Application of different drying techniques to fresh-cut salad waste to obtain food ingredients rich in antioxidants and with high solvent loading capacity

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Abstract: Wastes from iceberg salad fresh-cut processing were submitted to air-drying, freeze-drying, and supercritical-CO2-drying with or without ethanol as co-solvent. Drying was combined with grinding to obtain flours. Samples were analysed for macro- and micro-appearance, particle size, dietary fibre, polyphenol content, antioxidant activity, water vapour sorption, water and oil holding capacity. Air-drying produced a collapsed brown material allowing a flour rich in fibre (>260 g/kg) and polyphenols (3.05 mg GAE/gdw) with antioxidant activity(6.04 OD-3/min/gdw)to be obtained. Freeze-drying maintained vegetable structure and colour while partly retaining polyphenols(1.23 mg GAE/gdw).Supercritical-CO2-drying with ethanol as co-solvent, produced an expanded material able to entrap huge amounts of water and oil(43.2 and 35.2 g of water and oil for g of dry sample).Air-dried salad waste derivatives could be used as functional food ingredients, while supercritical-CO2-dried ones can be exploited as bulking agents and absorbers of oil spills or edible oils.

Dear Editor,

We send to your attention the research article entitled "**Application of different drying techniques to obtain food ingredients with target functionalities from fresh-cut salad waste**" by Stella Plazzotta, Sonia Calligaris and Lara Manzocco. Following, we report the abstract.

Wastes (external leaves and core) from iceberg salad fresh-cut processing were submitted to air-drying, freeze-drying, and supercritical-CO_2-drying with or without ethanol as co-solvent. Drying was differently combined with grinding to obtain flours. Samples were analysed for macro- and micro-appearance, particle size, dietary fiber, polyphenol content, antioxidant activity, water vapour sorption, water and oil holding capacity. Air-drying produced a collapsed material which underwent browning reactions but allowed a flour rich in fiber with high antioxidant activity to be obtained. Freeze-drying highly maintained vegetable structure and colour while partly retaining polyphenolic content. Finally, supercritical-CO_2-drying, using ethanol as co-solvent, produced an expanded material, which completely lost the original salad colour and was able to entrap huge amounts of water and oil. Salad waste flours could be used as food ingredients, bulking agents, adsorbents for oil spills and structuring agents for liquid oil.

Best regards,

Sonia Calligaris

ANSWERS TO REVIEWERS

Reviewer #1: According to my point of view a suggestion is listed below:
1. Key words should be: Iceberg salad; Drying techniques; Fibre; Polyphenol; Antioxidant activity; Food ingredients.
Keywords have been modified as suggested. "Iceberg salad" was not included in the keyword list as the latter should not exceed 5 words. The word salad is however present in the title (lines 1-2).

Reviewer #2:
1. In terms of "salad waste", were the external leaves shredded, sliced orchopped prior to drying?
Salad leaves and core were removed from salad heads simulating operations that are industrially carried out and chopped before treatment using a sharp knife.Details were added to the manuscript (lines 81-82).

2. Generally pretreatments may be applied to hot air drying to preserve the phenolic compounds, physical properties and microstructure as well as to shorten drying time (in some cases). Using hot air drying alone was certainly not a good choice for preparation of dried vegetables.
We definitely agree with the reviewer that hot-hair drying efficacy can be increased by a number of different pretreatments, including but not limited to dipping in acid solutions, osmotic treatments, US and IR. As stated in the paper aims (lines 63-65), the objective of the research was to evaluate the possibility to valorize salad waste by the application of different drying technologies. The latter were applied as a unique processing step, before or after salad grinding, in the absence of any other pre-treatment. To our knowledge, no prior studies were carried out about salad leaves drying. For this reason, we decided to test its efficacy in the absence of additional variables deriving from possible pretreatments.In the case the process had allowed interesting materials to be prepared (as it actually was demonstrated in the Results and Discussion section), direct drying of salad waste would have been certainly more simple and affordable in an industrial context. However, it is not excluded that possible evolution of the research activity could include the study of the effect and economic sustainability of the application of additional pretreatments before drying.

3. Also give drying time and RH during drying n°3.
This information was added in the text (lines 87-91, 115-117).

Give reason for hot air drying at 70
Air-drying temperature was selected in the range of temperatures generally reported in the literature to produce flours from fruit and vegetable materials. This information, supported by adequate references, was added in the text (lines 42-45).

4. Title should be changed as the term "target functionalities" is too wide. Please be more specific
Title was modified, as suggested by the reviewer (line 1-2). In particular, the generic terms "food ingredients with target functionalities" were substituted with more specific ones ("food ingredients rich in antioxidants and with high solvent loading capacity").

5. Explain "Drying was differently combined with grinding to obtain flours" in the abstract.
The sentence was simplified (line 10), removing the term "differently".

6. Explain "structuring agents for liquid oil" as stated in abstract and conclusion.

Abstract (line 19), Manuscript (lines 346-347) and Conclusion (lines 356-359) sections were improved to increase clarity.

7. What was the best drying technique recommended for this work and based on what criteria? This information should be stated in the abstract.

Results indicated that derivatives with different properties, and thus different potential use, are obtained depending on the applied drying techniques. For this reason, a unique criteria to compare efficacy of the different drying technologies was no presented. By contrast the peculiar advantages of their application were discussed. To better clarify this approach, abstract (lines 17-19) and conclusions (lines 355-356) were modified accordingly.

8. What was the form of sample, whole leaf or chopped leaf?

This information was added in the text (lines 81-82).

How to get an even drying if the whole leaves were subject directly to hot air drying as the thickness of midrib and leaf blade are different?

We agree with the reviewer that salad waste material presents an intrinsically high variability. In order to obtain representative samples, we decided to proceed as usually reported in the literature to dry leaf plant matrices (Nilnakara, S., Chiewchan, N., &Devahastin, S. (2009). Production of antioxidant dietary fibre powder from cabbage outer leaves. *Food and Bioproducts Processing*, 87, 301–307). In addition, variability was reduced by drying more leaves and performing analyses at least three times on two replicated samples.

Could freeze drying be conducted without any other preparation? Please also state the size of sample prior to drying.

Similarly to air drying, also freeze drying was applied as a unique processing step, in the absence of any other pre-treatment, which could have modified the final result. Information relevant to size of samples was added (lines 81-82).

9. Generally pretreatments may be applied to hot air drying to preserve the phenolic compounds, physical properties and microstructure as well as to shorten drying time (in some cases). Using hot air drying alone was certainly not a good choice for preparation of dried vegetables.
10. Also give drying time and RH during drying.°10. Give reason for hot air drying at 70

See answers 2 and 3.

11. Why the SCCD-EtOH sample contained TDF content similar to those of other samples? GenerallyEtOH could dissolve other components in the sample. The remaining part should be mainly plant cell wall components, which account for dietary fiber. Therefore the fraction of TDF content in the treated sample should be higher.

The mean value of TDF in SCCD-EtOH sample was actually higher than that of the other samples. In agreement with the reviewer comment, this could be attributed to the extraction of leaf waxes and other compounds (Table 2). However, although more analyses were performed on each sample, the difference in TDF values was not demonstrated to be statistically significant, probably due to the intrinsic variability of the vegetable material.

12. It seems like the "dry weight" was used for the basis of calculation. Was it the bone dry mass or just weight of "the dried sample", which still contained certain amount of water?

Dried samples were evaluated for residual moisture using AOAC gravimetric method (material and method section, $2.9, lines 155-156). Calculation are thus based on dried matter of dried samples.

13. How could the hot air dried samples possess higher phenolic content than the fresh samples and the freeze dried sample? Could the authors recheck the basis of calculation?

We have accurately check data. A possible explanation is the formation of novel compounds upon thermal treatment, such as Maillard reaction derivatives. Even if they are not polyphenols, they may actually react with Folin reagent leading to data shown in Table 2. Although it is one of the most used methods for determining TPC in vegetable matrices, Folin-Ciocalteau method is not specific for phenolic compounds. Rather, it measures the ability of both phenolic andnonphenolic compounds in alkaline medium to reduce the phosphomolybdic/phosphotungstic acid reagent to blue complexes thatare detected spectrophotometrically (V.L. Singleton, R. Orthofer, R.M. Lamuela-Raventos, Oxidants and Antioxidants,Pt A 299 (1999) 152–178). This information was added into the text (lines 306-307).

