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$^{87}Sr/^{86}Sr$ isotopic composition of wine: uses and limitations

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ABSTRACT

Isotopic $^{87}Sr/^{86}Sr$ fingerprints are increasingly applied to identify wine provenance, i.e., the geology of the source region. We present $^{87}Sr/^{86}Sr$ analyses of 27 samples of commercial wines from Central Europe, Southern Moravia, Czech Republic, done by thermal ionisation mass spectrometry (TIMS), and compare these data with isotopic compositions of bulk soil, bio-available soil fraction groundwater and precipitation. $^{87}Sr/^{86}Sr$ ratios of wines vary from 0.708 to 0.710 and correspond to those in groundwater and leachates from soils. We have divided the rocks (soils) into three groups according to their $^{87}Sr/^{86}Sr$ isotopic composition: (i) below that of precipitation (0.700–0.709); (ii) similar to precipitation (0.709–0.710); (iii) above that of open-area precipitation (> 0.710). We describe how bedrock from each group can affect the isotopic composition of juice, must (i.e., juice + pomace) or wine. Wines from South Moravia have very similar $^{87}Sr/^{86}Sr$, independent of cultivars and vintage. We conclude that the groundwater sample could be used as a tracer for bio-available Sr isotopic composition. We recommend analysing the rainwater and groundwater or/and leachate from soil together with wine samples and discuss the limitations of $^{87}Sr/^{86}Sr$ use in wine as a geological tracer. We offer a simple predictive model that connects the wine-leachate-bulk rock/soil Sr isotopic composition. To interpret isotopic data correctly, careful assessment of geochemical context, i.e., isotope systematics in bedrock and soil, is necessary.

KEYWORDS: Central Europe, Moravia, loess, soil, Sr isotopes, wine

INTRODUCTION

Verifying the geographical origin of agricultural products is becoming increasingly important, especially due to regional certification. Inorganic chemical analysis increasingly plays a key role in these purposes. While determining all processes influencing element concentrations in a given product is often complicated, the system is usually treated as a "black-box", and multivariate statistical analysis is used. On the other hand, isotopic ratios of heavy elements have the potential to distinguish provenance that may have been masked in examining elemental concentrations alone. Strontium is particularly well suited for this type of investigation. It is sufficiently abundant in rocks and plants, and its isotopic composition can provide a fingerprint of the nutrient sources. Strontium isotopes have been proposed and/or successfully used as a source area tracer for several foods, including wine (first suggested by Horn *et al*., 1993, juice (Rummel *et al*., 2010), or milk (Crittenden *et al*., 2007).

Since the early study by Horn *et al*. (1993), numerous studies have examined the applicability of radiogenic isotope ratios ($^{87}Sr/^{86}Sr$) of strontium to wine origin, as described by (Boari *et al*., 2008; Epova *et al*., 2019; Moreira *et al*., 2017; Tescione *et al*., 2018; Tescione *et al*., 2020). The chemical and biogeochemical behaviour of Sr is similar to that of Ca; however, in contrast to stable Ca isotopes ($^{44}Ca/^{40}Ca$), the $^{87}Sr/^{86}Sr$ isotope ratio varies considerably in natural environments, and this variability is geologically (i.e., geographically) systematic. Many authors suggest that radiogenic Sr isotopes could serve as an excellent fingerprinting tool for tracing the geographical origin of wine (Almeida and Vasconcelos, 2004; Horn *et al*., 1993; Marchionni *et al*., 2013; Petrini *et al*., 2015; Tescione *et al*., 2020), but the details of the Sr isotope connection between rock, soil and wine are not straightforward.

The final $^{87}Sr/^{86}Sr$ composition of the wine results from isotopic mixing of several components: soil bioavailable Sr originating from bedrock, Sr dissolved in groundwater, atmospheric input (rainwater and sea aerosols), and Sr from the anthropogenic additives such as agrichemicals. The Sr isotope ratio of products that combine several regionally different raw materials with different isotope compositions would be a weighted average, considering variability in the Sr concentration of each component. Reverse identification of two or more end members of such a mixture in a one-dimensional isotope system (such as $^{87}Sr/^{86}Sr$) is impossible (Albaréde, 1995). Still, the Sr isotopic composition of rainwater and seawater is known, as are the overall compositions of various rocks.

The Sr isotopic fingerprint is regionally (geologically) characteristic but not unique. Therefore, it cannot be used to identify the exact place of origin, but it can confirm (or reject) potential candidate sites and help identify potential candidate sites.

This study aimed: (1) To examine the Sr radiogenic isotopic composition of wines from South Moravia, Czech Republic and compare them with the isotopic composition of bulk disturbed soil samples, bio-available Sr, and a groundwater sample from a local well. (2) To present a simple model of $^{87}Sr/^{86}Sr$ isotopic composition of wine formulated from a geological point of view. (3) To assess the limitations of $^{87}Sr/^{86}Sr$ as a tool for determining the geographical origin of wine.

MATERIALS AND METHODS

1. Sample selection

Twenty-six samples of mono-cultivar variety and one blended (vino 9 sample) commercial wines from four wineries were selected for this study (Table 1). Samples were aliquoted from bottled wine. Due to the geographical location of the vineyards, we divided wine samples into four groups according to their location: A, B, C, and D (Figure 1B,C). Wines were produced from 11 different grape varieties grown in Southern Moravia, Mikulov (group A: 9 wines and C: 8 wines), and Velké Pavlovice (group B: 8 wines and D: 2 wines) subregions (Table 1, Figure 1). Sample selection was primarily based on the ability to match vineyard location with commercially produced wine accurately; inter-variety differences were of secondary concern. Loess forms the geological substratum for all samples. The studied vineyards were founded after 1989, and all four wineries use similar methods of wine production.

Group A is the main focus of this study, as sampling of potential isotopic reservoirs in the local environment is the most complete for this region. In 2020, soil samples were taken from the village of Sedlec u Mikulova locality (Group A) at 30, 60, 90, and 200 cm depth intervals directly from the vineyard. A groundwater sample from a 4 m deep well approximately 1.1 km from the soil sampling point was taken for comparison with soil leachates (Table 2, Figure 2). Soil samples were not collected from the other group localities. However, due to the similar bedrock, climatic and hydrological conditions, we infer that other sites' soil conditions and geochemical characteristics are similar to those of Sedlec u Mikulova (Group A).

The shallow vine roots of Group A Rheinriesling cultivar (RR) were observed to a depth of 40 cm, and the deep roots exceeded a depth of 170 cm.

2. Sample preparation

Chemical separations for Sr isotope analyses were conducted in the Ultra-Clean Laboratory at the Czech Geological Survey (CGS) in Prague. The analytical procedure is described in Erban Kochergina *et al*. (2022).

A water sample of approximately one litre was filtered (Chromafil Xtra CA-45/25 cellulose acetate, porosity 0.45 µm), acidified with HNO_3, evaporated at 100 °C, and the residue was dissolved in an H_2O_2–HNO_3 mixture, and re-dissolved in 2 M HNO_3.

