

Analysis of isotopic ratios for the detection of illegal watering of beverages

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Abstract

The applications of the stable isotope ratio analyses (D/H, ^{13}C and ^{18}O) in the assessment of watering in beverages such as wine and fruit juices are discussed in this study. The measurement of $^{18}\text{O}/^{16}\text{O}$ ratio in combination with D/H and $^{13}\text{C}/^{12}\text{C}$ ratio is mainly focused and the international official methods implementing stable isotope techniques will be mentioned. Furthermore, the ongoing study on a possible use of $^{13}\text{C}/^{12}\text{C}$ ratio of CO_2 to detect the addition of technical produced CO_2 in mineral sparkling waters is also presented. © 2007 Elsevier Ltd. All rights reserved.

Keywords: Food authentication; EU regulations; Stable isotope; Deuterium; $\delta^2\text{H}$; Carbon-13; $\delta^{13}\text{C}$; Oxygen-18; $\delta^{18}\text{O}$; Wine; Watering; Fruit juice; Carbon dioxide; CO_2

1. Introduction

In the last 30 years, the measurements of stable isotopes (i.e. non-radioactive isotopes) have demonstrated their convenience as a tool in the fight against fraud in the food products industry (Rossmann, 2001; Kelly, Heaton, & Hoogewerff, 2005). Several isotopic analyses are now official or standard methods in Europe and North America for testing the authenticity of food commodities, as will be discussed later. These methods are based on the measurement of stable isotope contents (^2H , ^{13}C , ^{18}O , ^{15}N , etc.) (Table 1) of a product or of a specific component, such as an ingredient or a target molecule of the product. The determinations, carried out using nuclear magnetic resonance (NMR) and/or isotope ratio mass spectrometry (IRMS), provide information on the botanical and geographical origins which are often considered important characteristics of many food products either by the consumer or by national and international regulations.

However, it is important to underline that only a multi-isotopic – of more than one element – and/or multi-component – on more than one “ingredient” – analysis permits a reliable assessment of the authenticity of a food product (Table 2).

2. Hydrogen and oxygen: natural isotopic fractionation

Fractionation of hydrogen and oxygen isotopes takes place along with the evaporation and condensation processes of the water cycle: water passing from its liquid state in the ocean to that of the vapour in the atmosphere moisture undergoes a strong isotope fractionation resulting in depletion in the heavier isotopes (deuterium or ^2H , oxygen-18 or ^{18}O) in vapour and clouds. The extent of the fractionation is dependent on the temperature.

As an air mass follows a trajectory from the source (oceans, seas) over continents it cools and loses water vapour along the way as precipitation. During this “rain-out” process, isotopically enriched rain is falling and the vapour becomes progressively depleted in its ^{18}O - and ^2H -content (Fig. 1). The effect of the sum of climatic geographic and topographic conditions results in a worldwide

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Nomenclature

AIJN	Association of the Industry of Juices and Nectars from Fruits and vegetables of the European Union, Brussels (Belgium)
AOAC	Association Of Analytical Communities, Gaithersburg (USA)
BEVABS	Bureau Européen des Vins, Alcools et Boissons Spiritueuses (European Office for Wine, Alcohol and Spirit Drinks), Ispra (Italy)
CEN	Comité Européen de Normalisation (European Committee for Standardization), Brussels (Belgium)
δ , delta	Relative scale for expression of isotope ratios. For example, for the $^{18}\text{O}/^{16}\text{O}$ ratio delta is defined: $\delta^{18}\text{O}_{\text{V-SMOW}} = \left[\frac{\left(\frac{^{18}\text{O}}{^{16}\text{O}} \right)_{\text{sample}}}{\left(\frac{^{18}\text{O}}{^{16}\text{O}} \right)_{\text{V-SMOW}}} - 1 \right] \times 1000$.
	δ values are expressed in parts per 1000 (‰, per mille)
EA-IRMS	Elemental Analyzer coupled to an IRMS

