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Isotope Analysis (^{13}C, ^{18}O) of Wine From Central and Eastern Europe and Argentina, 2008 and 2009 Vintages: Differentiation of Origin, Environmental Indications, and Variations Within Countries

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In this study, we compare the stable isotope composition of oxygen and carbon of wines from four Central and Southeastern European countries and from Argentina to study the similarities and differences in the isotope signatures and, thus, the potential of differentiation of the various wine-growing countries. We observe similar trends for wines from Austria, Slovenia, and Romania with respect to the vintages 2008 and 2009, which are absent in the Montenegrin and Argentinean samples. It is speculated that the weather develops similarly for Austria, Slovenia, and Romania, as these countries are positioned at a similar latitude and not too far away from each other (general central and eastern European weather situation), whereas Montenegro is not influenced by the latter being situated farther south and dominantly influenced by the Adriatic Sea. Investigations on further vintages are needed to test this assumption.

Keywords: oxygen isotope, carbon isotope, water, alcohol, authenticity, geographic origin

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INTRODUCTION

In the last century, several wine scandals occurred in the 70's and 80's in Europe, a few of them even had lethal consequences (http://www.spiegel.de/spiegel/print/d-13519771.html). To tackle this situation seriously affecting the consumers' confidence, rights, and health, in 1990, the EU commission passed a regulation and installed the EU-wine database. For this database, every wine-producing EU-country has to collect a certain number of grape samples in the vineyards every year, transform the grapes into wine by micro-vinification, and measure the isotope pattern of these samples by isotope ratio mass spectrometry (IRMS) and site-specific natural isotope fractionation nuclear magnet resonance (SNIF-NMR). Regulation EC No. 555/2008 amended and replaced the

earlier ones. Initially, the isotope investigations were solely intended for wine authenticity control, to identify any illegal addition of water or sugar (chaptalization and sweetening).

As the isotope ratio of most biogenic materials and especially plant tissue is influenced by their environmental conditions, the control of declared geographic origin can also be carried out by these analyses. As wine from certain wine-growing regions and also vineries receive far higher prices (e.g., sparkling wine from Champagne/France) than wine from other areas or wine producers, there is also a need to control the geographic origin. The investigation of the geographic origin of wine by analyzing the isotope pattern is founded on the idea that every region and locality has unique environmental conditions. Stable isotope measurements for the control of declared food origin investigate the isotope ratio of elements influenced by, e.g., weather and water availability, distance from the sea, altitude, soil, and natural and anthropogenic emissions (e.g., Rossmann, 2001; Camin et al., 2007; 2010; Horacek and Min, 2010; Horacek et al., 2010, 2015). The isotope ratio in precipitation is a result of the climate and geographic position (Bowen and Revenaugh, 2003). Water vapor evaporating from a water surface is isotopically depleted with respect to the water from which it emanates (Dansgaard, 1964). This isotopic fractionation is temperature-dependent, with a strong fractionation at low temperatures and minor fractionation at elevated temperatures (temperature effect). Water vapor migrating in clouds over a continent becomes isotopically successively more and more depleted in ^{18}O and ^{2}H (continental effect), as the heavy oxygen and hydrogen isotopes preferentially enter the liquid phase (rain, snow) and are, in this way, removed from the clouds (Gat and Gonfiantini, 1981).

For wine, the weather is a very important influencing factor, not only with respect to quality and quantity, but also regarding the stable isotope pattern. Martin and Martin (2003) identified that $\delta^{13}C$ is positively correlated with the mean temperature and negatively correlated with the amount of precipitation during the sugar accumulation and grape ripening period in wines. In addition, they showed that the wine water $\delta^{18}O$ value is positively correlated with the temperature and negatively with the amount of precipitation, but also additional influences by other climatic parameters. Furthermore, in Californian wines, a good correlation was found for $\delta^{18}O$ and the daily relative humidity in the 3 weeks prior to harvesting, for crop evapotranspiration in September, and for the average maximum daily temperatures from July (Ingraham and Caldwell, 1999). Other studies demonstrate the influence of relative humidity of a period of 30 days prior to harvest and the $\delta^{18}O$ of atmospheric humidity on the wine water $\delta^{18}O$ (Hermann and Voerkelius, 2008). For Italian wines, a statistical evaluation of the relevant influencing parameters (including geographical and climatic information) on the isotope values has been carried out by Camin et al. (2015); for Austrian wines, this has been preliminary done by Heinrich et al. (2016). A study on the differentiation of the geographic origin of wine from a border region applying a diverse combination of physico-chemical methods, to investigate strengths and limitations of these methods, and the potential of their combination has been carried out by Horacek et al. (2019a,b).

FIGURE 1 | Map indicating the countries of origin for the investigated wine samples.

