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FEATURE ARTICLE

History of reference materials for food and nutrition metrology: as represented in the series of BERM symposia

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Introduction

The establishment of a metrology-based measurement system requires the solid foundation of traceability of measurements to available, appropriate certified reference materials (CRMs). In the early 1970s, the first "biological" reference materials of Bowen's Kale and the Orchard Leaves and Bovine Liver Standard Reference Materials (SRMs) from the then National Bureau of Standards (NBS), Gaithersburg, MD, USA, were characterized solely for trace element content. Over the ensuing years, with the evolution and growth of international concepts of metrology-based measurements, the area of food and nutrition measurements has seen a steady evolution of available reference materials, and a much wider availability of analytes and food/nutrition-related matrix CRMs. From the perspective of being involved in this measurement area over these 30 plus years, it is useful to briefly look at the history of this growth of reference material activities in the area of food and nutrition measurements. This history is documented in the series of International Symposia on Biological and Environmental Reference Materials (BERM), which was initiated in 1983 with a specific focus on food-related reference materials.

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Metrology-based measurement system

The generation of high-quality analytical data requires a measurement system based on sound metrological concepts. The requirements for such a measurement system are:

1. Validated analytical methods that are appropriate in response to a defined purpose statement. This "fit for purpose statement" should include appropriate precision (repeatability) for expected use of the resultant data, adequate selectivity to ensure that only the desired analyte is contributing to the measurement signal, and the necessary sensitivity to quantitatively measure the analyte at expected levels in the samples of interest.
2. The accuracy of the measurement is assessed with appropriate external matrix standards (CRMs).
3. Assessment of the continued proficiency of the analysts practicing the methods, including establishment of appropriate quality control systems.

This paper focuses on the second part of this measurement system for food and nutrition metrology: in particular, the history of food matrix CRMs characterized for nutrient content, as represented within the proceedings of the series of BERM symposia, begun in 1983.

Food matrix CRMs for trace elements

Although the establishment of standardized weights and measures for commerce dates back many centuries, it was in the first part of the twentieth century that the NBS established a wide number of metallurgical and geological reference materials with known elemental content and established the worth of these materials in resolving major disagreements by analysts using similar or distinct techniques [1]. Throughout the 1950s, biological and nutritional roles of newly discovered essential "trace elements" were being elucidated and many analysts were publishing methods for their determination, which they claimed were applicable to biological materials, without testing them on real materials whose composition was known. In 1964, since the only available biological reference materials were a few commercial sera, and Kenworthy's [2] original orchard leaves reference material (which was characterized for 12 elements), Bowen at Reading University, UK, produced a large batch (91 kg) of dried finely ground powdered kale. Samples of this Bowen's Kale [3] material were widely distributed to analysts and a large amount of information on its elemental content was published. Bowen's Kale was followed by the Orchard Leaves (ca. 1970) and Bovine Liver (ca. 1971) SRMs produced by the NBS. In the mid-1970s, initiation of collaborative efforts on food-related SRMs (which continue to the present) between the NBS and the US Department of Agriculture (USDA), Beltsville, MD, USA, led to development of SRM 1569 Brewers Yeast. From these early "biological" CRMs, an expansion of reference materials related to food and nutrition measurements became available over the following decade. These new CRMs focused primarily on characterization of trace elements in a growing variety of biological and food matrices. In October of 1980, a workshop was held at the NBS to discuss needs for reference materials for organic nutrients in foods [4].