EU	European Union
IAEA	International Atomic and Energy Agency, Vienna (Austria)
IRMS	Isotopic Ratio Mass Spectrometry
NIST	National Institute of Standards and Technology, USA
OIV	Organisation (ex Office) Internationale de la Vigne et du Vin (International Organisation of Vine and Wine), Paris (France)
SNIF-(^2H)NMR [®]	site-specific natural isotopic fractionation – nuclear magnetic resonance of deuterium
TC/EA-IRMS	total conversion-elemental analyzer coupled to an IRMS
V-PDB	Vienna-Pee Dee Belemnite. International Standard Reference for the $^{13}\text{C}/^{12}\text{C}$ ratio
V-SMOW	Vienna-Standard Mean Ocean Water. International Standard Reference for the $^2\text{H}/^1\text{H}$ and $^{18}\text{O}/^{16}\text{O}$ ratios.

distinctive isotope pattern of the meteoric waters (Rozanski, Araguas, & Gonfiantini, 1993).

While water is the only source of hydrogen for the photosynthesis, oxygen is taken by plants from several sources, from atmospheric oxygen and carbon dioxide, and mainly from water pools from soil. Consequently, the ^2H - and ^{18}O -contents of the vegetation water of wines or fruit juices should reflect the place where the product comes from (Fig. 2).

Plant physiology also plays an important role in isotope fractionations, for instance with the relative aperture or closing of stomata to adapt to the availability of water in the plant environment (atmospheric and soil moisture).

The evapo-transpiration taking place during the maturation period is in fact a factor of isotopic fractionation: this process causes enrichment in heavy isotopes of both oxygen and hydrogen atoms of water of plants and fruits. The isotope ratios observed in plant water are positive relative to those of the corresponding ground water. Furthermore, an “aerial” plant as vine shows a stronger evapo-transpiration effect than sugar beet which grows underground and this will be recorded as enrichment in deuterium in sugars from vines comparing to sugars from beet. Such difference is the principle of SNIF-NMR method (site-specific natural isotope fractionation-NMR) that measures the enrichment of deuterium site by site on the alcohol from fermentation.

Table 1
Mean natural abundances, international references and other data for light stable isotopes used in food commodities authentication

Element	Isotope	Relative mean natural abundance (%) (Rossmann, 2001)	International reference and isotopic ratio (R)
Hydrogen	^1H	99.9855	V-SMOW (H_2O)
	$^2\text{H(D)}$	0.0145	$^2\text{H}/^1\text{H} = 1.5576 \times 10^{-4}$
Carbon	^{12}C	98.892	V-PDB ^a (CaCO_3)
	^{13}C	1.108	$^{13}\text{C}/^{12}\text{C} = 1.1237 \times 10^{-2}$
Nitrogen	^{14}N	99.6337	AIR (atmospheric nitrogen – N_2)
	^{15}N	0.3663	$^{15}\text{N}/^{14}\text{N} = 3.6765 \times 10^{-3}$
Oxygen	^{16}O	99.7587	V-SMOW or V-PDB
	^{17}O	0.0375	$^{18}\text{O}/^{16}\text{O} = 2.0052 - 2.0672 \times 10^{-3}$
	^{18}O	0.2039	

^a The PDB is no longer available. Other international reference materials (distributed by IAEA and by NIST) are used.

Table 2
Typical isotope ratio analyses in food stuff authentication and fraud assessment

Product	Adulteration	Analysis
Fruit juices	Watering	(D/H) and ^{18}O IRMS
Fruit juices	Sweetening	^{13}C IRMS and ^2H NMR
Honey	Addition of inverted and cane sugars	^{13}C IRMS
Aromas, flavours (vanillin, raspberry ketone, etc.)	Mislabelling (artificial ↔ natural)	^{13}C IRMS, ^{18}O IRMS, ^2H NMR
Alcoholic beverages	Mislabelling (geographical and botanical origins)	^{13}C IRMS, ^{18}O IRMS, ^2H NMR, ^2H and ^{18}O pyr IRMS
Wines	Watering	(D/H) and ^{18}O IRMS
Wines	Chaptalisation (addition of sugars)	^{13}C IRMS, ^2H NMR

