

HIGH-PRECISION BIOGENIC FRACTION ANALYSES OF LIQUID FUELS BY ^{14}C AMS AT HEKAL

Tamás Varga¹ • István Major¹ • Róbert Janovics¹ • Júlia Kurucz¹ • Mihály Veres² •
A J Timothy Jull^{1,3,4} • Mónika Péter⁵ • Mihály Molnár^{1*}

¹Isotope Climatology and Environmental Research Centre, Institute for Nuclear Research, Hungarian Academy of Sciences (Atomki), Debrecen, Hungary.

²Isotoptech Zrt Debrecen, Hungary.

³Department of Geosciences, University of Arizona, Tucson, AZ 85721, USA.

⁴AMS Laboratory, University of Arizona, Tucson, AZ 85721, USA.

⁵MOL Nyrt, Research and Development, Refining Product Development, Százhalombatta, Hungary.

ABSTRACT. The biocomponent ratio in liquid fuels as well as the usage of renewable resources for fuel consumption in the transport sector needs to be increased as a result of EU directive 2003/30/EC. Based on radiocarbon (^{14}C) measurements, it should be relatively simple and fast to measure the weight percentage of the fossil and biological sources by accelerator mass spectrometry (AMS) as recommended in the ASTM D 6866-12 and EN 16640 standards. In this study, a relatively easy and fast sample preparation and measurement method based on AMS measurements was developed at the Hertelendi Laboratory of Environmental Studies (HEKAL) using reference samples from the Hungarian MOL Nyrt. oil company. Considering the recent EU regulation for mixing rates of liquid fuels in the transport sector (0.7–2% biofuel content) and the projected higher rates (2–10% biofuel content), the method is applicable to determine fatty acid methyl ester (FAME) and/or hydrotreated vegetable oil (HVO) derived proportions of fuel blends with a 1σ uncertainty better than $\pm 0.3\%$ m/m.

KEYWORDS: AMS, biogenic fraction, liquid fuel, radiocarbon.

INTRODUCTION

The main aims of the EU 2003/30/EC directive are to increase sustainability, reduce fossil fuel dependence, and reduce the increase of atmospheric CO_2 levels (Chase et al. 2001; Oinonen et al. 2010). Renewable components or biofuels are produced by different biological processes, such as anaerobic digestion or fermentation from recent organic matter. In contrast, fossil fuels were produced by geological processes from prehistoric biological matter which does not contain radiocarbon (^{14}C). The chemical properties and composition of these biological components are generally quite similar to recently used fossil fuels. Due to the chemical and physical similarities of the biobased compounds to the fossil fuel, it is simple to intermix them without any major special modifications to the engine systems of vehicles (Deepanraj et al. 2011; Lawrence et al. 2011). On the other hand, due to these physical and chemical similarities, the post-production verification measurements of the bio/fossil mixing ratio is often quite difficult using common industrial analytical or chromatographic methods. Consequently, the independent determination of bio/fossil mixing ratio is a primary issue from both technical and the economic aspects. Measurement of fossil and biological fuel origin is highly effective using radiocarbon as a biogenic tracer (EU Directive 2003; Dijs et al. 2006; Norton and Devlin 2006; Norton et al. 2007; Yunoki and Saito 2009; Oinonen et al. 2010; Vrtiška and Šimáček 2016). The biogenic fraction determination based on ^{14}C content measurement is possible in different ways. The direct measurement of the ^{14}C activity of liquid fuels by liquid scintillation counter (LSC) is a fast method but burdened by some disadvantages. For example, the color of fuels causes different quenching properties in the scintillation cocktail and affects the measurement efficiency (Doll et al. 2017). Based on the measurement of radiocarbon content as $^{14}\text{C}/^{12}\text{C}$ and $^{14}\text{C}/^{13}\text{C}$ isotope ratio, the biogenic fraction is also measurable by accelerator mass spectrometry (AMS), where the number of ^{12}C , ^{13}C , and ^{14}C atoms can be counted directly (Bronić et al.