As the weather conditions play such a fundamental role for the isotope pattern of wine, the year of harvest is important since the weather varies from year to year (e.g., Christoph et al., 2003, 2004, 2015; Magdas et al., 2012; Philipp et al., 2018). The investigation of wine harvested in 2008 and 2009 is of relevance for wine control, as (I) wine is a commodity that is not only consumed as young wine, but also as aged wine, with the (suitable) aged wine becoming more esteemed and, thus, expensive, and (II) the environmental conditions do vary from year to year, but similarities between certain years are usually found (e.g., cold-humid vs. hot-dry weather) and, thus, past vintages show ranges of variations between years, relevant for control of wine without vintage information.

In the present study, wine samples from Austria, Slovenia, Romania, Montenegro, and Argentina (**Figure 1**) have been collected, processed, and analyzed for their carbon and oxygen isotope values. The isotope results of samples are compared with respect to vintages and with respect to the investigated samples from the other countries. The aim of this work was to investigate if any pattern exists in both vintages and the potential of discrimination of the samples from other origins. Furthermore, comparison of European wines with samples from Argentina shows whether there is a distinctive correlation of distance and isotope patterns of the wine samples. We hypothesize that we can differentiate the wine samples by their isotope patterns due to their geographic origin. The further away the geographic origin, the better the discrimination for identical vintages.

Wine Regions

Wine samples of the years 2008 and 2009 from Austria, Slovenia, Romania, Montenegro, and Argentina have been collected (**Table 1**) and analyzed. As some of these samples (from Austria and Slovenia) are the official ones for the EU wine database, which were harvested in selected vineyards by governmental collectors and vinified applying a standardized protocol, an assignment to a certain area, locality, or winery was not done to retain data confidentiality. The Romanian samples come from only two of the wine producing regions, Oltenia and Muntenia, both located in the very south of Romania (Magdas et al., 2012) and, therefore, do not represent the isotopic variation of the entire Romania, which is significantly larger (see Magdas et al., 2012). The Montenegrin wine samples come from "13. Jul PlantaŽe," the biggest winery in Montenegro, which produces around 50% of the entire Montenegrin wine production. Argentinean wine samples come from the Mendoza area, except for one sample from the Neuqen region.

SAMPLES AND METHODS

The wine samples from Austria, Romania, and Slovenia were collected, processed, and analyzed according to the EU regulation EC No. 555/2008 and the Compendium of the OIV (OIV-MA-AS312-06, 2001; OIV-MA-AS2-12, 2009). For the other countries (Argentina and Montenegro), commercially produced bottled wines were collected and analyzed. All samples were distilled using an automated distillation control system using cadiot distillation columns by Eurofins/Nantes, France. Distilled samples have a yield better than 90%. Carbon isotope ratio was analyzed of the distillated ethanol, oxygen isotope ratio of the wine water in bulk wine samples. The samples from Austria and Montenegro were analyzed at the Austrian Institute of Technology GmbH Tulln stable isotope laboratory. Argentinean wine samples were measured at the BLT Wieselburg stable isotope facility. Slovenian samples were processed at the Agricultural Institute of Slovenia and analyzed at the JoŽef Stefan Institute, Ljubljana and the Romanian samples at the National Institute for Research and Development of Isotopic and Molecular Technologies, Cluj-Napoca. Instrumentation details and descriptions can be found in **Supplementary Material 1**.

The results are expressed in the conventional δ-notation in ‰ with respect to the V-SMOW (Vienna-Standard Mean Ocean Water) and with respect to the V-PDB (Vienna-PeeDee Belemnite) standards for oxygen and carbon, respectively, which are as follows:

$$\delta X‰ = ((Rsample/Rstandard) - 1)x\ 1000 \quad (1)$$

where X is ^{13}C or ^{18}O and R is the ratio of $^{13}C/^{12}C$, or $^{18}O/^{16}O$, respectively. The enlarged reproducibility of measurements of $\delta^{13}C$ and $\delta^{18}O$ were better than ±0.3 and ±0.5‰, respectively, for all laboratories. For quality control and comparability of the results, identical or comparable certified standards and reference materials were analyzed together with the wine samples. Among the measured standards are V-SMOW ($\delta^{18}O = 0.0‰$) and SLAP ($\delta^{18}O = -55.5‰$) (both provided by the International Atomic Energy Agency (IAEA); and BCR 660 ($\delta^{13}C = -26.72‰$) and BCR 656 ($\delta^{13}C = -26.91‰$) [both produced by the Institute of Reference Materials and Measurements (IRMM)].

Statistical Evaluation

The statistical evaluation has been carried out using classifications by the linear discriminant analysis (LDA). The chemometric processing of experimental data was made using the SPSS Statistics 24 (IBM, USA). The algorithm behind this method is based on finding a linear combination among the analyzed variables, which separates the predefined classes of samples. By these combinations, a model is obtained, which is validated by the "leave-one-out" cross validation method. This method removes each sample from the sample-set and reclassifies it as an unknown. Outlier test was performed using the Grubbs test by the Graphpad Software (graphpad.com; USA) and orthogonal partial least squares discriminant analysis (OPLS-DA) by the SIMCA (https://www.sartorius.com/en/products/process-analytical-technology/data-analytics-software/mvda-software/simca; Germany).