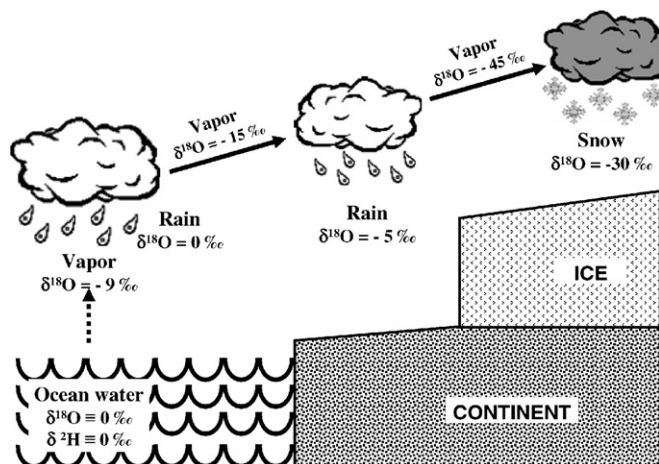


Fig. 1. Climatic and geographical effects on the isotopic fractionation of elements in water. The ^2H depletion process has the same trend than ^{18}O .

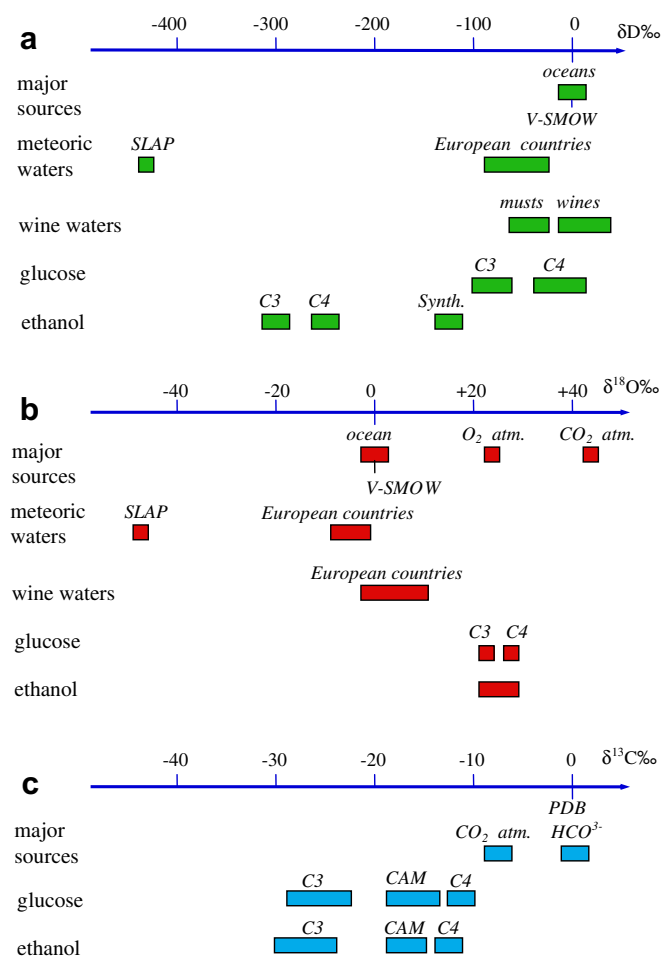
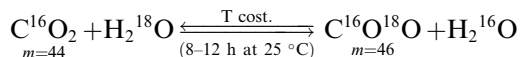


Fig. 2. Natural stable isotope abundances for hydrogen (a), oxygen (b) and carbon (c). Plants are classified as C3, C4 and CAM according to their photosynthetic path to incorporate CO_2 , due to the different physiologies. Typical C3 plants are grape, beet, rice and wheat. Among C4 plants are cane and corn. Examples of CAM plants are pineapple, cactus and vanilla.