Initiation of international symposia on biological reference materials

With the realization of this growing interest in food-related reference materials, in autumn of 1982, a tour of European scientists and laboratories involved in aspects of food-related reference material development was undertaken by Wayne Wolf, USDA, Beltsville, MD, USA. Midway through the tour (during discussions with Herbert Muntau, Ispra, Italy) it was realized that a focused meeting bringing together those scientists interested in "biological reference materials" was appropriate and timely. Thus, the initial meeting was organized in October of 1983 in Philadelphia, PA, USA, and was attended by about two dozen professionals from the USA and Europe who presented work and held discussions on biological reference materials: availability, uses, and needs for validation of nutrient measurement (BRM-1) [5]. At the meeting, Bowen [6] presented his extensive experience of 19 years with the Bowen's Kale material, which was still available. Presentations highlighted the extensive activity in clinical and biological reference materials by the NBS [7], and the more than 15 years of experience of the IAEA [8] in providing analytical quality control services, including production and characterization of eight biological reference materials characterized for minor and trace elements. The European Community, through its Community Bureau of Reference (BCR) in Brussels, had developed an extensive metrology program with a variety of reference materials. The BCR was beginning a series of projects of food-related reference materials [9], including development of some 20 homogeneous materials as candidate food reference materials, conducted by Herbert Muntau at the Joint Research Centre (JRC), Ispra, Italy [10]. An evaluation of the then-available certified biological reference materials for inorganic nutrient analysis [11] suggested that many available reference materials, such as Orchard Leaves and Bovine Liver, were really not good matrix matches for measurements of nutrient trace elements of interest in food materials. The preparation of a mixed-diet reference material was presented to address this issue of a more representative food material reference material [12]. Also presented at this meeting was the introduction by Agriculture Canada of a program to prepare a new set of 12 candidate agricultural reference materials being characterized for trace element content [13]. Many of these reference materials produced by Agriculture Canada were subsequently made available to the international community as reference materials marketed by the NBS (now the National Institute of Standards and Technology, NIST).

Although the majority of presentations at BRM-1 focused on aspects of availability, production, and characterization of biological reference materials characterized for trace elements, there was the beginning of interest in standards for organic nutrients [14], and the previously mentioned NBS organic nutrients workshop [4] was discussed and the details were reprinted in full in the BRM-1 proceedings [5].

This first symposium in 1983 was deemed to be very useful and was expanded to include environmental measurements, and the subsequent series of International Symposia on Biological and Environmental Reference Materials (BERM) was founded. These BERM symposia were initially held every 2 years, alternately in Europe and the USA, reflecting the core of reference material production activity, and the locations of its two active co-chairs, Wayne Wolf in the USA and Markus Stoeppler in Germany. The main focus of the BERM symposia has been to periodically bring together the producers of these reference materials. Although the BERM symposia have from the beginning included involvement with the clinical chemistry measurement community, this community has its own welldeveloped and very active measurement activities and thus the BERM symposia have focused on areas of food, nutrition, and environmental measurements beyond those of interest to clinical chemists. The proceedings of papers presented at each of the subsequent BERM symposia have been published as special issues of *Fresenius' Zeitschrift fur Analytische Chemie*, which has now become *Analytical and Bioanalytical Chemistry*. A look at the highlights of food- and nutrition-related metrology reflected in the published proceedings for each succeeding BERM symposium is a snapshot of the progress of the developing metrology addressed at each point in time.

BRM-2, Neuherberg, Germany, April 1986

The format for the BERM symposia from the beginning included general overviews of the new materials and activity occurring in each of the major providers: the NBS, the IAEA, and the European Community [15]. Preparation of a new database reported by the IAEA [16] included information on 60 internationally available biological reference materials. Mertz [17] presented an overview which stressed the importance of the accuracy of analytical data of nutritional relevance and the importance of the use of relevant reference materials in nutrition research. Okamoto [18] reported on a tea leaves reference material from the National Institute of Environmental Studies in Japan.

A food section presented papers on laboratory-prepared reference materials in nutrient analysis [19], the need for food reference materials in Latin America [20] and Asia [21], BCR [22] and FAO [23] programs in Europe, and a mixed-diet reference material [24, 25]. Interlaboratory trials on the analysis of proximates in foods carried out in Europe [26] and Sweden [27] clearly showed that laboratories produced widely different values for macronutrients in common foods and that reference materials of certified nutrient concentration were needed for these major food components. These studies led to increased activity within the BCR to produce these food matrix reference materials, and this was reported at subsequent BERM meetings. Southgate [28] reviewed the special features of nutritional analysis and their control with reference materials and criteria for production of food reference materials, and he presented the idea of representing the distribution of suggested reference materials within the matrix of the major food components of fat, carbohydrate, and protein. This idea was later adapted as the food triangle concept by AOAC INTERNATIONAL and by the NIST; this will be discussed later.