*Corresponding author. Email: molnar.mihaly@atomki.mta.hu.

2017). In this study, the biobased content of reference samples from the Hungarian oil company, MOL Nyrt., with predetermined mixing ratios were measured by the EnvironMICADAS AMS at the Hertelendi Laboratory of Environmental Studies (HEKAL) (Molnár et al. 2013). The main aim of the study was to develop a simple, fast, and reproducible sample preparation method for AMS ^{14}C measurement of liquid fuel samples.

METHODS

Properties of the Applied Bio- and Diesel Fuels

In this study, we used fuel blends that included fossil and biofuel mixtures. The different fuel types have quite similar physical and chemical properties, as shown in Table 1.

HVO (hydrotreated vegetable oils) are sulfur and aromatic compound-free, chemically hydrogenated vegetable oils, or paraffin hydrocarbon mixes. HVO is produced from raw vegetable oil or different triglycerides-rich material, such as used cooking oil and animal fat (Huber et al. 2007). These HVO compounds are known as renewable diesel fuels, while fatty acid methyl esters (FAME) are known as biodiesels, as these compounds are biodegradable and eco-friendly with low toxicity and vapor pressure. The leading process of producing FAME is transesterification of plant oils, fats and extraction from algae (Vyas et al. 2010; Nigam and Singh 2011). The production of HVO is less difficult from these waste materials than the production of FAME. As shown in Table 1, the properties of fossil and renewable liquid fuels from different sources are quite similar (Gonzalez et al. 2007; Aatola et al. 2009). Furthermore, differentiation by classical chemical methods is rather complicated, due to the colour and physical properties of the compounds, and differentiation by liquid scintillation counting is also difficult.

Mixed Fuel Test Samples from MOL

Fourteen different test samples with known composition and biocomponent mixing ratio were provided by MOL Nyrt. company for this study (Table 2). Note that FUEL_2 sample is EN 590 standard diesel fuel.

Table 1 Typical properties of different fuel components (Aatola et al. 2009).

Properties	HVO	FAME	EN 590:2004 diesel fuel
Density at 15°C (kg/m ³)	775...785	~885	~835
Viscosity at 40°C (mm ² /s)	~2.5...3.5	~4.5	~3.5
Cetane number	~80...99	~51	~53
Distillation range (°C)	~180...320	~350...370	~180...320
Cloud point (°C)	-5... -25	~-5	~-5
Heating value, lower (MJ/kg)	~44.0	~37.5	~42.7
Heating value, lower (MJ/l)	~34.4	~33.2	~35.7
Total aromatics (wt-%)	0	0	~30
Polyaromatics (wt-%)	0	0	~4
Carbon content (wt-%)	84.8	~76–81*	85.9
Oxygen content (wt-%)	0	~11	0
Sulfur content (mg/kg)	<10	<10	<10
Lubricity HFRR at 60 °C (µm)	<460	<460	<460
Storage stability	Good	Very challenging	Good

*Calculated carbon content. The carbon content was calculated from FAMES of different chain lengths by the molecular formula. The carbon content of the FAME slightly depends on the length of polycarbon chains and carbon number.

Table 2 Biocontent composition of test fuel samples.

Sample code	HVO (m/m%)	FAME (m/m%)	Biocomponent (m/m%)
FUEL_1	0.71	0.00	0.71
FUEL_2 (100% diesel)	0.00	0.00	0.00
FUEL_3	0.99	0.00	0.99
FUEL_4	0.47	0.00	0.47
FUEL_5	0.00	1.04	1.04
FUEL_6	0.94	1.07	2.01
FUEL_7	0.84	0.00	0.84
FUEL_8 (100% HVO)	100	0.00	100
FUEL_9 (100% FAME)	0.00	100	100
FUEL_10	2.81	0.00	2.81
FUEL_11	4.69	0.00	4.69
FUEL_12	9.40	0.00	9.40
FUEL_13	4.67	5.30	9.97
FUEL_14 (100% FAME)	0.00	100	100

The biocompound mass percentages were calculated using the weight of the added biocompound and the fossil fuel measured at the laboratory of MOL by gravimetric methods with a Sartorius 1702MP8 Electronic Analytical Balance, giving a precision of $\leq \pm 0.1$ mg. Certain samples contained only FAME or HVO as biocompounds, while other samples were mixed with both of them (FAME and HVO) (Table 2).