3. Measurement of $^{18}\text{O}/^{16}\text{O}$ ratio

The $^{18}\text{O}/^{16}\text{O}$ ratio can be measured in different ways, but all the official methods are based on the “equilibration method”, described by Epstein and Mayeda (1953), that takes advantage of the following equilibrium:



This classical method has been improved during the years and it is useful to measure the ^{18}O -content of water in a complex matrix. Once the carbon dioxide (CO_2) gas used for the equilibration has exchanged ^{18}O with the H_2O in wine/fruit juice, the headspace is pumped through a cold trap, to eliminate H_2O and eventual ethanol, and then introduced and analysed with the Isotope Ratio Mass Spectrometer. This mass spectrometer is capable to measure just a narrow range of masses, but with a very high precision.

The CO_2 gas from the sample is first ionised by the electronic source of the IRMS (Fig. 3). The ions of different masses (44, 45 and 46) due to the distribution of ^{18}O (and ^{17}O and ^{13}C) are separated during their flights by means of a magnetic field and collected by the mass detectors (Faraday cages) that proportionally generate electrical signals. The ratio of the electrical signals of masses 46 to 44 is given as a deviation of the isotopic ratio from a reference substance and calculated as delta value per mille (δ , ‰).

The $\delta^{18}\text{O}$ ‰ value of the sample is calculated for comparison to a water, chosen as working standard, already calibrated against the international standard V-SMOW.

4. Measurement of $^2\text{H}/^1\text{H}$ ratio

The analysis of $^2\text{H}/^1\text{H}$ of water in wine and fruit juice needs a long preparation in laboratory, which, in the original methods – considered for measurements in pure water – involves the use of reducing agents such as uranium, zinc alloy or chromium to obtain hydrogen (H_2) gas (Bigeleisen, Perlman, & Prosser, 1952; Coleman, Shepherd, Durham, Rouse, & Moore, 1982). These methods are used either as on-line or off line reduction methods, but their disadvantage for complex food matrix is that they necessitate an extraction step (distillation or inversed freeze-drying) to separate quantitatively, and without isotopic fractionation, the pure water from other components such as the organic compounds sugars, acids, lipids or ethanol. The determination of $^2\text{H}/^1\text{H}$ ratio in fruit juices by distillation, reduction and IRMS measurement is an officially accepted method (CEN/TC174 No. 110, 1996).

There is still an open discussion on the reliability of the various methods of reduction, especially for application to complex matrices. Alternative to the reduction methods described above, a technique similar to the equilibration used for $\delta^{18}\text{O}$ measurement has been tested successfully on beverages like fruit juice or wine (Bréas, Guillou, Reniero, Sada, & Tanet, 1996). The principle of the method is

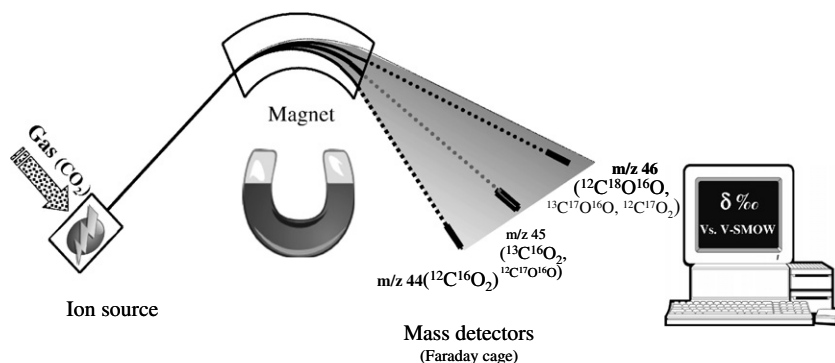


Fig. 3. Principles of the IRMS system set up for the $^{18}\text{O}/^{16}\text{O}$ measurement in water after equilibration with CO_2 .

based on the equilibration of the samples with hydrogen gas using a platinum catalyst.

Nowadays commercially available devices facilitate the operation of these equilibration procedures. This will most likely help to improve the comparability of results among laboratories and will allow proposing standard validated international methods for analysis of the isotopic content of water in food matrices.