The BERM symposia also usually included an analytical methodology session, which at this time focused exclusively on methods for elemental analysis.

BRM-3, Bayreuth, Germany, May 1988

At BRM-3 [29], general papers updated biological reference material activity at the IAEA [30], the National Institute of Environmental Studies (Japan) [31], the NBS [32], and the BCR [33], there were detailed reports on the preparation of 12 candidate agricultural reference materials from Agriculture Canada [34], and a database (COMAR) was described [35], all of which still focused almost exclusively on elemental content. There was the beginning of interest in identifying reference materials that could serve multipurpose needs, including both inorganic and organic components of interest to human nutrition and health in a total diet material [36], and a survey of selected available reference materials that could be used for organic food components [37]. Also reported were a single cell protein CRM [38] and edible oils and fats reference materials [39] from the BCR.

A BRM-3 session focusing on quality control and interlaboratory comparisons discussed needs for reference measurements [40], the role of statistics in quality assurance [41], and quality control samples in a collaborative study of trace elements in daily diets [42].

BERM-4, Orlando, FL, USA, February 1990

Although the BERM-4 [43] meeting formally incorporated the significant complementary activity in environmental reference materials in its title, highlights relating only to food and nutrition measurements are reflected below. This meeting reflected the increasing concerns about the consequences of both harmful and beneficial constituents in biological and environmental matrices which have led to the development of increasingly more complex analytical methodology and measurement systems applied to provide the thousands of "scientific measurements" per week used for decisions on the use of goods and food and on the environment. The basis of all of these decisions demands accuracy-based measurement systems, for which reference materials play a key role in transferring high analytical accuracy of primary metrology measurements to other laboratories [44]. REMCO—the ISO Council Committee on Reference Materials—plays an important role in providing an international forum and guidelines for producers of reference materials [45].

Revolutionary changes in measurement science had allowed the capacity to perform simultaneous multispecies measurements. An evaluation of the average concentrations for nine inorganic nutrient constituents in 24 food groups used principal component analysis, factor analysis, and cluster analysis to graphically depict the relationships between foods and candidate reference materials, in many cases indicating a poor match between the reference material and sample composition [46].

Further developments from the BCR in providing food matrix reference materials for nutritional analysis included the first candidate reference materials for total protein, total fat, available carbohydrates, and dietary fiber [47]. Most reference materials for macronutrients available at this time were freeze-dried specialized products that were not always compatible with ordinary food products. Thus, a homogeneous "fresh" canned meat material was produced and certified for macronutrient analysis [48]. An evaluation of the possible use of the US Food and Drug Administration's Total Diet as a possible CRM for proximate and organic nutrients such as vitamins [49] was proposed, and the preparation of SRM 1548 Total Diet was described [50].

A preliminary assessment of the homogeneity of ten new natural matrix agricultural reference materials was presented [51]. A total diet reference material from Finland was characterized for essential and toxic elements [52]. The new SRM 1845 Cholesterol in Whole Egg Powder was used in a nationwide study to update values for cholesterol content of eggs in USDA tables of food composition [53].

BERM-5, Aachen, Germany, May 1992

The opening session of BERM-5 [54] highlighted the increasing international awareness of the importance of measurement accuracy and the role of reference materials in achieving this accuracy. Papers were presented by representatives of the BCR program, Brussels, Belgium [55], the NIST [56], AOAC INTERNATIONAL [57], and the COMAR database of CRMs [58], followed by overviews of very active programs in China [59] and Poland [60]. This was followed by a session in which representatives of developing countries described their work [61–63]. This forum concluded that many of the reference materials available from the major producers were not representative of specific economic or environmental needs in their specific regions and were prohibitively expensive. These representatives expressed a strong interest in how they could produce their own reference materials.

The second day included a session on principles of reference material certification, including approaches to demonstrate homogeneity [64] and stability [65]. Potentially serious problems of comparability of reference values for similar matrix reference materials were highlighted by a paper comparing values for a series of reference materials certified for trace elements by different producers [66]. Certain elements are certified to considerably different levels of precision. For example, Pb is covered in 63 biological materials with uncertainties of 1–30% standard deviation, whereas Ni is covered in 40 materials with uncertainties of 10–60% confidence interval [66]. Reference materials for food microbiology were also covered at BERM-5 [67].