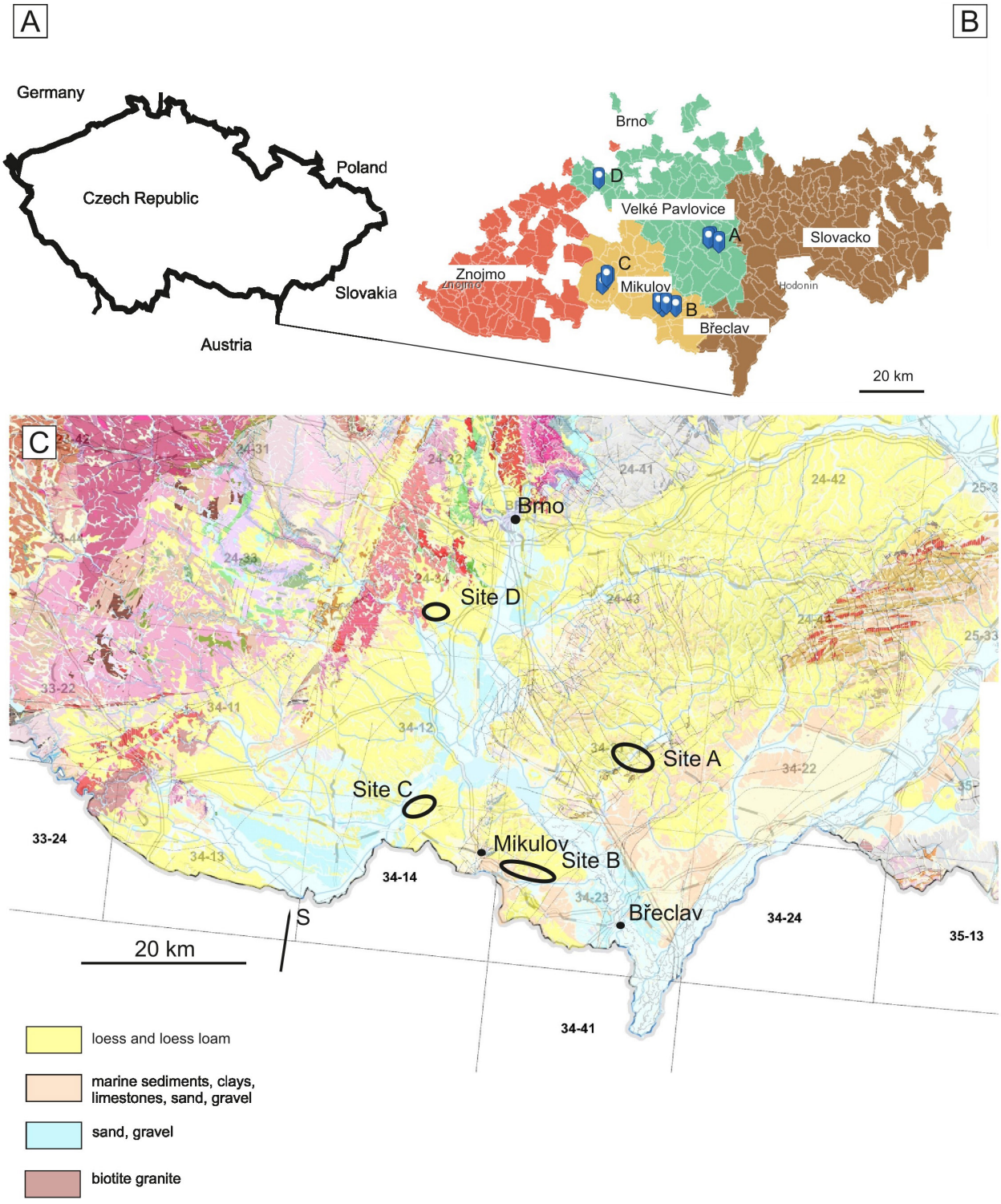


FIGURE 1. A. Location of the studied area in the context of Central Europe. B. Wine region "Morava" (Moravia) with sub-regions (Znojmo, Mikulov, Velké Pavlovice, Slovácko (mapavina.cz/online) with studied localities. C. Geological sketch of the Moravia Region, Czech Republic (https://mapy.geology.cz/geocr50/). All vineyards lie on loess bedrocks.

TABLE 1. Sr isotopic composition of wines from Southern Moravia, Czech Republic.

2 SE: two standard error, * two different bottles of vine.

	Winery	Sample	Locality	Year	Grape variety	Group	$^{87}Sr/^{86}Sr$	2 SE
Mikulov subregion	Ludwig	vino 25	Sedlec	2019	Chardonnay (Ch)	A	0.709165	0.000012
	Ludwig	vino 26	Sedlec	2019	André (An)	A	0.709063	0.000010
	Ludwig	vino 27	Sedlec	2019	Zweigelt (Zw)	A	0.708612	0.000008
	Ludwig	vino 28	Sedlec	2018	Welschriesling (RV)	A	0.709338	0.000010
	Ludwig	vino 11	Sedlec, Stolová hora	2019	Rheinriesling+Aurelius (Fr+Au)	A	0.709200	0.000014
	Vican	vino 2	Pod Mušlovem	2016	Blaufränkisch (Fr)	A	0.709549	0.000014
	Vican	vino 13	Pod Mušlovem	2017	Blaufränkisch (Fr)	A	0.709550	0.000012
	Vican	vino 15	Pod Mušlovem	2018	Blaufränkisch (Fr)	A	0.709493	0.000009
	Vican	vino 16	Pod Mušlovem	2019	Blaufränkisch (Fr)	A	0.709320	0.000012
	Ludwig	vino 8	Janův vrch	2018	Muscat Moravian (MM)	C	0.709324	0.000011
	Ludwig	vino 10	Stará hora	2019	Neuburger (Ng)	C	0.708392	0.000009
	Marcinčák	vino 3	Janův vrch	2017	Rheinriesling (RR)	C	0.709165	0.000014
	Marcinčák	vino 4	Janův vrch	2018	Rheinriesling (RR)	C	0.708952	0.000011
	Marcinčák	vino 5	Janův vrch	2019	Rheinriesling (RR)	C	0.708253	0.000011
	Marcinčák	vino 6	Slunečná	2016	Rheinriesling (RR)	C	0.709097	0.000012
	Marcinčák	vino 7	Slunečná	2017	Rheinriesling (RR)	C	0.709114	0.000011
	Marcinčák	vino 9	Slunečná	2018*	Rheinriesling (RR)	C	0.708443	0.000012
	Marcinčák	vino 12	Slunečná	2018*	Rheinriesling (RR) duplicate	C	0.708431	0.000012
Great Pavlovice subregion	Jedlička	vino 18	Terasy	2019	Rheinriesling (RR)	B	0.709414	0.000027
	Jedlička	vino 19	Terasy	2018	Rheinriesling (RR)	B	0.709600	0.000011
	Jedlička	vino 20	Terasy	2017	Rheinriesling (RR)	B	0.709402	0.000013
	Jedlička	vino 21	Terasy	2016	Rheinriesling (RR)	B	0.709638	0.000013
	Jedlička	vino 22	Veselí	2020	Müller-Thurgau (MT)	B	0.708988	0.000010
	Jedlička	vino 23	Veselí	2019	Müller-Thurgau (MT)	B	0.708960	0.000014
	Jedlička	vino 24	Veselí	2018	Müller-Thurgau (MT)	B	0.709352	0.000012
	Ludwig	vino 17	Kobylí	2019	Cabernet Moravia (CM)	B	0.709523	0.000011
	Ludwig	vino 14	Němčičky	2019	Solaris (Gm)	D	0.709498	0.000010
	Ludwig	vino 1	Němčičky	2020	Solaris (Gm)	D	0.708527	0.000010