5. Official methods for the detection of watering of beverages

Over the last 15 years, in Europe, measurements based on the determination of stable isotope ratios for the analysis of beverage commodities have been adopted as official methods by European Union (EU) regulations and by the International Organisation of Vine and Wine (OIV) (Calderone, Guillou, Naulet, 2003 for a review). Successively some countries as France and Mexico have adopted (or are still discussing) these techniques at national level also for other products, such as honey, aromas etc. (Table 3).

The method described above to determine the $^{18}\text{O}/^{16}\text{O}$ ratio of wine water using IRMS was officially put in force

within the EU in 1997 (E.C. Regulation No. 822/97). This analysis allows the detection and quantification of watering of wine but it needs the comparison of analytical data with those determined on genuine products from the same origin. In 1993 the European Parliament launched the *European Office for Wine, Alcohol and Spirit Drinks* (BEVABS) at the Joint Research Centre (JRC) to combat major frauds in the wine area. The JRC plays a crucial role by managing the European Wine Databank on authentic wines – as foreseen in EC Regulation No. 2729/2000, recently updated by Regulation No. 2120/2004 that has also taken into account the new Member States producers of wine who joined the EU on May 2005 – and by coordinating the network of official Member States laboratories involved in the IRMS and NMR determinations required for the EU Wine Databank. As the Reference Laboratory for these measurements, BEVABS has also a specific role for quality control, validation of the data and arbitration of disputes, as is shown by an example later in this paper.

Since 2002, also Mexican regulations have implemented the $^{18}\text{O}/^{16}\text{O}$ ratio determination for the assessment of wine watering (NMV-V-025-NORMEX-2002).

Table 3

Synopsis of official analyses internationally adopted that implement isotopic methods for the analyses/detection of water in beverages

EU (European Union)

European Commission Regulation No. 2676/90 determining Community methods for the analysis of wines – Official Journal of the European Communities No. L 272/64. (SNIF- ^2H NMR of ethanol from wine)

EC Regulation No. 822/97 amending E.C. Regulation No. 2676/90 – OJEC No. L117/10. ($^{18}\text{O}/^{16}\text{O}$ of water in wine)

OIV (Organisation International de la Vigne et du Vin)

Resolution ENO 2/96. Watering: detection method. Bulletin OIV 1997; 791–792: 44–65

CEN (Comité Européen de Normalisation)

CEN/TC174 No. 109, 1996: Determination of the stable oxygen isotope ratio ($^{18}\text{O}/^{16}\text{O}$) of water from fruit juices – Method using Isotope Ratio Mass Spectrometry

CEN/TC174 No. 110, 1996: Determination of the stable hydrogen isotope ratio ($^2\text{H}/^1\text{H}$) of water from fruit juices – Method using Isotope Ratio Mass Spectrometry

AOAC (Association Of Analytical Communities)

AOAC method, determination of sugar beet derived syrups in frozen concentrated orange juice- $\delta^{18}\text{O}$ measurements in water. Method 992.09

Mexican regulations

NMX-V-025-NORMEX-2002 bebidas alcohólicas – (...) – determinación de adición de agua en los vinos mediante la relación isotópica del oxígeno 18 ($\delta^{18}\text{O}_{\text{VSMOW}}$) por espectrometría de masas de isótopos estables – método de prueba

PROY-NOM-000-SCFI-2004 alimentos – jugos y néctares envasados. Denominaciones, especificaciones fisicoquímicas, información comercial y métodos de prueba

