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Comparison of Different Membrane Filtration Systems for Recovering Active Phenolic Compounds from the European Spread Macroalga *Sargassum muticum*

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Abstract

Sargassum muticum is an invasive macroalga that uncontrollably widely grows on the European Atlantic coasts. This macroalga is particularly rich in phenolics (phlorotannins) presenting biological activities such as antioxidant, radical-scavenging, antibacterial, photoprotective properties, etc. Among the diversity of metabolites produced by the species, phenolic compounds could constitute one of the potential ways of valorization of *Sargassum muticum*.

For the needs of the study, algae were harvested along a latitudinal gradient in Norway, Ireland, France, Spain and Portugal.

After extraction, the highest phenolic contents and bioactivities were measured for the samples coming from Norway and Portugal. Consequently, both these countries have been retained in the present work.

The extracts are composed of a "pool" of phenolic compounds of molecular weight varying, according to the literature, from 126 Da (phloroglucinol) to 650 kDa. But the most common compounds are comprised between 10 and 100 kDa. With the aim of concentrating and eventually fractionating (i.e. isolating the most active fractions) the phenolic extracts of *Sargassum muticum*, a few membranes of Molecular Weight Cut-Off 10 kDa were chosen. These membranes were tested by using different technologies: ultrafiltration, centrifugal tube and dialysis.

Results show that there is no significant activity in the permeates signifying that the retention rate is about 100 % for the active fractions. Concerning the retentates, results show clearly an increase of the total phenolic contents as well as antioxidant and radical scavenging activities. The best performances are obtained with the centrifugal tubes using 10 kDa membranes.

Introduction

Marine macroalgae spreading along the intertidal rocky areas, as invasive species, produce high biomass^[1] and can have an economic importance^[2]. Among the diversity which occur in marine environment, i.e. 1726 species of *Ulvophyceae*, 6758 *Florideophyceae* and *Bangiophyceae* and 2048 *Phaeophyceae*^[3], Brittany (France) is considered as a hot-spot region concerning macroalgae with nearly 700 species of macroalgae currently listed on the Breton littoral^[4].

The present study is focused on *Sargassum muticum*, (Yendo) Fensholt (1955). This brown macroalga was introduced accidentally on the European coasts in the 1970s with oyster spat imported from Japan^[5]. From its introduction point (United Kingdom), the alga spread along the Atlantic coast and en-

larged its repartition. It is now observed from Norway to Morocco^[6-9], and also on the French Mediterranean coasts^[10]. Its proliferation is due to intrinsic characteristics such as a great tolerance to the variations of temperature and salinity, fast growth, mode of reproduction and dispersal, as well as the favourable conditions met on the European coasts. This proliferation cannot be controlled and it hampers certain human activities in summer period^[9]. However, this abundant and under exploited marine living resource contains a lot of bioactive substances whose applications are numerous in food industry, cosmetics or pharmaceuticals^[11-23]. As demonstrated in the majority of the previously cited references, *Sargassum muticum* is particularly rich in phenolic compounds (phlorotannins) presenting biological activities such as antioxidant, radical-scavenging, antibacterial, photoprotection, etc. Industrialists are very interested by natu-

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ral antioxidants in food industry because of the restricted use of synthetic anti-oxidants suspected to have toxic effects^[24,25]. So, the extraction of phenolic compounds constitutes one of the main potential ways of valorization of *Sargassum muticum*.

Most often, phenolic extracts are too diluted to be used directly in the industry and so, they need to be concentrated. But the elimination of water by evaporation may denature the heat-sensitive phlorotannins. So, membrane processes can be used because they allow concentrating a sample at low or ambient temperatures. Some studies have been reported on the use of ultrafiltration to concentrate phenolic compounds from terrestrial plants^[26-31], on the use of nanofiltration to produce phenolic rich fractions from artichoke^[32] or orange press liquors^[33], even on the use of electrodialysis applied to phenolics from tea^[34]. All these articles cited as examples and others listed in the literature survey of Bazinet^[35] show that membrane techniques are efficient for the separation of phenolics but very few studies have been published on marine resources such as macroalgae, except the use of ultrafiltration and/or dialysis for fractionating phlorotannins from brown algae^[14,36,37].

