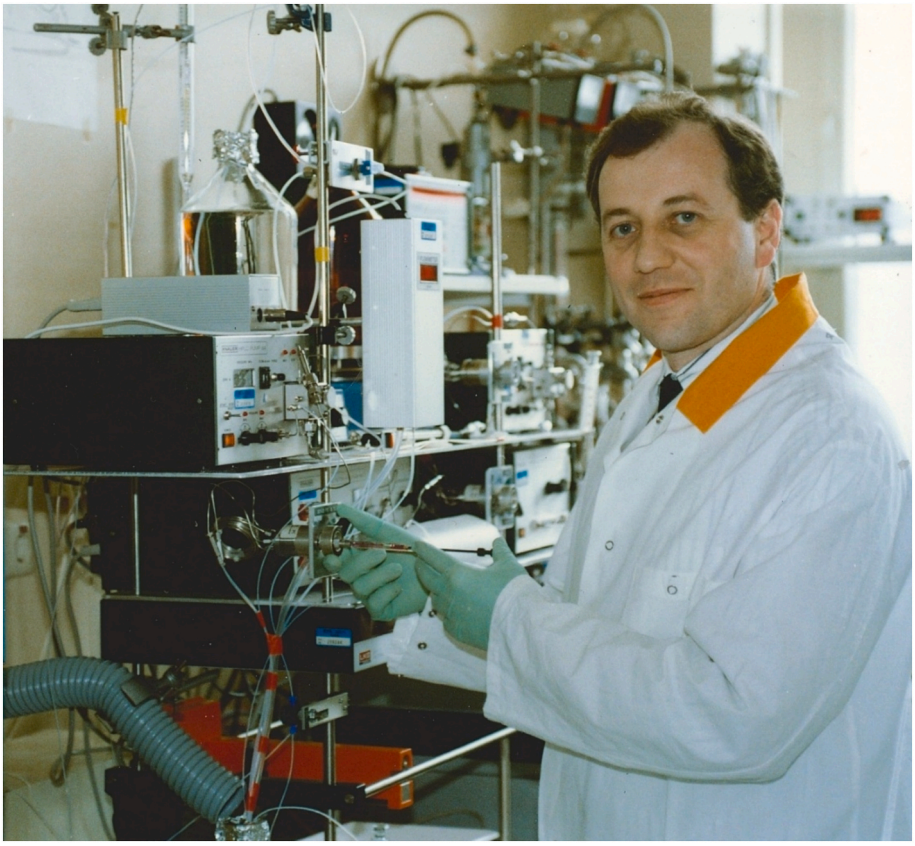

Fig. 1. Working on the HPLC - taken in 1990 in the FZJ laboratory.

After completing his dissertation in 1979, he remained with the Jülich research team and applied of radiochemical methods to create of radiopharmaceuticals for imaging cardiac metabolism; i.e., radiolabelled fatty acids, labelled with both SPECT and PET nuclides. This comprehensive research included studied radiolabelling at various positions, in vivo metabolic studies in mice, as well as pharmaceutical preparation for human use. In addition to this radiopharmaceutical development, he gained extensive knowledge in radiolabelling methods for heavy radiohalides [5]. At this time, he developed an early reputation for a series of pioneering work on ^{76}Br- and ^{123}I-labelled radiopharmaceuticals.

3. Postdoctoral experience

Shortly after his work on fatty acids, Heinz diversified his training in radiopharmaceutical science with a fellowship as a visiting professor in 1981 with Prof. William C. (Bill) Eckelman, at the George Washington University School of Medicine. His work with this pioneer of radiopharmaceutical science greatly expanded his understanding of the limitless potential of radiopharmaceuticals for study of biological processes in vivo [6]. He also learned at this time the benefits of close collaboration with medical colleagues, who can define clinical problems that radiopharmaceutical scientists can focus on solving. This one-year fellowship gave him a broader perspective of the field, many new personal connections, and several future research ideas.

4. Focus on ^{18}F-chemistry

Upon returning to FZJ from his fellowship, Heinz spearheaded the development of PET radiopharmaceuticals labelled with the fluorine-18, a central topic that was to remain throughout his career (Fig. 1). Although it was universally recognized that fluorine-18 was an ideal radiolabel for PET radiopharmaceuticals, chemical techniques for its incorporation into molecular structures was very limited at that time.

