特　別　講　演

第１日　１０月２日（月）

特別講演１　　阪上正信

放射化分析特別講演　　Frans De Corte

第２日　１０月３日（火）

特別講演２　　Vélonique Michel

1S01　放射能研究の源流を訪ね将来を思う

（金沢大・名誉教授）阪上　正信

あと一ヵ月余でX線発見１００年，それに続く四ケ月後に放射能発見１００年を迎える記念すべき本討論会で，その経緯とその後の発展の流れをたどることは意義深い。「ものごとの本質の理解を助けるものはその発展の歴史である」ともいわれる。 Cannnizzaroも The mind of a person learning a new science has to pass through all the phases which the science itself has exhibited in its historical evolutionと述べいる。

その際，ただ項目的に歴史を概観するのみでなく，「科学とは，それに没頭し，それを推し進めた人々の個性や人生と深く係わる人生の成果である」ともいわれるように，個々の科学者の生涯を顧みることも，いま研究にたずさわる人へのはげみと　教えともなる。

すべての科学的発見には，たとえ偶然と表面的にはみえても，それに至るいわば必然的偶然を生むべき科学的・社会的さらに人間的な準備熟成期間がある。地球創成のさいから存在しながら，目に見えないため非常に発見の遅れた放射能についても，同様に目に見えぬ空気成分の18世紀中葉の研究に始まり，赤外線・紫外線の発見と写真法の進展，電気と真空中（脱空気）での放電現象の研究，さらに蛍光・燐光体の検討がその基盤にある。

1895年11月ドイツWürzburgの W.K.Röntgenは Crookes放電管から黒紙を通しても蛍光板を光らせたX線を発見した。そしてその性格の丹念さでその性質を種々に研究した結果を論文に年末に書き上げ，１月１日に別刷となったそれをX線による透過写真とともに，各地の物理学者友人に送付した。フランスでそれをうけとった H.Poincaré はその返信のなかで，X線が陰極線のみで発生するものなのか，その原因が何であれ蛍光を発するものからも発生しないのか（陰極線のある放電管ガラス面は蛍光を発する）という彼の問いかけを述べ，フランスアカデミーでのその報告においてもこの考えを示唆した。この席に出席してこれを聞いた H.Becquerelは，祖父より３代パリーの自然史博物館応用物理部にあって蛍光物質の研究を重ねていたので，早速それを実験的に検討し始めた。まず日光をあてて，強い蛍光を発するウラニル塩について，黒紙に包んでもX線様の写真に感ずる放射線放出を認めたが，２月末パリーの天候が悪くかったので，日光に当てずに暗い机の引き出し内にウラニル塩を黒紙に包んだ写真乾板の上に載せて実験準備のため放置していたのをたまたま現像し，意外にも黒化像を認めた。さらに蛍光は発しない金属ウランについても検討しても同様の黒化像を認めた。これが1896年ウランの放射能発見の経緯である。

そのころポーランドから Parisに留学滞在し，物理学者 P.Curieとも1895年結婚し Mme Curie となっていた Marie Sklodowska は博士論文の題材としてこの研究に注目した。そして種々の物質の放射能の強さを相互に比較検討したが，そのなかで，ドイツ Erlangen の G.H.Schmidtとは独立にトリウムの放射能を発見し，またウランよりもその原鉱のピッチブレンドの放射能がかなり強いこと見出した。彼女はその原因をその中の未知物質によるのでないかと考え，娘時代に養われた根気強さで化学的分析手法と各部分の放射能測定を駆使して，1898年新元素PoとRaを発見した。そのあとヨアヒムスタールのウラン抽出廃棄物から４年にわたる努力を重ねて純粋な可秤量の塩化ラジウムを精製し原子量測定し，さらに金属ラジウムの調製した等は，いずれも歴史的な放射化学の第一歩であった。

一方ニュージランドからイギリス Cambridgeの J.J.Thomsonのもとに留学し空気の電離現象を研究していた E.Rutherford は，1898年27歳の若さでカナダ Montreal のMcGill大学教授に赴任した。そこでは新現象の放射能とそれからの放射線に注目しての研究に取組み，トリウムからの放射線が空気の流れの影響をうけて変動することから放射性気体のTh Emanation （トロン）を発見した。新婚後の数年間にわたるこの期間に，夜も研究室に赴いた当時の真摯な研究態度は，生まれて間もない一人娘をつれて夫人が里帰り中に，自宅より大学により近い寄宿先の大学の友人への次の言葉にも窺える。 I am seeking to know Nature, now I am a part of Nature, therefore, I should be able to infer her laws, and to make sure I don't make any mistake. I lay snares to her, so I go at any time,day or night, to see if I have caught her.'