Poster sessions included nutrition-labeling-related papers on certification of major components and elements in food reference materials [68], intercomparison and preparation of reference materials for vitamin analysis within the BCR program [69], and related papers on stability studies of vitamins in food reference materials [70] and dietary fiber in vegetables [71].

BERM-6, Kona, HI, USA, April 1994

BERM-6 [72] initiated a formal association with AOAC INTERNATIONAL under cosponsorship as a topical conference with its newly formed Technical Division on Reference Materials. This is of key importance for food and nutrition measurements as much of the history of validated analytical methods for food analysis lies within AOAC International.

The opening session as usual highlighted recent achievements of CRM producers, including presentations from the European Community Measurement and Testing Program (formally the BCR), the National Research Council of Canada, the NIST, the Institute for Reference Materials and Measurements, the IAEA, REMCO, US Pharmacopeia, and AOAC INTERNATIONAL. All of these included some aspects of food-related CRMs.

A key paper focusing on global perspectives for secondary reference materials for food and related biological samples [73] recognized that although there were very many reference materials for inorganic components (60 reference materials for Zn content) in parts of the world, there was an existing paucity in other areas, particularly in developing countries, which are craving for these analytical quality control tools. Only a handful of available reference materials were characterized for components such as proximates and major minerals, let alone organic food constituents, which are basic requirements for food analysis laboratories. There was also a widely recognized global need for more secondary reference materials and the ability to produce these regionally [73].

Other presentations focused on food-related topics such as reference materials for food analysis laboratories in India [74], strawberry and cabbage candidate reference materials from Finland [75], a spinach reference material [76], proposed SRM 1846 Infant Formula [77], and bovine kidney and bovine muscle reference materials [78]

A new food matrix organizational system was proposed by a Food Definition subcommittee of AOAC INTERNATIONAL to define reference materials applicable to all food matrices [79]. This system describes a food matrix by its location in one of nine sectors in a triangle, with each point defined as representing 100% and the opposite side representing 0% of the normalized contents of each of the three major food components of fat, carbohydrate, and protein [80]. This concept had been adapted from the food triangle idea presented by Southgate [28] at BRM-2. This same scheme can be used to select a few food matrices representing each sector for development of a series of reference materials representing all foods.

BERM-7, Antwerp, Belgium, April 1997

At BERM-7 [81], the NIST reported on recent SRMs for organic and inorganic nutrients in food matrices [82], representing their distribution across the food triangle [79]. Since the NIST did not have the resources to measure every nutrient, collaboration was begun with the food industry and selected outside laboratories to value-assign such reference materials. Certification of SRM 1846 Infant Formula was a collaborative process [83] which included values for a wide range of vitamins. SRM 2383 Baby Food Composite also included values for vitamins, provided in collaboration with a large number of outside laboratories. SRM 1544 Fatty Acids/Cholesterol in Frozen Food Composite was the initial SRM with a value-assigned fatty acid profile. To further fill out the food matrix triangle, a processed meat SRM (SRM 1546) was under development and work to assign proximate values to available Agriculture Canada reference materials was under way [82]. The application of this paradigm to use data from a number of laboratories in value assignment, while being able to broaden the scope of assigned values, has introduced additional between-laboratory variability and thus led to wider expanded uncertainty limits for these components.

A report on sampling constants for niacin content in a dry material, SRM 1846 Infant Formula, suggested that use of subsample sizes of less than about 2–3 g can introduce significant variation into determinations for organic nutrients [84].

Other issues encompassed general areas covering increased global activities and needs in developing countries [85], including food- and nutrient-related topics. There were thought-provoking presentations on the proper use of CRMs, including the increased need for working matrix CRMs [86] and a lack of information on how to use CRMs [87]. The most outstanding trend in analytical chemistry over the past 60 years to provide instruments to cover nearly every analytical need has been a major driving force providing needs for CRMs, as instrument operating technicians replaced chemists. The instruments need calibration and there needs to be validation of the results and the achievement of traceability using reference materials [88].