RESULTS

A short summary is given in **Table 1** and graphically shown in **Figures 2**, **3A,B**. Austrian wine samples from 2008 have $\delta^{13}C$ values ranging from −25.8 to −29.7‰, and from 2009, from −25.3 to −30.0‰. The $\delta^{18}O$ values of the Austrian samples are between −3.0 and +1.8‰ for 2008 and between −2.6 and +3.1‰ for 2009 (**Figures 2**, **3A,B**). Average values for 2008 are

TABLE 1 | Number (*n*) of wine samples per year and country, average $\delta^{13}C$ and $\delta^{18}O$ values, and standard deviation for 2008 and 2009.

	n	*n*	$\delta^{13}C$		$\delta^{18}O$		$\delta^{13}C$		$\delta^{18}O$	
Country	**2008**	**2009**	**Av $\delta^{13}C$ 2008**	**STDEV**	**Av $\delta^{18}O$ 2008**	**STDEV**	**Av $\delta^{13}C$ 2009**	**STDEV**	**Av $\delta^{18}O$ 2009**	**STDEV**
Austria	50	50	−28.0	±0.8	−1.3	±1.1	−28.2	±0.9	0.3	±1.3
Slovenia	20	22	−28.4	±0.8	1.4	±2.1	−28.0	±1.0	2.0	±2.4
Romania	9	14	−25.7	±0.8	3.5	±0.8	−27.0	±1.0	4.0	±1.7
Montenegro	3	3	−27.4	±0.7	8.0	±0.2	−27.8	±0.2	8.1	±0.2
Argentina	6	5	−27.2	±0.5	2.8	±2.1	−27.2	±0.3	2.0	±0.8

−28.0 ± 0.8‰ and −1.3 ± 1.1‰ and for 2009, −28.2 ± 0.9 ‰ and 0.3 ± 1.2‰ for ^{13}C and ^{18}O, respectively. The Austrian samples from the same year form a cluster with a slightly positive correlation of the investigated parameters. The cluster of the year 2008 shows an about 1.5‰ lower value than the cluster from 2009, but almost no difference in the $\delta^{13}C$ averages. The presented data are the first published combined C- and O-isotope results of wine from Austria.

The Slovenian samples range in $\delta^{13}C$ from −29.5 to −27.1‰ for the year 2008 and between −29.9 and −26.6‰ for the year 2009. Oxygen isotope values vary between −3.0 and +6.7‰ for 2008 and −2.0 and +5.3‰ for 2009 (**Figures 2**, **3A,B**). The 2008 and 2009 average values for $\delta^{13}C$ are −28.4 ± 0.8‰ and −28.0 ± 1.0‰, respectively. For $\delta^{18}O$, the average values are 1.4 ± 2.1 and 2.0 ± 2.4‰ for 2008 and 2009, respectively. The Slovenian sample results of the same year are falling into two clusters, one cluster hosting the "enriched" isotope samples and the other one, the "depleted" samples. The two investigated parameters are clearly positively correlated and separated from each other by a 1.5–2‰ gap in the oxygen isotope values. In addition, for the Slovenian samples, the vintage 2008 have lower $\delta^{18}O$ values compared with 2009, and also, the $\delta^{13}C$ is lower. The presented values are in good agreement with the data (wine samples from 1996–1998) published by Ogrinc et al. (2001). In the mentioned publication, the two clusters described above have also been identified ("enriched" cluster: coastal area, "depleted" cluster: Sava and Drava areas), however, for the investigated years, these clusters overlap. This fact might be due to the annual weather variations, but also, the larger sample set investigated by Ogrinc et al. (2001) might have influenced the outcome to some extent. For the vintages 2008 and 2009, it seems to be a complete separation as two separate clusters are clearly present, however, this is a speculation as the exact geographic origin of the wine samples is not revealed.

The Romanian samples vary in $\delta^{13}C$ from −27.2 to −24.4 and −28.7 to −24.8‰ for the years 2008 and 2009, respectively. In $\delta^{18}O$, the Romanian samples range between 1.4 and 4.2 and 1.3 and 6.7‰ for the years 2008 and 2009, respectively (**Figures 2**, **3A,B**). Average $\delta^{13}C$ values are −25.7 ± 0.8 and −27.0 ± 1.0‰ for 2008 and 2009, and average $\delta^{18}O$ values are 3.5 ± 0.8 and 4.0 ± 1.7‰ for 2008 and 2009, respectively. In addition, for the Romanian samples, a significant difference in the average values is present for the 2008 and 2009 clusters. The data fit very well with the published data by Magdas et al. (2012).