14. As described in Line 157, the water and oil holding capacities were calculated based on 1 g of dried sample. Therefore, the unit presented in Tables 2 and 3 should be for WHC and OHC should be per gram dried sample not dry weight.

We thank the reviewer for correcting this error. Table 1 and 2were revised accordingly.

15. Rewrite lines 344-345. Do both products really have a potential to be used as those claimed in the mentioned sentence? Have to make this clear.

The text was modified to increase clarity (lines 355-359).

16. The output from this work was only to improve the technological properties, in terms of water and oil holding capacities, of the products from salad waste.

As indicated in the introduction (lines 24-32), salad waste has very low nutritional value and represents nowadays an environmental burden and a company cost. Based on this consideration, it is our impression that also the possibility of turning it intoa value-added derivative with some technological function could be regarded as a successful valorization strategy.

Editor's corrections

Please check that your manuscript is conform to the instructions to authors of LWT Food Science and Technology, and in particular:

-Reference style:

o Give issue numbers for all or none of the Journals

References were modified accordingly (lines 368-475).

-Use SI units, and in particular

o The authors' guide clearly mentions that "%" is not accepted unit for concentration and composition and yet you have extensively used %. All the % units used for concentration/composition must be changed to direct unit of g/kg or g/L as appropriate.

oexpress concentrations in g/L or mL/L or g/kg, not w/w, v/v, w/v ppm etc

oexpress pressures in Pa pascals, not atm (L85)

oexpress centrifugal force in x g, not rpm (L155). The actual official unit is the m/s2 calculated as omega square x radius (omega: angular speed in radian per second (1 rpm = 0.105 radian/second)); to have it in g divide by 9.8 m/s2) . There is usually an abacus with the centrifuge.

Unitswere modified accordinglyalong the text.

oLatin binomials in italics, including in references;

o Allium cepa NOT Cepa L356

Text was modified accordingly (lines 369, 386, 393, 415, 425, 442).

- Highlights should be short, active sentences that "convey the core findings of the article", as written in the instructions for authors, i.e. contain the most significant results, not a summary of the study: rewritenotably highlights 1 & 5

Based on the instruction for authors, a number of highlights from 3 to 5 is recommended.For this reason, we decided to omit highlights 1 and 5 and maintain highlights 2-4 which contained significant results relevant to the main effects of the three drying technologies assessed in the research.

- The Abstract should contain tangible, quantitative, results, not only generalities

Abstract was added with quantitative data relevant to fiber content, polyphenol concentration, antioxidant activity, water holding capacity and oil holding capacity (lines 13-14, 15, 17).

L99: NL?

Text wascorrected (line 109).

Table 4: number of replicates and SDs?

Analyses were carried out at least three times in two replicated experiments (line 221-222). The standard deviation for polyphenol HPLC data was added in Table 3.

- Tables 1 and 3 should be merged.

According to Editor’s suggestion, data initially shown in Table3were merged to dataoriginally shown in Table 1 and 2.

- When you quote numbers, make sure you use the minimum number of significant digits or decimal places, as explained for example in Taylor, J. R. (1997). Error analysis: The study of uncertainties in physical measurements Sausalito, CA: University Science Books. Particularly, the following rules must be applied: 1.The mean cannot be more accurate than the original measurement. 2. The mean has the same significant digits as the standard deviation which determines the number of significant digits. 3. Standard deviation has been rounded to one significant digit (first value different to zero).

- Fig 1: The keys of symbols must be at the end of captions text and not all over the graph or below the x-axis. For titles of axes use the convention "Entity (unit). Please follow the format for this journal.

Figure 1 was modified as suggested.

- Fig 2: increase font sizes for the axes (numbers and axes titles).

Figure 2 was modified as suggested.

***Highlights (for review)**

1. Air-drying allows salad flours rich in fibre and antioxidants to be obtained
2. Freeze-drying partly maintains fresh salad structure, colour and polyphenols
3. Supercritical-CO_2-drying with ethanol as co-solvent produces highly porous flours

Application of different drying techniques to fresh-cut salad waste to obtain food ingredients rich in antioxidants and with high solvent loading capacity

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Abstract

Wastes from iceberg salad fresh-cut processing were submitted to air-drying, freeze-drying, and supercritical-CO_2-drying with or without ethanol as co-solvent. Drying was combined with grinding to obtain flours. Samples were analysed for macro- and micro-appearance, particle size, dietary fibre, polyphenol content, antioxidant activity, water vapour sorption, water and oil holding capacity. Air-drying produced a collapsed brown material allowing a flour rich in fibre (>260 g/kg) and polyphenols (3.05 mg GAE/g_{dw}) with antioxidant activity(6.04 OD^{-3}/min/g_{dw})to be obtained. Freeze-drying maintained vegetable structure and colour while partly retaining polyphenols(1.23 mg GAE/g_{dw}).Supercritical-CO_2-drying with ethanol as co-solvent, produced an expanded material able to entrap huge amounts of water and oil(43.2 and 35.2 g of water and oil for g of dry sample).Air-dried salad waste derivatives could be used as functional food ingredients, while supercritical-CO_2-dried ones can be exploited as bulking agents and absorbers of oil spills or edible oils.

Keywords: Drying techniques; Fibre; Polyphenol; Antioxidant activity; Food ingredients

1. Introduction

Salad represents about 50% of the entire fresh-cut market in Europe and US (Cook, 2015; Rabobank International, 2010). Although highly convenient, fresh-cut salad processing produces huge amounts of wastes. To this regard, a recent survey in a large Italian fresh-cut company revealed that, in a standard iceberg salad process, up to 40% of the initial salad weight is wasted due to removal of core and external leaves (Plazzotta, Manzocco, & Nicoli, 2017).

Salad waste can be exploited as soil conditioner or composted. However, these waste management strategies can absorb only limited amounts of salad residues, due to the risk of pathogen development and nitrate enrichment in soil and water. Biogas production from salad waste is also possible but with

low yields, requiring thus co-digestion with other organic wastes in centralized plants with high transport and disposal costs (Zheng, Phoungthong, Lü, Shao, & He, 2013).

Salad waste management would thus require redirection towards more profitable valorisation strategies. The latter are based on exploitation of vegetable waste to produce value-added derivatives, such asfood-grade dried materials and flours, rich infibre and antioxidants (Galanakis, 2012; Ferreira et al., 2015). Dried salad waste derivatives are expected to be microbiologically stable and have lower volume, reducing packaging, storage and transport issues (Ahmed, 2010; Karam, Petit, Zimmer, Baudelaire, & Marie, 2016).

The main drawback of salad waste drying lays in the cost of water removal from a material containing more than 900 g/kg moisture (Strumillo & Adamiec, 1996). Nevertheless, different drying techniques could be exploited to increase process affordability. Air-drying is based on the contact of wet materials with a hot air flow. Temperatures usually applied during air-drying to produce flours from fruit and vegetable wastes are generally in the range 65-90 °C (Ferreira et al., 2015; Nilnakara, Chiewchan, & Devahastin, 2009). The processis energy intensive and is associated to material shrinkage, hardness, poor appearance, reduced ability to rehydrate and bioactive loss. On the other hand, it is the most commonly applied food drying technique and has limited investment costs (Ratti, 2001; Strumillo & Adamiec, 1996). On the contrary, freeze-drying produces high-quality dried products, due to water removal by sublimation of ice crystals. However, equipment is costly, drying rates are low and much energy is consumed for freezing and vacuum phases(Ratti, 2001).Novel drying techniques, such as supercritical-CO_2-drying, have been claimed to increase environmental sustainability of traditional drying processes. In this case, water is slowly removed from the food material by a continuous supercritical-CO_2 flow. Temperature and pressure conditions are mild (20-50 °C and 10-20 MPa), guaranteeing a good bioactive retention. Moreover, co-solvents such as ethanol can be used to significantly reduce drying time(Brown, Fryer, Norton, Bakalis, & Bridson, 2008). Supercritical-CO_2-drying avoids the formation of vapour-liquid interfaces, allowing product structure to be preserved (Brown et al., 2008; García-González, Camino-Rey, Alnaief, Zetzl, & Smirnova, 2012). Investment and running costs are high but they could be counterbalanced using non-toxic carbon dioxide, which leaves no residues and can be recycled (Viganó, Machado, & Martínez, 2015).

Although drying of salad waste is costly, it could lead tangible profit due to the reduction of company waste management costs and the development of an eco-friendly image, highly appreciated by consumers(Vermeir & Verbeke, 2006).

