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THE POTENTIAL OF BIOGENIC FRACTION ANALYSIS BY RADIOCARBON IN FOOD, DRUG, AND COSMETIC PRODUCTS

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ABSTRACT. Biobased content analysis is a well-established, analytically independent, standardized method to determine the biobased content of fuels and plastics, based on differences of the specific radiocarbon (^{14}C) activity of fossil and recent biogenic compounds. This biogenic content analysis can be useful for the producers as a quality assurance tool, for the customers as feedback about the truly biobased products and for the control organizations as an independent analytical tool to prove the biological origin. More than 100 commercially available foods, cosmetics, and drug samples have been used for biobased carbon content analysis by accelerator mass spectrometry (AMS) ^{14}C measurement to demonstrate the potential of this technique. Our results show that this measurement technique is a unique tool for the determination of biocontent in foodstuff and medical products. Most of the tested materials were nearly or completely biobased (≥ 98 pMC), and no completely fossil-based final product was detected. The lowest biogenic compound was measured in a vanilla aroma flavor. In 45 of the 102 samples selected a wide range (2–98%) presented fossil-based carbon content. The method can be applied for monitoring raw materials and final products for biobased content in the industry and consumer protection as well.

KEYWORDS: biobased content, biogenic fraction, drug, food, radiocarbon.

INTRODUCTION

Nowadays, the demand for biobased materials is increasing rapidly. In many fields, the traditional, fossil carbon containing, petrochemical-based materials, such as fuels and plastics can be replaced by biogenic compounds, not only with the same materials and molecules but with alternatives as well (Bastioli 2001; Gill et al. 2022; Palstra and Meijer 2014; Jou et al. 2015; Santos et al. 2019). The demand for these biobased materials has increased not only at the industrial side but at the side of the customers and consumers as well (Hermann et al. 2011; European Commission 2019; Pires et al. 2015; Santos et al. 2019; Pandey and Singhal 2021; Popp et al. 2021). For instance, it is observable not only in the plastic market but in the cosmetics and food market as well, as there is an increasing demand for vegan and biobased products (Wirsenius et al. 2010; Bozza et al. 2022). These efforts may later affect other markets, for example, the drug industry. The depletion of non-sustainable and fossil sources can also increase the demand for bioproducts in the fuel industry and in several other fields.

Although, the "bio" name is not well defined in different markets, as sometimes bio means that the material is biodegradable, but it can also be made from fossil material, sometimes means the material is biobased (natural), or both, but for regulation, control and quality assurance of these materials, independent and reliable analytical tools are needed to prove the completely biological origin. As fossil and biobased materials can have the same molecular composition,

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classical chromatographic and spectroscopic chemical analytical tools are not able to distinguish them. Classical chromatography can only identify different signatures, not being able to quantify the biogenic fractions (Silva et al. 2021). Isotope analytical tools, such as accelerator mass spectrometry or isotope ratio mass spectrometry can resolve this issue, by measuring the carbon isotopic composition. Radiocarbon-free, fossil materials can be distinguished from recent, modern biological materials, that have a well-defined $^{14}C/^{12}C$ isotope ratio, close to an equilibrium state with the atmospheric isotopic composition of CO_2 due to the photosynthesis (Monteiro et al. 2008; Oinonen et al. 2010; Buczyńska et al. 2013; Krištof and Logar 2013; Bronić et al. 2017; Varga et al. 2018; Santos et al. 2019; Delli Santi et al. 2021; Pironti et al. 2020).

^{14}C is a cosmogenic radionuclide, which is produced by secondary cosmic-ray neutrons interacting with nitrogen in the atmosphere. The ^{14}C produced rapidly oxidized in the atmosphere to CO_2, where it becomes well-mixed there and enters to the carbon cycle (Lal and Suess 1958; Damon 1968; Karlen et al. 1968; Kutschera 2019, 2013).

The applied method in our study is well-defined and standardized for plastic and fuel samples (ISO 16620-2:2019, 2019; ASTM D6866, 2020), but the literature and investigations are lacking in the field of food, cosmetics and pharmaceuticals industry. Sakamoto et al. (2002) published radiocarbon data about food products but focused only on the fossil content of flavors using liquid scintillation counting method.

The principle behind the standard technique is that the fossil materials (fossil oil and gas) do not contain radiocarbon, since they completely decayed in these materials, due to the long geological storage and 5700±30 years half-life. In contrast, the recent, modern biological materials have a well-defined radiocarbon content and $^{14}C/^{12}C$ ratio, which is close to the equilibrium with the atmospheric CO_2 (Suess 1955; Hua et al. 2021). In this way, the recent biological materials are naturally radioactively labelled by the atmospheric $^{14}CO_2$. Before the industrial era, the $^{14}C/^{12}C$ ratio of atmospheric CO_2 was close to an equilibrium, which was mainly influenced by small changes in cosmic radiation. During the industrial era, the $^{14}C/^{12}C$ ratio was affected by fossil emissions, which diluted the natural signal in an anthropogenic manner.