The biocontent of the test mixtures was defined by MOL in connection with the recent EU regulation for mixing rates of liquid fuels in the transport sector (0.7–2% biofuel content) and the projected higher rates (2–10% biofuel content). In this way the method developed can be tested to determine FAME and/or HVO derived proportions of fuel blends, which are very close to the recently used commercial fuels.

As the weight percent of reference sample mixtures were known, it was possible to test and evaluate the ultimate accuracy and performance of the complete AMS ^{14}C measurement based biocontent analyses at HEKAL. The samples were stored in sealed glass vials until the AMS sample preparation, at +5°C.

Preparation and Combustion for AMS Analyses

The samples were prepared without filtration or any other physical pretreatment. 4–5 mg of the samples was dropped onto MnO_2 powder in a borosilicate glass combustion tube (Duran, 150 mm long, 9 mm O.D.) then attached to a vacuum system, cooled in advance by dry ice and alcohol slush before being evacuated using a vacuum pump. After a short pumping time, the combustion glass tubes were sealed using a gas torch. Finally, all of the sealed tube samples were combusted at 550°C, for 24 hr following a simplified MnO_2 based combustion method, developed in HEKAL (Janovics 2015). A compact vacuum line was used to quantitatively extract and purify the CO_2 from the combusted samples. Combusted sealed tube samples were attached and cracked up in the vacuum line (ultimate vacuum is $< 1 \times 10^{-2}$ mbar, by dry scroll pump) by a special glass tube cracker supplied with a needle (Janovics 2015). The water vapor was trapped with isopropyl alcohol-dry ice mixture trap (at -78°C) and the CO_2 was frozen into a next trap by liquid nitrogen at -196°C . The pressure of collected CO_2 gas sample at room

temperature was measured by a high precision MKS Baratron pressure transducer in a known volume to determine the exact amount of the C yield/content. The purified CO₂ samples were graphitized by the sealed tube Zn-based graphitization method (Rinyu et al. 2013) and their ¹⁴C activity was measured by AMS at HEKAL (Molnár et al. 2013). For each test sample, 4–7 parallel repetitions were prepared and measured to examine the reproducibility of the method. In this study, IAEA C-9 standards were also prepared along with the samples for quality control and chemical blank investigation. After the AMS analyses, pMC (percent modern carbon, where 100 pMC_{abs} which refers to a specific ¹⁴C activity concentration of 226 Bq/kg C, normalized to AD 1950 and –25‰ δ¹³C_{VPDB}), was used for the calculation of the biocomponent fraction. MICADAS Bats data reduction software was used for the evaluation of the results (Wacker et al. 2010) including δ¹³C isotope fractionation correction.

Determination of the Biocontent Ratio

The radiocarbon-specific activity of fossil fuel carbon is zero, which refers to 0 pMC (hereafter pMC_F), as its ¹⁴C content has completely decayed away (half-life is 5700 ± 30 yr) during long geological storage. The percent modern carbon in the bio-components (hereafter pMC_B) is similar to the value of the atmospheric CO₂ (close to 100 pMC) (Berhanu et al. 2017) as carbon in plant organic compounds are result of photosynthesis of the atmospheric CO₂. Total ¹⁴C activity (A_T) and total mass (m_T) of the fuels are the sum of the ¹⁴C activity and mass of the fossil fuel component (A_F and m_F, respectively) and the ¹⁴C activity and mass of the biogenic fuel component (A_B and m_B, respectively):

$$A_T = A_F + A_B \quad \text{and} \quad m_T = m_F + m_B \quad (1)$$

As the fossil fuel total ¹⁴C is zero (A_F = 0), thus the total ¹⁴C activity of the fuel can be defined from its bio content:

$$A_T = A_B \quad (2)$$

The total ¹⁴C activity (normalized to stable isotope fractionation and to AD 1950) can be expressed as a product of the percent modern carbon (pMC), total carbon content (m), and carbon mass concentration (c), as:

$$A_T = \text{pMC}_T \times m_T \times c_T = \text{pMC}_B \times m_B \times c_B = A_B \quad (3)$$