なおこの放射性気体に関する報文は,1900 年初頭の Phy.Mag. 誌に掲載されたが，ドイツ Halleの大学物理学教室のE.Dornはこれに注目し，その研究方法で教室にある諸放射性物質を検討した。 それは丁度E.Rutherfordは婚約者と結婚のためニュージランドに一時帰国し研究中断の頃であった。そしてRutherfordはウランからは放射性気体の放出はなさそうだとしていたが，ラジウムを含むと考えられるバリウム塩からは別種の放射性気体のRa Emanation(ラドン）が放出されるのを発見した。

これらに続く放射性気体に関連して観測される励起２次放射能（放射性沈積物）の性質およびその放射能の減衰速度の研究は，歴史的に大きな役割を果たした。これらの面ではRutherfordの研究室に入ったMcGill大卒業のHarriet Brooks女史の協力研究も忘れてはならず，さらにイギリスOxfordから来て1900年から1903年まで Rutherford と協同研究した化学者 F.Soddyによる Emanationの化学的性質およびトリウムからのThXの化学的分離とその壊変生長の研究は，1900年の W.Crookesによるウランからの UXの化学的分離とその壊変生長とともに，放射能と化学元素つまり原子の変換を結びつけるようになる化学的手法による大きな寄与である。

19世紀の後期，ドイツ北東部 WolfenbütelのGymnasium の教師 J.Elster と H.Geitel は，空気の電離とその電気伝導度に興味をもち，簡単な箔検電気器を用いて種々な条件でのその測定変化を研究していた。両人は幼時から仲がよく，その研究からは光電管を開発しそれで太陽光の変動測定を行い，またはじめての熱電子管も作成している。そして一般環境についても，雷・降雨そして日食，さらにヨーロッパの緯度・高度の異なる各地に旅した際にも，気象条件も配慮し空気電気伝導度の変化を測定している。その中で地下室や洞窟では空気電気伝導度が高いことを見出し，1901年その原因探究のために，電圧をかけた針金にいわゆる励起放射能（放射性沈積物）を捕集し，その減衰特性から一般環境中にもラドンが存在することを発見した。そしてこのことをもととして，実験的なElsterと理論的なGeitel両人を初めとする種々の天然環境放射能の諸研究が進んだ。Rutherfordもその著作(1913)のなかで，両人をこの面での先駆者第一人者として，類のない貢献をしたと述べている。

本講演では時間的制約もあり，その後の原子核現象としての放射能研究，人工放射能，核分裂の発見，原子力利用とその環境影響の歴史にふれることはできないが，ほぼ上述の20世紀初頭までの放射能研究の源流を，私が学会・委員会等に渡航のみぎり心して訪れた次頁各所の現況を示しつつ，資料とともに，相共に歴史の旅をしていただくこととする。