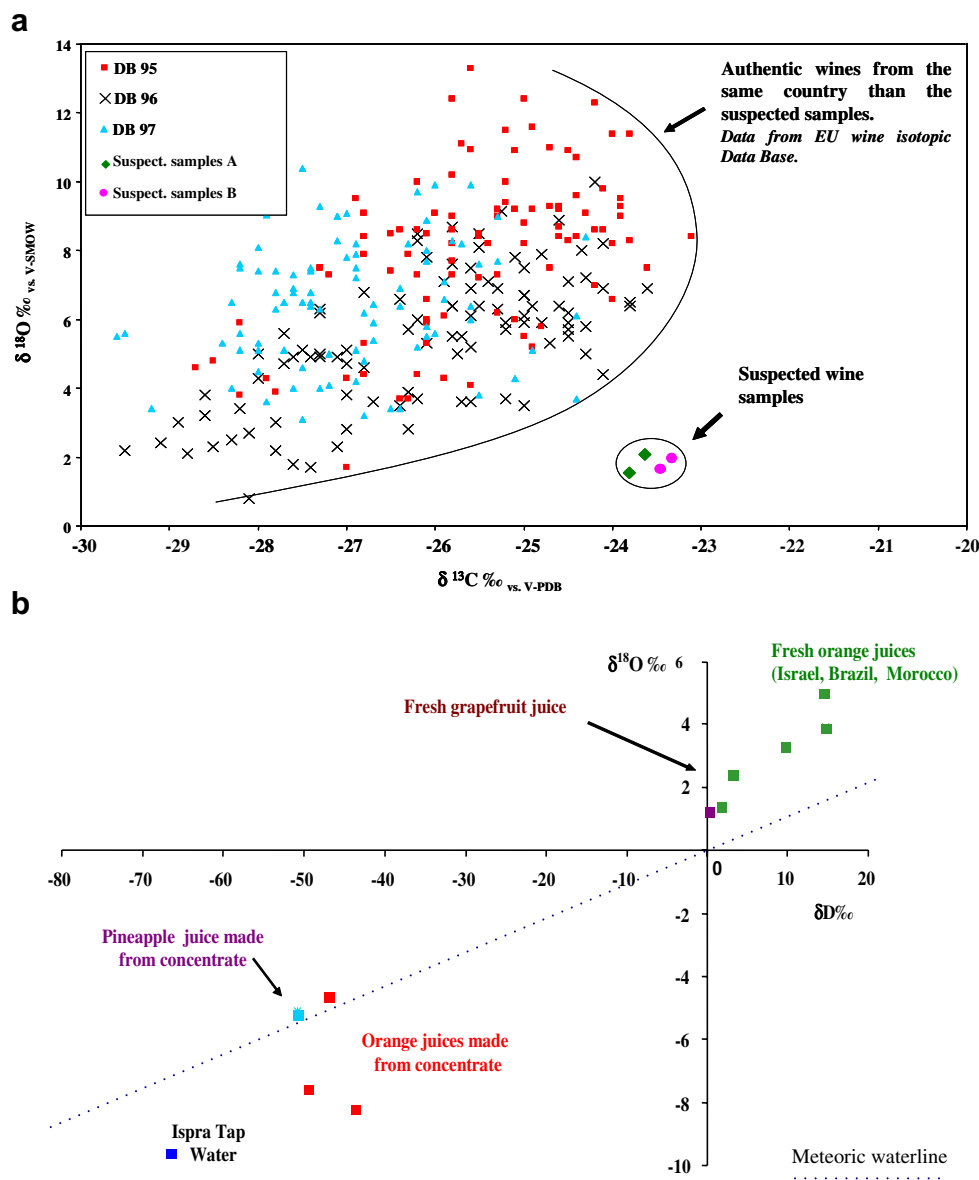
Both, the industry, Comité Européen de Normalisation (CEN) and the Association Of Analytical Communities (AOAC) have adopted the analysis of $^{18}\text{O}/^{16}\text{O}$ and $^2\text{H}/^1\text{H}$ ratios of water for the assessment of fruit juices (CEN/TC174 No. 109 and 110, 1996) and in frozen orange juice (AOAC Method 992.09).

6. Examples of applications

6.1. Wine

A real case of multi-isotopic and multi-component analysis of wine (^{13}C measured on the distilled ethanol from wines, ^{18}O determined on the water of the wine) for detection of adulteration is shown in Fig. 4a.