Later nuclear emissions increased the ratio, as atmospheric nuclear bomb tests until 1963 nearly doubled the amount of radiocarbon in the atmosphere worldwide. After the Partial Test Ban Treaty (1963), the atmospheric ^{14}C amount has constantly decreased over time due to atmosphere-ocean-biosphere CO_2 exchange, which is measurable in the atmosphere and living materials, for example in yearly tree rings, is called the "atmospheric radiocarbon bomb-peak" (Hua et al. 2013, 2021; Graven et al. 2017; Turnbull et al. 2017). This anthropogenic input of ^{14}C can be used as a calibration curve for radiocarbon dating of recent materials (Hua et al. 2021). As the recent standard for biofuels shows, the recent measured atmospheric ^{14}C content is around 100 pMC, approaching pre-bomb atmospheric ^{14}C concentrations (ASTM D6866, 2020). This 100 pMC is valid for the terrestrial biosphere, but this value may vary in the aquatic biosphere. Due to the mixture with pre-aged carbon pools, the carbon sources for fishes and aquatic animals in the sea and ocean typically have a lower $^{14}C/^{12}C$ ratio, which can cause them to appear older than their true age, presenting different specific radiocarbon activity compared to the atmospheric value (marine reservoir effect or MRE) (Philippsen 2013; Fernandes et al. 2016; Larsen et al. 2018; Alves et al. 2022; Svyatko et al. 2022).

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Although radioactive labelling by ^{14}C and usage of this isotope in the food, drug and medical-related sciences has also been known for a long time, the estimate of the biobased content of these materials and final products is not widespread, mainly due to different scientific approaches (Libby et al. 1964; Bergmann et al. 2012; Rinyu et al. 2019). For this reason, more than 100 commercially available foods, cosmetics and drug samples have been investigated for biobased content analysis by accelerator mass spectrometry radiocarbon measurement to demonstrate the potential of this approach. Biobased content analysis can be useful for producers as a quality assurance tool, for the costumers as feedback about the truly biobased products and for the control organizations as an independent analytical tool to prove the biological origin.

MATERIALS AND METHODS

Samples

Food Samples

We have selected 46 food samples for biobased content analysis. In some cases (4 of 46), different fractions were separated physically and then prepared and analyzed in parallel, to investigate the differences between the possible sub-fractions. In total, 51 different samples were measured. All the food samples were commercially available from the (Hungarian) market. In this demonstration study, we selected sweeteners, food colorings, chocolates, cake jelly, soft drinks and children's drinks, sports drinks, energy drinks, jam, margarine, hazelnut cream, gummy candy, and fiber syrup samples. In the case of soft drinks and chocolates, we have also investigated traditional and sugar-free products. The detailed list of the selected 46 food types, fractions and descriptions are shown in Supplementary Table S1.

Drug Samples

For this study, 29 individual drug samples were selected for biobased content analysis. For similar reasons as in the case of food samples, eight of the drug samples were separated for different fractions to investigate the differences in the bio-content ratio of the fractions. In total, with the sub-fractions, 37 different drug samples were measured. Several types of drug products were selected, such as fever and pain relievers, hormones, lozenges, vitamins, probiotics, etc. A detailed list of the selected 31 drugs and description is shown in Supplementary Table S1. Most of the samples were available commercially, but some of the medicines are only available with a prescription.

Cosmetics and Skin Care Product Samples

Fourteen cosmetic and skin care product samples have been selected for radiocarbon measurement in this study. We have selected ointment, soaps, cream deodorant, toothpastes, shampoos, hand sanitiser and teeth whitening powder for the investigation. All the samples were commercially available. Most of the samples have been selected as "bio" products, supposedly produced without treatment of industrial chemicals. The detailed sample list of the selected 14 products with a description is shown in Supplementary Table S1.

Sample Processing and AMS Measurement

The liquid phase samples, such as soft drinks and energy drinks were dried at 70°C in glass flasks on a laboratory hot plate. The samples were not dried until mass constancy, the drying step was applied only to reduce the sample size and make them sample denser. The solid samples have not been prepared before the next step.

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Then, about 3–4 mg sample and ~300 mg MnO_2 reagent were weighed into glass test tubes, depending on the expected carbon concentration, to gain ~1 mg C after the preparation. Then, the glass tubes were flame sealed under vacuum ($<2*10^{-5}$ mbar). The samples in the vacuum-sealed tubes were combusted at 550°C for 12 hr in a laboratory furnace. The oxygen for the combustion in the sealed tube was provided by the MnO_2 reagent. Then the glass tubes were cracked in a vacuum system to capture the water by dry ice-isopropyl alcohol mixture and the produced pure CO_2 by liquid nitrogen (LN) trap at –196°C after the combustion step. The applied combustion method and the description of the dedicated vacuum line are further detailed in Janovics et al. (2018).

After the gas purification, the captured pure CO_2 samples were graphitized with the sealed tube graphitization method (Rinyu et al. 2013). About 1 mg carbon as CO_2 gas was trapped by LN into a glass reduction tube which contained 10 mg TiH_2 and 60 mg Zn powder, and in a smaller glass tube 4.5 mg iron catalyst. Then the glass tubes were flame sealed under vacuum ($<2*10^{-5}$ mbar) and heated first to 500°C for 3 hr to release the hydrogen from the TiH_2, followed by a second step to 550°C for 5 hr to increase the efficiency of iron catalyst at the end of the graphitization process.