Thus, the mass ratio of biogenic fraction in a mixed fuel (m_B/m_T) can be calculated by the following equation, based on the radiocarbon content and the mass balance:

$$m_B / m_T = (c_T \times \text{pMC}_T) / (c_B \times \text{pMC}_B) \quad (4)$$

where c_T and c_B is the carbon concentration of the total mixture and biogenic component, respectively, while pMC_T and pMC_B is the pMC values of total mixture and fossil component, respectively.

According to the assumptions of the ASTM D6866-12 standard, if the carbon percentage of fossil (c_F) and biobased (c_B) components are close to each other, their ratio is close to 1. With this assumption, Equation 4 can be further simplified to:

$$m_B / m_T = \text{pMC}_T / \text{pMC}_B \quad (5)$$

Furthermore, the ASTM D6866-12 standard makes an assumption to the specific ¹⁴C activity of the commonly used biocomponent to fix it as a constant value (about 105% as pMC_B for all kind of biocomponents, for the year 2010). The theory behind is that nearly all new biobased

products are produced in a post-bomb environment, all pMC values (after correction for isotopic fractionation) must be multiplied by 0.95 (as of 2010) to better reflect the true biobased content of the sample. Considering the effect of the 1960s bomb peak, the value is not 100 pMC (F) ($1F = 100 \text{ pMC}_{\text{abs}}$), but a little higher value (1.05 F, 105 pMC), which means practically a multiplication by 0.95 (as a reciprocal of $\sim 1.05 \text{ fM}$) in these equations ($1/\text{pMC}_B = 0.95$). Using this further assumption the standard applies the next simplified equation for the bio/fossil mass ratio calculation for mixed fuels:

$$m_B / m_T = \text{pMC}_T \times 0.95 \quad (6)$$

In practice, different biocompounds may have varying ^{14}C (pMC_B) values, depending on whether only FAME, only HVO, or FAME and HVO together were added to the blend. The actual pMC_B is also dependent on the year of biogenic production, because the specific radiocarbon activity of the atmospheric carbon level was constantly decreasing since the 1960s, after the bomb-peak (Hua et al. 2000; Quarta et al. 2005). Hence, more precise biocontent value may be obtained using Equation 4 or 5 if it is possible measure directly the pMC_B of the biocompounds used instead of relying on the fixed (assumed) constant pMC_B value assumed by the ASTM D6866-12 standard (Equation 6). In this study, we present results according to ASTM D 6866-12 also (see Equation 6) instead of the later versions of the standard (etc ASTM D 6866-16 and ASTM D 6866-18). At the time of the measurements and the evaluation of our results only the ASTM D 6866-12 standard was available. ASTM D 6866-12 standard was widely applied around the world by many other studies that is why we also aimed to highlight the possible related problems as well.

RESULTS AND DISCUSSION

Carbon Contents

The extracted CO_2 gas from the samples was measured in a calibrated volume to determine the carbon content (Measured c_T) of the blends as described in the Methods section. Using the biocontent composition and measured c_T data of the pure, non-mixed bio (FUEL_8, FUEL_9) and fossil fuel (FUEL_2) samples, an expected c_T was calculated for the mixed blends. Then, comparing the expected and measured c_T of mixed samples, the carbon (C) yield was determined, which was $>98\%$ in every case, as shown in Table 3.

As Table 3 shows, the pure FAME samples have the lowest c_T . The exact carbon content value is important, if we use Equation 4 for the determination of biocomponent ratio, because this data affects the final result.

