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論文概要
(Summary of the Thesis/Dissertation)

Doctoral Program in Life Science Innovation
School of Integrative and Global Majors
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1. Title of the Thesis/Dissertation :
Geographical Traceability of Tunisian Olive Oils Based on the Elemental and Stable Isotope Composition

2. Summary (800 – 1,000 words in English)

The protected geographical indications (GI) are powerful sustainability and marketing tools that have extensively been applied in the EU to protect, promote and valorize foodstuffs originating from specific geographical areas. In order for a foodstuff to register as a protected GI in the national legislation, the causal link showing how the characteristics of a particular region affect a product in a way that other regions cannot must be justified. Different geographical traceability methods of Tunisian olive oils are suggested in the literature. They are mainly based on the olive oil's biochemical composition (fatty acids, polyphenols, triacylglycerols, sensory profile, etc…) and molecular markers. These compounds can be useful in verifying the varietal origin or investigating the adulteration with cheaper vegetable oils. However, it is necessary to use tracers that reflect the provenance characteristics, in other words, the geology and climate in the case of geographical origin traceability. The main objectives of this research are the establishment of a database and the development of a traceability methodolgy for the Tunisian olive oils based on the multielemental and stable isotopic composition coupled to chemometrics.

The first part of this study represents a pilot study conducted using the multielemental profiling approach by ICP-MS. Despite the long and established use of multielements in geographical traceability of olive oils in other countries, their validity as markers of the provenance factors (e.g. soil geochemical composition) is not proven, in contrast to the information available for stable isotopes whose variability in olive oils according to origin was correlated with climatic and geographical parameters. Therefore, the link between the olive oils elemental composition and the geochemical soil composition (determined by XRF and LA-ICP-MS) was investigated. The results of this pilot study showed the predominant geochemical source of the elements in the olive oils based on their associations in a latent space according to Goldschmidt rule and their correspondence with the provenance soils composition. These results proved that a multielemental profiling approach is a valid and promising geographical traceability tool. However, the total concentration of the elements in the soils on which the olive trees are grown cannot predict the elemental composition of the derived olive oils. This will direct us towards exploring additional environmental factors that may impact the oils' multielemental profile.

Additionally, the results pointed out limitations of the analytical method based on the microwave-assisted extraction of the multielements. For this reason, additional research was conducted dealing with the optimization of the multielements extraction from the olive oils. Three methods are compared among which one was originally proposed in this research, with a special focus on the performance criteria: precision and method detection limits. The results of this comparative study have shown that the ultrasound-assisted extraction performed better than the other two microwave-based methods.

The next part of the thesis is focused on two aspects: (i) the geochemical and edaphic characterisation of the main 11 olive oil producing regions to deepen the understanding of the relationship between the elemental composition of olive oil and its environment and (ii) the creation of an extensive national database of multielements and stable isotopes of C and O (using IRMS) in Tunisian olive oils from the main 11 producing regions with a view of testing the geographical discriminatory power of these two tracers.

For this reason, additional parameters have been involved including the soil characteristics (pH, EC, OM and $CaCO_3$), the bioavailable fraction of the soil elements in addition to their total concentration and the climatic parameters. The objective is to identify origins with unique elemental markers that can become strongly recognized by their elemental signature and therefore can become GI areas. The results showed a good consensus between the geochemical and edaphic characteristics pointing out some geochemical processes; and that the regions with similar outcrop lithology presented similar soil chemical characteristics. When considered all together at the same time and in the same model, out of the 11 regions, only 2 regions presented distinct soil characteristics whereas the rest of the regions which were located above similar outcrop lithologies could hardly be distinguished. The same observations apply to olive oil multielemental composition. This fact highlights the limitation of the traceability approach based on chemical analytical database when similar driving factor (here is the geology) prevails in the regions to be discriminated. The study of the relationship between the olive oil elemental composition and bioavailable fraction of elements in soil revealed that Ti, Fe, Ni and Ba presented significant positive Spearman correlation coefficients. This is the first study addressing and reporting such significant correlations which show the effective link of olive oil inorganic composition to its environment and consequently provides an additional piece of evidence for the validation of this traceability approach and also the elements that can establish strong and well-established traceability.

The last part of this research studies the isotopic composition of C and O of olive oils from the same 11 regions and compares the performance of the isotopic and elemental profiling as a traceability tool for intra-(inside Tunisia) and inter-(Tunisia vs Italy) country scales. The main climatic and geographical factors that determine the isotopic signature of Tunisian olive oils have been identified and significant Pearson correlations have been proven which will allow the future quantitative prediction of their isotopic composition based on regressions. Concerning the intra-country discriminatory power, both tracers performed similarly with difficulties encountered whenever the drivers are similar between the regions to be discriminated. Our results also showed that the combination of the elemental and isotopic fingerprinting approach can increase the discriminatory power of the model. As for the inter-country, multielements showed a better efficacy than stable isotopes to discriminate Tunisian and Italian oils. This finding may be explained to the geographical proximity of the two countries and the similar climatic conditions prevailing in southern Italy and Tunisia. In the future, an integrated work involving researchers, agricultural authorities and olive producers should be carried out in the highly potential GI areas to materialize the objective of valorizing olive oil as a key bioresource in Tunisia.