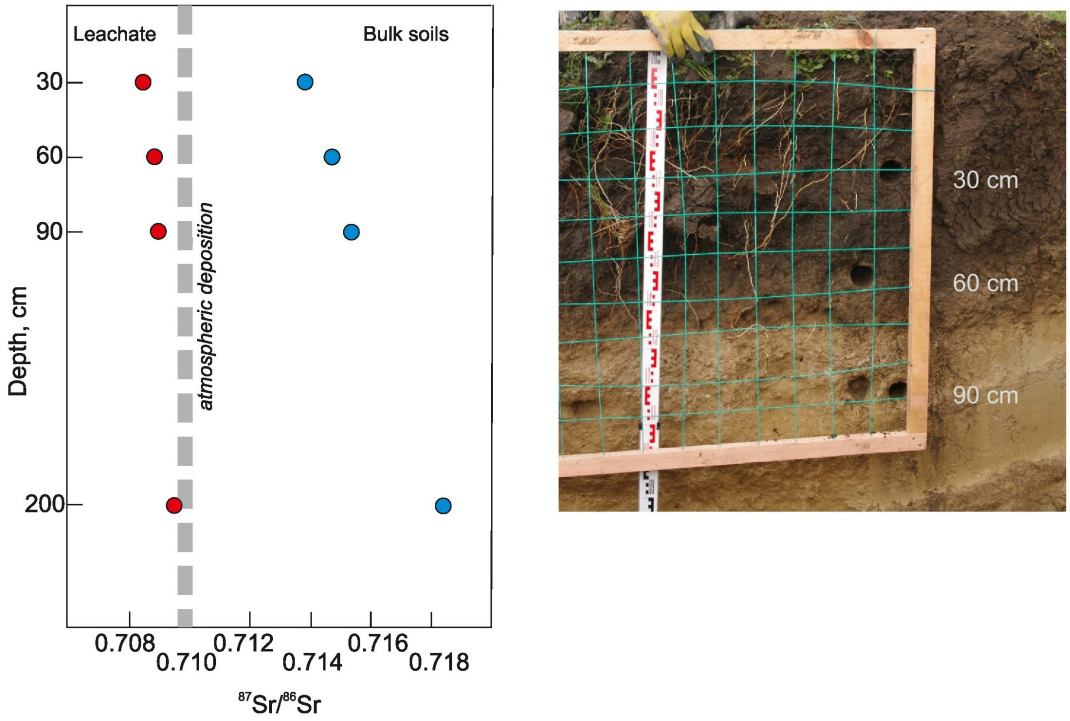


FIGURE 2. Comparison of $^{87}Sr/^{86}Sr$ isotopes in bulk soils and their leachate. Field photographs of the studied soil samples from Sedlec u Mikulova, Southern Moravia.

TABLE 2. $^{87}Sr/^{86}Sr$ composition of leachate soils and bulk soil samples from Sedlec, Mikulov sub-region.

Sample	Depth, cm	Locality	Sample preparation	$^{87}Sr/^{86}Sr \pm SE$
VN30	30	VN	leachate	0.708444 ± 14
VN31	30		bulk sample	0.713801 ± 14
VN60	60	VN	leachate	0.708821 ± 18
VN60	60		bulk sample	0.714684 ± 12
VN90	90	VN	leachate	0.708958 ± 14
VN90	90		bulk sample	0.715323 ± 12
VN200	200	VN	leachate	0.709460 ± 10
VN200	200		bulk sample	0.718377 ± 16
groundwater from the local well				0.709219 ± 14
open-area precipitation from Erban Kochergina *et al.* (2021)				0.7097 – 0.7098

Soil samples were air-dried in the laboratory until their weight no longer decreased. Dry samples were sieved with a clean 2 mm stainless steel sieve, homogenised, and milled to analytical fineness using an agate mortar (for details, see Kochergina *et al*., 2017). This fraction was used for chemical analyses (Erban Kochergina *et al*., 2022).

To obtain the bio-available soil phase, we used only the first extraction step of the sequential extraction procedure described by Sutherland and Tack (2002), which was then analysed for Sr isotopes. Soil samples (1.00 ± 0.01 g) were leached in 40 mL of 0.11 M acetic acid for 16 h. For bulk $^{87}Sr/^{86}Sr$ analyses, approximately 100 mg of powdered soil samples were dissolved (Erban Kochergina *et al*., 2022).

3. Strontium isotopes

Strontium fractions were isolated from the bulk matrix using Sr-spec resin (Triskem Intl.), and isotopic measurements were performed on a Triton Plus Thermal-Ionization Mass Spectrometer (TIMS) manufactured by Thermo Scientific and housed at CGS, using a single Ta filament assembly. The $^{87}Sr/^{86}Sr$ ratios were corrected for mass fractionation, assuming $^{86}Sr/^{88}Sr = 0.1194$. The external reproducibility of Sr is given by repeated analyses of the NIST SRM 987 reference material, $^{87}Sr/^{86}Sr = 0.710257 \pm 0.000023$ (2σ, n = 94), identical to the values presented by Marchionni *et al* (2016), $^{87}Sr/^{86}Sr = 0.710251 \pm 0.000010$ (2σ, n = 20). The results are also in agreement with published data in GeoReM (Jochum and Nohl, 2008), a Max-Planck Institute online database (http://georem.mpch-mainz.gwdg.de), where the NIST SRM 987 measured by TIMS show $^{87}Sr/^{86}Sr = 0.710224 \pm 0.000479$ (2σ, n = 1703). The soil NIST 2709a reference material yielded an $^{87}Sr/^{86}Sr$ value of 0.708179 ± 0.000032 (2σ, n = 23), leached sample of soil NIST 2709a reference material yielded an $^{87}Sr/^{86}Sr$ value of 0.707906 ± 0.000010 (2σ, n = 4). Replicate analysis of wine from Slunecna (2018) from two different commercial bottles yielded similar $^{87}Sr/^{86}Sr$ (Table 1, vino 9 and vino 12 samples), confirming the method's reliability.

RESULTS

We determined strontium isotopes in 27 wine samples (Table 1, Figure 3). The $^{87}Sr/^{86}Sr$ ratios for the groups are as follows: Group A wines 0.7086–0.7096 (mean 0.7093), Group B wines 0.7090 – 0.7093 (mean 0.7094), Group C wines 0.7083–0.7093 (mean 0.7088), Group D wines 0.7085, and 0.7094.

We analysed 10 samples of Rheinriesling (RR) cultivar from three localities: Januv vrch (3 samples from 2017, 2018, 2019), Slunecna (3 samples from 2016, 2017, 2018) and Terasy (4 samples from 2016, 2017, 2018, 2019). These samples show differences in Sr isotopic composition according to vintage. The most heterogeneous RR samples are from Januv vrch with a maximum $^{87}Sr/^{86}Sr \sim 0.7092$ in 2017, $^{87}Sr/^{86}Sr \sim 0.7090$ in 2018, and a minimum $^{87}Sr/^{86}Sr \sim 0.7083$ in 2019; the relative standard deviation (RSD) is 549 ppm. A similar trend, where older samples have more radiogenic composition, was observed in samples from Slunecna. Samples from 2016 and 2017 are almost identical, with $^{87}Sr/^{86}Sr \sim 0.7091$, and a sample from 2018 has $^{87}Sr/^{86}Sr \sim 0.7084$. The RSD for this locality among sampled vintages is 441 ppm. Samples of RR from Terasy are less heterogeneous (0.7094–0.7096), the differences in $^{87}Sr/^{86}Sr$ are in the fourth decimal place, and the RSD for four samples is 150 ppm.