In this study, with the aim of fractionating the phenolic extracts of *Sargassum muticum* (i.e. isolating the fractions with the highest antioxidant activity), a few membranes of Molecular Weight Cut-Off (MWCO) 10 kDa were chosen. Indeed, the extracts from *Sargassum* are composed of a “pool” of phenolic

compounds of Molecular Weight (MW) varying from 126 Da (phloroglucinol) to 650 kDa (polymers) with most of the common compounds comprised between 10 and 100 kDa^[38]. It is well known that the molecular size has a role in the bioactivity level of polyphenolics, although some authors claim that all the MW are active after ultrafiltration of extracts from *Sargassum* with membranes 10kDa^[39,40]. So, in this study, different membrane based separations commonly used at the laboratory scale, i.e., ultrafiltration, membrane centrifugal tube and dialysis, were tested in order to validate the feasibility of the separation of active phlorotannins from *Sargassum muticum*.

Material and Methods

Biological Material and Collection

Sargassum muticum is a pseudo-perennial species which settled in rocky shores in intertidal and subtidal zones in Brittany^[9]. The perennial part is the main axis which persists all along the year. The annual parts are the flexible laterals visible from spring to summer.

For the needs of the study, individuals of *Sargassum muticum* were harvested along a latitudinal gradient from Norway, Ireland, France, Spain to Portugal on three different sites in each country (Table 1).

Table 1: Sites of Collection of Samples of *Sargassum muticum*.

Country	Region	Site	Code	Latitude	Longitude	Hydrodynamism
Norway (N)	Øygarden	Solberg	N1	36°35'N	4°47'W	Semi-exposed
	Øygarden	Kraekjebaerholmen	N2	36°35'N	4°50'W	Semi-exposed
	Øygarden	SauØyna	N3	36°35'N	4°51'W	Semi-exposed
Ireland (I)	Connaught	Clifden	I1	53°40'N	10°11'W	Exposed
	Munster	Spanish point	I2	52°85'N	9°44'W	Semi-exposed
	Munster	MiltownMalbay	I3	52°84'N	9°44'W	Semi-exposed
France (F)	Brittany	Grand Dellec	F1	48°21'N	4°34'W	Exposed
	Brittany	Le Minou	F2	48°20'N	4°36'W	Exposed
	Brittany	Pointe du diable	F3	48°21'N	4°33'W	Exposed
Spain (S)	Galicia	Museo del Mar	S1	42°13'N	8°46'W	Semi-exposed
	Galicia	Caboes tai	S2	42°11'N	8°48'W	Exposed
	Galicia	Caboudra	S3	42°20'N	8°49'W	Exposed
Portugal (P)	Algarve	Queimado	P1	37°49'N	8°47'W	Semi-exposed
	Algarve	Almogave	P2	37°39'N	8°48'W	Semi-exposed
	Algarve	Zambujeira	P3	37°33'N	8°47'W	Semi-exposed

The collection of samples was done in April-May at the same level of physiological development (end of the growth phase). Only the apical part was collected to minimize the environmental impact. After sampling, epiphytes were removed and the thalli were rinsed with fresh water before freeze-drying and powder grinding^[41].

Extraction and Purification of Phenolic Compounds

Phenolic compounds were extracted from freeze-dried algal powder by a mixture of acetone: water (50: 50) at 40°C for 2 hours in the dark. The extract was then centrifuged, filtered and vacuum evaporated before adjusting the volume of the raw extract to 50 mL with distilled water^[16].

Analysis of the Total Phenolic Content and Activity Tests of the Extracts/Fractions

The Folin-Ciocalteu colorimetric method modified for microplates by Sanoner *et al*^[42] was used to measure the total phenolic content of extracts/fractions.

Two types of activities have been evaluated:

Radical-scavenging activity with the DPPH test on microplates by following the method described by Fukumoto and Mazza^[43]. The results were expressed by the IC50 index, which is the concentration (mg/mL) of phenolic compounds necessary to inhibit 50% of the reducing reaction of the DPPH. So, a low value of IC50 corresponds to a high radical scavenging activity.

Antioxidant activity with the β -carotene bleaching test on microplates was carried out following the methodology de-

scribed by Kaur and Kapoor^[44]. Butyl Hydroxyanisole (BHA) was used as a positive control, whereas ethanol was the negative control. An Antioxidant Activity Coefficient (AAC) was then calculated as follows:

$$AAC = ((As(120) - Ac(120)) / (Ac(0) - Ac(120))) * 1000$$

As: Absorbance of the sample at 120 min

Ac: Absorbance of the control (ethanol) at 0 or 120 min

The AAC700 corresponds to the concentration of the extract (mg/mL) necessary to have an AAC of 700, which is the value obtained with the positive control as described by Le Lann *et al*^[12]. So, a low value of AAC700 corresponds to a high antioxidant activity.