A key achievement in this regard was the development of the Kryptofix2.2.2 method for radiofluorination by Heinz and co-workers at FZJ. This radiolabelling methodology was based on earlier macroscopic fluorination chemistry by a group in Hamburg, and enabled the efficient and large-scale nucleophilic no-carrier-added radiosynthesis of ^{18}F-labelled compounds [7]. It has since developed into the universally-accepted standard methodology for high-specific radiofluorination of radiopharmaceuticals.

In collaboration with Kurt Hamacher and Gerhard Stöcklin at FZJ, this technique was applied for the first-ever multi-Curie (GBq) synthesis of 2-deoxy-2-[^{18}F]fluoro-D-glucose ([^{18}F]FDG). It can arguably be stated that the availability of large quantities of [^{18}F]FDG from a single batch production made possible the growth of clinical PET, since it greatly reduced production costs and lack of availability for this universally used radiopharmaceutical. The new method streamlined production method facilitated the first automated synthesis module [8], which was prototype for today's commercially available ^{18}F-synthesizers.

The “Kryptofix method” was also used for preparation of n.c.a. ^{18}F-fluoroalkylation and ^{18}F-fluoroacylation agents [9] as well as direct radiofluorination of arenes [10]. The utility of these are demonstrated by the synthesis of [^{18}F]fluoroethylspiperone [11] which was used at the Max Planck Institute for Neurological Research (MPI) Cologne [12] for PET studies of cerebral dopamine D_2 receptor density.

Besides nucleophilic radiofluorination, Heinz was also active in electrophilic radiofluorination of PET radiopharmaceuticals [10]. In this context, he worked on radiotracer development for imaging protein synthesis, which were gaining importance in tumour diagnostics, and identified 2-[^{18}F]fluoro-L-tyrosine as a promising candidate whose development required the full spectrum of bench-to-bedside translation. Following extensive chemistry and preclinical studies in mice, clinical

studies in collaboration with the MPI Cologne, demonstrated its utility as a protein synthesis tracer. Unfortunately, tumour uptake was primarily affected by transport-mediation rather than by protein incorporation [13,14]. For this fundamental research, however, he received the Mallinckrodt Award of the German Nuclear Medicine Society (DGN) in 1990.

5. Research experience in the clinical setting

In 1991, Heinz accepted an Associate Professorship for Nuclear Chemistry and Radiopharmacy at the Clinic for Nuclear Medicine (Director Prof. Christoph Reiners) in the University (Gesamthochschule) of Essen, Germany. The transition to this highly clinical environment provided new perspectives to his career development. At Essen he was able to collaborate with cardiologists, psychiatrists and neurologists on clinical research projects [15,16]. In part refocusing on radioiodinated radiopharmaceuticals during that time, he developed the first bimodal tracers for a hybrid combination of imaging techniques. He validated the suitability of kinetics in humans for iodine-123 SPECT tracers (such as IMT and β-CIT) by comparing them with their ^{124}I-labelled PET analogues, thereby justifying their application on patients with brain tumours and Parkinson's disease, respectively [16–18]. He demonstrated the utility of longer-lived positron emitters like iodine-124 for study of extended biokinetics and to perform preclinical ex vivo studies on animals [19]. While in Essen he was also deeply involved in the establishment of a new cyclotron facility and managing associated regulatory challenges.

6. Merging his scientific experiences at Forschungszentrum Jülich

In 1996, Heinz returned to the FZJ and succeeded Gerhard Stöcklin as Full Professor at the UoC and as director of the Institute of Nuclear Chemistry at FZJ (Fig. 2). He restructured the institute, established a new research group for the pharmacological evaluation of radiotracers, and strengthened collaborations with clinical researchers both within the FZJ, e.g. with Karl-Josef Langen on preclinical and clinical evaluation of [^{123}I]iodo- and [^{18}F]fluoroamino acids for tumour imaging, and at surrounding university hospitals [20,21]. To address clinical needs, it was necessary to optimize the synthesis of known radiopharmaceuticals with improved automation. Examples are preparation of [^{18}F]altanserin [22] and 2-[^{18}F]fluoroethyl-L-tyrosine ([^{18}F]FET) [23], which is now the leading diagnostic agent for brain tumours [21]. Additionally, new radiotracers were successfully developed with “bench-to-bedside” approaches, including the adenosine A_1 ligand [^{18}F]CPFPX [24] and the four stereoisomers of [^{18}F]fluoroproline [25].