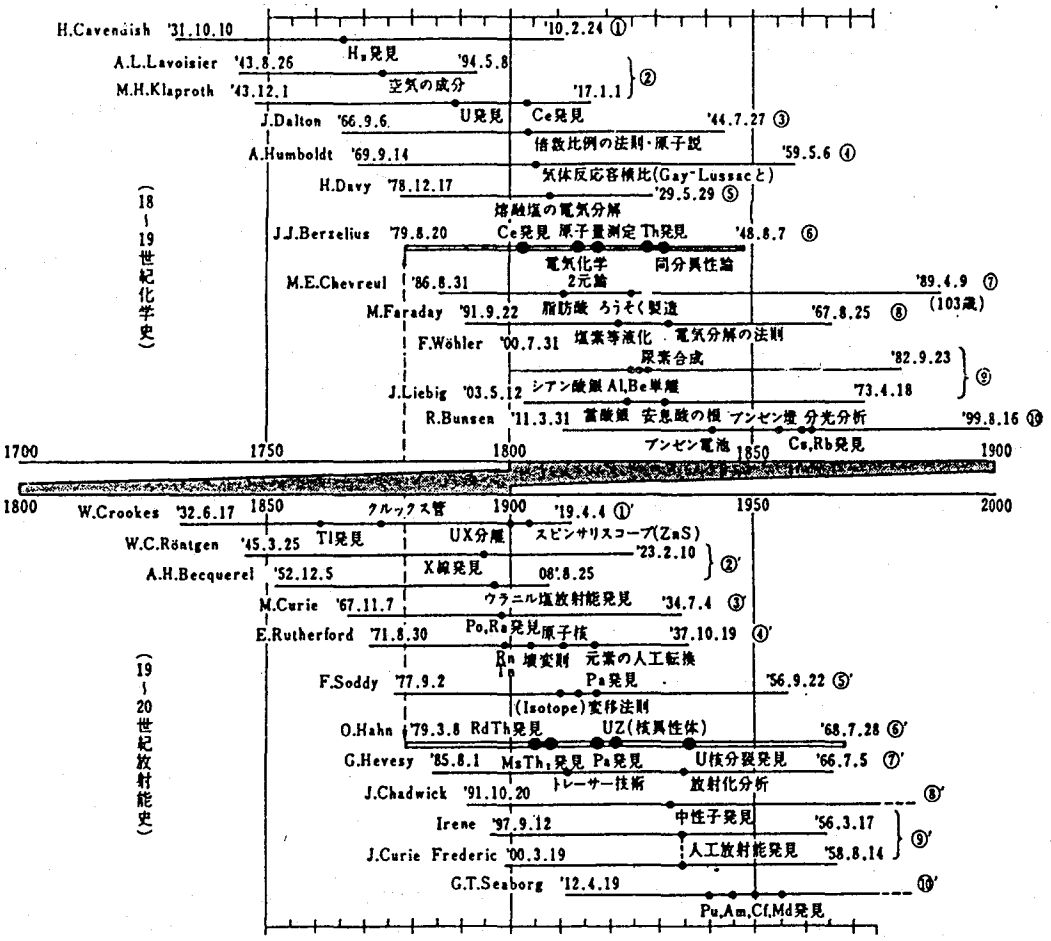
OHP・スライドで示す訪れた歴史関連の各所：　〔 〕国と地名，　( ) 訪問年

〔ドイツ Würzburg〕・Würzburg大学Röntgen Labor, W.K.Röntgenと関係資料　('73,'95)
〔 〃　Lennep〕　・ W.K.Röntgen生誕地，レントゲン博物館　('73)
〔フランス Paris〕
・Jardin des Plantes内のCuvier館，H.Becquerel 放射能の発見　('81,'95)
・Ecole Municipale de Physique et Chimie (現 Ecole Superieure de Physique et Chimie Industrelles de la Ville de Paris)
Curie 夫妻 Po, Ra 発見(G.Bemont 協力),Ra精製単離と関係資料 ('67,'81,'95)
・Institut du Radium, Pavillion Curie; rue Pierre et Marie Curie
1914年設立，実験室・展示，庭園（現在 Curie研一部として使用）('67,'95)
・the Panthéon (caisse nationale des monuments historique et des sites)
地下室霊廟に1995年５月　Curie 夫妻移葬。　('95)
〔カナダ Montrèal〕
・McGill大学，Macdonald Building，もと研究実験室，現在は図書館　('82,'95)
the Rutherford Collection(the E.Rutherford Phy.Building)　('95)
・rue Saint-Famille 1702 (もとNo.152) Rutherford一家旧宅　('95)
〔ドイツ Halle〕
・Halle 大学物理教室，1890年建設，Ernst Dorn教授と関係資料　('91)
・Deutsche Akademie der Naturforscher Leopoldina(Univ. Bibiothek)Rn　('91)
〔ドイツ Wolfenbütel〕
・Große Schule, Elster und Geitel Haus, Herzog August Bibiothek ('73,'81)
〔チェコ Jachymov (Joachimstal)〕
・旧鉱山，ウラン抽出工場跡地，キュリー記念碑公園，（旧東独鉱産地も）('93)
〔オーストリヤ Wien〕
・Institut für Radiumforsachung,1910年設立　('83,'95)
・Hygiene Institut und Bundesanstalt für Lebensmittel-u. ｳﾗﾝなど旧試料 ('95)
〔ポーランド Warszawa〕
・Freta 街 Muzeum Marii Sklodowskiej Curie，生誕地　('95)
・Krakowskie Przedmiescie 通り66，J.J.Boguskie旧実験室，
1890-1901年 M.Curieがここで実験をする機会を持った。　('95)