The carbon isotopic composition ($^{14}C/^{12}C$ isotopic ratio) of solid graphite samples produced from food, drug, skincare and cosmetic products were then measured by the EnvironMICADAS accelerator mass spectrometer (AMS) in the INTERACT laboratory of the Institute of Nuclear Research, Hungary, Debrecen (Molnár et al. 2012, 2013a, 2013b). The AMS measures the carbon isotopic composition the introduced graphite samples, where $^{13}C/^{12}C$ ratio, needed for the stable-isotope correction, is measured simultaneously with the $^{14}C/^{12}C$ ratio. For the data evaluation, the dedicated Bats^{TM} data reduction software was used (Wacker et al. 2010). The raw radiocarbon ($^{14}C/^{12}C$) results are expressed in pMC units (percent Modern Carbon). The pMC unit is generally used for environmental sample, 100 pMC is equal to a specific radiocarbon activity of 0.226 Bq/g carbon, which is equal with the hypothetical specific activity of atmospheric carbon of year 1950 (Stuiver and Polach 1977; Stenström et al. 2011).

Biogenic Fraction Calculation

The specific ^{14}C activity of fossil components is 0 pMC (zero), as fossil materials do not contain radiocarbon due to the relatively short half-life of the ^{14}C isotope and the long geological storage of these materials. In contrast, recent natural bio-, and plant-based materials have a well-measurable specific ^{14}C activity, is close to 100 pMC. However, their actual value depends on the growth year and decreased since the maximum in 1963–1964 (Hua et al. 2021).

The calculation method of biobased content is standardized for fuel and plastic samples (ISO 16620-2:2019, 2019; ASTM D6866, 2020), which defines the gross ^{14}C activity (A_T) and total weight (m_T) of the sample as depending on the weight (m_F) and ^{14}C activity (A_F) of the added fossil component and weight (m_B) and ^{14}C activity (A_B) of the added biocomponent:

$$A_T = A_F + A_B \; and \; m_T = m_F + m_B \tag{1}$$

As the specific ^{14}C activity of the fossil component by the standard method is zero (A_F=0 pMC), the gross ^{14}C activity of the sample depends on the biocomponent:

$$A_T = A_B \tag{2}$$

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The A_T can be expressed from the specific ^{14}C activity of the sample (in pMC units, normalized for 1950 and stable isotope corrected value):

$$A_T = pMC_T \times m_T \times c_T = pMC_B \times m_B \times c_B = A_B \tag{3}$$

Where the pMC_T is the specific ^{14}C activity of the sample in pMC unit, m_T is the total weight of the sample, c_T is the carbon concentration of the sample (m/m%), what can be expressed from the specific ^{14}C activity of the biocomponent (pMC_B), weight (m_B) and carbon concentration (c_B) as A_B.

The mass ratio of the biological component in the sample (m_B/m_T) can be calculated based on the following equation:

$$m_b/m_T = \left(pMC_T \times c_T\right) / \left(pMC_B \times c_B\right) \tag{4}$$

Where the c_T is the carbon concentration (m/m%) of the sample and the pMC_T is the specific ^{14}C activity of the sample. When the carbon concentration in the fossil and biogenic components are close to equal, then the ratio is (close to) 1. Based on this assumption, the equation can be simplified as:

$$m_b/m_T = pMC_T / pMC_B \tag{5}$$

The equation is further simplified in the case of unknown samples, when the source materials are unknown, only the final product can be measured, as in the case of commercial food and drug samples. In this case, one has to apply an assumption (atmospheric correction factor, REF) about the pure biobased component's ^{14}C specific activity.

$$\frac{m_b}{m_T} = (pMC_T/REF) * 100 \tag{6}$$

The REF in the ASTM D866 (2020) is 100 for 2020, so the biobased component ratio is practically equal to the measured pMC_T. On the other hand, REF depends on the year of the origin, as the specific ^{14}C activity of the contemporary pure biogenic materials in that year. REF is decreasing with time, due to the declining trend of the atmospheric bomb-peak. In former standards, the REF for unknown samples was higher, for instance the REF was 102 for 2015 (ASTM D6866, 2020). In case of the analyzed material is produced in 2015, but the year of the origin is unknown and the latest REF (100) would be applied, Eq. (6) results in 102% biocomponent ratio. Every result higher than 100 pMC is considered to be completely biogenic. The ASTM D6866 uncertainty is considered to be 3%.