Analysis of the Extracts by Nuclear Magnetic Resonance (¹H NMR)

With the aim of verifying the presence of phenolic compounds in the crude and purified extracts, we analysed our extracts/fractions with ¹H NMR. Then, the extracts/fractions were mixed with deuterized methanol (MeOD) at room temperature and were analysed with a spectrometer BRUKER DRX 400 using the standard pulse sequences available in the Bruker software (Bruker, Wissembourg, France).

Chemical shifts were expressed in ppm. NMR profiles unambiguously indicate the presence of phenolic compounds

within the extracts, with peaks between 5.5 and 6.5 ppm.

Molecular Weight Separation of Phenolic Compounds

In order to concentrate under mild conditions the phenolic extracts, membrane filtration systems were chosen. Three different processes (Ultrafiltration, Membrane centrifugal tubes and Dialysis) often used at the laboratory scale were selected. All the experiments were carried out by using membranes of MWCO 10 kDa that may normally retain the most common phenolic compounds whose MW is comprised between 10 kDa and 100 kDa.

Ultrafiltration

The ultrafiltration system was a Minimate Tangential Flow Filtration capsule (PALL) of 50 cm² in polyethersulfone (Figure 1A). The filtration was carried out at a pressure of 2.10⁵ Pa and a temperature of 20°C with membranes of MWCO 10 kDa.

Membrane Centrifugal Tubes

Membrane centrifugal tubes (Figure 1B) equipped with membranes in regenerated cellulose (Hydrosart) of surface area 3.9 cm² and MWCO 10 kDa were used at a centrifugal acceleration of 3000 g and a temperature of 15°C.

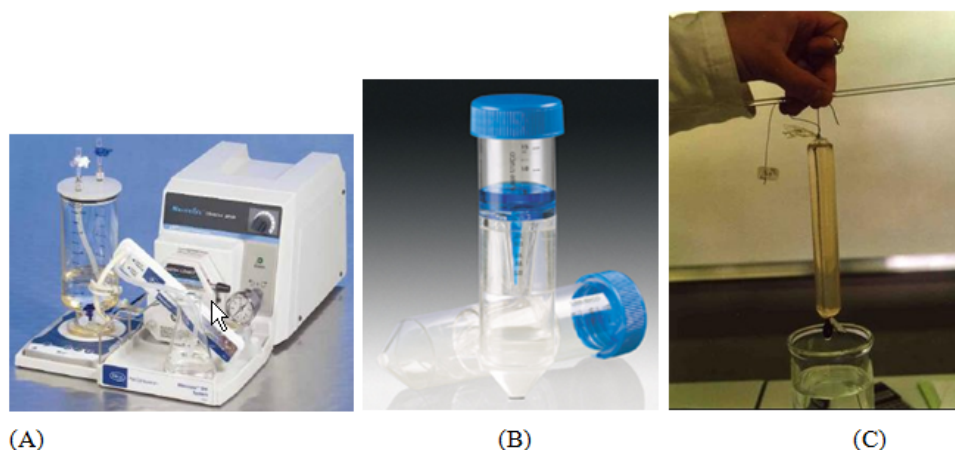


Figure 1: Different membrane filtration systems: (A) Ultrafiltration, (B) Membrane centrifugal tubes, (C) Dialysis.

Dialysis

Membranes in cellulose ester of MWCO 10 kDa constituted the dialysis chamber filled with the extracts. The dialysis bag was put for 28 hours in distilled water (renewed 3 times) (Figure 1C).

Results

Phenolic Content of the Crude Extracts

The total phenolic contents (expressed in % of Dry Matter DM) of the crude extracts have been evaluated for the 3 sites in each country^[17]. It can be observed on Figure 2 that the mean values are the highest for Norway (7.27 %) and Portugal (5.58 %). These results are confirmed by the measurement of the radical-scavenging activity (Figure 3) which is on average the lowest for Portugal (1.65 mg/mL) and Norway (1.62 mg/mL). The best values IC₅₀ = 0.791 mg/L for the site 2 in Portugal (P2) and IC₅₀ = 0.738 mg/L for the site 2 in Norway (N2) are very close to the positive control (0.590 mg/mL)).