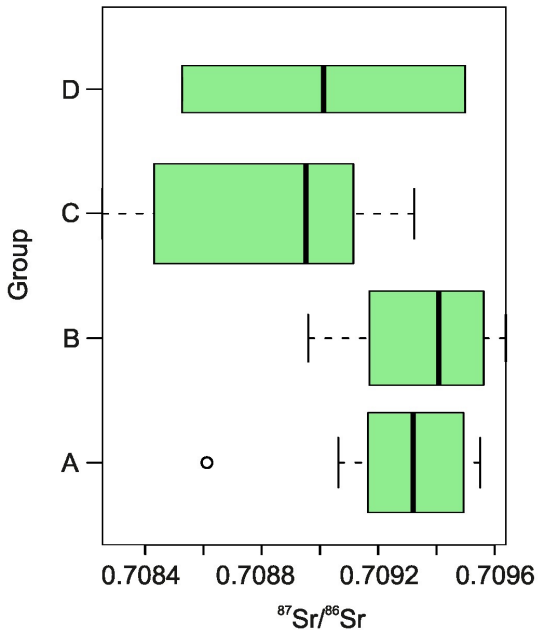


FIGURE 3. Strip boxplot (Janoušek *et al.*, 2016) showing a compilation of $^{87}Sr/^{86}Sr$ values for selected wine samples from different groups, as defined in the Material and methods section (Figure 1B).

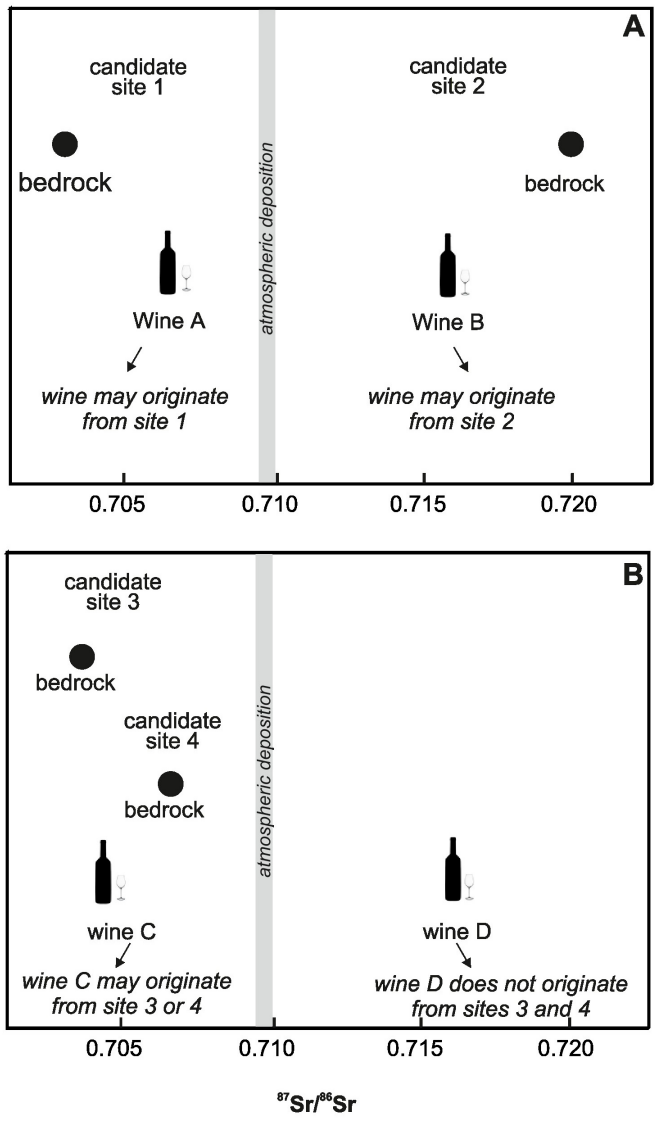


FIGURE 4. Wine origin derived from the $^{87}Sr/^{86}Sr$ systematics in bedrock and rainwater, a schematic illustration.

The model considers simple scenarios, whereby different Sr isotope signatures of the bedrock and overlapping ranges of wine $^{87}Sr/^{86}Sr$ ratios due to partial uptake of atmospheric Sr (adopted from Erban Kochergina *et al.*, 2021). This model can also be used to determine the origin of the wine. Knowing the composition of the wine and the bedrock candidates, we can determine the putative origin of the wine: (i) if the Sr isotopic signatures of bedrock/bulk soil at two candidate sites plot in opposite directions relative to the $^{87}Sr/^{86}Sr$ ratio of rainfall (~ 0.710), the Sr isotope analysis of the plant (wine) sample shows the probable place of origin (Figure 4A); (ii) if the $^{87}Sr/^{86}Sr$ ratio of the plant (wine) sample is more/less radiogenic than local rainfall, compared to the two candidate sites, it is likely that neither of the candidate sites was the place of the plant (wine) origin (Figure 4B); (iii) if the $^{87}Sr/^{86}Sr$ ratios of wines and two candidate sites are similar, there is no way to indicate the likely place of origin (Figure 4B).

A sample of the Blaufränkisch (Fr) cultivar from 2019 from Pod Muslovem also shows less radiogenic Sr isotopic composition (~ 0.7093) than older samples from 2016 to 2018, with $^{87}Sr/^{86}Sr$ ~ 0.7095. The RSD for four samples is 133 ppm. We do not observe any systematic variations among the grape varieties (Table 1).

Strontium isotopes for bulk soil samples and their leachates, together with data for groundwater and rainwater, are presented in Table 2. The $^{87}Sr/^{86}Sr$ ratio of the bulk soil samples increases with depth, from 0.7138 at 30 cm to 0.7184 at 200 cm. The leached soil samples have lower isotopic ratios. However, they mirror the bulk trend of $^{87}Sr/^{86}Sr$ in the soil profile: the isotope ratio of the samples increases with the depth, from 0.7084 to 0.7095.

The sample of the groundwater from the well shows $^{87}Sr/^{86}Sr$ ~ 0.7092, almost identical to the average of the $^{87}Sr/^{86}Sr$ of the bio-available fraction and the average of rainwater in Central Europe (Erban Kochergina *et al.*, 2021; Novák *et al.*, 2023; Novak *et al.*, 2020a; Novak *et al.*, 2020b).

DISCUSSION

1. Geology of the Morava viticulture region

National and EU legislation codifies wine production areas in the Czech Republic. Accordingly, there exist two regions, "Morava" (Moravia) with four sub-regions (Znojmo, Mikulov, Velké Pavlovice, Slovácko, Figure 1) and "Čechy" (Bohemia) with two sub-regions (Mělník, Litoměřice).