RESULTS AND DISCUSSION

Food Samples

Forty-six food samples were measured for biobased radiocarbon analysis by accelerator mass spectrometry and 5 from the selected samples were separated for subsequent radiocarbon measurements, thereby, different fractions totalizing 51 food samples. Most of the samples, 35 samples from 51 (69%), presented values higher than 98% biobased content, so these samples can practically be considered as close or completely biobased materials (Figure 1). Eight samples presented pMC values between 50 and 98 pMC (16% of the selected food samples), which shows several food materials can have greater fossil contribution (Figure 1). Most of

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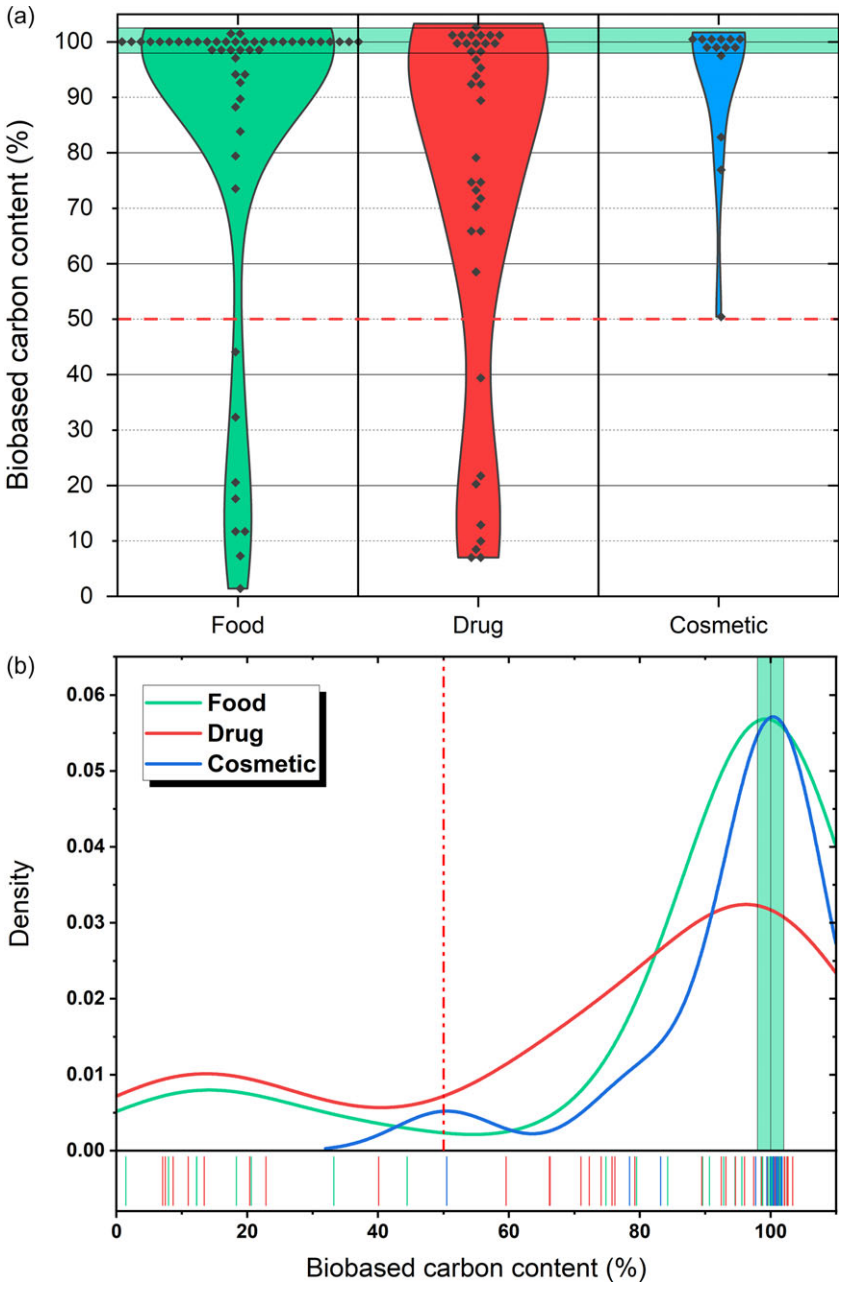

Figure 1 Results on violin plot (a) for the selected food, drug, and cosmetic product samples and (b) density distribution of these data. The green horizontal and vertical bar at (a) and (b) shows the interval of near or completely biobased (98–102%) value and the red dashed horizontal and vertical line at (a) and (b) shows the 50% biobased carbon content.

these samples were sugar-free soft drinks or energy drinks. For instance, the carbon content of a sugar-free cola product can be even less than 80% biobased (precisely 74.3±0.3% pMC), however, the original soft and energy drinks (not sugar-free) were considered as completely

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biobased (close to 100 pMC). Eight samples have been presented less than 50 pMC. The lowest biobased carbon content was obtained in a vanilla flavoring that presented 1.4±0.04 pMC (Figure 1). Considering the 3% uncertainty of the ASTM D6866 standard, this value is close to being pure fossil.

Our results show that the carbon content of some of the samples can be more than half fossil-based, this indicated a significant contribution of fossils to the final products. Food material analyzed has shown to be a mixture of biobased and petrochemical-based products in some cases and 100% biobased in other cases. No completely fossil-based (~0 pMC) was observed. The detailed list and results are shown in Table 1 and Figure 1.