このほか Parisでの Curieの研究所と関連する人々として，ノルウェーOslo大学に戻り教授ともなった Ellen Gleditsch女史(1879-1968) の遺品，助手として後に新元素Frを発見した M.Perey女史(1909-1975) のStrasbourgの記念室，またロシヤの地球化学者で旧ソ連になって1922年Ra研究所設立も導いた V.I.Vernadsky(1863-1945) のMoscow記念室等の諸資料，さらにMontrèalにも赴いてRutherfordと研究しやがて核分裂も発見したO.Hahnの生涯（Isotope News誌1975年３月号－1976年11月号）も訪れて味わい深いものがあった。

なおわが国の放射能研究はほぼ1910年頃から始まるが，これについては明年３月より半年にわたり日本化学会化学会館で展示がある予定なので参考となるであろう。

歴史は螺旋状にある程度の類似で進行し，「前事不忘，後事之師」の言もある。ここに19世紀末から20世紀にかけの放射能研究に登場する諸人物の生涯の業績を，それからほぼ100年前に登場した諸人物の生涯の業績と比較してた図を作って下に示す。①と①'，②と②'，……と諸科学者の組み合わせを考えると，興味ある類似比較ができる。それでは次の世紀の放射能研究はどうなるであろうか。20世紀のおとし子の核兵器の廃絶，剣を鋤への道は必須な将来となったが，そもそも万物の存在し諸物質の中心である原子核の廃絶はあるべくもなく不可能である。それを対象としプローブとする核化学，放射化学はその基礎として不可欠である。小環境の核反応ターゲットから地球・宇宙におよぶ環境，水溶液化学から非水溶液の熔融塩・火山，化学種の問題，そして廃棄物処理処分の課題等々など，その寄与が次世紀に期待され，また地味な研究から意外な新発見もあるのでなかろうか。



THE ORIGIN OF RADIOACTIVITY STUDY AND ITS EARLY DEVELOPMENTS WITH ITS FUTURE

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With the proximity of the centennial of the discovery of radioactivity, by showing the status of various memorial places and their materials, a historical review of radioactivity studies is presented with some comments in future.

1S02

FUNDAMENTAL ASPECTS, INSTALLATION AND PRACTICAL APPLICATIONS OF k_0-STANDARDIZED NEUTRON ACTIVATION ANALYSIS

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The concept of k_0-standardized reactor neutron activation analysis (k_0-NAA) was launched in the mid 1970s[1] with the aim to overcome the drawbacks of the classically applied methods: the experimental workload of the relative standardization, the inaccuracy of the absolute standardization and the inflexibility of the single-comparator standardization. The final goal when developing k_0-NAA was to arrive at a standardization method which is generally applicable, experimentally simple, accurate, flexible with respect to both the irradiation and counting conditions and suitable for computerization.