The Morava region, with its four sub-regions, is a continuous area in Southern Moravia, roughly around and between the cities of Brno, Uherské Hradiště, Hodonín, Břeclav, and Znojmo. The region straddles the NE–SW trending contact of two large geological units, the Bohemian Massif in the west and the Western Carpathians in the east. This main boundary also controls the orientation of almost all geological units in the area, which, in map view, forms parallel bands.

Sedimentary rocks such as loess deposits, siliciclastic deposits, Pliocene marine and continental sediments of the Vienna Basin, mudstones, sandstones, and carbonates predominate in the region. The NE part of Znojmo and Velké Pavlovice sub-regions is formed by Cadomian igneous (granites) and various Variscan metamorphosed units (orthogneiss), partly covered by Upper Paleozoic sediments (Šimíček *et al*., 2021). Loess and its derivatives cover about 20 % of the Moravian surface and are an important part of the European loess belt (Šimíček *et al*., 2021).

The studied vineyards are located on loess deposits (Figure 1C), which are more than 20 m thick (Šimíček *et al*., 2021; Waroszewski *et al*., 2021). Source regions for Pleistocene aeolian loess deposits of these sediments are expected to be in the Variscan part of the Bohemian Massif (Lisá and Uher, 2006).

2. The "terroir "concept and modern geochemistry

Schaller (2017) described the modern concept of terroir in agriculture and concluded that many variables contribute to the landscape imprint on a particular product. Ideally, at least some of those variables would be a set of quantifiable parameters that could be unambiguously tested. Wine-growing areas are characterised by natural environmental conditions, such as climate, soil conditions, and the human factor, which is decisive in selecting variety and agrotechnical practices. The "soil-plant-atmosphere" connection significantly determines the character and results of a certain vineyard (Santos *et al*., 2020).

Modern geochemistry could provide a powerful toolkit for wine "terroir" specification. Trace element composition of wines (Kment *et al*., 2005), such as Sr (Epova *et al*., 2019), could be characteristic of different viticulture areas. However, Sr isotopic composition is only effective in complex geological investigations. The composition of the local rainwater (Figure 6) is an essential parameter contributing to the overall isotopic characteristics of the terroir. Analysis of a groundwater sample gives an overview of the isotopic composition of the available Sr and simplifies the investigation with the complicated geological structure of the bedrock. Information on $^{87}Sr/^{86}Sr$ of bio-available Sr in the local soil and $^{87}Sr/^{86}Sr$ composition of wines (or juices or musts) of different vintages could mark the characteristic limit values of $^{87}Sr/^{86}Sr$ for a specific locality. Together, these parameters allow identification of wine's geographical origin and authenticity.

Santos *et al*. (2020) assume that all factors can influence wine quality and composition. Therefore, future wine studies could focus on these factors and their impact on $^{87}Sr/^{86}Sr$. Despite numerous publications about wine's $^{87}Sr/^{86}Sr$ composition, there is not yet sufficient evidence to connect the wine-soil-bedrock signatures. Unfortunately, a large proportion of the previously measured Sr isotopic dataset was measured on instruments (single collector ICP-MS, even optical emission spectrometers ICP-OES) that are not specifically intended for precise isotopic ratio measurements. For the most part, those instruments lack the mass resolution required to resolve neighbouring peaks and make it difficult to collect the entire peak in the detector at once (i.e., have a "flat-topped" peak). Therefore, these data are largely unusable, given the relatively small differences in isotopic signatures of the geochemical reservoirs involved. Consequently, we agree with Epova *et al*. (2019) and Rosner (2010) that only isotopic analyses at TIMS and MC-ICP-MS quality levels are comparable to geological studies, with precision and accuracy. Only these data should be used for modelling rock-soil-water-wine processes and determining the origin of wines.

3. Sr isotopic composition of bedrock in Southern Moravia: marine sediments, flysch sediments, granites

The geology of the studied Moravian region is very heterogeneous. In addition to loess sediments, other rocks with contrasting $^{87}Sr/^{86}Sr$ composition also occur in South Moravia, such as marine sediments, granitic rocks, and flysch. The whole rock Sr ratios of the marine sediments can thus be estimated from the published global marine Sr evolution curve for the Phanerozoic (McArthur *et al*., 2012). In general, seawater $^{87}Sr/^{86}Sr$ was < 0.709 during the Mesozoic and Cenozoic (McArthur *et al*., 2012), similar to or lower than local rainwater $^{87}Sr/^{86}Sr = 0.7097$ (Novak *et al*., 2020a; Novak *et al*., 2020b).

Cretaceous to Neogene flysch sediments, characterised by a heterogeneous $^{87}Sr/^{86}Sr$ composition (Mason *et al*., 1996), range from 0.7078 to 0.7170 for the Eastern Carpathian samples. Modern $^{87}Sr/^{86}Sr$ of granitic rocks of the Bohemian Massif are more radiogenic than seawater carbonates, flysch, and loess sediment, with $^{87}Sr/^{86}Sr$ compositions of up to 0.730 (Janoušek *et al*., 2022; Janoušek *et al*., 2020). In some cases, values are extremely high (over 0.8; (Erban Kochergina *et al*., 2021; Hasalová *et al*., 2008). The heterogeneous $^{87}Sr/^{86}Sr$ composition of Southern Moravia bedrock allows us to test the hypothesis that such heterogeneity will be reflected in the $^{87}Sr/^{86}Sr$ of wines.

A recent study (Erban Kochergina *et al*., 2021) focused on the relationships between the Sr isotopic composition of the bedrock, water input, and plant tissue. The authors examined different tree species (oak, pine, spruce) from the contrasting geological sites without evolved soil horizons, for example, quarries and rock outcrops. The authors offered a simplified model for determining plant origin based on Sr isotopic ratios. We used this model to describe the relationship between the isotopic composition of wine, rainwater and bedrock (Figure 4). The determinative factor for this model is the relative Sr isotopic compositions of rock and rainwater. While the isotopic composition of the local rainwater varies between 0.709 and 0.710, similar to sea spray $^{87}Sr/^{86}Sr$, we can divide all rocks into two groups: (i) rocks with $^{87}Sr/^{86}Sr$ lower than the local rainwater composition and (ii) rocks with more radiogenic $^{87}Sr/^{86}Sr$ than the local rainwater. The authors suggest that the plant's final $^{87}Sr/^{86}Sr$ composition is located on the mixing line between $^{87}Sr/^{86}Sr$ of rainwater and $^{87}Sr/^{86}Sr$ of bedrock.

The tendency of plant material to have isotopic compositions, which are a mixture of bedrock and atmospheric deposition (rain) compositions, may be used to find the origin of a wine sample from among a set of putative localities. The isotopic composition of rainfall is uniform relative to the highly variable contribution derived from the local geology. Because of this, the relative positions along the $^{87}Sr/^{86}Sr$ axis where the wine sample, precipitation, and bedrock of the various candidate localities plot determine whether the origin of the wine sample can be identified. This is shown in the following three cases, each involving two candidate sites: (i) The bedrock of the candidate sites falls on opposite sides of the rainfall value. Since the wine sample is a mixture of precipitation and bedrock isotopic values, the locality plotting on the opposite side of the rainfall value from the wine sample can be excluded (Figure 4A). (ii) If there are two sites with bedrock values on the same side of the rainfall value and similar to the wine sample, the wine sample could have originated from either of the two sites (Figure 4B). (iii) If there are no putative localities with bedrock on the same side of the rainfall value, there is high confidence that none of the sites is the actual origin (Figure 4B).