Mean measured ^{14}C concentration of the selected samples 86±30 pMC (percent Modern Carbon, short%). The minimum value is 1.4±0.04, while the maximum is 102.5±0.3 pMC. While the standard shows that the 100% biobased content corresponds to 100 pMC, we measured ^{14}C concentrations higher than 100 pMC in several samples, where the excess ^{14}C was greater than the uncertainty of the measurement (~0.3 pMC in modern samples around 100 pMC). These materials are likely older food products (more than 1–2 years old), the biobased materials for these final products have been collected a couple of years ago when atmospheric ^{14}C concentrations were higher because of their temporal proximity to the peak ^{14}C production caused by atmospheric nuclear tests. Older food products can have higher specific radiocarbon activity due to the higher value of the radiocarbon bomb peak at those times. In this case Eq. (6) will give an apparent biobased carbon content significantly higher than 100%, which corresponds to pure (100%) biobased carbon, as the carbon was a product of biological activity before 2020 AD.

Our results show that commercial food products can have a great many fossil source contributions, especially household raw materials, such as some of liquid sweeteners, food coloring and aroma materials. Presumably, these materials with higher fossil carbon content cause the higher fossil contribution observed in sugar-free soft and energy drinks, which can be lower than 80% biobased. The biologically based sweeteners (stevia, erythritol, and xylitol) presented completely biobased materials by the results obtained. There was neither any significant difference between the different layers (surface and inner layer) of the milk and dark chocolates, nor in the different subsamples of gummy candy, all these samples were completely biobased.

Drug Samples

Twenty-nine drug samples were measured for biobased content by AMS. In 8 case of the drug samples, different layers or subfractions were prepared parallel, thereby, 37 different samples were measured. A detailed list of the samples and results is shown in Table 2 and Figure 1. The fossil- and bio-based contributions and their distribution are similar to what was observed in the case of food products. We observed several nearly or completely biobased drug materials (>98 pMC), 14 out of 37 samples (38%) (Figure 1). These samples were fish oil, hormone, vitamin, probiotic, organic selenium, activated carbon and lozenge samples. For fifteen of the thirty-seven samples,^{14}C results varied between 50 and 98 pMC (41% of the drug samples) (Figure 1). This is a higher ratio than what was observed in the selected food samples. It has to be stated, however, that our sample selection is not representative, these are rather randomly selected, and the sample number and the selection were not predetermined. Most of these drug samples presented more than 75% biobased content (pMC), and several samples show a higher

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Table 1 List and results of the analyzed food products for biobased content

No.	Lab code	Name	Description	Biobased carbon content (pMC%)*
1	DeA-32507	Sweetener-1	Stevia powder	100.27 ± 0.25
2	DeA-32509	Sweetener-2	Xylitol powder	98.75 ± 0.26
3	DeA-32511	Sweetener-3	Erythritol	99.61 ± 0.30
4	DeA-32512	Sweetener-4	Liquid sweetener	20.61 ± 0.13
5	DeA-32513	Sweetener-5	Stevia pill	94.54 ± 0.29
6	DeA-32514	Sweetener-6	Solid sweetener pill	12.28 ± 0.10
7	DeA-32517	Sweetener-7	Liquid sweetener	33.21 ± 0.17
8	DeA-32527	Food coloring-1	Red color	12.23 ± 0.09
9	DeA-32551	Food coloring-2	Green color	18.33 ± 0.14
10	DeA-32552	Food coloring-3	Yellow color	7.96 ± 0.07
11	DeA-32553	Food coloring-4	Blue color	44.42 ± 0.16
12	DeA-32554	Vanilla aroma	Food flavoring	1.43 ± 0.04
13	DeA-32515	Milk chocolate-1	Surface layer	100.78 ± 0.26
14	DeA-32516	Milk chocolate-1	Inner layer	101.00 ± 0.26
15	DeA-32519	Milk chocolate-2	Surface layer	100.85 ± 0.26
16	DeA-32520	Milk chocolate-2	Inner layer	101.02 ± 0.27
17	DeA-32521	Milk chocolate-3	Surface layer	100.06 ± .027
18	DeA-32522	Milk chocolate-3	Inner layer	101.52 ± 0.30
19	DeA-32517	Dark chocolate-1	Orange flavoring, surface layer	101.10 ± 0.27
20	DeA-32518	Dark chocolate-1	Orange flavoring, inner layer	100.93 ± 0.26
21	DeA-32526	Cake jelly-1	Colorless	100.44 ± 0.30
22	DeA-32527	Cake jelly-2	Red	100.50 ± 0.25
23	DeA-32540	Cola drink-1	Original, no sugar-free	101.41 ± 0.24
24	DeA-32542	Cola drink-2	Sugar-free	74.79 ± 0.28
25	DeA-34767	Cola drink-3	Sugar-free, lemon flavoring	84.25 ± 0.27
26	DeA-34768	Cola drink-4	Sugar-free	79.50 ± 0.27
27	DeA-32507	Soft drink-1	Mixed fruit flavoring	100.37 ± 0.29
28	DeA-32538	Soft drink-2	Carbonated, orange flavor	98.53 ± 0.27
29	DeA-34765	Soft drink-3	Lemon flavoring	100.41 ± 0.31
30	DeA-34766	Soft drink-4	Sugar-free, lemon flavoring	90.62 ± 0.29

Table 1 (*Continued*)