That the introduction of k_0-NAA turned out to be a successful enterprise, was not only due to the fact that the phenomena of (n,γ) activation [signal excitation], radioactive decay and measurement of gamma-radiation [signal detection] are well-understood and can be accurately described, but also to the determination of the fundamental nuclear data being based on parallel but independent measurements performed by research teams at two (and in later years even more) institutes, involving different experimental conditions (and thus leading to the detection and elimination of systematic errors): the Institute for Nuclear Sciences/Gent (F. De Corte) and the Central Research Institute for Physics/Budapest (A. Simonits). In the first instance, this refers to the measurement, according to the "activation method", of k_0-factors, which are in fact compound nuclear data containing molar masses, isotopic abundances, 2200 ms^{-1} cross sections and gamma-intensities for both the analyte and the comparator [for which $^{197}Au(n,\gamma)^{198}Au$ with E_γ = 411.8 keV is chosen]. At present, k_0's with an accuracy of better than 2% are available for the relevant gamma-lines of 122 analytically interesting radionuclides formed by (n,γ) reaction [see Ref.[2] (and references therein), where these and the hereafter following topics are dealt with].

Next to the k_0-factors, a large effort was spent to determine the corresponding Q_0-values (= ratio of resonance integral to 2200 ms^{-1} cross section), in order to make a correction for the contribution of epithermal activation. At the same time, user-friendly procedures were developed for measuring f, the thermal-to-epithermal neutron flux ratio, in the irradiation facility at hand. Also, a great deal of the attention went to the elaboration of correction procedures for taking into account the non-ideality of the epithermal neutron flux distribution, approximated by a $1/E^{1+\alpha}$ shape: this not only involved the experimental determination of the factor α but also the calculation, from the resonance parameter data, of the so-called effective resonance energies (E_r). In this context, considerable efforts were spent to develop an elegant and sufficiently accurate procedure for in-situ and simultaneous monitoring of f and α, for which it was proposed to make use of the "bare triple-monitor"-method based on the reactions $^{197}Au(n,\gamma)^{198}Au$, $^{94}Zr(n,\gamma)^{95}Zr$ and $^{96}Zr(n,\gamma)^{97}Zr$ [involving simple coirradiation and measurement of Al-0.1%Au and Zr foils or wires].

A next topic considered in great detail was the calibration of the Ge detector. As to the determination of the full-energy peak detection efficiency (ε_p) for extended samples measured close to the detector, a situation usually encountered in the practice of NAA, a semi-empirical method was developed for the conversion of the experimentally measured "reference" ε_p-curve ($\log\varepsilon_p$ versus $\log E_\gamma$, measured for point sources at large distance from the detector), based on the concept of the so-called effective solid angle Ω which takes into account source-detector geometry, detector response and gamma-attenuation. Thus, to achieve this goal, the geometric configuration of

source and detector must be known and, with respect to the latter, this occasionally requires a fine-tuning which should be performed by experimentally controlling the accuracy of the ε_p-conversion. In addition to this, it was found necessary to develop correction procedures for true-coincidence effects (summing-in and/or summing-out) of cascading gamma-rays when measuring close to the detector. This not only required the analysis of the decay schemes, but also necessitates the experimental determination of the peak-to-total ratio P/T [in order to calculate the total detection efficiency $\varepsilon_t=\varepsilon_p/(P/T)$, needed in case of summing-out effects]. Especially for these topics related to detector calibration, it was a necessity to develop computer codes [SOLANG and COIN for VAX; SOLCOI for PC].

In addition to the above mentioned main domains of k_0-NAA [nuclear data; calibration of irradiation facility; calibration of detector], extensive work was done with respect to quality control and quality assurance. This was based on the evaluation of the final accuracy both via error propagation calculation and via the analysis of a variety of (certified) reference materials. It was concluded that, in average conditions of irradiation and counting, the inaccuracy introduced by k_0-standardization is not exceeding 4%. Next to this, attention was paid to the traceability of the method, an aspect which is of great importance in certification analysis.

Eventually, much effort was spent in the development of software to handle the numerous calculations in k_0-NAA. Whereas at first this was restricted to home-made computer codes for mainframe [SINGCOMP, SOLANG etc. for VAX], it finally resulted in the availability of KAYZERO/SOLCOI, a software package for PC commercialized by DSM Research (Geleen, The Netherlands).

Based on the above outlined methodology, the task of the analyst wanting to install k_0-NAA is restricted to the application of the following user-friendly procedures: i) the characterization of the irradiation facility [investigation of the constancy of the neutron flux during irradiation, a prerequisite in all absolute NAA-standardizations (including k_0); determination of the flux parameters f and α]; ii) the characterization of the Ge detector [determination of ε_p in reference conditions; investigation of the accuracy when converting $\varepsilon_{p,ref}$ to actual analytical geometries; determination of P/T]. This should then simply be combined with the available nuclear data library and the use of an adequate software package - not forgetting to perform a quality assurance via the analysis of some reference materials.