^{14}C Analysis of Fuel Mixtures by AMS

For estimation of the biocomponent ratio, the calculated biocomponent ratio was determined from the calculated mass percentage and measured carbon content of blends. The measured ^{14}C content results of fuel blends between 0.7 and 9.4% biofraction ratio are shown in the Table 4 in pMC units. Based on these data, the biobased content ratio was determined using different assumptions (as given by Equations 4–6) within a weight precision of $\pm 0.2 \text{ m/m } \%$, except for the samples FUEL_12, 13, and 14, where we could estimate the biocomponent ratio within a precision of $0.3 \text{ m/m } \%$. The pMC of the pure FAME samples (FUEL_9, FUEL_14) are lower than 100 pMC, because FAME contains a methyl group originated from fossil sources that are usually used in the production process due to the cost-efficiency (Holmgren et al. 2007).

Comparisons between the estimated and real mixing ratios value can be made, as the exact biocomponent ratio value was known. The results obtained using the different assumption (as

Table 3 Carbon content and yield of preparation.

Code	Replication	Measured c_T (m/m %)	$\pm c_T$ (%)	Expected c_T (m/m %)	C yield (%)
FUEL_1	5	84.62	0.49	85.99	98.41
FUEL_2 (Pure diesel)	5	84.77	0.62	— *	100*
FUEL_3	6	84.81	0.61	85.89	98.74
FUEL_4	6	84.69	0.97	85.89	98.60
FUEL_5	5	85.27	0.58	86.69	98.36
FUEL_6	6	85.26	0.60	85.78	99.39
FUEL_7	4	85.07	0.10	85.89	99.05
FUEL_8 (Pure HVO)	7	83.81	0.81	—*	100*
FUEL_9 (Pure FAME)	5	76.05	0.21	—*	100*
FUEL_10	4	84.26	0.65	85.87	98.13
FUEL_11	4	84.54	0.99	85.85	98.47
FUEL_12	4	84.45	0.36	85.80	98.43
FUEL_13	4	84.38	0.26	85.33	98.89
FUEL_14 (Pure FAME)	4	72.45	4.81	— *	100*

*Expected c_T was calculated using the measured c_T of the pure bio- and fossil fuel compounds and biocontent composition of blends.

Table 4 Results estimated by ^{14}C AMS measurement and different calculation methods.

Code	Calculated biocomponent ratio (m/m%)	pMC ($\pm 1\sigma$)	Equation 4 (m/m%) ($\pm 1\sigma$)	Equation 5 (m/m%) ($\pm 1\sigma$)	Equation 6 (m/m%) ($\pm 1\sigma$)
FUEL_1	0.70	0.78 ± 0.16	0.76 ± 0.16	0.75 ± 0.15	0.74 ± 0.15
FUEL_2	0.00	0.05 ± 0.06	Pure DIESEL		
FUEL_3	0.98	1.13 ± 0.13	1.10 ± 0.13	1.09 ± 0.13	1.07 ± 0.12
FUEL_4	0.46	0.42 ± 0.17	0.41 ± 0.17	0.40 ± 0.16	0.40 ± 0.16
FUEL_5	0.93	0.86 ± 0.13	0.99 ± 0.15	0.88 ± 0.13	0.82 ± 0.12
FUEL_6	1.90	1.85 ± 0.09	1.95 ± 0.10	1.83 ± 0.15	1.76 ± 0.08
FUEL_7	0.83	1.04 ± 0.07	1.02 ± 0.10	1.00 ± 0.06	0.99 ± 0.07
FUEL_8	100.00	103.97 ± 0.46	Pure HVO		
FUEL_9	100.00	97.56 ± 0.43	Pure FAME		
FUEL_10	2.78	2.83 ± 0.11	2.74 ± 0.15	2.72 ± 0.11	2.69 ± 0.10
FUEL_11	4.64	4.62 ± 0.20	4.48 ± 0.28	4.44 ± 0.19	4.39 ± 0.19
FUEL_12	9.31	9.48 ± 0.16	9.19 ± 0.25	9.12 ± 0.16	9.01 ± 0.15
FUEL_13	9.43	9.23 ± 0.09	9.62 ± 0.14	9.14 ± 0.13	8.77 ± 0.09
FUEL_14	100.00	97.05 ± 0.36	Pure FAME		
Average relative difference			± 0.10	± 0.12	± 0.19

