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Special Issue dedicated to Professor Hanns-Ludwig Schmidt on the occasion of his 85th birthday

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EDITORIAL AND TRIBUTE

Special Issue dedicated to Professor Hanns-Ludwig Schmidt on the occasion of his 85th birthday

This Special Issue of *Isotopes in Environmental and Health Studies* honours Hanns-Ludwig Schmidt, a pioneer of stable isotope science. H.-L. Schmidt has had a most distinguished career in isotope research. It started with the synthesis of radioactive tracer compounds and led to a wide range of fundamental, methodical and applied research on the stable isotope composition of the main elements of the biosphere, principally $^{13}\text{C}/^{12}\text{C}$, $^{15}\text{N}/^{14}\text{N}$, $^2\text{H}/^1\text{H}$, $^{18}\text{O}/^{16}\text{O}$ and $^{34}\text{S}/^{32}\text{S}$. That career has been an awe inspiring long one, 55 years [1] and counting, as is documented by his contribution to a review on the causes and implications of *in vivo* isotope fractionations [2] in this Special Issue. Young stable isotope researchers may wonder how such a career could have unfolded, what circumstances and coincidences – academic and other – influenced and guided it, and which interactions with colleagues and students accompanied and enriched it. For that reason H.-L. Schmidt was asked by the Editors of this Special Issue to contribute a personal account of his career. After some hesitation with regard to the appropriateness of such a contribution, H.-L. Schmidt agreed and presented us with a concise and lively account of his walks of life with isotopes [1].

H.-L. Schmidt and his students and co-workers have contributed many fundamental findings to several fields of basic and applied stable isotope research, in plant biochemistry, human and animal physiology and ecology, environmental science, archaeology, paleoscience, forensics and food authenticity proofing. That diversity is also reflected in the contributions to this Special Issue by colleagues and by former students and co-workers [2–13]. Schmidt's own work included early studies on photosynthetic carbon isotope discrimination [14,15], which provides clues on the affiliation of plant species to photosynthetic types, and on limitations of discrete steps in the transfer of CO_2 from the air to RuBisCO in both C_3 and C_4 plants, and on the enzymatic mechanisms underlying post-photosynthetic carbon isotope discrimination phenomena. The occurrence of inter- and intra-molecular isotope distribution in natural compounds was known at least since the famous paper of Abelson and Hoering [16]. Schmidt and his group provided a theoretical framework explaining such phenomena by connecting observations with specific enzyme reactions during synthesis and degradation processes in autotrophic or heterotrophic organisms [2,17–21]. One such finding with wide implications was the heterogeneous carbon isotope pattern in glucose [22] with a ^{13}C enrichment in the positions C-3 and C-4 which is caused by an equilibrium isotope effect on the aldolase reaction [23]. This pattern, also detectable in leaf soluble sugars and transitory starch [24], is responsible for the ^{13}C depletion of acetogenic lipids and the relationship between $\delta^{13}\text{C}$ values of the fermentation products ethanol or acetic acid and that of the source sugar ([10] and literature cited therein), and contributes to the ^{13}C enrichment of dark-respired CO_2 in plants ([25, see also [9,11]) and in connection with further isotope effects

(e.g. [26]) to inter- and intra-molecular diversity of carbon isotope composition in plants (e.g. [27–29]). A general ^{13}C depletion of methoxyl groups derived from SAM was discussed to be caused by an isotope effect on formation or transfer of CH_3 groups [30]; an application of this ^{13}C depletion of methoxyl groups in climatology is one topic of this Special Issue [13].

H.-L. Schmidt and his group applied naturally ^{13}C enriched food or macronutrients based on C_4 plants not only for breath tests [31], but also to follow digestion, absorption and oxidation of macronutrients *in vivo* in human subjects and animals [32,33]. Later on, work dealing with amino acid metabolism based on natural tracers, and on ^{13}C and ^{15}N labelled substances became possible [34,35] as a result of methodological developments (e.g. GC-c-IRMS, derivatisation of amino acids) initiated by H.-L. Schmidt and his co-workers. The paper of Hammon et al. [8] in this issue is one example for the influence of that early research on recent investigations of animal physiology. The influence of glucose metabolism (glycolysis, pentose phosphate pathway, etc.) on the glucose isotopologue pattern in human blood is topic of the article by Marsch et al. in this issue [6].

Fundamental studies on nitrogen and oxygen isotope fractionations in the formation and decomposition of nitrate [36] were honoured with a scientific prize and provided a methodology for tracing sources of nitrate in aquifers [37]. A review on the inter- and intra-molecular systematics of hydrogen isotopes [21] was highly influential for the field of paleoclimatology [38]. The dependency of isotope ratios of organic substances used in paleoclimatology on environmental drivers is also subject of one article in this Special Issue (alkanes [12]). A second article deals with the stability of intra-molecular carbon and hydrogen isotope ratios in methoxyl groups during decomposition processes [13]. The theoretical framework developed by H.-L. Schmidt and his co-workers [18,21] was further on used to elaborate ‘systematic rules for the isotopic characteristics’ of phenylpropanoids based on the knowledge of biosynthetic pathways and specific reaction mechanisms involved [39, p. 1094]. The work of Brenna et al. [5] in this issue reports on intra-molecular characteristics of organic molecules in following the stereochemical way of the hydrogen atoms through an ene-reductase reaction.