It goes without saying that the application of k_0-NAA is especially beneficial in case of multi-element determination or even panoramic analysis: coirradiation of one monitor with the sample, followed by some 3 or 4 measurements with varying decay times then leads to the possibility of obtaining results - concentrations or detection limits - for about 60 elements. But k_0-NAA turned out to be a competitive and manageable analytical tool in many fields (environmental, biomedical, industrial, etc.), including for instance the investigation of materials which are difficult to bring into solution such as a variety of polymers (e.g. for the determination of catalyst residuals). Eventually, it is worth mentioning that k_0-NAA is successfully used as a calibration and even a reference method for other analytical techniques such as XRF. This is possible because in k_0-NAA all sources of uncertainties are well-known and can be accurately quantified.

References:

1. A. Simonits, F. De Corte, J. Hoste, Single-comparator methods in reactor neutron activation analysis, J. Radioanal. Chem., 24 (1975) 31-46.
2. F. De Corte, A. Simonits, Vade mecum for k_0-users, Addendum to the KAYZERO/SOLCOI software package (Version 3.0), DSM Research Report R94/11492, December 1994, 49 pp.

2S01 Influences of the Fossilization Processes on the Fundamentals of Radiometric Dating

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For establishing the chronology of the hominid evolution during the Pleistocene, the uranium-series disequilibrium (U–Th: $^{230}Th/^{234}U$) and the electronic spin resonance (ESR) dating are usually used. The results of these radiometric dating applied to the archaeological fossil bone, dentine and enamel depend on the preservation of samples. In fact, it is important to determine the fossilization processes in order to attain a better interpretation of the dating data.

The U–Th method is based on the radioactive daughter products of thorium-230 (half-life: 75.2 ky). Modern bone contains less than 0.1 ppm of uranium and no thorium. During burial, the organic matrix decay in the bone causes a reduction of the soluble uranyl complex (VI) present in groundwater to the insoluble U (IV) form. Uranium appears to be adsorbed by the mineral phase of bone and the growth of thorium activity begins. In that case, the fractionation between U and Th is produced and at the present day, the measurement of the activity ratios $^{230}Th/^{234}U$ and $^{234}U/^{238}U$ for closed system yields age of the fossil. The dating range of this method is considered to be about 350 ky.

The ESR dating of fossil bone was introduced by Ikeya in 1975. The ESR technique is based on the measurement of the amount of the trapped electrons in crystal defects. These trapped electrons have been created by natural radiation resulting from the decay of uranium, thorium and potassium in the material and also from the environment. ESR dating consists of (i) measurement of accumulated dose or equivalent dose (DE) by the method of artificial additive doses using ESR and (ii) evaluation of annual dose-rate (Da) by analysis of radioactive elements in the sample and its surroundings. The age is given by the relationship: Age = (DE) / (Da). In making the age calculation, it is necessary to make some assumption about the history of uranium uptake which is of great importance. For that, two models have been suggested by Ikeya: the *early uptake* (EU) assumes that the present day uranium content was acquired in the sample very soon after burial and the *linear uptake* (LU) assumes a continuous U-accumulation.

These two dating methods are related to the stability of the mineral during burial and to the uranium uptake history. Therefore, in order to date with better certainty the archaeological levels of the Lazaret karstic cave (France), the history of bones and teeth from *Cervus elaphus* jaws that were discovered in the C continental infilling (6 m depth) is evaluated by chemical, microscopic and structural analyses. The clay gravel deposits divided into three levels (CI, CII and CIII) are located between a beach deposit B at the bottom and a stalagmitic flowstone E at the top.