The aim of the present work was to investigate the possibility to valorise fresh-cut iceberg salad waste by turning it into dried materials and flours via traditional (air-drying and freeze-drying) and novel (supercritical-CO_2-drying with or without ethanol as co-solvent) drying techniques. Dried salad waste and flours were analysed for macro- and micro-appearance, particle size, dietary fibre, polyphenol content, antioxidant activity, water vapour sorption, water and oil holding capacity. Results were discussed to suggest possible uses of salad waste submitted to different drying and grinding processes in food and non-food sectors.

2. Materials and methods

2.1. Salad waste preparation

A 10-kg batch of iceberg salad (*Lactuca sativa* var.*capitata*) was purchased at the local market and stored overnight at 4 °C. Outer leaves and core were manually removed from salad heads, simulating operations that are industrially carried out during fresh-cut salad processing. Salad waste amounted to351±35g/kg of the entire processed salad, with external leaves representing the majority of the overall waste (274±23 g/kg).Salad leaves were washed with flowing water (18±1 °C) and sanitized 20 min in a chlorinated bath containing 200 mg/L of NaClO with a100g/L salad/water ratio. Leaves were then rinsed with flowing water and centrifuged in a manual kitchen centrifuge (mod. ACX01, Moulinex, France) for 1 min (Manzocco, Ignat, Bartolomeoli, Maifreni, & Nicoli, 2015)Salad waste was manually chopped in homogeneous pieces (about 5 x 5 cm) with a sharp knife and immediately submitted to drying.

2.2. Salad waste drying

Air-drying

Salad waste (1 kg) was spread on a perforated tray in single layers and dried at 70±0.5 °C at a relative humidity in the drying chamber in the range 55-65%, using an air-drying oven (UM100, Memmert, Schwabach, Germany). During each experiment 3-5 g of the sample was taken out at various intervals to determine its moisture content. The air-drying oven operated until the mass of the sample reached the equilibrium value.

Freeze-drying

Salad waste (1 kg) was dried in single layers and frozen at -30 °C for 24 h and then freeze dried for 72 hat 4053 Pa by using the pilot plant model Mini Fast 1700 (Edwards Alto Vuoto, Milan, Italy).

Supercritical-CO_2-drying

An amount of 5 g salad waste was dried usingsupercritical-CO_2-drying with or without previous substitution of salad water with ethanol. In this case, salad waste was immersed(100 g/L) in pure ethanol (J.T.Baker, Centre Valley, USA) for 24 h twice. During this time, water was progressively removed from salad leaves, as indicated by monitoring the decrease in the alcoholic degree of the ethanol solution by a lab alcoholmeter (Alcolyzer plus, Anton Paar, Graz, Austria). Additional samples were prepared by grinding (MC3001, Moulinex, China) the salad waste submitted to ethanol substitution and subsequently removing excess solvent by vacuum filtration before supercritical-CO_2-drying.Supercritical-CO_2-drying was performed by using a plant developed at the Department of Agricultural, Food, Environmental and Animal Sciences (University of Udine), previously described by Manzocco, Valoppi, Calligaris, Andreatta, & Nicoli (2017). Sample was placed inside the reactor in which CO_2 was then pressurized at 11±1 MPa and 45 °C. The outlet flow through the reactor was set at 6.0L/min. This flow was selected since allowing drying time to be minimized while maintaining the structural integrity of the material as visually assessed. Samples in which water had been previously substituted with ethanol were considered dried when ethanol was no more detectable in the gaseous outlet. Decompression from 11 MPa to atmospheric pressure was then carried out in 30 min. In the case of samples not submitted to water substitution with ethanol, at increasing drying times, samples were removed from the reactor and weighted. The end of the drying process was set in correspondence of a residual moisture in the sample lower than 50 g/kg. Drying time was of 2.5, 5.0 and 1.5 hours for salad waste in which water was substituted with ethanol, for samples containing water and for ground samples, respectively.

2.3. Salad waste flour

Dried salad waste was finely ground using a ball mill (MM2, Retsch, Hann, Germania) for 15 min.

2.4. Sample storage

Dried salad waste and flours were stored at 20 °C in sealed aluminized aseptic bags until use.

2.5. Particle size distribution

An amount of 20 g of flour was sieved on a set of sieves with mesh sizes of 500, 250, 125, 63 and 20 µm (Endecotts Ltd, London, UK). The amount of flour remaining in each sieve was weighted and expressed with reference to the initial flour weight (g/kg).

2.6. Colour determination

Colour was determined using a tristimulus colorimeter (Chromameter-2 Reflectance, Minolta, Osaka, Japan) equipped with a CR-300 measuring head. The instrument was standardized against a white tile. Colour was expressed in L*, a* and b* Hunter scale parameters (Chen, Zhu, Zhang, Niu, & Du, 2010).

2.7. Image acquisition

Images were acquired using an image acquisition cabinet (Immagini& Computer, Bareggio, Italy) equipped with a digital camera (EOS 550D, Canon, Milan, Italy) placed on an adjustable stand, positioned 45 cm above a black cardboard base where samples were placed. Light was provided by 4 100 W frosted photographic floodlights, in a position allowing minimum shadow and glare. Images were saved in *jpeg* format resulting in 3456 x 2304 pixels.

2.8. Optical and electronic microscopy

Samples were observed at room temperature using a Leica DM 2000 optical microscope (Leica Microsystems, Heerburg, Switzerland). The images were taken at 200Xmagnification using a Leica EC3 digital camera and elaborated with the Leica Suite Las EZ software (Leica Microsystems, Heerburg, Switzerland).

For scanning electron microscopy, samples were mounted on aluminium sample holders and sputter coated with 10 nm of gold using a Sputter Coater 108 auto (Cressington Scientific Instruments, Watford, United Kingdom). The aluminium holder was transferred to the SEM unit (EVO 40XVP, Carl Zeiss, Milan, Italy), which was at ambient temperature and under vacuum. Samples were imaged using an acceleration voltage of 20 kV and SmartSEM v. 5.09 (Carl Zeiss, Milan, Italy) application software was used to capture images of the samples. Images were taken at 1000X magnification and saved in *tiff* format resulting in 1696 x 2048 pixels.

2.9.Moisture content

Moisture content was calculated according to AOAC gravimetric method (AOAC, 1997).

158 *2.10. Water vapour sorption*

159 Samples (2 g)were inserted into dried weighting bottles and transferred into desiccators containing
160 distilled water. Sample weight increase was monitored for 5 h during rehydration.

161

162 *2.11. Water and oil holding capacities*

163 Dried salad waste leaves (2 g)were immersed into water or sunflower oil for 24 h at room temperature
164 under gentle mixing. Samples were accurately drained on a wire mesh for 10 min.

165 In the case of flours, an accurately weighted amount of sample was inserted into tared 2-mL Eppendorf
166 tubes and added with 2 mL of distilled water or sunflower oil. Tubes were stirred using a vortex
167 (Vortex 1, Ika, Milan, Italy) three times for 30 s and centrifuged at 1327 x g 30 min (Mikro 20, Hettich
168 Zentrifugen, Tuttlingen, Germany). The sediment obtained after centrifugation was weighted. Water
169 and oil holding capacities were calculated as g of water or oil held by 1 g of dried sample.

170

171 *2.12. Total dietary fibre*

172 Total dietary fibre (TDF) was calculated according to the AOAC international method (AOAC, 1997)
173 using a total dietary fibre assay kit (TDF-100A, Sigma-Aldrich, St. Louis, Missouri, USA).

174

175 *2.13. Preparation of salad waste extract*

176 An amount of 10 g of salad waste, trimmed with a sharp knife, or flour were extracted by reflux with
177 boiling water for 60 min applying a dilution of 250 g/L and 50 g/L respectively. Extracts were cooled
178 at room temperature, vacuum filtered thorough Whatman no. 1 filter paper (Maidstone, UK), freeze-
179 dried at -50 °C and stored in a desiccator containing P_2O_5 at room temperature until use.