This simplified model is confirmed by previously published results: Isotopic composition of wine from the Vulture volcanic rocks from Italy has a radiogenic Sr isotopic composition of ~ 0.708 (Marchionni *et al*., 2013), for example, on the mixing line between $^{87}Sr/^{86}Sr$ of the rock (~ 0.707), rainwater with $^{87}Sr/^{86}Sr$ ~ 0.710 (Rao *et al*., 2015), and/or seawater aerosol ~ 0.7092 (Kuznetsov *et al*., 2012). More radiogenic Sr in wine samples from the Portuguese wine region Douro with $^{87}Sr/^{86}Sr$ ~ 0.729 (Almeida and Vasconcelos, 2004) falls along the mixing line between bulk soil $^{87}Sr/^{86}Sr$ ~ 0.732 and atmospheric input (0.709 − 0.710).

However, numerous existing rock types have Sr isotopic composition similar to atmospheric input ~ 0.709−0.710 (Braschi *et al*., 2018; Epova *et al*., 2019; Marchionni *et al*., 2016). In this case, determining the wine's regional identity becomes more complicated.

3.1. Loess bedrock Sr isotopic composition

Loess is formed by clay, sand, and silt cemented by carbonates. The amount of well-soluble carbonate and its ratio to clay minerals dominate the wine's final $^{87}Sr/^{86}Sr$ composition. Waroszewski *et al*. (2021) found that samples with a higher amount of CaO and Sr content had less radiogenic $^{87}Sr/^{86}Sr$; the ratio of each loess component could affect the $^{87}Sr/^{86}Sr$ composition of the bioavailable end member. Šimíček *et al*. (2021) examined loess–palaeosol sequences of Moravia and found that in loess samples, quartz minerals predominate over feldspars, phyllosilicates (mica, chlorite, clay minerals), carbonates, amphiboles, and garnets, with Ca and Sr being associated with carbonates in the medium to coarse silt and clay fractions. Due to the presence of phyllosilicates (commonly with high Rb/Sr), the bulk rock Sr isotopic ratios are likely to be high, resulting in radiogenic soil compositions, i.e., $^{87}Sr/^{86}Sr$ = 0.7184 in the deepest C-horizon. However, the carbonate matrix is the most enriched Sr end member in loess, and its Sr isotopic compositions are 0.7084 – 0.7095 (leached samples, Table 2). Such significant differences between bulk rock silicate and the available leached fraction are not uncommon. Similar results were obtained with carbonate (Nádaskay *et al*., 2019), carbonatite (Rapprich *et al*., 2017), and soil samples (Braschi *et al*., 2018; Marchionni *et al*., 2016; Petrini *et al*., 2015).

4. $^{87}Sr/^{86}Sr$ composition of the studied soil samples

4.1. Bulk soil vs. leached soil Sr isotopic composition

Soils, the primary source of the bio-available Sr, consist of two main components: inorganic matter, resulting from the weathering of their parent rocks, and organic matter. The $^{87}Sr/^{86}Sr$ composition of soils can differ from that of bedrock. Therefore, soil analyses are more useful in studies of wine origin. Our studied soils represent a quite predictable situation: The $^{87}Sr/^{86}Sr$ of the bulk soil at all depths is higher than the rainwater composition (> 0.7097); the $^{87}Sr/^{86}Sr$ of the available phase is lower than the rainwater composition (< 0.7097). The isotopic composition of soil leachates corresponds to those of the studied wines. Therefore, comparing bulk soils and wine compositions can lead to incorrect conclusions.

4.2. Variability of soil $^{87}Sr/^{86}Sr$ with depth

Observed differences in bio-available $^{87}Sr/^{86}Sr$ from the leached soil phases in different soil horizons (depth) are determined by the amount of available water and the weathering of minerals (e.g., carbonates and clay minerals) and their variable mixing proportions. We assume that the root length could play a major role, as young plants will likely get the Sr from a shallower depth than old ones. Perhaps this is one reason why Almeida and Vasconcelos (2004) noted the minor differences in the isotopic composition between wines from "young" and "old" vineyards.

4.3. Leached soil vs. groundwater Sr isotopic composition

The average isotopic composition of the bio-available Sr from studied soils is almost identical to the groundwater composition. Accordingly, we suggest that groundwater Sr isotopic composition is a good tool to determine the depth-averaged $^{87}Sr/^{86}Sr$ of the bio-available soil phase. In other words, one sample represents the average isotopic composition of bio-available Sr over all soil profiles. In some cases, the groundwater sample could be more meaningful than the soil sample, for example, in cases of a very heterogeneous geological basement.

5. $^{87}Sr/^{86}Sr$ isotopic composition of Moravian wine

Almeida and Vasconcelos (2004), Tescione *et al*. (2020), and Marchionni *et al*. (2016) conclude that the winemaking process does not affect the $^{87}Sr/^{86}Sr$ composition of the wine. These studies also confirm that the differences in $^{87}Sr/^{86}Sr$ isotope ratios are the result of heterogeneity of the soil/bedrock composition and, therefore, Sr isotope composition could be a good tool to trace the origin of the wine. Taking these results into account, we were able to rule out the influence of winemaking on the $^{87}Sr/^{86}Sr$ isotope heterogeneity of the Moravian wines. It also allows us to compare samples from different wineries.

The $^{87}Sr/^{86}Sr$ of the wine samples (Figure 3) from Groups A and B are almost identical and comparable to groundwater and bio-available $^{87}Sr/^{86}Sr$. The Group A and B wines belong to different sub-regions of Mikulov and Velké Pavlovice, respectively. This indicates that the geographical/historical divisions into wine (sub-) regions do not correspond to the chemical composition of wines, resulting from the locality's geological characteristics. The low heterogeneity of the wine samples is comparable to the changes in the $^{87}Sr/^{86}Sr$ with depth.

Group C and D samples have lower $^{87}Sr/^{86}Sr$ ratios than Groups A and B. However, these differences are insufficient to distinguish production from these localities.

We cannot distinguish any variations among the grape varieties (Table 1) and vintages (Figure 5). In previous studies, Durante *et al*. (2015) and Petrini *et al*. (2015) observed that the $^{87}Sr/^{86}Sr$ of wines of different vintages are homogeneous. Epova *et al*. (2019) described time-dependent changes in the Sr concentration and the $^{87}Sr/^{86}Sr$ ratio in wines from a winery of Pomerol AOC winery produced in 1965–2015. The authors hypothesise that this could result from geochemical processes in soils associated with Sr mobility. The slight deviation in the composition of the South Moravian wine $^{87}Sr/^{86}Sr$ from similar rock layers can be explained by (i) past and present soil management, (ii) the age of the vineyards, and (iii) the role of water. Water stress could play a key role in the $^{87}Sr/^{86}Sr$ composition of wines. We hypothesise that if plants use water from the lower horizons in dry years, the Sr radiogenic ratio could be shifted to more radiogenic (higher Sr ratios) results. This assumption can only be confirmed with a long-term dataset (e.g., Epova *et al*., 2019) from a winery with a long history and/or a combination of $^{87}Sr/^{86}Sr$ and light isotopes (O, C, H).