The Lazaret excavated fossil bones and dentine contain high concentration of U, in the range of about 10 to 55 ppm. However, fossil enamels concentrations are low and average about 1 ppm. The uranium content of the surrounding Lazaret clay deposits range between 2 and 8 ppm. The microscopic analysis indicates that fossil bone and dentine tissues are very porous by the presence of the relatively well preserved haversian canals and Tomes's canals, respectively. Therefore bone and dentine have a very great reactive surface and exchanges with the hydrologic and sedimentary environments are

more significant than for enamel which is impermeable. Furthermore, the inorganic phase of bone, dentine and enamel compared to the modern samples is characterized by X-ray diffraction (XRD) and infrared spectroscopy (IR). It is mainly consisting of carbonate-substituted hydroxyapatite (OHAp: $Ca_{10}(PO_4)_6(OH)_2$). The calcite is also detected by XRD in most bones and dentine. Calcite precipitates in the porous spaces and cracks of the bones and dentine, particularly as a consequence of bacterial and fungal processes. Chemical analyses show that the uranium concentration appears to be associated to the apatite content rather than $CaCO_3$ content. The most significant change in IR spectra of bones, dentine and enamels concerns the large H_2O bands which mask the OH stretching band at 3567 cm^{-1}. The amplitude of the H_2O stretching and bending band at 3430–3300 cm^{-1} and 1650 cm^{-1}, respectively, is higher in fossil bones and dentine than in the fossil enamels. Besides, fossil samples have lost most of their organic matter and as a consequence contain less H_2O than modern samples. Then, uranium content seems also to be associated to the organic matter amount present during decomposition of samples.

The determined $^{230}Th/^{234}U$ and $^{234}U/^{238}U$ of studied samples are represented in the graphical solution of the U–Th age equation. The data are scattered and range between 60 000 and 200 000 y. The U–Th ages represented according to the stratigraphic position of the sample in the deposit, decrease with depth which is anomalous. According to the ESR and U–Th dating of the beach deposit B and the flowstone E, there is a early uranium uptake for same bones and dentine from lower levels of the continental infilling CII, and uranium leaching for some bones from the upper levels CIII. They lead to get respectively younger and older U–Th ages. The U–Th method applied to bone, dentine and enamel is consequently largely sensitive. Therefore, it is useful to apply another ESR method which has a different principle.

The ESR (EU) and (LU) ages are represented according to the stratigraphic position of the sample in the deposit. The bone and dentine ESR ages are in the range of 35 and 100 ky and are lower than enamel ages which are in the range of 60 and 200 ky. The bone and dentine U–Th ages are higher than the corresponding ESR ages. According to the high uranium contents of fossil bones and dentine, the age disparity is associated to their lower equivalent dose (DE) compared to the enamel values. The recrystallization and the carbonation of bones and dentine are clearly demonstrated by comparison of XRD and IR spectra. These structural and chemical changes would have affected the dating ESR signal (clock). Nevertheless, the chemical and crystallographic fossil enamel analyses, realized by Rictveld refinement of powder X-ray diffraction data and the decomposition of infrared absorption bands, mark the great stability of the mineralogical phase and provide evidence of the ESR reliability results. The Rictveld method yields cell parameters with great precision. A good correlation has been found between the α cell parameter and the CO_3^{2-} content. During fossilization, the CO_3^{2-} –for– PO_4^{3-} substitution at the apatitic B-site was slight for enamels considering their geological age (Middle Pleistocene). Then enamel is stable and better for dating than bone and dentine. On the other hand, fluorine content is used as permeability marker. The low fluorine content of samples from the lower levels are correlated to the younger U–Th age. This indicates a stagnation of groundwater which corroborates with a recent uranium uptake. The fluorination degree is related to permeability of the containing sediments rather than with the age. In lower levels the stagnation of water has consequently caused the U–Th and ESR dates more recent than in the upper levels.

By the combining ESR/U–Th method introduced by Grün *et al.* (1987), the uranium diffusion parameters were determined within enamel, tending to show that the true age is situated between the two uptake uranium modes (EU) and (LU) for the upper levels and close to the LU mode for the lower levels. The proposed chronology is as follows: the level CII was assigned to (170 $\pm$ 20) ky (isotopic stage 6) and the upper beds of the CIII level to (130 $\pm$ 15) ky (the end and the beginning of stage 6 and 5 respectively).

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