given by Equations 4–6) are quite similar for all the tested sample mixtures, as shown in Table 4 and Figure 1. The R^2 value is higher than 0.99 in each case, showing that all of the estimations made from the measured ^{14}C activity correlate very well with the calculated biocomponent ratios. Clearly, the different assumptions (in Equations 4–6) gave quite accurate results with relatively low error and good agreement in each case. The lowest difference between the calculated and the estimated biocontent values is given by Equation 4, which used the real

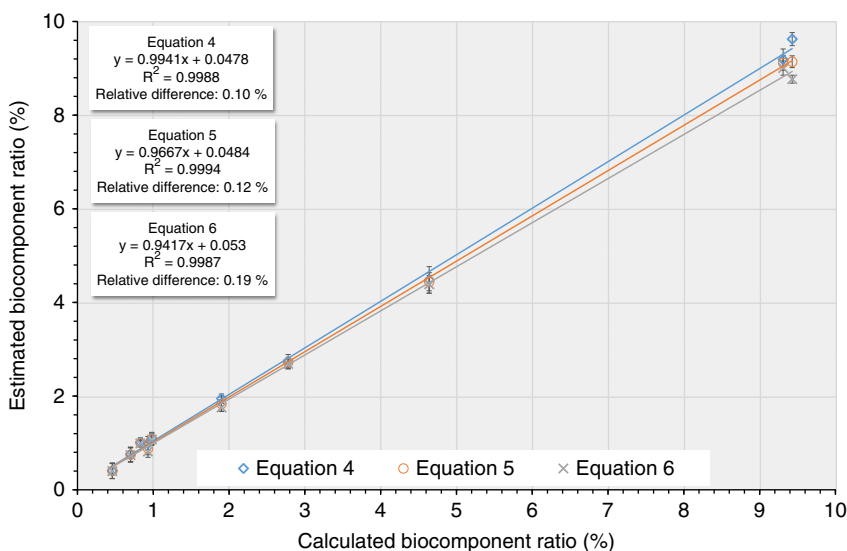


Figure 1 Correlation of the mixtures' estimated biobased content to the calculated biofraction.

measured carbon concentrations for the calculation of the biocontent. This confirms that fewer assumptions and more measured parameters give better results. On the other hand, this approach also have higher reported error bars at the end of the whole calculation due to the propagation of known uncertainties of the applied more measured parameters. Lower errors are found for the simpler calculations (Equations 5 and 6) however, these can underestimate error ranges, as the uncertainty of the applied assumptions are not taken into account.

The simplified biocontent calculation methods result in underestimated error ranges and they gave almost twice the scatter compared to Equation 4. From a practical point of view, all of the estimated biocarbon content shows a good agreement with the calculated values, however the simplified approaches (i.e. Equations 5 and 6) also resulted in a bit higher deviations from the real, expected values. If the exact carbon content was not used for the determination of biocomponent ratio, the final value can be underestimated, because the carbon concentrations of the different components are not the same, and this can affect the final results. These results show that the AMS measurement and the sample preparation techniques developed are precise enough to determine the biobased content of fuel blends in the range of 0–10% bio content. Quarta et al. (2013) pointed out that $\delta^{13}\text{C}$ mass fractionation correction for fossil/biogenic mixtures is complex and can give rise to problems when AMS technique is applied, however, when the biogenic content is lower than 10%, this gives only a negligible change in the uncertainty (Quarta et al. 2013; Palstra et al. 2014).