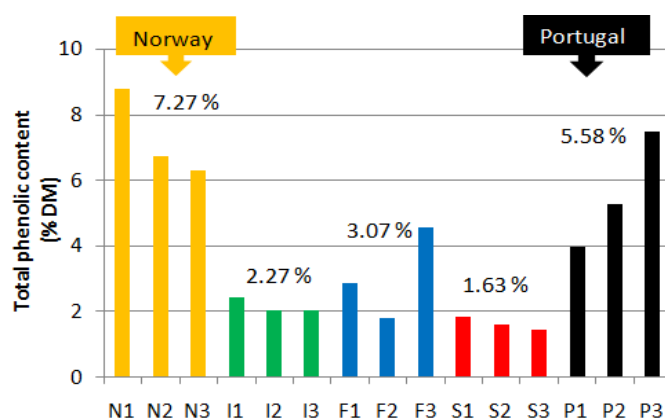


Figure 2: Total phenolic content (expressed in % of Dry Matter DM) in the raw extracts (N: Norway, I: Ireland, F: France, S: Spain, P: Portugal) Numbers 1,2,3 correspond to the three sites sampled by country.

For that reason, the rest of this study will be focused on the phenolic extracts from site 1 Norway (N1) and site 3 Portugal (P3).

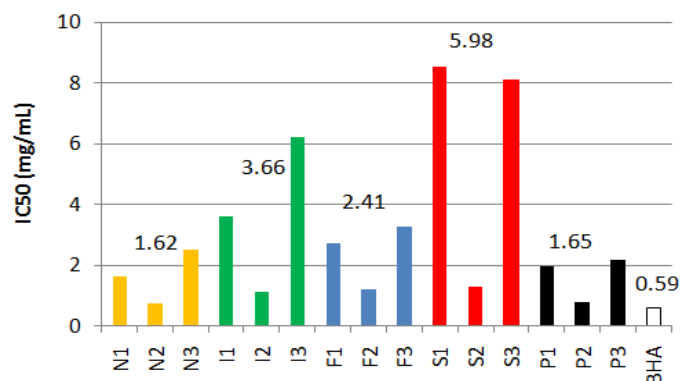


Figure 3: Radical-scavenging activities in the crude extracts (N: Norway, I: Ireland, F: France, S: Spain, P: Portugal, BHA: Butyl Hydroxy-anisole) Numbers 1,2,3 correspond to the three sites sampled by country.

Fractionation of Phenolic Compounds by Membranes

The raw extracts have been concentrated by ultrafiltration, centrifugal tube and dialysis, implemented with membranes of MWCO 10 kDa.

Only negligible amounts of phlorotannins were detected in the permeates, whereas some retentates were highly enriched in phenolic compounds, especially after concentration with centrifugal tube. In this latter case, it has been reached a maximum total phenolic concentration in the retentate of 73.5% for Portugal (Figure 4).

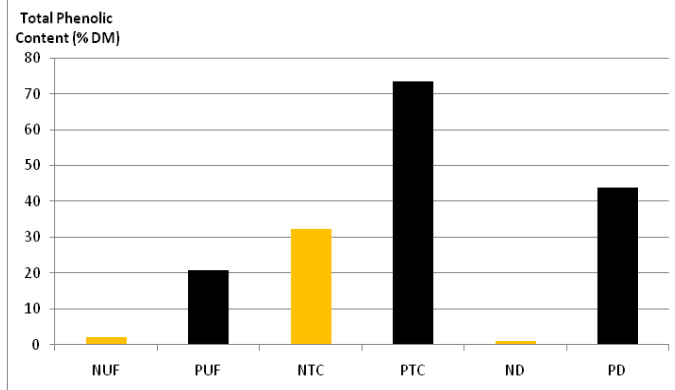
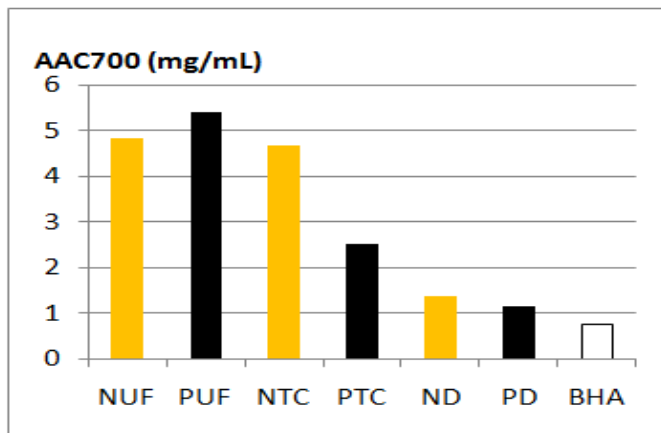
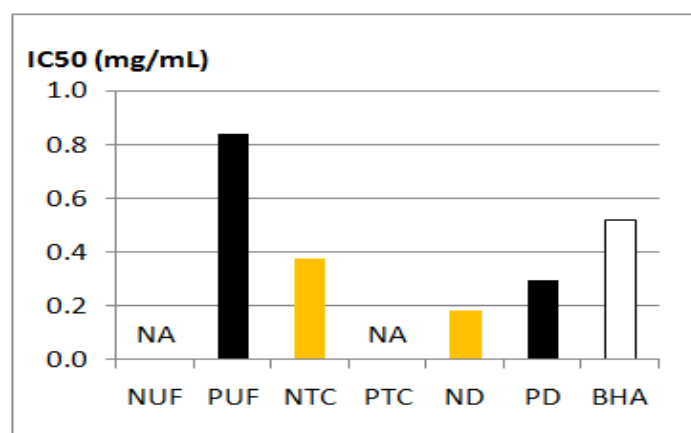