No.	Lab code	Name	Description	Biobased carbon content (pMC%)*
31	DeA-34774	Children's drink	Mixed fruit flavoring	100.42 ± 0.30
32	DeA-32509	Ice tea	Peach flavoring	99.86 ± 0.32
33	DeA-32511	Tonic	Original, no sugar-free	100.00 ± 0.28
34	DeA-32536	Sports drink	Non-carbonated isotonic drink	100.52 ± 0.24
35	DeA-32546	Energy drink-1	High caffeine content	100.20 ± 0.26
36	DeA-34769	Energy drink-2	Tutti frutti flavor	99.93 ± 0.31
37	DeA-34770	Energy drink-3	Tutti frutti flavor, sugar-free	92.82 ± 0.30
38	DeA-34771	Energy drink-4	Sugar-free	89.59 ± 0.29
39	DeA-34772	Energy drink-5	Mixed fruit flavoring	95.58 ± 0.30
40	DeA-34773	Energy drink-6	Tutti frutti flavor	101.25 ± 0.32
41	DeA-32548	Jam-1	Peach flavor, with sweetener	101.06 ± 0.30
42	DeA-32549	Jam-2	Blueberry flavor, with sweetener	102.49 ± 0.27
43	DeA-34775	Hazelnut cream-1	Hazelnut cocoa cream	101.01 ± 0.31
44	DeA-38570	Hazelnut cream-2	Sugar-free	100.42 ± 0.28
45	DeA-34776	Margarine-1	Traditional margarine	100.99 ± 0.21
46	DeA-34777	Margarine-2	Traditional margarine	101.20 ± 0.31
47	DeA-34778	Gummy candy-1	Subsample, white part	100.18 ± 0.32
48	DeA-34779	Gummy candy-1	Subsample, green part	100.49 ± 0.30
49	DeA-34780	Gummy candy-2	Fruit flavor	100.37 ± 0.31
50	DeA-34781	Multivitamin	Bio multivitamin candy	99.55 ± 0.31
51	DeA-38569	Fibre syrup	Honey flavor	100.83 ± 0.27

*Higher than 100 pMC values does not mean higher than 100% biocontent ratio, it only shows that the organic material used for the produced commercial material was produced in a year or years before 2020 with a higher atmospheric ^{14}C, but the calculation here does not take into account the variable higher atmospheric ^{14}C in case of unknown samples with unknown growth periods of bio-products. These materials also can be considered as 100% biobased.

Table 2 List and results of the analyzed drug for biobased content.

No.	Lab code	Name	Description	Biobased carbon content (pMC%)*
1	DeA-38558	Activated carbon-1	Pill, surface layer	102.51 ± 0.29
2	DeA-38559	Activated carbon-1	Pill, inner layer	100.96 ± 0.25
3	DeA-38116	Allergy medicine-1	Pill	92.43 ± 0.22
4	DeA-38556	Allergy medicine-2	Pill	95.99 ± 0.29
5	DeA-38557	Antibiotic-1	Film tablet	72.28 ± 0.23
6	DeA-38563	Antibiotic-2	Film tablet	66.28 ± 0.21
7	DeA-38130	Blood platelet aggregation inhibitor	Pill	20.36 ± 0.11
8	DeA-38564	Chewable tablet	Pill, iron	89.44 ± 0.27
9	DeA-38109	Cranberry concentrate-1	Pill, inner layer	102.14 ± 0.22
10	DeA-38110	Cranberry concentrate-1	Pill, surface layer	75.76 ± 0.19
11	DeA-38115	Effervescent tablet	For respiratory disease	79.24 ± 0.21
12	DeA-38106	Fever and pain reliever-1	Pill	13.41 ± 0.08
13	DeA-38111	Fever and pain reliever-2	Film tablet, surface layer	7.44 ± 0.06
14	DeA-38112	Fever and pain reliever-2	Film tablet, inner layer	22.86 ± 0.11
15	DeA-38114	Fever and pain reliever-3	Film tablet, paracetamol	10.97 ± 0.07
16	DeA-38125	Fever and pain reliever-4	Pill, paracetamol	7.04 ± 0.06
17	DeA-38119	Vasoprotective drug-1	Film tablet, surface layer	76.19 ± 0.22
18	DeA-38120	Vasoprotective drug-1	Film tablet, inner layer	100.47 ± 0.25
19	DeA-38571	Fish oil	Capsule, oil content	103.35 ± 0.27
20	DeA-38117	Hormone-1	Progesterone, capsule, inner layer	101.23 ± 0.22
21	DeA-38118	Hormone-1	Progesterone, capsule, surface layer	100.9 ± 0.24
22	DeA-38113	Lozenge-1	Throat lozenge	100.68 ± 0.23
23	DeA-38123	Lozenge-2	Throat lozenge	99.49 ± 0.22
24	DeA-38124	Lozenge-3	For stomach acid treatment	93.14 ± 0.24
25	DeA-38127	Lozenge-4	Throat lozenge, orange flavoring	97.39 ± 0.25
26	DeA-38555	Nausea prevention drug	Pill	40.08 ± 0.18
27	DeA-38567	Ointment	Haemorrhoid ointment with shark liver oil	8.65 ± 0.08
28	DeA-38562	Organic selenium	Pill	102.66 ± 0.29

Table 2 (*Continued*)