The wine samples from Montenegro have $\delta^{13}C$ values between −28.1 and −26.8‰ and −28.0 and −27.5‰ for the years 2008 and 2009, respectively. The $\delta^{18}O$ values are within 7.4 and 8.7 and 7.5 and 8.7‰ for the years 2008 and 2009, respectively (**Figures 2**, **3A,B**). The sample average $\delta^{18}O$ is 8.0 and 8.1‰ and the sample $\delta^{13}C$ are −27.4 ± 0.7 and −27.8 ± 0.2‰ for the years 2008 and 2009, respectively. The values cluster at more or less the same place for the two investigated years. The Montenegrin samples show a very homogenous pattern. These are, to our knowledge, the first published data for the Montenegrin wine. They are in agreement with the isotope results of the wine from the coastal area of Croatia (Christoph et al., 2004).

The Argentinean samples range in $\delta^{13}C$ from −28.1 to −26.7‰ and −27.6 to −26.8‰ for the years 2008 and 2009, respectively. In $\delta^{18}O$, the values are between 0.8 and 6.5 and 1.0 and 3.2‰ for 2008 and 2009, respectively (**Figures 2**, **3A,B**). Average $\delta^{18}O$ values are 2.8 ± 2.1 and 2.0 ± 0.8‰ and average $\delta^{13}C$ values are 27.2 ± 0.5 and 27.2 ± 0.3‰ for 2008 and 2009, respectively. The samples from both years cluster more or less at the same range (between −27.5 and −26.5‰ for carbon and 0.8 and 3.2‰ for oxygen), with the exception of one sample from 2008 possessing an entirely different $\delta^{18}O$ value coming from a different wine-growing area in Argentina. Grubbs outlier test confirmed the value as an outlier (Z: 2.51775358743, Significant outlier. $P < 0.05$, Critical value of Z: 2.35472945013). If this sample is ignored, the average values are almost identical for both years. Nevertheless, the small sample set has to be considered with respect to the completeness, as obviously, an increase in the samples might significantly influence the result. The outlier sample was produced in another wine-growing region (Neuqen) of Argentina than the other samples, which came from Mendoza, the largest Argentinean wine production area. If we disregard that sample, the isotope results are more or less identical for both years. The isotope values are well in agreement with the literature data (Di Paola-Naranjo et al., 2012, Christoph et al., 2015). The single "outlier" datum is a sample from the southernmost Argentinean wine-growing region (Neuqen area) possessing a significantly different environmental conditions than the other Argentinean wine producing areas.

Statistical evaluation by LDA shows differing results for the vintages 2008 and 2009. Whereas for 2008, an average correct classification of around 80% (84.1% initial classification, 79.5% cross-validation) is achieved for the samples from the

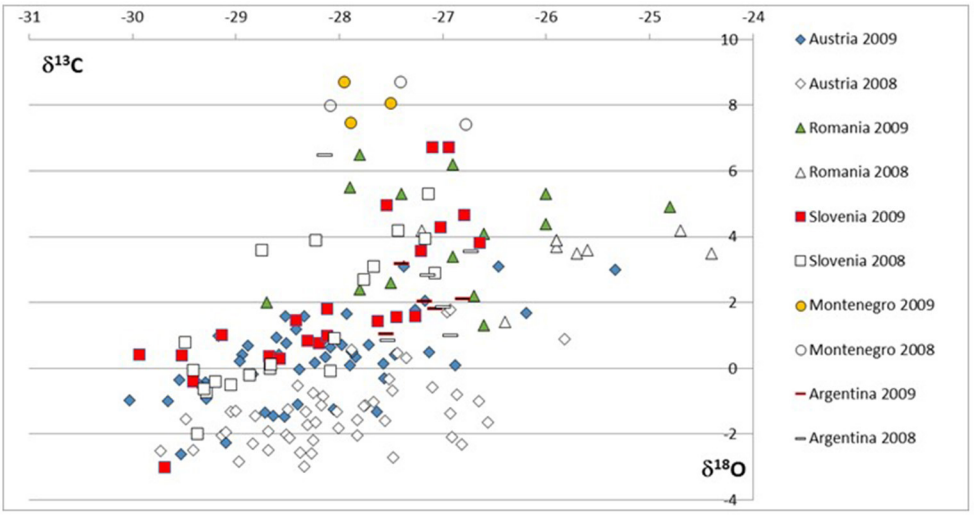

FIGURE 2 | $\delta^{13}C$ vs. $\delta^{18}O$ of the investigated wine samples (from Austria, Slovenia, Romania, Montenegro, and Argentina) with linear trend lines for both vintages (2008 and 2009).

five investigated countries (**Figure 4**), the correct classification decreases to around 55% (56.4% initial classification, 54.3% cross-validation) in 2009 (**Figure 5**). Taking into account only the three bigger sample sets (with sample numbers of nine and above for each vintage) from Austria, Romania, and Slovenia increases the results to almost 90% (89.9% initial classification, 88.6% cross-validation) for 2008 (**Figure 6**) but to only above 60% (62.8% initial classification, 62.8% cross-validation) for 2009 (**Figure 7**). Combining both vintages of the investigated samples from all five countries result in ca. Sixty-five percentage correct classifications (67.0% initial classification, 65.4% cross-validation; **Figure 8**). For details, see **Supplementary Material 3**. OPLS-DA demonstrated a significance in the differentiation between the samples of the respective countries [$P < 0.0001$ for all countries including both vintages (**Supplementary Material 2**)] and also a significance in the differentiation of both vintages ($p < 0.00348$), due to the differences in the oxygen isotope values. Significantly lower $\delta^{18}O$ values were observed in 2008 compared to 2009.