Figure 4: Total phenolic content in the retentates (N: Norway, P: Portugal, UF: Ultrafiltration, TC: Centrifugal Tube, D: Dialysis).

Concerning the radical-scavenging and the antioxidant activities, the most active retentates are obtained after dialysis with excellent IC₅₀ and AAC700 close to positive control BHA (Figure 5A and Figure 5B) for Norway and Portugal.

The analysis of the retentates by ¹H-NMR has shown that the three processes (ultrafiltration, centrifugal tube, dialysis) are efficient to concentrate phlorotannins. But the most accurate peaks are obtained after dialysis (Figure 6) meaning a better purity of the concentrated extract.



(A) **Figure 5:** Activities of the retentates obtained in extracts issued from populations of *Sargassum muticum* collected in Norway and Portugal. (A) Radical scavenging activity, (B) β-carotene bleaching activity, (N: Norway, P: Portugal, UF: Ultrafiltration, TC: Centrifugal Tube, D: Dialysis, BHA: Butyl Hydroxyanisole)

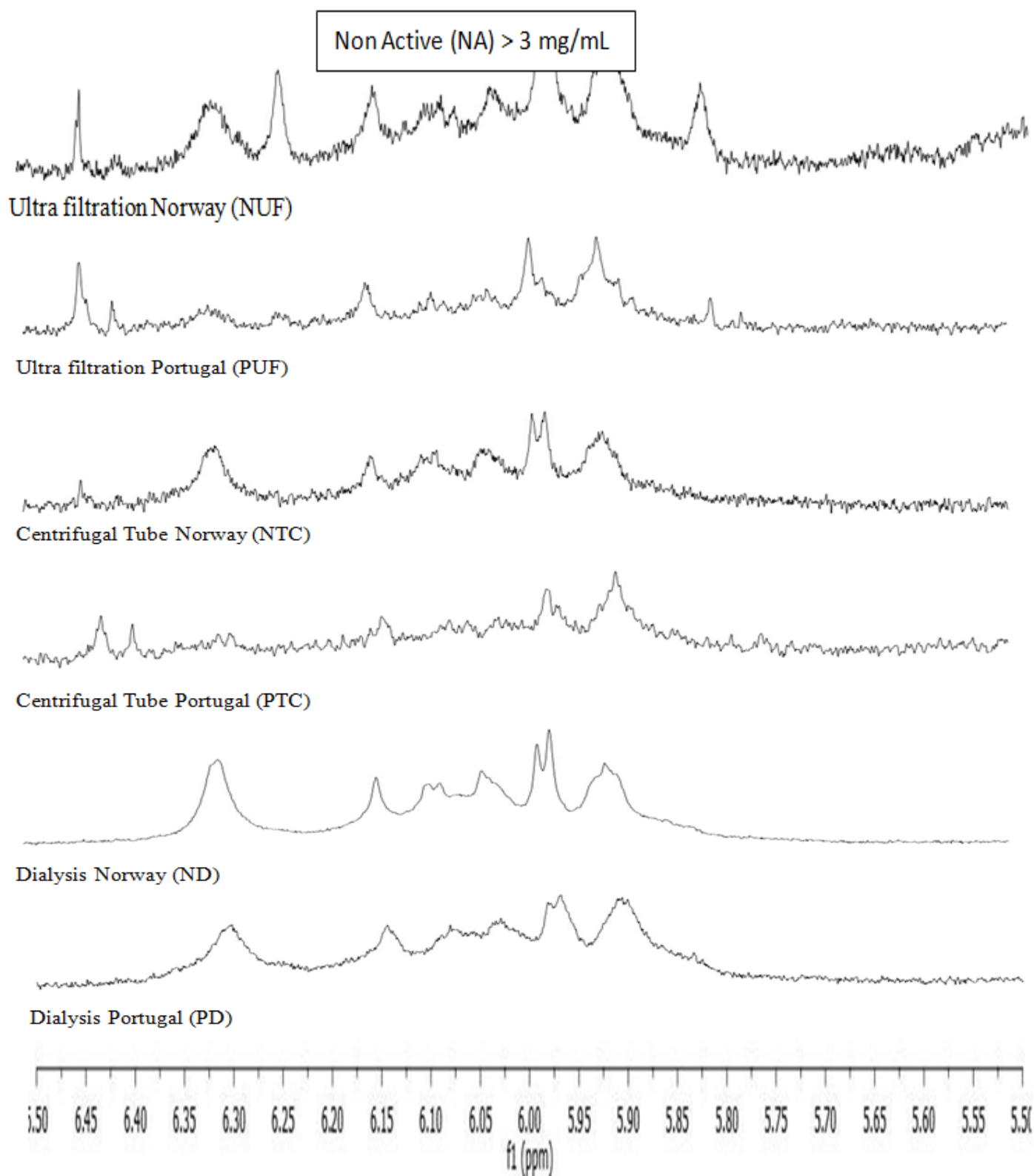


Figure 6: ^1H -NMR analysis of the retentates with enlargement of the spectra in the aromatic area between 5.5 and 6.5 ppm (phenolic compounds).

Discussion

The industrial valorization of the invasive brown algae has already been studied^[45] but there is still much to do. In particular, phenolic compounds (phlorotannins) from brown macroalgae are not industrially exploited and commercialized for their antioxidant properties. So, it is important for future industrial applications to determine the phenolic contents in brown mac-

roalgae *Sargassum muticum* (taking into account the variability of the resource), to measure the activities according to the molecular size of the phlorotannins and to evaluate the feasibility of the concentration by membrane technologies.

Populations of *Sargassum muticum* collected in Norway and Portugal have higher total phenolic contents and higher radical-scavenging activities than populations from Ireland, France or Spain. This is in accordance with previous studies