In parallel to these initiatives, he continued development of new radiofluorination methods to expand the range of clinically available ^{18}F-radiotracers. A major initiative was n.c.a ^{18}F-labelling of electron-rich aromatic compounds on large activity scales, which was generally only possible via nucleophilic methods starting from [^{18}F]fluoride [26] and build-up reactions, e.g. via ^{18}F-fluoroarylation [27]. Together with Johannes Ermert he studied the use of iodonium compounds to enable the direct synthesis of electron-rich arenes [28–30]. He also established a new pathway for the synthesis of amino acids with ^{18}F-incorporation into aromatic positions, such as [^{18}F]fluoro-L-DOPA and 2-[^{18}F]fluoro-L-tyrosine [31].

The availability of several particle accelerators at the FZJ and the expertise of Syed M. Qaim in nuclear chemistry provided broad research opportunities for study of nuclear data and reactions, and optimization radionuclide production. This field of research remained another major focus of Heinz throughout his scientific career, leading to the development of numerous production methods for radionuclides. Notably, he contributed to the production of non-standard positron-emitting nuclides like ^{45}Ti [32], ^{52}Mn [33], ^{55}Mn [34], ^{64}Cu, ^{67}Cu [35], ^{75}Br, ^{76}Br [36], ^{120}I, ^{124}I [37], all of which are potentially medically relevant [19].

Fig. 2. Taken in the middle of his scientific career around 1996.

He also developed new labelling methods, for selenium-73, enabling the n.c.a. synthesis of both aliphatic and aromatic selenium compounds [38]. Heinz also designed innovative complexes of radiomanganese [39,40] that use the isotopic pairs to combine the positron emitter ^{52g}Mn and stable paramagnetic ^{55}Mn for synergistic integration of the high sensitivity of PET with the excellent spatial resolution of MRI.

His broad scientific curiosity and the diverse interdisciplinary opportunities compelled him to vigorously support the working group at UoC that he sustained. With a primary mission of teaching and training, that group remains focused on the use of nuclear methods in geological and cosmological research. At FZJ he also collaborated with plant biologists, enabling the application of PET in plant research on transport phenomena upon photosynthesis [41].

Thanks to the outstanding achievements of Heinz and the Institute of Nuclear Chemistry that he led, the management of FZJ, together with the Helmholtz Association of German Research Centres, decided to construct a new laboratory building for his radiochemical research, equipped with a state-of-the-art level-IV cyclotron with p-, d- and α-beam options and GMP-compliant laboratories. This is a fitting legacy for a productive scientist like Heinz to leave for successive researchers.

7. Activities following retirement

Since his retirement in 2015, Heinz is spending more time with his wife and family, especially his grandchildren, to somewhat make up for the many hours he spent in laboratory and office (Fig. 3). Nevertheless, he still continues to dedicate himself to radiopharmaceutical chemistry, publishing existing research results and working on advisory

Fig. 3. Taken in 2015 at his farewell symposium at FZJ.

committees and as reviewer. As the field attracted researchers from diverse backgrounds, inconsistent terminology became a challenge. Heinz strongly advocated for standardized nomenclature in radiochemistry and radiopharmacy. Under his and Anthony Gee's leadership, international experts revised the nomenclature rules to better fit the evolving discipline. The resulting guidelines, published as a reference by the Society of Radiopharmaceutical Sciences (SRS), are now widely accepted [42,43]. He also authored a brief history of the SRS [44].

CRediT authorship contribution statement

Johannes Ermert: Writing – original draft, Conceptualization. **Stephen Moerlein:** Writing – review & editing.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work the authors used ChatGPT in order to improve the language. After using this tool, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

Declaration of competing interest

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

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