180

181 *2.14.Totalpolyphenolic content*

182 Total polyphenolic content (TPC) was determined using Folin-Ciocalteau reagent (Singleton & Rossi,
183 1985). The reaction mixture contained 100 µL of salad waste extract solubilised in water (0.1 g/mL),
184 500 µL of the Folin-Ciocalteau reagent, 4 mL of water and 2 mL of a sodium carbonate-water solution
185 (0.15 g/mL). After 2 h reaction at ambient temperature, mixture absorbance was read at 750 nm using
186 US-Vis spectrophotometer (Shimadzu UV-2501PC, UV-Vis recording spectrophotometer, Shimadzu
187 Corporation, Kyoto, Japan). A calibration curve was made with standard solutions of gallic acid in the
188 range 0.1–1000 mg/L (R^2=0.99). Results were expressed as mg of gallic acid equivalents per g of dry
189 weight (mg GAE/g_{dw}).

2.15. HPLC

Freeze-dried extracts (10 mg)were dissolved in 1 mL of distilled water, filtered through a 0.45 µm membrane filter (GVS, Meckenheim, Germany)and analysed using a HPLC system equipped with a Prostar 230 pump (Varian, Walnut Creek, USA) and a Prostar 330diode array detector(Varian, Walnut Creek, California, USA). To this aim, 20 µL sample was injected in a C18 column (Alltima, 5 microns, 250 x 4.6 mm, Grace, Lokeren, Belgium). The mobile phase was waterwith 50 mL/L formic acid (Fluka, St. Louis, Missouri, USA) (solvent A) and HPLC grade methanol (Chromasol≥99.9%, Sigma-Aldrich St. Louis, Missouri, USA)(solvent B) at a flow rate of 1 mL/min. The linear gradient started with 10% B in A to reach 20% B at 25 min, 50% B at 40, 50% B at 45 min and 90% B at 60 min. Chromatograms were recorded at 335nm. Data elaboration was performed by Polyview program (v.5.3).Phenolic compounds identification was based on their UV spectra and retention times (DuPont, Mondin, Williamson, & Price, 2000; Llorach, Barberàn, & Ferreres, 2004; Tomás-Barberán, Loaiza-Velarde, Bonfanti, & Saltveit, 1997). Chicoric acid was quantified (Lee & Scagel, 2013) using an external standard while other compounds were quantified as 3-O-caffeoylquinic acid by comparison with external standard (Sigma-Aldrich, St. Louis, Missouri, USA).

2.16. Chain-breaking activity (DPPH· assay)

The chain-breaking activity (CBA) was measured following the bleaching rate of 2,2-diphenyl-1-picrylhydrazyl (DPPH·) in the presence of the sample. 3mL of 6.1×10^{-5} M DPPH· methanol solution was used. The reaction was started by the addition of 150 µL of salad waste extract solubilised in water (0.1 g/mL). DPPH· bleaching was followed at 515nm (UV-2501PC, UV-Vis Recording Spectrophotometer, Shimadzu Corporation, Kyoto, Japan) at 25 °C for 10 min. DPPH· bleaching rate was proportional to sample concentration. The following equation was chosen to obtain the reaction rate of DPPH· bleaching, *k*(Manzocco, Mastrocola, & Nicoli, 1999):

$$\frac{1}{A^3} - \frac{1}{A_0^3} = 3kt \qquad (1)$$

where A_0 is the initial optical density (OD) and A is the optical density at increasing time, t. The chain-breaking activity was expressed as k/g of dry weight ($OD^{-3}/min/g_{dw}$). The chain-breaking phenolic ratio (CBP) was also determined dividing the chain-breaking activity of the sample by its phenol content ($OD^{-3}/min/mg_{GAE}$).

2.17. Data analysis

Analyses were carried out at least three times in two replicated experiments. Analysis of variance ($p<0.05$) and linear regression analysis were performed using R (The R foundation for statistical computing, v.3.1.1). Goodness-of-fit was evaluated based on $R^2$and p-values.

3. Results and discussion

3.1. Characterization of salad waste

External leaves of iceberg salad from fresh-cut processing presented the typical green colour and a moisture level exceeding900 g/kg (Table 1 and 2). Salad waste resulted particularly rich in fibre, in agreement with nutritional databases relevant to edible salad (USDA, 2016). By contrast, salad waste polyphenol content resulted about 4 times lower than that reported by Llorach et al. (2004). Different factors, including agronomic practices, salad variety and extraction solvent, could significantly affect polyphenol quantification (Llorach, Martìnez-Sànchez, Tomàs-Barberan, Gil, & Ferreres, 2008). Nevertheless, due to its polyphenol content, comparable to that of grape marc, salad waste can be considered an always-available and cheap source of antioxidants (Table 2) (Bonilla, Mayen, Merida, & Medina, 1999).

3.2. Characterization of dried salad waste

Water content of salad waste makes it microbiologically unstable, posing critical management issues. To increase its stability, dehydration could be performed, as proposed for other vegetable wastes (Annadurai, Juang, & Lee, 2002; de Oliveira et al., 2009). Air-dried (AD), freeze-dried (FD), supercritical-CO_2-driedsamples were thus prepared. The latter were produced in the absence (SCCD sample) or presence (SCCD-EtOH sample) of ethanol as co-solvent.

Drying techniques exerted different effects on salad waste colour (Table 1). AD sample appeared brown, due to enzymatic and non-enzymatic reactions, prevailing in the initial and advanced phases of the process, respectively (Adam, Mühlbauer, Esper, Wolf, & Spiess, 2000). FD leaves maintained the original colour, confirming the ability of freeze-drying to minimize quality damage (Argyropoulos, Heindl, & Mu, 2011). Similarly, SCCD samples resulted green, suggesting this technology as a valid alternative to freeze-drying (Brown et al., 2008). Interestingly, SCCD-EtOH sample completely lost the original colour, probably due to pigment extraction during salad immersion into ethanol. In fact, the SCCD sample, which had not been immersed in ethanol, highly retained the original colour (Table 1). Pigment extraction by supercritical-$CO_2$was probably negligible since the pressure here applied (<12 MPa) was lower than that required for chlorophyll extraction (>25 MPa) (Guedes et al., 2013).

The drying technique strongly affected sample physical structure, as shown by visual appearance and microscopic analyses (Table 1).AD samples resulted severely shrunk, since water evaporation created intense capillary tensions in cellular structure(Ahmed, 2010). On the contrary, FD samples maintained

cellular organization thanks to water removal by sublimation of ice, which provides structural rigidity (Ratti, 2001). However, no clear morphology of cells was revealed by SEM, probably due to the presence of the typical wax protective layer on vegetable surface. SCCD samples appeared completely collapsed. This phenomenon was prevented by adding co-solvents during drying (Table 1). Cells of SCCD-EtOH sample were actually visible and appeared even swallowed in microscopic images. Similar effects were also observed in carrot slices and can be attributed to tissue expansion during CO_2decompression (Brown et al., 2008). In addition, in SEM image of SCCD-EtOH sample, no protective wax layer was evident onto sample surface, probably due to its solubilization in the supercritical-CO_2 flow (Roy, Goto, Kodama, & Hirose, 1996).

To better assess the effects of drying treatments on salad waste properties, the ability of the dried leaves to interact with water vapour was evaluated (Figure 1A).All samples showed a progressive vapour adsorption upon maintenance in a moisture-rich atmosphere. The evolution of vapour sorption was significantly affected by the drying technique. AD and SCCD samples showed a slow vapour uptake, probably due to their dense microstructure(Table 2)(Argyropoulos et al., 2011; Ratti, 2001).A faster water vapour sorption was observed for FD sample, which well maintained structure (Table1). The expanded SCCD-EtOH sample (Table 1) showed the fastest and highest vapour uptake. These findings suggest that drying-induced structure deeply affects the ability of samples to interact with solvents. To confirm this hypothesis, samples were analysed for water and oil holding capacity (WHC, OHC). AWHC much higher than OHC was observed for all samples (Table1), being vegetable waste rich in hydrophilic polysaccharides (Ferreira et al., 2015). SCCD-EtOH sample showed the highest WHC and OHC values(Table 1). Excellent rehydration properties were also observed for carrot slices submitted to supercritical-CO_2-drying using ethanol as co-solvent (Brown et al., 2008). Rehydration ability was attributed to the capacity of supercritical-drying with ethanol as co-solvent to beget highly porous materials, favouring water capillary adsorption. Interestingly, the amount of water held by 1 g of SCCD-EtOH sample resulted much higher than that originally present in the fresh salad waste tissue (*circa* 16 g H_2O/g_{dw}, as computed based on moisture content, Table 2).The capacity of SCCD-EtOH sample to absorb water beyond the amount entrapped in the native plant tissue could be attributed to the expanded structure obtained by supercritical-CO_2-drying and to water solvation of polysaccharides, which would favour sample swallowing. By contrast, oil adsorption did not promote swallowing of sample, which retained *circa* 16 g oil/g_{ds}, indicating that oil simply substituted voids left upon water removal.