DISCUSSION

Austria

Despite the rather small size of the entire wine-growing area in Austria, the wine isotope values are very heterogeneous as the wine-producing regions in Austria are situated north, east, and south of the Eastern Alps, which have a strong influence on the regional weather. Nevertheless, the general weather trend (as a main influencing factor for the C- and O-isotope ratios) influences the vintages and resulted in significantly higher $\delta^{18}O$ values for the year 2009 than 2008, although there is some overlap. Still, it is to be expected that most Austrian wines of these years can be assigned to the respective year (most likely due to the oxygen isotope value) as demonstrated by the statistical evaluation, where almost 70% of the vintages were correctly classified (**Supplementary Material 3**). The carbon isotope values, on the other hand, are very similar and do only show very moderate differences, despite the variation in $\delta^{18}O$. The most likely explanation is a larger amount of precipitation in September 2008 (or generally within the last 4 weeks prior to harvest; Christoph et al., 2003) resulting in lower $\delta^{18}O$ ratios for the 2008 vintage.

Slovenia

The results of the Slovenian wines fall clearly into two clusters, as described above. Many of the 2009 vintage samples have higher $\delta^{18}O$ values than the 2008 vintage, although the shift toward higher values in 2009 is lower in its magnitude than for the Austrian samples of the same vintage. This indicates that the weather condition responsible for the variation of the oxygen isotopes in Austrian wines also influenced the Slovenian vintage (**Figures 2**, **3A,B**), but to a slightly lower extent than the Austrian samples (see **Table 1**). However, as the trend lines of the two vintages are very similar in Slovenia, it seems that in addition to the variation in $\delta^{18}O$, some positively correlated trend also occurs in the $\delta^{13}C$ values of the Slovenian wine samples, possibly indicating a slightly higher draft stress condition, resulting in higher $\delta^{13}C$ ratios for the 2009 vintage as well. By initial classification and cross-validation, ca. Eighty percentage of the Slovenian samples were correctly assigned to the respective vintages (**Supplementary Material 3**).

Romania

In addition, the $\delta^{18}O$ values of the Romanian samples show the influence of the general weather condition in 2008 and 2009 with lower values in 2008 and higher in 2009, with a similar shift between the 2 years as for the Slovenian wine samples. However, in 2009, the samples show a larger spread of values, indicating