CONCLUSION

A relatively easy and fast method was used at the MTA ATOMKI Hertelendi Laboratory of Environmental Studies for liquid biofuel biocontent analyses. The method was effectively used for 14 fuel samples with a 1σ uncertainty to better than $\pm 0.3 \text{ m/m } \%$. The precision and reproducibility of our method give similar precision to published techniques (Norton et al. 2007; Oinonen et al. 2010), even in the quite low mixing ratio range ($<1\%$). The preparation time was significantly reduced by using a sealed-tube combustion method with MnO_2 . Thanks to the low volume vacuum line, the gas purification time on the vacuum system can be performed in less

than 10 min/sample. The bottlenecks of the AMS measurement technique are the sample preparation, combustion, and graphitization whereas the time of a normal AMS measurement itself is fixed. Using the specified MnO_2 combustion method combined with the sealed tube graphitization, all of the samples can be combusted and later graphitized altogether (up to 30 samples/normal working day). All things considered, the complete analyses of 30 unknown samples is feasible within 3 normal working days. These results show that the AMS ^{14}C measurement technique is an excellent method for determination of biobased content in fuel blends, even below 1.0 m/m %.

ACKNOWLEDGMENTS

The research was supported by the European Union and the State of Hungary, co-financed by the European Regional Development Fund in the project of GINOP-2.3.2-15-2016-00009 "ICER".

REFERENCES

- Aatola H, Larmi M, Sarjoavaara T, Mikkonen S. 2009. Hydrotreated vegetable oil (HVO) as a renewable diesel fuel: trade-off between NO_x , particulate emission, and fuel consumption of a heavy duty engine. *SAE International Journal of Engines* 1(1):1251–62.
- ASTM D6866-12. 2012. *Standard Test Methods for Determining the Biobased Content of Solid, Liquid, and Gaseous Samples Using Radiocarbon Analysis*, West Conshohocken, PA: ASTM International.
- Berhanu TA, Szidat S, Brunner D, Satar E, Schanda R, Nyfeler P, Battaglia M, Steinbacher, Hammer S, Leuenberger M. 2017. Estimation of the fossil fuel component in atmospheric CO_2 based on radiocarbon measurements at the Beromünster tall tower, Switzerland. *Atmospheric Chemistry and Physics* 17:10753–66.
- Bronić KI, Barešić J, Horvatinčić N, Sironić A. 2017. Determination of biogenic component in liquid fuels by the ^{14}C direct LSC method by using quenching properties of modern liquids for calibration. *Radioation Physics and Chemistry* 137:248–53.
- Chase TN, Pielke RA, Kittel TGF, Zhao M, Pitman AJ, Running SW, Nemani RR. 2001. The relative climatic effects of land cover change and elevated carbon dioxide combined with aerosols: a comparison of model results and observations. *Journal of Geophysical Research* 106 D23:31685–91.
- Deepanraj B, Dhanesh C, Senthil R, Kannan M, Santhoshkumar A, Lawrence P. 2011. Use of palm oil biodiesel blends as a fuel for compression ignition engine. *American Journal of Applied Sciences* 8(11):1154–8.
- Dijs IJ, Windt1 van der E, Kaihola L, Borg van der K. 2006. Quantitative determination by ^{14}C analysis of the biological component in fuels. *Radiocarbon* 48(3):315–23.
- Doll CG, Wright CW, Morley SM, Wright BW. 2017. Analysis of fuel using the direct LSC method determination of bio-originated fuel in the presence of quenching. *Applied Radiation and Isotopes* 122:215–21.
- EN 16640. 2017. European standard: Bio-based products – Bio-based carbon content – Determination of the bio-based carbon content using the radiocarbon method.
- EU Directive. 2003. 2003/30/EC Directive of the European Parliament and of the Council of 8 May 2003 on the Promotion of the Use of Biofuels and Other Renewable Fuels for Transport. OJEU L123. 17 May 2003.
- Gonzalez YM, Caro P de, Thiebaud-Roux S, Lacaze-Dufaure C. 2007. Fatty acid methyl esters as bio-solvents of epoxy resins: a physicochemical study. *Journal of Solution Chemistry* 36: 437–6.
- Holmgren J, Gosling C, Marinangeli R, Marker T, Faraci G, Perego C. 2007. New developments in renewable fuels offer more choices. *Hydrocarbon Process* 86:67–72.
- Hua Q, Barbetti M, Jacobsen GE, Zoppi U, Lawson EM. 2000. Bomb radiocarbon in annual tree rings from Thailand and Australia. *Nuclear Instruments and Methods in Physics Research B* 172:359–65.
- Huber GW, O'Connor P, Corma A. 2007. Processing biomass in conventional oil refineries: Production of high quality diesel by hydrotreating vegetable oils in heavy vacuum oil mixtures. *Applied Catalysis A* 329:120–9.
- Janovics R. 2015. Radiokarbon alapú mérési módszerek fejlesztése és alkalmazásai nukleáris környezetellen-őrzéshez [PhD dissertation]. University of Debrecen. p 49–58. URL: <http://w3.atomki.hu/PhD/these/Janovics%20R%C3%B3bert/disszertacio.pdf>
- Lawrence P, Mathews PK, Deepanraj B. 2011. Effect of prickly poppy methyl ester blends on CI engine performance and emission characteristics. *American Journal of Environmental Sciences* 7(2):145–9.