Investigations of isotopic changes during decomposition of collagen from bones [40,41] or tracing the origin of marble from the Pergamon altar provided original contributions of H.-L. Schmidt and his group to archaeology [42], and studies on mechanisms involved in the formation and decomposition of humic material [43–45] contributed to the biogeochemistry of soil organic matter transformations. Furthermore, H.-L. Schmidt gave the first insight into sulphur isotope fractionation in the metabolism of higher plants and animals [46].

The modern forensics and archaeology tools kit includes multi-element stable isotope analysis techniques as essential methods. This tool is often applied to human tissues such as teeth, bones or hair for inferring temporal changes in the isotopic composition of foodstuffs and beverages – which are determined by regional/geographic or cultural conditions – and, hence, present clues for geographical movements of ancient and modern man or changes of dietary habits. In fact H.-L. Schmidt and collaborators [47] have published one of the first measurements of $\delta^{13}\text{C}$ values in human hair in relation to dietary protein. In this issue, the paper describing changes of multi-element stable isotope composition of the hair of German air force pilots during their stay in the USA presents a new case study in this field of research [7], and the study of the dispersal of fast-spreading invasive gobies along man-made river bank structures is an example of an application in invasive species ecology [4]. Isotopic signals in body tissues can also be altered by health conditions that affect isotope fractionation in metabolism, and may present evidence, for example, of starvation or malnutrition. The development of substance-specific and intra-molecular isotope analysis has greatly expanded the interpretative power of multi-element stable isotopic tools in forensics. Similar approaches were first applied for origin assignment of heroin and cocaine [48,49], and are still widely used to trace the origin of drugs (natural, semi-synthetic or synthetic), explosives and other compounds of forensic interest. The knowledge of the relation

between δ values of proteins in animals and their food discussed above could be used to indicate illegal feeding of meat and bone meal to ruminants [50,51]. From the beginning, H.-L. Schmidt and his group have contributed much to the methodical, theoretical and empirical foundations of this diverse field of stable isotope research.

In a methodical sense, the works on forensics, archaeology and human and animal biology have very close relationships with isotope methods that are used in food authenticity control, another rapidly expanding applied field of stable isotope research to which H.-L. Schmidt and his co-workers have contributed since the beginnings of works in this area [52]. The work by Schneider et al. [3] in this issue is a contribution to this topic, and documents the importance of ^{13}C discrimination phenomena in dairy cattle metabolism for understanding C isotope signals in milk components. Very early on H.-L. Schmidt pointed onto the necessity to perform multi-element and compound specific, and not only bulk isotope analyses, to check for intermolecular isotopic correlations, and in some cases to perform even positional specific (intra-molecular) isotope analyses, considering the biochemical pathways which had formed the substances and left a typical stable isotope pattern [53]. In this issue, the paper dealing with balsamic vinegar authenticity [10] is rooted in that early work of H.-L. Schmidt.

The basic stable isotope research of H.-L. Schmidt and his co-workers provided much of the scientific background for the adoption of stable isotope methodology as an official method for routine controls by the food industry and public food control authorities, for addressing questions of geographical authenticity of food, especially for products with a protected designation of origin (PDO), or for customs and subsidies purposes. The contribution of H.-L. Schmidt to instrumental and methodological developments is documented by several publications starting with measurement of $\delta^{18}\text{O}$ and $\delta^{15}\text{N}$ in nitrate [54], measurement of $\delta^{18}\text{O}$ and (site-specific $\delta^{13}\text{C}$) of organic compounds with an EA-IRMS system [55], first steps towards a measurement of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ of GC eluates [56], development of add-ons for additional simultaneous measurement of $\delta^2\text{H}$ and $\delta^{34}\text{S}$ by combustion EA-IRMS [57] and finally $\delta^{18}\text{O}$ measurement of nitrogen-containing substances [58].

Hanns-Ludwig Schmidt has been at the origin of much of present-day stable isotope research in practically all fields of science that use the stable isotope methodology. He has contributed most significantly to a wide range of isotope methodologies, to the development of theoretical frameworks for understanding isotope effects, and to the use of empirical knowledge of the isotope composition of biological materials for understanding biological, (agro-)ecological and environmental problems. In doing so, he has also inspired a great number of students and a great number of fellow scientists. Wherever we encounter a publication using stable isotopes, chances are high that there is an 'element of H.-L. Schmidt' in it (even when papers do not directly reference his work). We are greatly indebted for his scientific legacy, and we look forward to having him around and to listen to his advice for many years to come.

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