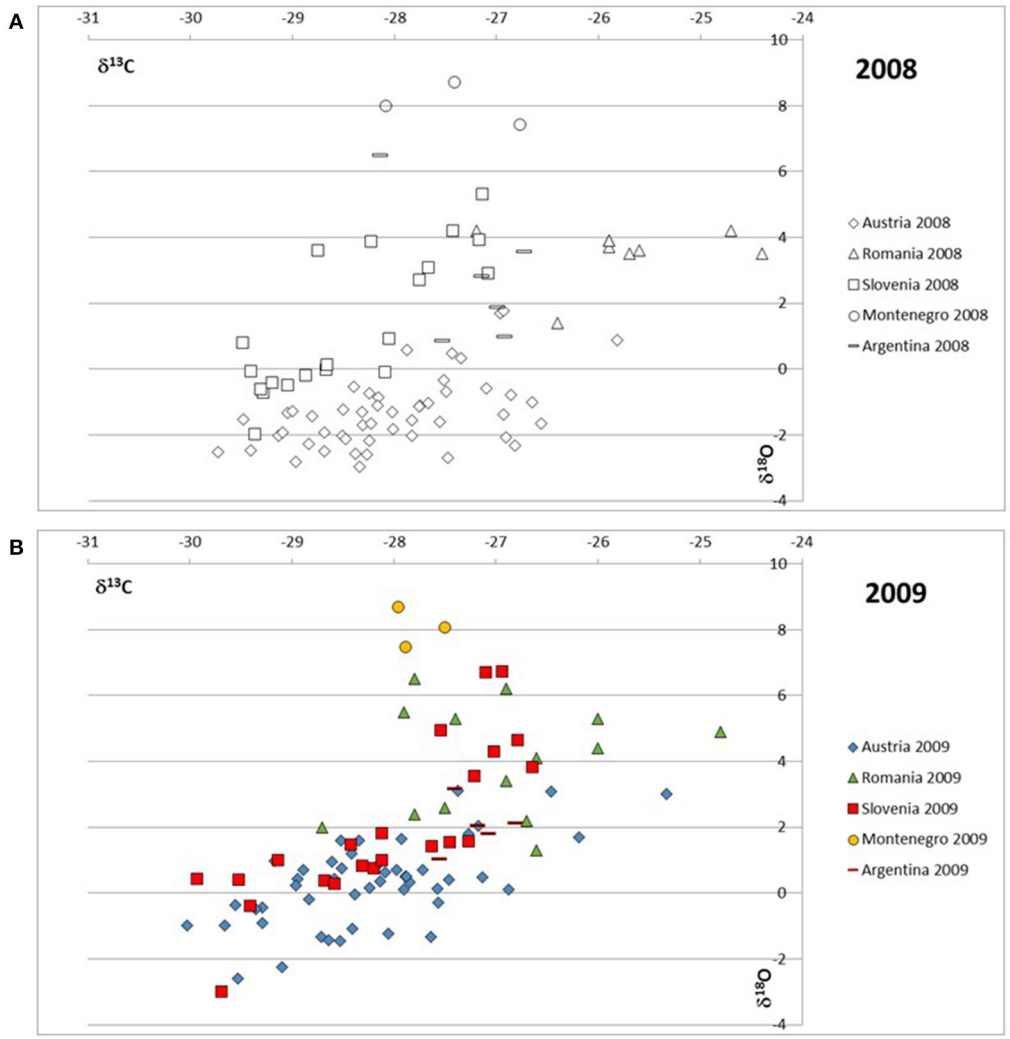

FIGURE 3 | (A) $\delta^{13}C$ and $\delta^{18}O$ values of vintage 2008. For this year, an almost complete discrimination is achieved for the samples of the investigated countries (except for Argentina). **(B)** $\delta^{13}C$ and $\delta^{18}O$ values of vintage 2009. In 2009, there is a trend to more extreme values (high and low) for the samples from Austria, Slovenia, and Romania with respect to 2008. Due to this broadening, the point clouds overlap.

a differentiation in the regional weather conditions in Romania. This assumption is also supported by the $\delta^{13}C$ values that have a smaller range in 2008 than 2009.

Montenegro

The Montenegrin wine samples show very similar values for both years, indicating stable and homogenous conditions during the investigated years and suggest that Montenegro was not influenced by the general weather conditions of central and eastern Europe. This lead to having identical trends in the wine isotopes in the three countries discussed above, or that such weather conditions did not significantly influence the vine and wine isotope pattern due to stable agricultural conditions as water supply and isotopic composition of water available to the vines. It has to be taken into account that the samples are just from one producer and, thus, do not reflect the variation among the produced Montenegrin wines. On the other hand, however, this producer accounts for about 50% of the Montenegrin wine production and the samples demonstrate homogenous conditions resulting in homogenous isotope values for the investigated vintages.

Argentina

The Argentinean wine samples generally show similar values for both investigated vintages, with one outlier coming from a separate wine area (Neuqen). The $\delta^{18}O$ values are low if we take into account the warm and arid climates of most wine-growing regions in the western Central and Northern provinces of Argentina. This can be explained by the irrigation with river water from high mountains, which might lead to a significant lowering of the oxygen isotope values (Gómez-Alonso and García-Romero, 2010), as the river water coming from the High

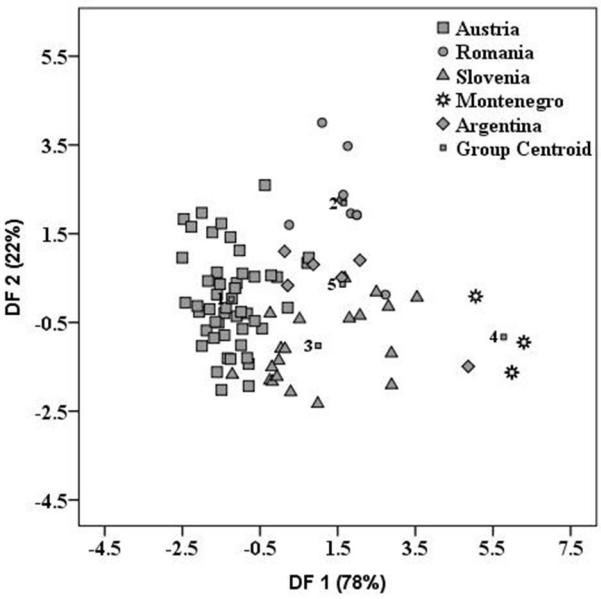

FIGURE 4 | LDA evaluation of the investigated samples from Austria, Slovenia, Romania, Montenegro, and Argentina for 2008.