The interesting ability of dried salad wastes to interact with water and oil suggeststheir possible exploitation as ingredients in dried instant foods (e.g. soups, noodles, meat).

3.3. Characterization of salad waste flours

The possibility to valorise salad waste by turning it into flours was studied. The attention was focused on AD, FD and SCCD-EtOH salad wastes. SCCD sample was not considered since characterised by a collapsed structure with low WHC and OHC (Table 1, Figure 1A). AD and FD samples were ground to flour with a 95% yield. On the contrary, grinding yield of SCCD-EtOH sample resulted<10%, possibly due to the difficulty in grinding an expanded tissue. The flour was thus obtained by grinding salad waste after ethanol substitution before supercritical-CO_2-drying. All salad flours presented most particles in the range 200-250 µm (Table 2). However, a lower size particle fraction was observed in flours from AD and FD samples, confirming their grinding to be particularly efficacious. Samples showed similar moisture and fibre content (Table 2). The latter resulted higher than that of rice (210 g/kg) and oat (150 g/kg) bran(USDA, 2016), suggesting the possible suitability of salad flours as ingredients to increase fibre content of foods (e.g. instant foods, bakery products).

Drying treatment significantly affected both polyphenol content and antioxidant activity of flours(Table 2).AD flours showed the highest polyphenol content and antioxidant activity, which resulted significantly higher than those of fresh sample ($p<0.05$) (Table 2). This can be attributed to the formation of partially-oxidised polyphenols and Maillard reaction products able to react with Folin-Ciocalteau reagent and with a prominent antioxidant action (Mrkìc, Cocci, Dalla Rosa, & Sacchetti, 2006). Freeze-drying allowed polyphenol content and antioxidant activity of fresh salad waste to be partly retained (Table 2). Due to the low process temperature and almost complete absence of oxygen, degradation reactions are minimized during freeze-drying (Michalska, Wojdyło, Lech, Łysiak, & Figiel, 2017).Nevertheless, phenols could be enzymatically oxidised upon enzyme de-compartmentalization during freezing (Chang, Lin, Chang, & Liu, 2006). SCCD-EtOH flour presented a phenolic content lower than that of FD sample, probably due to partial polyphenol extraction by supercritical-CO_2. The latter is actually applied for polyphenol extraction from vegetable matrices (Cavalcanti, Navarro-Díaz, Santos, Rostagno, & Angela, 2012; Gadkari, Balarman, & Kadimi, 2015).

HPLC was performed for polyphenol qualitative (Figure 2) and quantitative (Table 3) analyses. HPLC profile of fresh salad waste revealed the presence of different phenolic acids, in agreement with literature (Llorach et al., 2004). The main identified phenolic acid was dicaffeoyltartaric acid (chicoric acid) (peak 7), followed bycaffeoyltartaric acid (peak 2) and 5-O-caffeoylquinic acid (peak 4). The

latter can isomerise in warm aqueous phase, leading to 3-O-caffeoylquinic acid (peak 1) and 4-O-caffeoylquinic acid (peak 3) (Llorach, Carlos, Tomás-Barberán, & Ferreres, 2003). Flavonoid compounds, such as luteolin derivatives (luteolin 7-O-glucuronide, peak 9) and quercetin derivatives (quercetin 3-O-glucuronide, peak 10) were also detected. Independently on the applied technology, salad waste drying always promoted a severe decrease in the intensity of peaks relevant to naturally occurring polyphenols (Table 3). However, AD flour chromatogram also showed an intense peak at low retention times(5.8 min), probably ascribable to Maillard reaction compounds, which can account for the high antioxidant activity of this flour (Table 2) (Mrkìc et al., 2006). Drying technology thus affected not only content but also composition of flour phenols(Table 3) and, consequently, their chain-breaking activity. This was confirmed by the chain-breaking phenolic ratio (CBP, Table 2) that allows comparison of antiradical activity of samples with different phenolic content (Manzocco et al., 1999). AD flours showed the highest CBP, confirming the high antioxidant activity of compounds formed during air-drying. FD and SCCD-EtOH flours presented CBP similar to that of fresh samples (Table 2), suggesting supercritical-CO_2-drying as a suitable technology for producing high-quality dried products (Brown et al., 2008).

Salad waste flours were then evaluated for their water vapour sorption (Figure 1B).As expected, vapour uptake of flours, which have high absorptive surface, was higher than that observed in the not-ground samples (Figure 1A). Flour vapour uptake was in the order AD<FD<SCCD-EtOH, in accordance with decreasing sample structural collapse upon drying (Table 1). SCCD-EtOH flour also showed the highest WHC and OHC values (Table 2).Moreover, SCCD-EtOH flour presented a similar tendencytointeract with water and oil (Table 2). It can be inferred that performing grinding before supercritical-CO_2-drying allowed obtaining an extremely porous flour with excellent solvent-loading capacity and in which absorption would be mainly driven by capillary forces rather than chemical interactions. Large amounts of different solvents could be thus easily embedded into the pores of SCCD-EtOH flour. This property could have interesting practical relevance, suggesting the possible exploitation ofthis flour asoil spill absorber or bulking agent in food formulations. It could also be used to structure liquid oil, leading to the development of innovative materials, such as oleogels, able to simulate technological performances of fats while reducing saturated fatty acid content.

4. Conclusions

Salad waste drying represents a possible strategy to valorise this critical industrial discard by obtaining derivatives rich in fibre and antioxidant compounds with tailored physico-chemical properties.

Thesecan be steered by exploiting different drying mechanisms such as evaporation, sublimation or supercritical-fluid extraction. In this latter case, grinding before drying and using ethanol as co-solvent allowed obtaining a flour with excellent ability to absorb both water and oil.

In particular, air-dried materials and flours from salad waste could be exploited as functional food ingredients, while supercritical-dried ones as bulking agents or oil absorbers. The latter could be applied not only to absorb oil spills but also edible oils, thus begetting novel materials, such as oleogels, able to simulate the technological performance of fats while having a much lower saturated fatty acid content. The selection of the drying technology should be driven by proper considerations about target use, process costs and product sustainability. In addition, salad waste derivatives intended for food-use should be accurately assessed for safety aspects such as microbial quality and presence of contaminants deriving from cultivation practises, as well as for their sensory properties and technological performances.

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References

Adam, E., Mühlbauer, W., Esper, A., Wolf, W., & Spiess, W. (2000). Quality changes of onion (*Allium cepa*) as affected by the drying process. *Food / Nahrung*, *44*, 32–37.

Ahmed, J. (2010). Drying of vegetables: principles and dryer design. In *Handbook of Vegetables and Vegetable Processing* (pp. 279–298). Hoboken: Wiley-Blackwell.

Annadurai, G., Juang, R. S., & Lee, D. J. (2002). Use of cellulose-based wastes for adsorption of dyes from aqueous solutions. *Journal of Hazardous Materials*, *92*, 263–274.

AOAC (1997). Official methods of analysis. Washington, DC: Association of Official Analytical Chemists.

Argyropoulos, D., Heindl, A., & Mu, J. (2011). Assessment of convection, hot-air combined with microwave-vacuum and freeze-drying methods for mushrooms with regard to product quality. *International Journal of Food Science & Technology*, *46*, 333–342.

Bonilla, F., Mayen, M., Merida, J., & Medina, M. (1999). Extraction of phenolic compounds from red grape marc for use as food lipid antioxidants. *Food Chemistry*, *66*, 209–215.

Brown, Z. K., Fryer, P. J., Norton, I. T., Bakalis, S., & Bridson, R. H. (2008). Drying of foods using supercritical carbon dioxide-Investigations with carrot. *Innovative Food Science & Emerging Technologies*, *9*, 280–289.

Cavalcanti, R. N., Navarro-Díaz, H. J., Santos, D. T., Rostagno, M. A., & Angela, M. A. (2012). Supercritical carbon dioxide extraction of polyphenols from pomegranate (*Punica granatumL.*) leaves: chemical composition, economic evaluation and chemometric approach. *Journal of Food Research*, *1*, 282–294.

Chang, C. H., Lin, H. Y., Chang, C. Y., & Liu, Y.C. (2006). Comparisons on the antioxidant properties of fresh, freeze-dried and hot-air-dried tomatoes. *Journal of Food Engineering*, *77*, 478–485.