Based on the above, we assume that the $^{87}Sr/^{86}Sr$ for wines from these sub-regions of the Moravian region vary from 0.708 to 0.710, comparable to the soil leachate and groundwater composition. However, without a systematic study of the isotopic composition of wines, soils, and water from each vineyard, we cannot identify the exact origin of Southern Moravian wine. A combination with other radiogenic (e.g., Nd, Pb) or stable (O, C, H, N, S) isotopes would possibly make this possible.

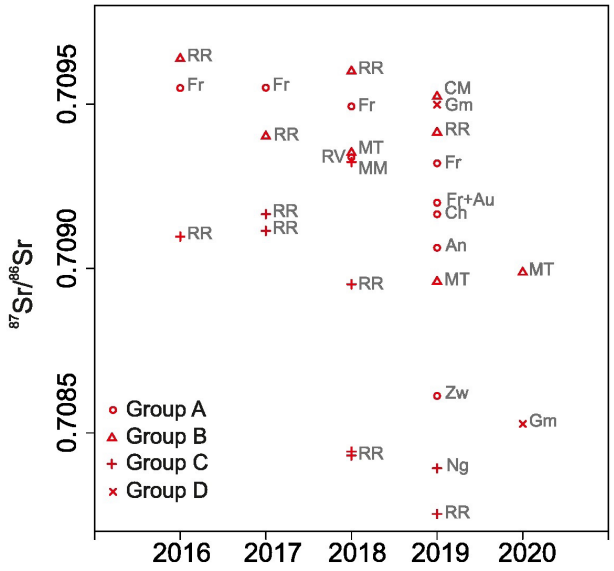


FIGURE 5. $^{87}Sr/^{86}Sr$ of wines from different vintages. There are no correlations between the isotopic composition of the wine, year or grape variety. The abbreviations for varieties are listed in Table 1.

6. Are Sr isotopes a good tracer for wine origin?

The indistinguishable isotopic compositions of grape juices, must, and wine enables us to see connections between them (Tescione *et al*., 2020). Therefore, two wine samples from the same winery, vintage, and locality should have an identical Sr isotopic composition. We can also recognise two samples from geologically different localities (e.g., basalts and granites, Figure 4). In addition, we describe the geological limitation of the connection between bedrock, soil, and wine Sr isotopic composition.

6.1. Rainwater–bedrock $^{87}Sr/^{86}Sr$ mixing line

Using the published isotopic compositions of wines and bedrock, we have simulated two cases for the relationship between bedrock and wine (Figure 6A). The first situation is if the isotopic composition is similar to the bedrock. It can occur when the bedrock contains high amounts of soluble Sr, such as carbonates. The Sr input from the rainwater is insignificant due to the very low Sr content in the rainwater (Novak *et al*., 2020b). The opposite situation, in which the Sr isotopic composition of wine is identical to atmospheric deposition, is unrealistic since bedrock always contains a higher Sr content than rainwater. The final wine composition depends on the ratio of rainwater and soluble Sr from the rock and their isotopic compositions.

Graustein and Armstrong (1983) suggest that less than 25 % of the strontium in vegetation comes from the weathering of underlying rock and more than 75 % from atmospheric transport. Therefore, the amount of soluble Sr of the bedrock plays a key role in $^{87}Sr/^{86}Sr$ of wine, for example, if the bio-available Sr in the soil decreases, the role of rainwater (or irrigation) will increase.

6.2. The relationship between rainwater and the wine product

Raiber *et al.* 2009 compared the Sr isotopic composition of rainfall and basalt groundwater in south-eastern Australia. The freshest basalt groundwater has an $^{87}Sr/^{86}Sr$ signature close to the whole-rock strontium isotope ratios of the basalts (0.7045). However, down-gradient groundwater becomes more radiogenic and approaches those of local rainfall. The authors concluded that the influence of rainwater on the $^{87}Sr/^{86}Sr$ of the basalt groundwater is much greater than that of basaltic weathering.

6.3. Is it possible to predict $^{87}Sr/^{86}Sr$ of wine knowing its geological basement?

The model in Figure 6B describes the possible interactions between bedrock leachate and wine. It must be emphasised that no natural material on the Earth can have $^{87}Sr/^{86}Sr$ lower than 0.69899 (basaltic achondrite best initial BABI (Papanastassiou and Wasserburg, 1969)). Rosner (2010) pointed out that an $^{87}Sr/^{86}Sr$ below the modern mantle (0.702) is not possible for natural samples. The upper $^{87}Sr/^{86}Sr$ limit is not defined; Erban Kochergina *et al*. (2021) analysed granitic rock with a modern $^{87}Sr/^{86}Sr \sim 1.7$.

Based on the $^{87}Sr/^{86}Sr$ composition, we could categorise all possible bedrocks (or bulk soils) into three groups: rocks with an isotopic composition below the rainwater composition (0.702–0.709, process I in Figure 6B), rainwater-like composition (0.709–0.710, process II in Figure 6B), and rocks with a higher isotope composition than the rainwater composition (in our model > 0.710, process III, IV, V in Figure 6B).

Rocks with an $^{87}Sr/^{86}Sr$ lower than 0.709 are common and are often used for cultivated wine, such as sea carbonates and basalts (Figure 6B, I trend). Such rocks have a high amount of Sr. We presume that the leachate soil fraction Sr would have isotopic ratios similar to or higher than bulk rock/soil but never higher than the local rainwater. The rocks with the lowest $^{87}Sr/^{86}Sr$ are ocean island basalts, such as Hawaiian basalts. These rocks have $^{87}Sr/^{86}Sr$ vary from 0.7033 to 0.7044 (Bryce *et al*., 2005), with extractable soil phases having $^{87}Sr/^{86}Sr$ varying from 0.7036 to ~ 0.710 (Chadwick *et al*., 2009). Therefore, we conclude that worldwide wine samples would have $^{87}Sr/^{86}Sr$ isotopic ratios higher than 0.703 and wine with such a low isotopic composition (0.703 – 0.705) is rare in continental Europe.

If the bedrock of the studied locality has $^{87}Sr/^{86}Sr$ close to that of the rainwater (Figure 6B, II-Trend), the bedrock and wine sample's isotopic composition is not sufficient to model the geographical origin or distinguish two samples of wine with similar composition. In this case, it is necessary to know the exact composition of rainwater and the bio-available soil fraction. However, a statistically significant number of samples and/or other analyses are still required.

Rocks with more radiogenic Sr compositions (e.g., granites) contain numerous minerals with different Sr/Rb ratios and Sr concentrations. Their differing reactivity during the weathering is determinative of the composition of the leachate and the end product, wine. There are three possible situations. Process III is widespread: the leachate has an isotopic composition between the bulk rock and rainwater (0.720 and 0.709). Another very common situation where the mineral with higher $^{87}Sr/^{86}Sr$ (e.g., mica with high Rb/Sr) than the bulk rock dissolves faster than other minerals (IV-process). In this case, the process described above would influence the composition of leachate and wine. Therefore, comparing wines with the bedrock composition or literature and geological map data is incorrect and may lead to wrong conclusions. The V-process was described by Blum *et al*., (2002), who showed that plants' root mycorrhizal systems can directly affect apatite weathering. Apatite is a common Rb-free mineral, so its isotopic composition is constant through geologic time compared to other minerals (feldspars, mica) and can be below 0.709. During the V-process, the plant could have a lower $^{87}Sr/^{86}Sr$ than the rainwater, whereas the bulk rock has a more radiogenic $^{87}Sr/^{86}Sr$ composition, which is higher than the rainwater. In the example described by Blum *et al*. (2002), the apatite has a $^{87}Sr/^{86}Sr$ composition between that of silicate rock and atmospheric deposition, and, therefore, it is C-process in our model.