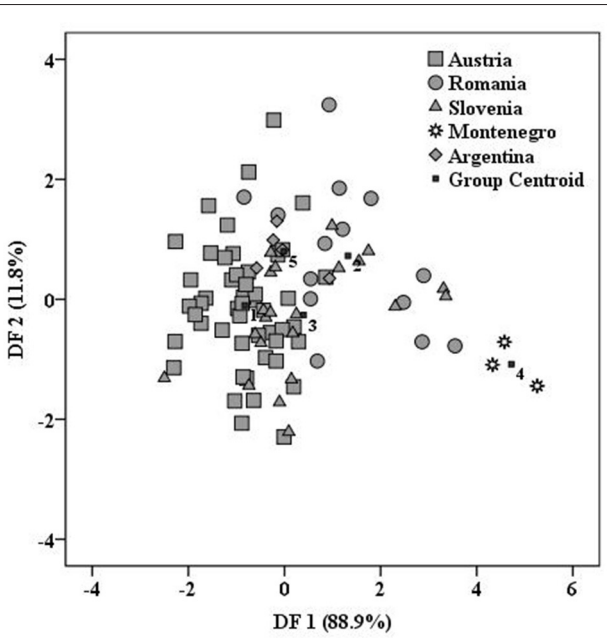

FIGURE 5 | LDA evaluation of the investigated samples from Austria, Slovenia, Romania, Montenegro, and Argentina for 2009.

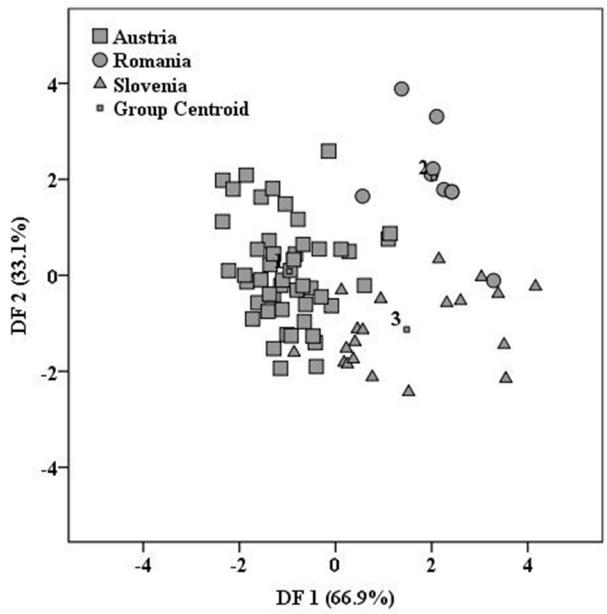

FIGURE 6 | LDA evaluation of the investigated samples from Austria, Slovenia, and Romania for 2008.

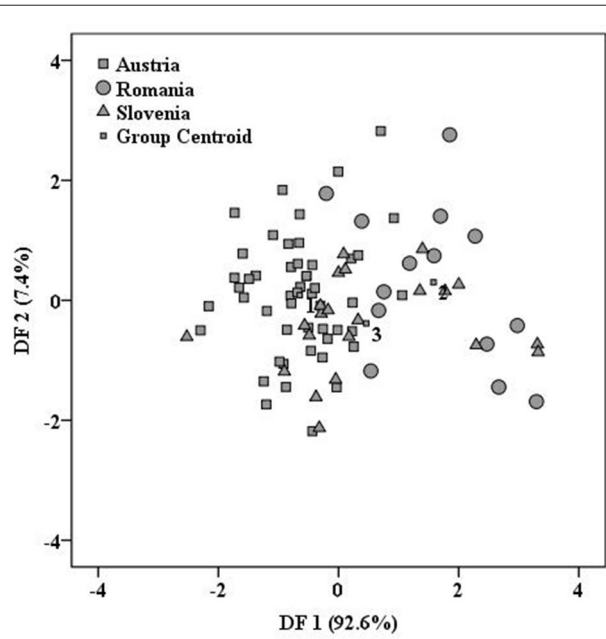

FIGURE 7 | LDA evaluation of the investigated samples from Austria, Slovenia, and Romania for 2009.

Andes mountain range has low isotope values, and the general climate situation in Central and Northern Argentina. The sample from the Neuqen area exhibiting a significantly higher $\delta^{18}O$ value indicates (but this needs to be verified with more samples of different vintages) the use of local water in this area enriched in oxygen isotopes.

Regarding the differentiation of geographic origin for the 2008 vintage, the investigated samples from the different regions only slightly overlap (**Figures 2**, **3A,B**). This indicates that, given certain environmental and weather conditions, there is a high potential to differentiate geographic origin by measuring $\delta^{13}C$