This case was submitted to BEVABS by an official EU Member State which had some suspicion about the authenticity of products declared as originating from another EU Member State. The year of production was not indicated for these products, so the isotopic parameters were compared with the values contained in the EU Wine Databank for the three last harvests of the declared country of origin of these wines. The single interpretation of the $\delta^{18}\text{O}$ or $\delta^{13}\text{C}$ values shows that the analysed data of the suspicious samples are still within the range of the databank values. However the combination of the results of both isotope ratios clearly indicates a wrong declaration. The comparatively positive ^{13}C -content observed for the suspicious wines is generally explained as addition of C4 sugar (e.g. cane sugar) used for enrichment of the alcohol grade, which



Graphs adapted after (Guillou, Reniero, 2001).

Fig. 4. Examples of stable isotope analyses applied to water in wines (a) and fruit juices (b). Graphs adapted after (Guillou & Reniero, 2001).

was not a practice allowed for the declared country of origin. Lower $\delta^{18}\text{O}$ values are generally indicative of watering of wines. In that particular case the explanation would thus be an extension of volume by addition of water and sugar (chaptalisation) to maintain the alcohol grade level.

6.2. Fruit juices

The IRMS determination of ^2H - and ^{18}O -content of water of juice is one of the most widely known applications permitting the distinction between the direct juices and the juices made from concentrates by re-dilution with “tap” water (Bricout, 1978; Bricout & Koziat, 1987). An example of recognition between these two categories of fruit juice is shown in Fig. 4b. In this graphic elaboration, the samples of fruit juices obtained from concentrated juice reconstituted with water are clearly separated from the fresh fruit juices. The advantage of the combined measurement of the $\delta^2\text{H}$ and the $\delta^{18}\text{O}$ values is the correlation of the values: juices that are not re-diluted with tap water must plot above the so called global meteoric waterline (GMWL) defined by Craig (1961) as $\delta^2\text{H} = 8 \times \delta^{18}\text{O} + 10$. As reference, a sample of tap water, Ispra Tap Water, $\delta^2\text{H} = -65\text{‰}$ and $\delta^{18}\text{O} = -10\text{‰}$, was also measured and plotted on the graph.

The fruit juice industry has accepted this method for the identification of re-diluted juices and has published indicative ranges of values for isotope contents of genuine fruit juices (Association of the Industry of Juices and Nectars from Fruits and vegetables of the European Union, AIJN, Code of practice for evaluation of fruit and vegetable juices).

7. Ongoing studies on stable isotopes applied to the water for beverage authentication

In recent years, both, our laboratory (BEVABS) and other research groups have studied the correlation between the $^{18}\text{O}/^{16}\text{O}$ ratios of water and that of ethanol resulting from the fermentation of sugars from the corresponding fruit juices or must (wine). This approach may help to improve the sensitivity for detection of added water, and/or for checking the geographical origin of fruit juices and wines (Jamin, Guérin, Rétif, Lees, & Martin, 2003; Calderone, 2004).

In Fig. 5 the ^{18}O -contents of genuine wine samples, in ethanol and in water, are correlated with each other. The wines, from vintage 1998, belong to the EU Wine DB. The $\delta^{18}\text{O}$ values of water were measured as usual, by IRMS after equilibration; the $\delta^{18}\text{O}$ values of the ethanol were measured on the distillates of the wine samples, by using a new technique, still under validation (Calderone, Reniero, & Guillou, 2006), the Total Conversion-Elemental Analyzer coupled to an IRMS (TC/EA-IRMS).

Fig. 5 offers a good correlation between ethanol and water and therefore may be used as a further method for the detection of added water. The wide range of the $\delta^{18}\text{O}$ values clearly shows the effect of the origin samples: the wine samples from colder/continental countries (Germany, Austria, part of French samples) are concentrated in the lowest part (depletion in ^{18}O), while the samples from warmer/peninsular countries (Greece, Spain, Portugal, part of Italian samples) are more in the upper zone (enrichment in ^{18}O).