Chen, Z., Zhu, C., Zhang, Y., Niu, D., & Du, J. (2010). Postharvest biology and technology effects of aqueous chlorine dioxide treatment on enzymatic browning and shelf-life of fresh-cut asparagus lettuce (*Lactuca sativa L.*). *Postharvest Biology and Technology*, *58*, 232–238.

de Oliveira, A. C., Valentim, I. B., Silva, C. A., Bechara, E. J. H., de Barros, M. P., Mano, C. M., & Goulart, M. O. F. (2009). Total phenolic content and free radical scavenging activities of methanolic extract powders of tropical fruit residues. *Food Chemistry*, *115*, 469–475.

DuPont, M. S., Mondin, Z., Williamson, G., & Price, K. R. (2000). Effect of variety, processing, and storage on the flavonoid glycoside content and composition of lettuce and endive. *Journal of Agricultural and Food Chemistry*, *48*, 3957–3964.

Ferreira, M. S. L., Santos, M. C. P., Moro, T. M. A., Basto, G. J., Andrade, R. M. S., & Gonçalves, E. C. B. A. (2015). Formulation and characterization of functional foods based on fruit and vegetable residue flour. *Journal of Food Science and Technology*, *52*, 822–830.

Gadkari, P. V., Balarman, M., & Kadimi, U. S. (2015). Polyphenols from fresh frozen tea leaves (Camellia assamica L.,) by supercritical carbon dioxide extraction with ethanol entrainer-application of response surface methodology. *Journal of Food Science and Technology*, *52*, 720–730.

Galanakis, C. M. (2012). Recovery of high added-value components from food wastes: Conventional, emerging technologies and commercialized applications. *Trends in Food Science and Technology*, *26*, 68–87.

García-González, C. A., Camino-Rey, M. C., Alnaief, M., Zetzl, C., & Smirnova, I. (2012). Supercritical drying of aerogels using CO_2: Effect of extraction time on the end material textural properties. *The Journal of Supercritical Fluids*, *66*, 297–306.

Guedes, A. C., Gião, M. S., Matias, A. A., Nunes, A. V. M., Pintado, M. E., Duarte, C. M. M., & Malcata, F. X. (2013). Supercritical fluid extraction of carotenoids and chlorophylls a, b and c, from a wild strain of *Scenedesmus obliquus* for use in food processing. *Journal of Food Engineering*, *116*, 478–482.

Karam, M. C., Petit, J., Zimmer, D., Baudelaire, E., & Marie, C. (2016). Effects of drying and grinding in production of fruit and vegetable powders : A review. *Journal of Food Engineering*, *188*, 32–49.

Lee, J., & Scagel, C. F. (2013). Chicoric acid: chemistry, distribution, and production. *Frontiers in Chemistry*, *1*. http://doi.org/10.3389/fchem.2013.00040.

Llorach, R., Barberàn, T., & Ferreres, F. (2004). Lettuce and chicory by-products as a source of antioxidant phenolic extracts. *Journal of Agricultural and Food Chemistry*, *52*, 5109–5116.

Llorach, R., Carlos, E. J., Tomás-Barberán, F. A., & Ferreres, F. (2003). Valorization of cauliflower (*Brassica oleracea L.* var. *botrytis*) by-products as a source of antioxidant phenolics. *Journal of Agricultural and Food Chemistry*, *51*, 2181–2187.

Llorach, R., Martìnez-Sànchez, A., Tomàs-Barberan, F. A., Gil, M. I., & Ferreres, F. (2008). Characterisation of polyphenols and antioxidant properties of five lettuce varieties and escarole. *Food Chemistry*, *108*, 1028–1038.

Manzocco, L., Ignat, A., Bartolomeoli, I., Maifreni, M., & Nicoli, M. C. (2015). Water saving in fresh-cut salad washing by pulsed light. *Innovative Food Science and Emerging Technologies*, *28*, 47–51.

Manzocco, L., Mastrocola, D., & Nicoli, M. C. (1999). Chain-breaking and oxygen scavenging properties of wine as affected by some technological procedures. *Food Research International*, *31*, 673–678.

Manzocco, L., Valoppi, F., Calligaris, S., Andreatta, F., & Nicoli, M. C. (2017). Exploitation of κ-carrageenan aerogels as template for edible oleogel preparation. *Food Hydrocolloids*, *71*, 68–75.

Michalska, A., Wojdyło, A., Lech, K., Łysiak, G. P., & Figiel, A. (2017). Effect of different drying techniques on physical properties, total polyphenols and antioxidant capacity of blackcurrant pomace powders. *LWT-Food Science and Technology*, *78*, 114–121.

Mrkìc, V., Cocci, E., Dalla Rosa, M., & Sacchetti, G. (2006). Effect of drying conditions on bioactive compounds and antioxidant activity of broccoli (*Brassica oleracea L.*). *Journal of the Science of Food and Agriculture*, *1566*, 1559–1566.

Nilnakara, S., Chiewchan, N., & Devahastin, S. (2009). Production of antioxidant dietary fibre powder from cabbage outer leaves. *Food and Bioproducts Processing*, 87, 301–307.

Plazzotta, S., Manzocco, L., & Nicoli, M. C. (2017). Fruit and vegetable waste management and the challenge of fresh-cut salad. *Trends in Food Science & Technology*, *63*, 51–59.

Ratti, C. (2001). Hot air and freeze-drying of high-value foods: A review. *Journal of Food Engineering*, *49*, 311–319.

Roy, B. C., Goto, M., Kodama, A., & Hirose, T. (1996). Extraction of essential oils and cuticular waxes from peppermint leaves. *Journal of Chemical Technology & Biotechnology*, *67*, 21–26.

Singleton, V. L., & Rossi, J. A. J. (1985). Colorunetry of total phenolics with phosphomolybdic-phosphotungstic acid reagents. *American Journal Of Enology and Viticulture*, *16*, 144–158.

Strumillo, C., & Adamiec, J. (1996). Energy and quality aspects of food drying. *Drying Technology*, *14*, 423–448.

Tomás-Barberán, F. A., Loaiza-Velarde, J., Bonfanti, A., & Saltveit, M. E. (1997). Early wound- and ethylene-induced changes in phenylpropanoid metabolism in harvested lettuce. *Journal of the American Society for Horticultural Sciences*, *130*, 466–476.

Vermeir, I., & Verbeke, W. (2006). Sustainable food consumption: Exploring the consumer "attitude-behavioral intention" gap. *Journal of Agricultural and Environmental Ethics*, *19*(2), 169–194.

Viganó, J., Machado, A. P. D. F., & Martínez, J. (2015). Sub- and supercritical fluid technology

applied to food waste processing. *The Journal of Supercritical Fluids*, *96*, 272–286.

Zheng, W., Phoungthong, K., Lü, F., Shao, L. M., & He, P. J. (2013). Evaluation of a classification method for biodegradable solid wastes using anaerobic degradation parameters. *Waste Management*, *33*, 2632–40.

Web references

Cook, R. (2015). Trends in the marketing of fresh produce and fresh-cut products, Department of Agriculture and Resource Economic, University of California Davis. http://ucanr.edu/datastoreFiles/234-2822.pdf.Accessed 28.06.17.

Rabobank International. (2010). The EU fresh-cut fruits and vegetables market. Fresh convenience conference. London. http://www.freshconveniencecongress.com/resources/documents/1308561709cindyvanrijswick.pdf. Accessed 28.06.17.

USDA (2016). Food composition databases. https://ndb.nal.usda.gov/ndb/. Accessed 28.06.17.

Figure captions

Figure 1. Adsorption of water vapour of dried salad waste leaves (A) and salad waste flours (B) submitted to air-drying (▲), freeze-drying (■), and supercritical-CO_2-drying without (**X**) or with (◆) ethanol. ds = dry sample.

Figure 2. HPLC profiles of water extracts of fresh salad waste (Fresh) and flour samples obtained by air-drying (AD), freeze-drying (FD) and supercritical-CO_2-drying using ethanol as co-solvent (SCCD-EtOH). Peak identification: (1) 3-O-caffeoylquinic acid; (2) caffeoyltartaric acid; (3) 4-O-caffeoylquinic acid; (4) 5-O-caffeoylquinic acid; (5) caffeic acid derivative; (6) isochlorogenic acid; (7) chicoric acid; (8) caffeic acid derivative; (9) luteolin 7-O-glucuronide; (10) quercetin 3-O-glucuronide. AU = arbitrary units.