showing that the biochemical composition (proteins, lipids, carbohydrates) of *Sargassum muticum* is linked to the geographical site and to the hydrodynamic conditions^[41]. It can be supposed that the severe environmental conditions at the extremities of the latitudinal gradient promote the production of phlorotannins, considered as defensive compounds against the UV or the variations of temperature. However, for the countries (France, Ireland, Spain) where phenolic content is not enough, *Sargassum muticum* could be cultivated under stressful conditions (metabolic orientation).

The most common phenolic compounds have a MW between 10 and 100 kDa^[46], that seems to be the case with *Sargassum muticum* from Norway and Portugal while no phlorotannins were detected in the permeates of the membranes 10 kDa. The phenolic compounds of low MW (less than 2000 Da) are very rare in temperate Sargassaceae^[14] whereas these small compounds are very abundant in tropical Sargassaceae (South Pacific)^[47].

The NMR spectra of the retentates are quite similar for ultrafiltration, centrifugal tube and electrodialysis, which suggest the presence of the same type of phenolic compounds. But a further study using HPLC for determining the size repartition of phlorotannins would be necessary, as it was demonstrated in another study on fish protein hydrolysates^[48]. Then, the concentration by tangential flow filtration will be possible at the industrial scale, either by ultrafiltration or nanofiltration as it was proved to be interesting in the Magnoliophyta, mate^[49]. And finally, fractionation will also be possible on the condition of using membranes of MWCO higher than 10 kDa.

Conclusion

This study has shown that the concentration by membranes of the phlorotannins from *Sargassum muticum* is possible. In this case, the main advantage of the membranes is to recover phenolic compounds without denaturing the other molecules. It sees an opportunity to develop an algorefinery process consisting in the recovery of the main target molecules (phlorotannins) and the potential secondary target soluble compounds (proteins, polysaccharides, pigments, alginates, etc), as well as the solid fraction. To reach this objective, the combination of membranes with mild extracting technologies, such as enzymatic hydrolysis for instance, is a promising route^[50].

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