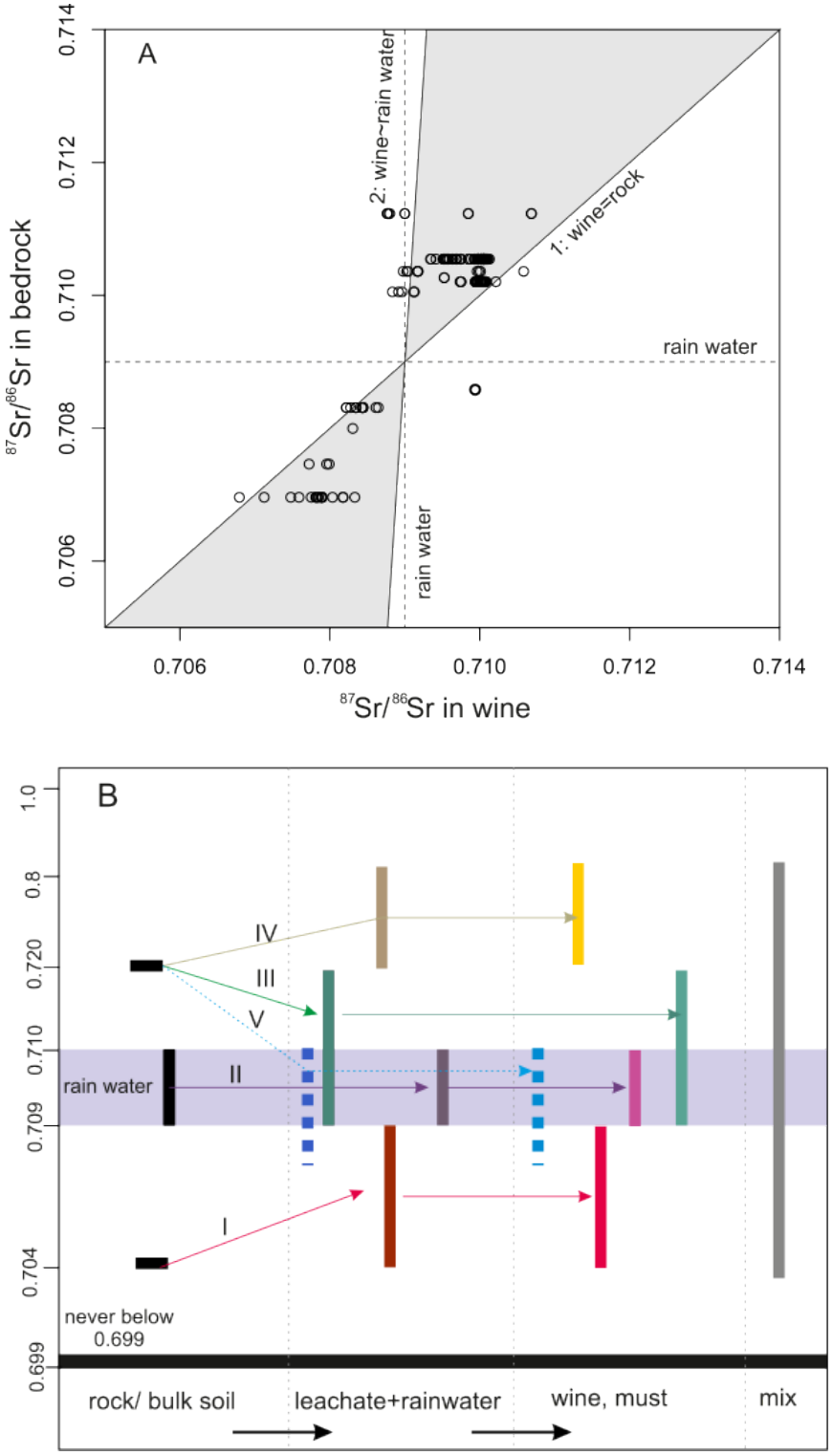


FIGURE 6. Possible model of bulk rock–rainwater mixing.

A. The diagonal line is a mixing line, for the case of wine composition = bulk rock composition and atmospheric input is negligible. The almost vertical line is another possible mixing line, where the atmospheric input is more influential. Data are from Petrini *et al.*, 2015, Almeida and Vasconcelos, 2004; Boari *et al.*, 2008; Braschi *et al.*, 2018; Marchionni *et al.*, 2013; Petrini *et al.*, 2015; Tescione *et al.*, 2020, Tescione *et al.*, 2018. Horizontal arrays are the result of several wine samples being paired with the same bulk rock analysis, in many cases these are imprecise and adopted from geological maps. B. The model, linked bulk rock composition (first column) with leachate (second column) and wine (third column) composition. The last column shows that in the case of mixing samples from different geological localities, the final wine composition could vary and would not correspond to any geological locality. In this case, soils and rocks behave similarly, in the sense that the shift in isotopic ratio during the leaching process is determinative of wine isotopic ratio.

We suggest that the V-process is very unlikely for wine production. However, we cannot rule out this unrealistic situation.

If fruit from different sites with different bedrocks were used during the winemaking process, the blended product could have a wide range in its $^{87}Sr/^{86}Sr$ isotopic composition. Hence, all models and conclusions would be incorrect. In this case, the grape and grape juice analyses could be used as end members in the winemaking process modelling.

CONCLUSION

This paper presents the first and original $^{87}Sr/^{86}Sr$ data on wine from the Moravian region in the Czech Republic. We compare its isotopic composition with that of bulk soil and its bio-available soil fraction from 30, 60, 90, and 200 cm depths.

The studied soils are heterogeneous in their $^{87}Sr/^{86}Sr$ composition; bulk soil composition varies from 0.7138 to 0.7222. The bio-available soil fraction varies from 0.7082 to 0.7105, with the average composition similar to the groundwater from the local well. We conclude that the groundwater sample is a good tracer for bio-available Sr.

We offer a simple model that connects the wine-leachate-bulk rock/soil Sr isotopic composition. We recommend analysing the rainwater and groundwater and leachate from soil together with wine samples. We show that there are situations in which the composition of the bulk rock can differ significantly from that in the wine. In the case of bedrocks with $^{87}Sr/^{86}Sr$ below the rainwater composition, we do not need to provide the soil leaching tests. In this case, the isotopic composition of wine would vary from 0.703 to rainwater $^{87}Sr/^{86}Sr$.

The $^{87}Sr/^{86}Sr$ for wines from South Moravia with sedimentary bedrocks vary from 0.708 to 0.710, which correlates to the $^{87}Sr/^{86}Sr$ in soil leachate. We do not observe substantial differences within and between the cultivars and vintages. Due to the similarity in the isotopic composition of the bedrocks of studied vineyards, we cannot distinguish between different wine sub-regions of Southern Moravia.

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