- Molnár M, Janovics R, Major I, Orsovski J, Gönczi R, Veres M, Leonard AG, Castle SM, Lange TE, Wacker L, Hajdas I, Jull AJT. 2013. Status report of the new AMS ^{14}C sample preparation lab of the Hertelendi Laboratory of Environmental Studies (Debrecen, Hungary). *Radiocarbon* 55(2–3): 665–76.
- Nigam PS, Singh A. 2011. Production of liquid bio-fuels from renewable resources. *Progress in Energy and Combustion Science* 37:52–68.
- Norton GA, Devlin SL. 2006. Determining of modern carbon content of biobased products using radiocarbon analysis. *Bioresource Technology* 97:2084–90.
- Norton GA, Hood DG, Devlin SL. 2007. Accuracy of radioanalytical procedures used to determine the biobased content of manufactured products. *Bioresource Technology* 98:1052–6.
- Oinonen M, Hakanpaa-Laitinen H, Hamalainen K, Kaskela A, Junger H. 2010. Biofuel proportions in fuels by AMS radiocarbon method. *Nuclear Instruments and Methods in Physics Research B* 268:1117–9.
- Palstra WLS, Meijer HAJ. 2014. Biogenic carbon fraction of biogas and natural gas fuel mixtures determined with. *Radiocarbon* 56(1):7–28.
- Quarta G, Elia MD, Valzano D, Calcagnile L. 2005. New bomb pulse radiocarbon records from annual tree rings in the Northern Hemisphere Temperate Region. *Radiocarbon* 47(1):27–30.
- Quarta G, Calcagnile L, Giffoni M, Braione E, Marisa D. 2013. Determination of the biobased content in plastics by radiocarbon. *Radiocarbon* 55(2–3):1834–44.
- Rinyu L, Molnár M, Major I, Nagy T, Veres M, Kimák Á, Wacker L, Synal HA. 2013. Optimization of sealed tube graphitization method for environmental ^{14}C studies using MICADAS. *Nuclear Instruments and Methods in Physics Research B* 294:270–5.
- Vrtiška D, Šimáček P. 2016. Prediction of HVO content in HVO diesel blends using FTIR and chemometric methods. *Fuel* 174:225–34.
- Vyas AP, Verma JL, Subrahmanyam N. 2010. A review on FAME production processes. *Fuel* 89:1–9.
- Wacker L, Christl M, Synal HA. 2010. Bats: A new tool for AMS data reduction. *Nuclear Instruments and Methods in Physics Research B* 268:976–9.
- Yunoki S, Saito M. 2009. A simple method to determine bioethanol content in gasoline using two-step extraction and liquid scintillation counting. *Bioresource Technology* 100:6125–8.