No.	Lab code	Name	Description	Biobased carbon content (pMC%)*
29	DeA-38128	Pregnancy vitamin-1	Gelatine capsule, mixed vitamins, minerals and trace elements, surface layer	71.00 ± 0.19
30	DeA-38129	Pregnancy vitamin-1	Gelatine capsule, mixed vitamins, minerals and trace elements, inner layer	66.18 ± 0.21
31	DeA-38560	Pregnancy vitamin-2	Film tablet, mixed choline, selenium, iron, folic acid and vitamin D, surface layer	59.55 ± 0.23
32	DeA-38561	Pregnancy vitamin-2	Film tablet, mixed choline, selenium, iron, folic acid and vitamin D, inner layer	94.61 ± 0.29
33	DeA-38121	Probiotic-1	Capsule, surface layer	74.06 ± 0.21
34	DeA-38122	Probiotic-1	Capsule, inner layer	100.85 ± 0.36
35	DeA-38107	Vitamin C-1	Capsule, surface layer	102.09 ± 0.21
36	DeA-38108	Vitamin C-1	Capsule, inner layer	98.59 ± 0.22
37	DeA-38126	Vitamin D	Pill	102.63 ± 0.24

*Higher than 100 pMC values does not mean higher than 100% biocontent ratio, it only shows that the organic material used for the produced commercial material was produced in a year or years before 2020 with a higher atmospheric ^{14}C, but the calculation here does not take into account the variable higher atmospheric ^{14}C in case of unknown samples with unknown growth periods of bio-products. These materials also can be considered as 100% biobased.

fossil contribution (<75 pMC). Probiotic, antibiotic, and pregnancy vitamin samples have lower than 75 pMC measured ^{14}C values, the lowest was a pregnancy vitamin (59.5±0.2 pMC) (Figure 1). Lower than 50 pMC values were observed as well (8 from the 37 samples, 22%). Generally, the fever and pain reliever samples presented more fossil-based content, as the most fossil-based fever and pain reliever sample had 7 pMC, while the samples with the highest ^{14}C still gave 23 pMC. Presumably, the active ingredient of these drugs is easier/cheaper to produce from fossil, petrochemical sources. A nausea prevention drug, a blood platelet aggregation inhibitor, and ointment, also show less than 55% biobased content (Table 2; Figure 1).

As in the case of food product samples, we could observe nearly or completely biobased samples (>98 pMC), and we could not observe any completely fossil-based material. The lowest biobased content in the drug samples ("fever and pain reliever-2", 7.04±0.06 pMC) was still higher than the lowest among the investigated food products ("vanilla aroma", 1.4±0.04% pMC) (Table 2).

We observed differences between the fractions and layers of some selected drug samples. While both fractions of Vitamin C are completely biobased (>98%), but there is more than a 3% difference between different fractions' ^{14}C content. It seems the surface layer of the investigated vitamin is produced from several years' older material, which is labelled more by the bomb ^{14}C (Table 2).

In the case of the investigated cranberry concentrate, fever and pain reliever-2, vasoprotective drug-1, probiotic-1, and pregnancy vitamin-2 drug samples, the surface layer was more fossil-based than the inner material of the samples. In these cases, the biobased content of the samples was lower than 80%, and it was even lower than 10% in the case of the surface layer of fever and pain reliever 2 (7.4±0.06%) (Table 2).

In the case of pregnancy vitamin-1, the inner layer is less biobased (66.2±0.21%) than the surface coating layer, but this layer is also lower than 80% (Table 2).

We did not observe bio content differences between the surface and inner layers of the hormone 1 and activated carbon subsamples (Table 2).

Due to the marine reservoir effect (MRE), it could be possible that there are lower radiocarbon levels in fish-based drug materials. Nonetheless, we could not observe the reservoir effect in the fish oil sample. The absence of reservoir effect does not mean it is not originating from fish, as assumed, however, those fishes could have been fed with fresh, recent fodder (terrestrial) material. We could detect quite low biobased content in the ointment-3 samples, which is a hemorrhoid ointment with shark liver oil. The biobased carbon content of this material is only 9%, probably not caused by a marine reservoir effect but cause a high amount of fossil material mixed with the bio-compounds.

Cosmetics and Skin Care Product Samples

Fourteen cosmetics and skin care product samples were measured for biobased content by accelerator mass spectrometry. A detailed list of the samples and results is shown in Table 3 and Figure 1. In this section, no separate sub-samples were prepared, and only the bulk sample materials were used and measured. Most of the samples appear nearly or completely biobased (>98 pMC), 10 of the 14 samples (71%), which ratio is higher than compared to the food and drug products (Figure 1). Only one sample, an ointment (Baby cream), was close to

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Table 3 List and results of the analyzed cosmetic and skin care products for biobased content