Table captions

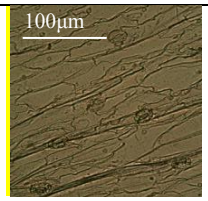
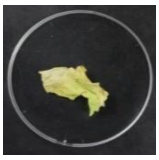
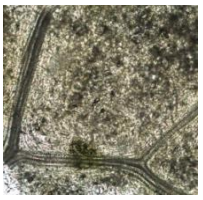
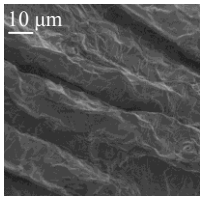
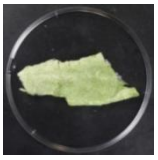
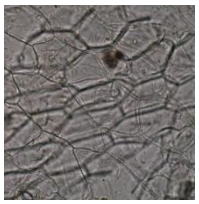
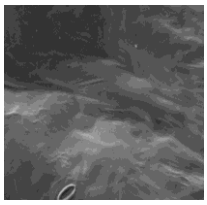
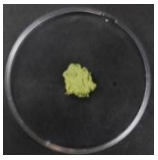
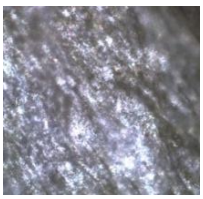
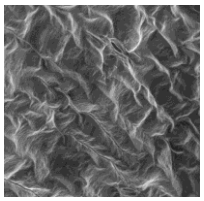
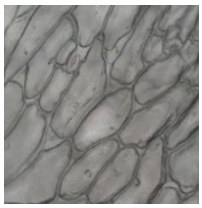
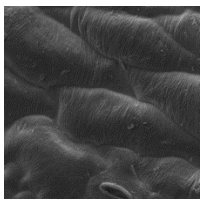
Table 1. Visual appearance, hunter scale colour parameters (L*, a*, b*), microscopic images (optical and SEM), water and oil holding capacities (WHC, OHC) of fresh salad waste leaves and dried samples obtained using air-drying (AD), freeze-drying (FD), and supercritical-CO_2-drying without (SCCD) or with (SCCD-EtOH) ethanol.

Table 2. Particle size distribution, moisture, total dietary fibre (TDF), total phenolic content (TPC), relevant chain-breaking activity (CBA), chain-breaking phenolic ratio (CBP) and water and oil holding capacities (WHC, OHC) of fresh salad waste and of flour samples obtained using air-drying (AD), freeze-drying (FD), and supercritical-CO_2-drying with ethanol as co-solvent (SCCD-EtOH).

Table 3. Quantification of phenolic compounds identified by HPLC in fresh salad waste (Fresh) and in flour samples obtained using air-drying (AD), freeze-drying (FD), and supercritical-CO_2-drying with ethanol as co-solvent (SCCD-EtOH).

Table

Table 1. Visual appearance, hunter scale colour parameters (L*, a*, b*), microscopic images (optical and SEM), water and oil holding capacities (WHC, OHC) of fresh salad waste leaves and dried samples obtained using air-drying (AD), freeze-drying (FD), and supercritical-CO_2-drying without (SCCD) or with (SCCD-EtOH) ethanol.

Salad waste	Visual appearance	Colour			Microscopy		WHC	OHC
		L*	a*	b*	Optical	SEM	(g H_20/g_{ds})	(g oil/g_{ds})
Fresh		71.4±1.3^{b}	-16.9±1.2^{e}	31.6±1.4^{a}		n.d.	n.d.	n.d.
AD		61.2±0.3^{c}	-1.5±0.2^{b}	23.2±0.1^{b}			5.3±0.6^{b}	1.1±0.2^{b}
FD		75.6±0.3^{b}	-11.2±0.1^{d}	18.4±0.2^{c}			7.5±0.4^{b}	2.1±0.4^{b}
SCCD		53.4±2.0^{d}	-5.2±0.7^{c}	15.9±2.2^{c}			4.2±0.9^{b}	1.0±0.4^{b}
SCCD-EtOH		85.0±2.4^{a}	-0.2±0.1^{a}	8.7±0.4^{d}			37.1±1.1^{a}	16.3±1.7^{a}

$^{a, b, c, d}$ In the same column, mean values indicated by different letters are statistically different ($p<0.05$); n.d. = not determined; ds = dry sample.

Table 2. Particle size distribution, moisture, total dietary fibre (TDF), total phenolic content (TPC), relevant chain-breaking activity (CBA), chain-breaking phenolic ratio (CBP) and water and oil holding capacities (WHC, OHC) of fresh salad waste and of flour samples obtained using air-drying (AD), freeze-drying (FD), and supercritical-CO_2-drying with ethanol as co-solvent (SCCD-EtOH).

Salad waste sample	Particle size (g/kg)		Moisture (g/kg)	TDF (g/kg)	TPC (mg GAE/g_{dw})	CBA (OD^{-3}/min/g_{dw})	CBP (OD^{-3}/min/mg_{GAE})	WHC (g H_20/g_{ds})	OHC (g oil/g_{ds})
	200-250 µm	<200 µm							
Fresh	n.d.	n.d.	941 ± 12^{nc}	16.1 ± 2.0^{nc}	1.84 ± 0.02^{b}	6.04 ± 0.79^{b}	4.17 ± 0.54^{bc}	n.d.	n.d.
AD flour	942 ± 9^{b}	61 ± 4^{a}	40 ± 1^{a}	266 ± 4^{a}	3.05 ± 0.08^{a}	27.03 ± 1.60^{a}	8.87 ± 0.13^{a}	9.1 ± 0.7^{c}	2.3 ± 0.4^{b}
FD flour	928 ± 1^{b}	80 ± 6^{a}	46 ± 2^{a}	266 ± 4^{a}	1.23 ± 0.01^{bc}	4.01 ± 0.05^{b}	3.22 ± 0.02^{c}	12.5 ± 0.6^{bc}	3.2 ± 0.3^{b}
SCCD-EtOH flour	996 ± 3^{a}	2 ± 1^{b}	39 ± 8^{a}	272 ± 3^{a}	0.84 ± 0.01^{c}	3.38 ± 0.08^{b}	4.04 ± 0.06^{bc}	43.2 ± 0.4^{a}	35.2 ± 0.7^{a}

[a, b, c] In the same column, mean values indicated by different letters are statistically different ($p<0.05$); [nc] not computed in statistical analysis; n.d. = not determined; dw = dry weight; ds = dry sample.

Table 3. Quantification of phenolic compounds identified by HPLC in fresh salad waste (Fresh) and in flour samples obtained using air-drying (AD), freeze-drying (FD), and supercritical-CO_2-drying with ethanol as co-solvent (SCCD-EtOH).

Phenolic compounds (mg/g_{dw})	Retention time (min)	Sample			
		Fresh	AD flour	FD flour	SCCD-EtOH flour
3-O-caffeoylquinic acid	10.5±0.1	0.014±0.004	0.002±0.001	0.010±0.001	ND
Caffeoyltartaric acid	12.8±0.2	0.158±0.003	0.060±0.003	0.074±0.004	0.023±0.002
4-O-caffeoylquinic acid	21.5±0.1	0.012±0.001	ND	ND	ND
5-O-caffeoylquinic acid	22.5±0.1	0.074±0.003	ND	0.003±0.001	ND
Caffeic acid derivative	23.3±0.1	0.036±0.002	ND	0.002±0.001	ND
Isochlorogenic acid	33.0±0.3	0.007±0.001	ND	ND	ND
Chicoric acid	38.1±0.2	0.187±0.002	0.044±0.002	0.040±0.006	0.002±0.001
Caffeic acid derivative	38.5±0.1	0.007±0.001	ND	ND	ND
Luteolin 7-O-glucuronide	42.4±0.1	0.007±0.001	ND	ND	ND
Quercetin 3-O-glucuronide	42.8±0.1	0.011±0.001	ND	ND	ND