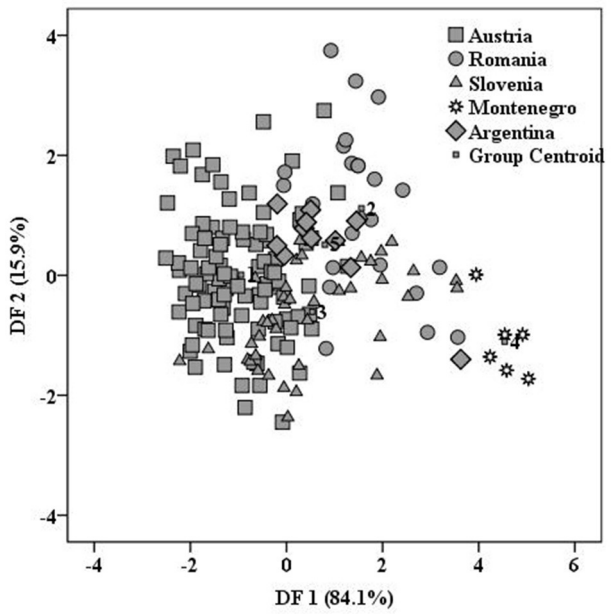

FIGURE 8 | LDA evaluation of the investigated samples from Austria, Slovenia, Romania, Montenegro, and Argentina for both investigated vintages 2008 and 2009.

and $\delta^{18}O$ with respect to the wine from Austria, Slovenia, Romania, Montenegro, and Argentina, even if no further indication of regional origin or exact locality is known. This is quite remarkable, as southern Austrian and northeastern Slovenian wine-growing areas are adjacent and a significant overlap of the data should be expected.

This overlap, though, occurs for the Austrian and Slovenian (supposedly the Drava and Sava cluster) samples of the 2009 vintage, enabling an incomplete differentiation of this vintage from these two areas only (**Figures 3A,B**). Furthermore, the year 2009 seems to have caused stronger regional differences within Austria, Slovenia and Romania, as the samples of these countries show a larger spread of values with respect to 2008. Still, a differentiation of geographical origin can be achieved, if additional information of origin (region, locality, winery) will be given for the investigated samples.

Due to its southern and coastal position, Montenegrin wine always (with respect to the investigated vintages and samples) can be differentiated from wine of the other countries investigated, but literature data indicates an overlap with wine from adjacent regions in Croatia (Christoph et al., 2004).

Argentinean wine samples exhibit quite homogenous values, but the "outlier sample" from a separate wine-growing area indicates that a much larger variations in the Argentinean wine isotope values can be expected, if a more comprehensive study includes samples from every wine-growing area of this large country.

The Argentinean samples overlap with the samples of the other countries investigated, except for the samples from Montenegro. This demonstrates that despite a large distance between different origins, wine samples from these areas can still sometimes have similar isotope patterns. However, the more parameters are investigated, the less likely is such a similarity (Di Paola-Naranjo et al., 2012). Identified trends and influences need to be verified by further vintages, to unequivocally classify them as constantly existing besides the annual weather variations.

As the general pattern of lower $\delta^{18}O$-values in 2008 and higher in 2009 occurs for the Austrian, Slovenian, and Romanian samples, we assume the existence and influence of a "general central and eastern European weather situation for the investigated relevant period resulting in these features. The Montenegrin samples do not show any influence of this phenomenon, potentially indicating that it is absent in Southern Montenegro. However, it has to be taken into account that the Montenegrin samples only represent one winery (although one of the biggest in Europe) and might not be representative for all of Montenegro.

CONCLUSIONS

The two investigated vintages from Austria, Slovenia, and Romania show significant congruent variations in the isotope pattern with lower $\delta^{18}O$ values in the 2008 vintage (Figure 1B). This indicates the existence and influence of a "general central and eastern European weather situation." Dominantly, the oxygen isotope value is more relevant for geographic differentiation. The Romanian samples also show a significantly larger variation in $\delta^{13}C$ in 2009 indicating stronger variable specific climatic conditions in the different wine-growing areas in that vintage. Montenegrin wine samples have similar values for both investigated years and are, thus, not influenced by the proposed weather situation. The Montenegrin wine can easily be distinguished by its enriched isotope values. The Argentinean wine samples significantly overlap with other investigated samples, evidencing the possibility of similar patterns despite the large distances between the compared regions. The investigation of further parameters as $(D/H)_I$ and $(D/H)_{II}$ by SNIF-NMR, Sr-isotopes, and trace element pattern can reduce these similarities. For 2008, the investigated samples from the different Central and Southern European countries show almost no overlap and can, thus, be nicely discriminated. For 2009, the samples have a larger overlap. The isotope variations between 2008 and 2009 are significant and enable identification of vintages in most cases for the investigated wines from Austria and Romania.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**, further inquiries can be directed to the corresponding author/s.

AUTHOR CONTRIBUTIONS

MH designed the project, analyzed the Austrian, Montenegrin, and Argentinian wine samples, and wrote the manuscript. NO analyzed the Slovenian samples and contributed to the manuscript. AM analyzed the Romanian samples, performed the statistical evaluation, and contributed to the manuscript. DW provided the Argentinean wine samples and contributed to the manuscript. SS, VM, and AM provided the Montenegrin wine samples and contributed to the manuscript. FČ analyzed Slovenian wine samples. RE organized vinification of Austrian grape samples. SW and WP analyzed Austrian wine samples. All authors contributed to the article and approved the submitted version.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fsufs.2021.638941/full#supplementary-material

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.