BERM-8, Bethesda, MD, USA, September 2000

In the 4 years since 1996, the NIST had released or had in preparation nine new food matrix SRMs characterized for nutrient content [89], with the driving force behind these being the Infant Formula Act of 1980 [90] and the Nutrition Labeling and Education Act of 1990 [91]. In addition, the NIST had added additional proximate and fatty acid information to a suite of already available reference materials. The NIST reevaluated its process of assigning values to reference materials and identified three categories of assigned values: certified, reference, and information values [89].

BERM-8 [92] included an overview report of 25 years of reference material activity at Agriculture and Agri-Food Canada [93]. This activity included the investigation of some 58 agricultural/food starting materials, of which 12 became reference materials distributed by the NIST. There was also a report of activity in reference material development since 1972 at the Joint Research Centre, Ispra, Italy [94]. Although most of this Ispra activity focused over the years on environmental measurements, some of the early work from 1974 included foodstuff materials certified for trace elements.

The focus on measurements for trace elements in food/biological matrix reference materials has expanded to include the challenges from speciation analysis [95]. This irreversible trend in analytical chemistry has led to the appearance of CRMs with value assignment for a variety of elemental species components [96]. These include arsenic species; methylmercury, and alkyl lead compounds, for example. Organic components such as selenomethionine in candidate reference materials were reported [97].

A key issue for nutrients in food CRMs is the long-term stability of these components. Stability of up to 60 months was reported for a range of nutrient concentrations in a frozen mixed-food control material [98].

BERM-9, Berlin, Germany, June 2003

BERM-9 [99] demonstrated that a large need for new reference materials exists, especially for food analysis. The recent availability of the first CRMs in areas such as detection of genetically modified organisms and microbiological reference materials for quality control of water and foods had generated a large amount of research and major analytical developments [100]. Sharpless et. al. [101] reported on the availability over the last decade of a suite of ten food matrix SRMs, developed at the request of the food industry to populate the nine-sectored food triangle developed by AOAC INTERNATIONAL, as described earlier [79, 80]. The trend towards characterizing elemental species was represented by a report on selenomethionine contents of NIST wheat reference materials [102]. The current focus on foods had shifted from nutrient issues to predominately concern with food safety and components, either accidentally or deliberately incurred, of potential negative health impact.

BERM-10, Charleston, SC, USA, June 2006

By the time of BERM-10 [103], the focus of new activity in food-related CRMs was expanding beyond just extending the range of availability of different types food matrices to include dietary supplements and the use of CRMs to improve the quality required of analytical data. Chen et al. [104, 105] described the development of a liquid chromatography–mass spectrometry method for determination of multiple water-soluble vitamins in dietary supplements. A presentation on improving data quality in food composition databases in the EuroFIP program presented extensive documentation of the use of available CRMs for nutrient food matrices in the program [106]. A survey of reference materials used in the USDA National Food and Nutrient Analysis Program for determination of the nutrient composition of foods compiled 690 assigned values for individual nutrients from certificates of analysis for 63 available food CRMs [107]. The specified uncertainty interval in each case was expressed as a percentage of the assigned value, giving an uncertainty interval percentage for each value, as shown in Fig. 1. Across all nutrients, 63.5% of the uncertainty intervals were less than 10% of the assigned value, 25.5% were between 10 and 20% of the assigned value, and 11% were greater than 20% of the assigned value. These high uncertainty intervals (more than 10%) in over one third of the best available CRMs are likely to be indicative of the precision and accuracy that can be obtained by current measurement systems for these components. These data suggest that care must be taken in choosing CRMs to monitor food composition analysis, including evaluating what levels of uncertainty are required in assigned values and which analytical measurements systems for food components need closer examination and improvement.