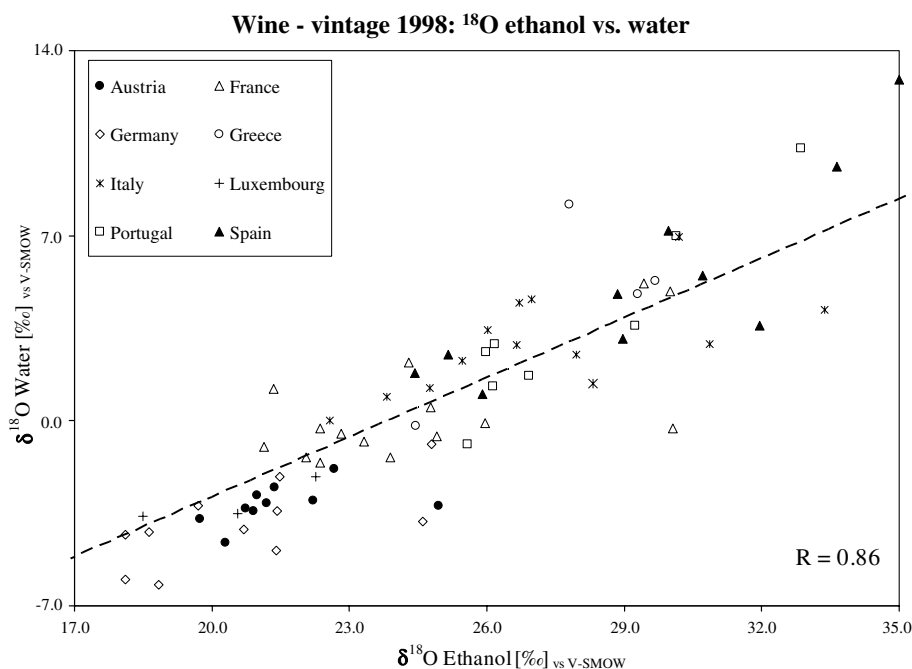


Fig. 5. Combination of $\delta^{18}\text{O}$ values of ethanol and water from analyzed wines.

Table 4
 ^{13}C -content of headspace CO_2 from samples of sparkling water on the market

Sample	Origin/bottling plant	$\delta^{13}\text{C}$ CO_2
Naturally sparkling 1	Vergèze, France	−4.4
Naturally sparkling 2	Gerolstein, Germany	−3.3
Naturally sparkling 3	Nepi (VT), Italy	−1.1
Naturally sparkling 4	Riardo (CE), Italy	−0.4
Naturally sparkling 5	Roma, Italy	−42.1
Carbonated 1	San Pellegrino Terme (BG), Italy	−5.1
Carbonated 2	Casolino al Piano (CO), Italy	1.3
Carbonated 3	San Giorgio in Bosco (PD), Italy	−29.2
Carbonated 4	Crodo (VB), Italy	−54.2

Values of $\delta^{13}\text{C}$ are expressed in ‰ vs. V-PDB.

Re-elaboration after Calderone et al. (2007).

An interesting application for bottled sparkling water is the control of the origin of the CO_2 on the basis of the analysis of the $^{13}\text{C}/^{12}\text{C}$ ratio of headspace CO_2 in the bottles. The declaration “Naturally sparkling” prohibits the use of technical produced CO_2 . Technical produced gases can be produced, for instance, by the combustion of hydrocarbons and are characterized by low $\delta^{13}\text{C}$ values (Boner & Förstel, 2001).

In Table 4 $\delta^{13}\text{C}$ values of CO_2 of different bottled waters are listed. The water sample Naturally sparkling 5 (sold on the market) was evidently mislabelled as can be seen by the low $\delta^{13}\text{C}$ value of the headspace CO_2 −42.1‰. The comparison of sample Naturally sparkling 5 to samples Carbonated 3 and Carbonated 4 is an extra evidence of the false or incorrect labelling (Calderone, Guillou, Reniero, & Naulet, 2007).

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