dw = dry weight; ND = not detected

Figure

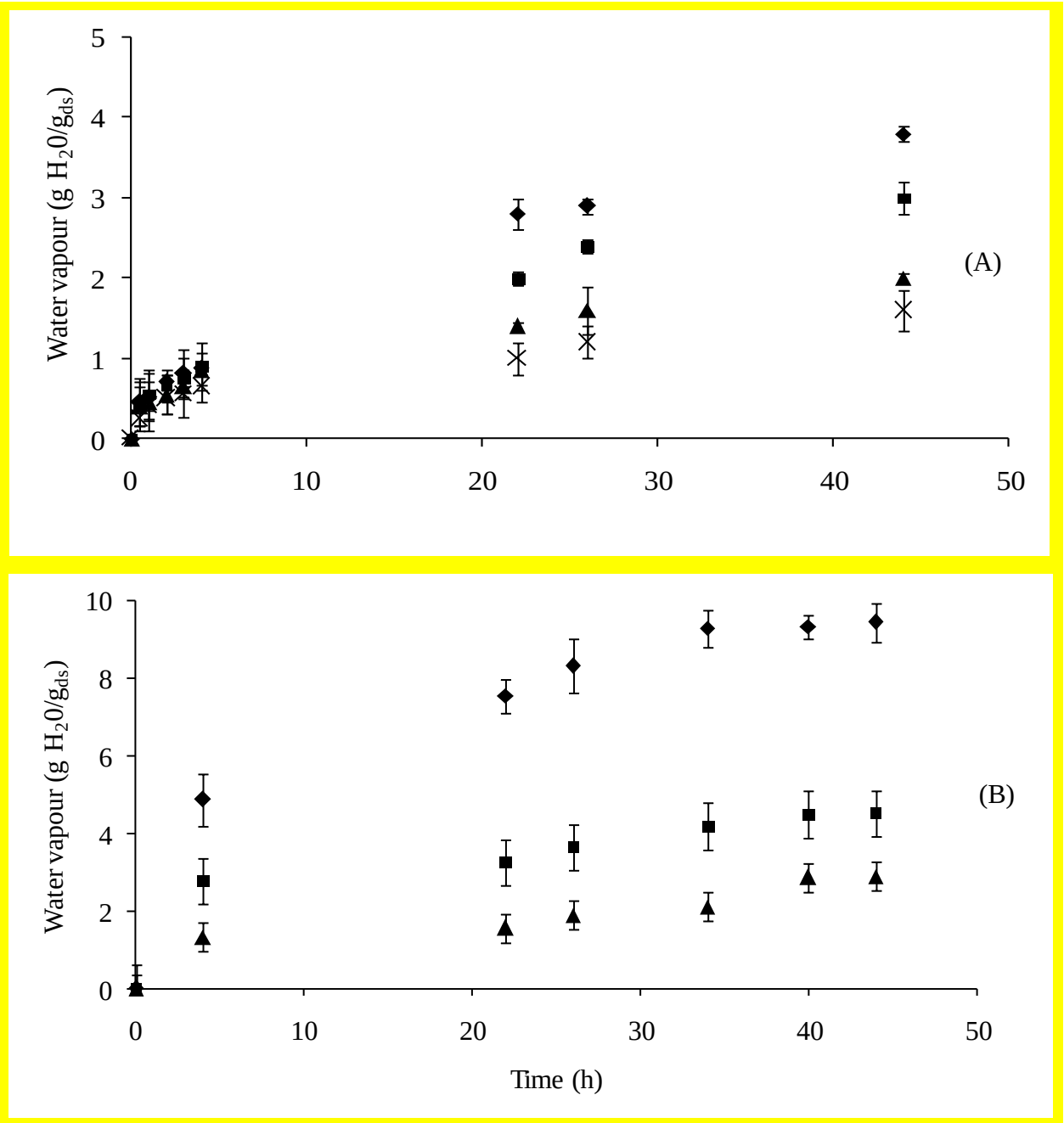


Figure 1. Adsorption of water vapour of dried salad waste leaves (A) and salad waste flours (B) submitted to air-drying (▲), freeze-drying (■), and supercritical-CO_2-drying without (**X**) or with (◆) ethanol. ds = dry sample.

Figure

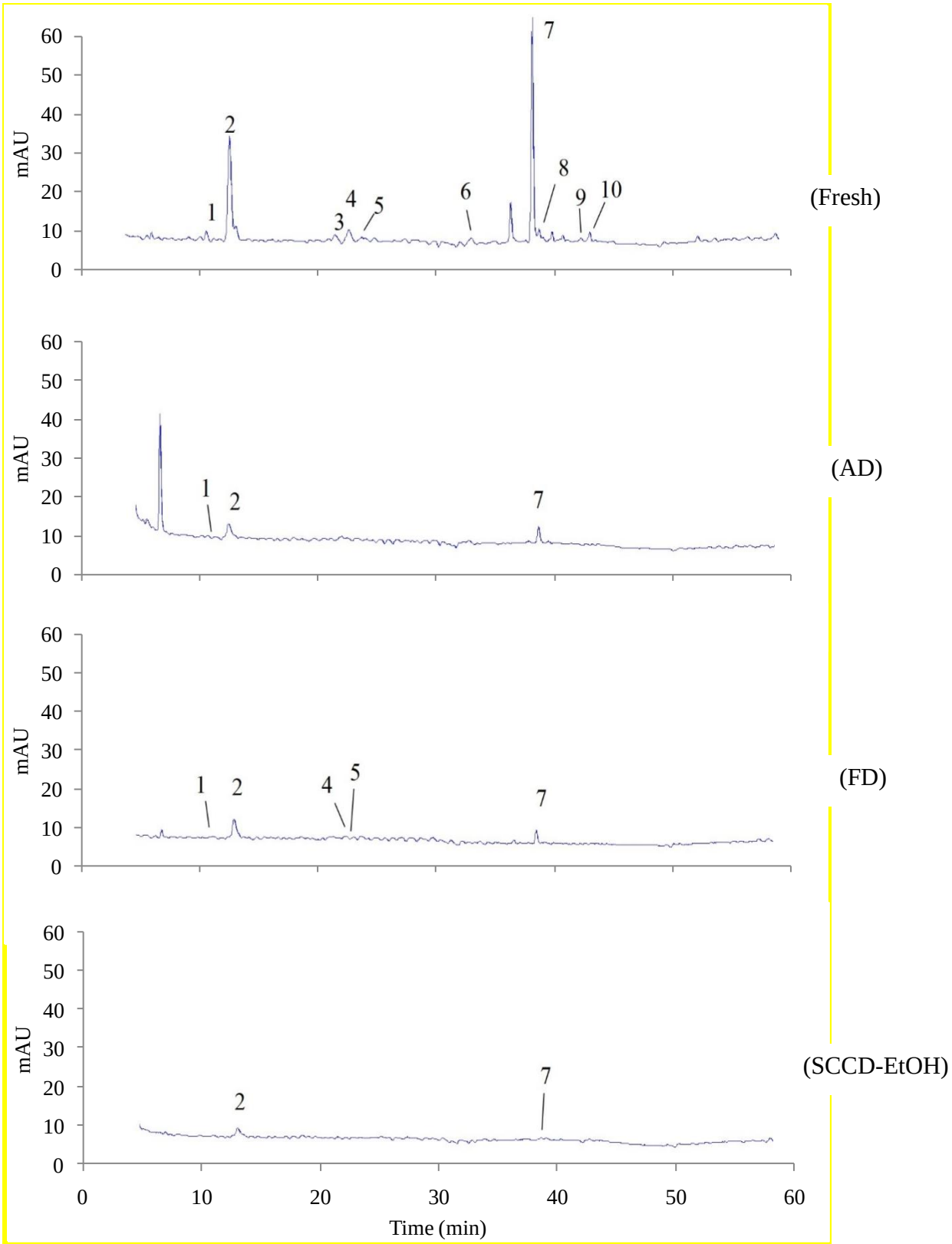


Figure 2. HPLC profiles of water extracts of fresh salad waste (Fresh) and flour samples obtained by air-drying (AD), freeze-drying (FD) and supercritical-CO_2-drying using ethanol as co-solvent (SCCD-EtOH). Peak identification: (1) 3-O-caffeoylquinic acid; (2) caffeoyltartaric acid; (3) 4-O-caffeoylquinic acid; (4) 5-O-caffeoylquinic acid; (5) caffeic acid derivative; (6) isochlorogenic acid; (7)

chicoric acid; (8) caffeic acid derivative; (9) luteolin 7-O-glucuronide; (10) quercetin 3-O-glucuronide. AU = arbitrary units.

Response to Technical Check Results

TECHNICAL:

1) Research papers should not exceed 5500 words including abstract and references but excluding figures, tables and their captions.

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