Several papers presented at BERM-10 were published in a special issue of *Analytical and Bioanalytical Chemistry* [108] that was devoted to current analytical issues in food safety and nutrition, including discussion of production of

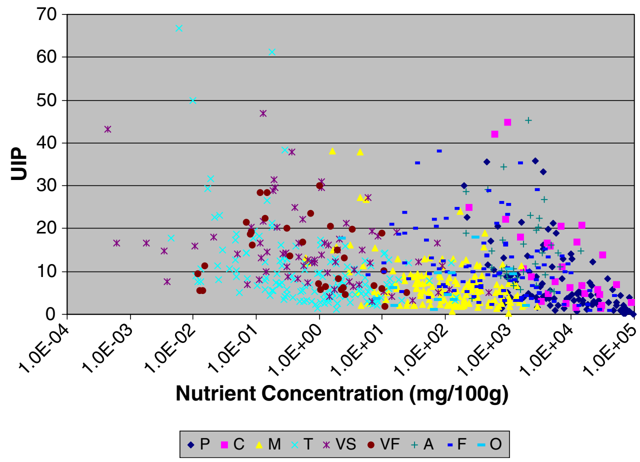

Fig. 1 Uncertainty interval percentages (*UIP*) [100×(uncertainty interval/value)] for certified and reference concentrations in certified reference materials as a function of nutrient concentration. *P* proximates, *C* carbohydrates, *M* minerals, *T* trace elements, *VS* water-soluble vitamins, *VF* fat-soluble vitamins, *A* amino acids, *F* fatty acids, *O* other (see Table 1 in [107] for the nutrients included in each group)

new CRMs for dietary supplements, which complemented BERM-12 presentations. This issue contained a paper presented at BERM-10 describing the use of CRMs to evaluate measurement systems for the nutrient composition of foods [109]. Over a 6.5-year period, a number of CRMs were submitted as blind samples to contract analytical laboratories as controls in the routine sample stream to generate food composition data for the USDA Nutrient Database for Standard Reference [110]. Overall, a total of 2,554 values were reported by nine laboratories for 259 certified or reference concentrations in 26 CRMs [109]. This unique data set was analyzed to gain some insight into the actual analytical practice of this representative set of contract analytical laboratories. Each value (X) from the laboratories was converted into a Z' score, reflecting the difference from the assigned value (X_a) related to the assigned uncertainty interval CRM value:

$$Z' = (X - X_a)/\mathrm{UI},$$

where UI is the uncertainty interval.

In this case a Z' score of $|1|$ or less indicated a value within the assigned uncertainty for the CRM. Z' scores greater than $|3|$ were over 3 times the uncertainty range. For some nutrients (Na, folate, dietary fiber, pantothenic acid, thiamin, tocopherols, carotenoids, and unsaturated fatty acids) more than 20% of the Z' scores were greater than $|3|$. The overall data set indicated that each of the laboratories performed some nutrient analyses especially proficiently, but none of the laboratories were proficient in all analyses. Care must be taken by clients of nutrient analysis laboratories to validate data sets using external measures such as including blinded CRMs or other matrix-matched control materials. These results were a first evaluation to open a dialogue on this topic and suggest that a further look at measurement systems for some of these nutrients is warranted.

BERM-11, Tskuba, Japan, October 2007

Food-related presentations at BERM-11 [111] focused on dietary supplements and speciation. A suite of three dietary supplement SRMs containing bitter orange was certified for levels of five alkaloids and caffeine by the NIST and several collaborating laboratories [112]. Methylmercury was certified in a cod fish tissue CRM using species-specific isotope dilution mass-spectrometric analysis [113]. A group from the National Metrology Institute of Japan used a highly precise isotope dilution inductively coupled plasma sector field mass spectrometry method for analysis of low concentrations of Pb in rice flour reference materials [114]

Future for food CRMs

Future trends for food and nutrition reference material activity will be a widened scope of food-related matrices with a broader scope of food components (such as a wider range of organic constituents and speciated components), new food types, and a fast-growing area of dietary supplement reference materials. New areas such as the widening spread of active proficiency testing programs, adoption of specific reference materials by instrument manufacturers, use of chemometric approaches, and even the possibility of replacing clinical testing with total DNA analysis will impact on future reference material activity for biological, food, and nutrition measurements [115]. The quality of available CRMs (with lower uncertainty values) will be further addressed. Most importantly there is a need and opportunity to improve the status of teaching in the academic community to bring emerging food and nutritional professionals up to speed about the importance of quality assurance in general and the role of reference materials in particular in providing the quality measurements required to address growing local, regional, and global policy issues.

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