No.	Lab code	Name	Description	Biobased carbon content (pMC%)*
1	DeA-27174	Cream deodorant	With shea butter	97.68 ± 0.26
2	DeA-27188	Hand sanitizer	With alcohol	78.41 ± 0.26
3	DeA-38565	Ointment-1	Baby cream	101.65 ± 0.25
4	DeA-38566	Ointment-2	Stretch mark cream	99.97 ± 0.30
5	DeA-38568	Ointment-3	Baby cream	50.47 ± 0.19
6	DeA-27175	Ointment-4	Baby cream	101.61 ± 0.27
7	DeA-27187	Shampoo-1	70% natural origins and 99% biodegradable	83.18 ± 0.30
8	DeA-27201	Shampoo-2	Shampoo soap, 100% natural origins	100.98 ± 0.29
9	DeA-27041	Soap-1	Marigold, 100% natural origins	99.40 ± 0.24
10	DeA-27042	Soap-2	Facial cleansing soap	100.40 ± 0.25
11	DeA-27173	Soap-3	100% natural origins	101.21 ± 0.29
12	DeA-27177	Teeth whitening powder	With activated carbon	101.75 ± 0.30
13	DeA-27176	Toothpaste-1	With black charcoal	100.79 ± 0.27
14	DeA-27180	Toothpaste-2	Vegan	100.50 ± 0.32

*Higher than 100 pMC values does not mean higher than 100% biocontent ratio, it only shows that the organic material used for the produced commercial material was produced in a year or years before 2020 with a higher atmospheric ^{14}C, but the calculation here does not take into account the variable higher atmospheric ^{14}C in case of unknown samples with unknown growth periods of bio-products. These materials also can be considered as 100% biobased.

50% (50.5±0.2%), indicating a considerable fossil portion. The remaining three samples (cream deodorant, one shampoo and one hand sanitiser) have shown results between 50 and 98% biobased (Figure 1). As shown in Table 3, most of the ointments, soaps, shampoos, toothpastes are completely biobased, however, we emphasize that but our sample selection was not representative for the whole/global market, and the number of samples selected was not high enough to give a general conclusion about the market of biobased products.

The materials analyzed are complex mixtures from quite different ingredients which could have completely different sources, and the relative contributions of these materials are proportionately added to the product's overall biobased content. The applied sample preparation and methods of analysis were not compound-specific, so the measured biobased content is representative of the whole sample, not of its specific ingredients. Due to the relatively simple method used in this study, it was not possible to determine which compound gives the fossil contribution to the entire sample.

CONCLUSION

Nowadays where the demand for renewable, sustainable, and biobased materials increases and the green chemistry is gaining ground, a reliable analytical method is needed for monitoring and control the production and market of biobased materials. Recently, several companies worldwide have set a goal to decrease or eliminate fossil-based materials in their products. This paper shows that the ^{14}C-AMS biobased carbon content analysis is an appropriate and reliable technique for an accurate determination of the biogenic composition of various food, drug, cosmetic raw, and final products as well. Although some standardized methods define precisely the completely biobased materials' specific radiocarbon activity, fully biobased materials that are several years older have higher ^{14}C activity due the declining bomb-peak. In addition, completely biobased materials which have been produced at high fossil-loaded areas (for instance, close to busy roads) could contain a lower specific radiocarbon concentration compared to products originating from less anthropogenic influence areas. Despite all the disturbing effects, the method presented here can determine even a low amount (few%) of fossil contribution in many types of samples and is also able to detect if a sample is completely biobased or fossil-based and vice versa. The limitation of the present method is that it only provides information about the origin of the carbon content, not other components, but this simply can be taken into account in most cases. However, this method does not indicate whether the materials are biodegradable and harmful or not. Our approach is only applicable to biogenic carbon content analysis, showing the origin of the carbon content (recent biological or fossil). This presented method cannot distinguish which compound of the product contributes more fossil contribution to the sample but provides information for the whole bulk sample as we applied bulk analyses. If specific compounds are to be investigated, then the compound must be separated from the product and ^{14}C analysis has to be done on it, separately.

Our results show that several foods, drugs and cosmetic products may contain fossil-based materials in a significant amount, even over 50%. Almost completely fossil-based materials have been observed in some food and drug products. The lowest biobased content was observed in some sweeteners, food colorings, and fever and pain reliever samples, even as low as 10%.

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Our selected sample groups were not representative of the global or Hungarian market on a whole in this study, but they demonstrate the potential of ^{14}C-AMS biogenic carbon content analysis for edible, drinkable substances and skin care product samples and show an overview of biobased content of these commercial products. As demonstrated, the ^{14}C-AMS method can be valuable for the industrial sector, but also to the public sector and private customers. With the evolution of cheaper methods, such as laser-based radiocarbon analysis, the measurement may become even more widely affordable. This independent, well-defined and tested analytical method could be a unique tool not only for plastics and fuel industry (as recently already applied) but for the non-traditional biobased content analysis of food, drug and cosmetic products. These results can be used to support green procedures, greening processes of food and other products and green chemistry and provide important information to the stakeholders. The demand for green, biobased, and vegan products in the cosmetic industry is also increasing rapidly, so this method could be a unique tool for manufacturers to prove the 100% biological origin of their products. The approach can be further tested on different, non-traditional radiocarbon samples to gain complete information about the regional or global markets, but the results shown here demonstrate the potential of the AMS and radiocarbon-based biocomponent analysis well, and there are applications to almost all fields of industry using fossil- and biobased organic materials in their products.

SUPPLEMENTARY MATERIAL

To view supplementary material for this article, please visit https://doi.org/10.1017/RDC.2023.98

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