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METABÓLITOS SECUNDÁRIOS E COR DAS FOLHAS DE *Bauhinia forficata Link* e *Piper aduncum L.* SUBMETIDAS À SECAGEM EM LEITO FIXO E FLUIDIZADO

Secondary metabolites and colour of Bauhinia forficata Link and Piper aduncum L. leaves subjected to fixed-bed and fluidised-bed drying

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Resumo

Grande parte dos compostos oriundos das plantas medicinais são sensíveis aos processos de secagem. Dessa forma, é importante a adequação das condições de secagem, como temperatura e velocidade do ar, visando um processo eficiente, e que não influencie negativamente a qualidade do produto. Assim, objetivou-se avaliar o efeito de diferentes sistemas de secagem sobre a cor e os compostos das folhas de *B. forficata* e *P. aduncum*. Realizou-se a secagem nas temperaturas de 40, 50 e 60°C em leito fixo e fluidizado. Avaliou-se a cor das folhas frescas e secas, posteriormente feita a extração dos compostos, com etanol. O estudo foi conduzido seguindo um delineamento inteiramente casualizado em esquema fatorial 3 × 2 e quatro repetições. Conclui-se que a condição de 60°C em ambos os sistemas de secagem e 40°C em leito fluidizado resultou em maiores concentrações dos compostos oriundos das folhas de *B. forficata*. Enquanto para os extratos de *P. aduncum*, as maiores concentrações dos compostos foram na temperatura de 60°C em ambos os sistemas de secagem, e 50°C em leito fixo. Após a secagem, as folhas de *B. forficata* ficaram mais esverdeadas, amarelas, e claras, e apresentaram cores mais intensas, porém com menor tonalidade. Já as folhas de *P. aduncum* ficaram mais avermelhadas, azuis e escuras, expondo cores menos intensas, porém com maior tonalidade.

Palavras-chave: Extração de compostos. Pata de vaca. Falso jaborandi. Propriedades terapêuticas.

Abstract

Most compounds from medicinal plants are sensitive to drying processes, so it is important to adjust the drying conditions, such as temperature and air speed, aiming at an efficient process that does not affect the quality of the product. Thus, the objective was to evaluate the effect of different drying systems on the colour and compounds of *B. forficata* and *P. aduncum* leaves. Drying was performed at temperatures of 40, 50 and 60 °C on a fixed bed and on a fluidised bed. The colour of fresh and dried leaves was evaluated, and then the compounds were extracted with ethanol. The study was conducted following a completely randomized design in a 3 × 2 factorial scheme and 4 replications. Temperatures of 50 and 60 °C led to different results from those obtained in the drying of *B. forficata* leaves at 40 °C. For the extracts of *P. aduncum,* at temperature of 50 °C, there were differences in the intensities of the compounds in both drying systems compared to the other temperatures tested. After drying, B. forficata leaves became more greenish, yellower, and lighter, exhibiting more intense colours, but with less shade, while

P. aduncum leaves became more reddish, bluer and darker, exhibiting less intense colours, but with more shade.

Keywords: Compound Extraction. Orchid tree. False jaborandi. Therapeutic Properties.

INTRODUCTION

Medicinal plants have been used for therapeutic purposes worldwide, and medicines derived from their active principles are still one of the alternatives for many people seeking the prevention or treatment of diseases, especially in regions with deficit of health infrastructure.

Among the numerous medicinal plants present in Brazil are *Bauhinia forficata* and *Piper aduncum*. *B. forficata*, popularly known as 'pata-de-vaca', is widely used in the production of herbal medicines, largely used as a natural remedy for treating diabetes (Salgueiro *et al*., 2016). *P. aduncum*, also known as 'falso jaborandi', is used as an antiseptic for stopping haemorrhages and in the treatment of ulcers, genital and urinary organ diseases (VOLPE *et al*., 2015) and diarrhoea (MARCHESE *et al*., 2006).

Generally, leaves are the part of medicinal plants that most concentrate the therapeutic properties and, at the time of harvest, these organs have a high moisture content, an undesirable factor because among so many factors it facilitates the attack of microorganisms, which leads to the degradation of the product, consequently compromising the quality of the active principles and causing instability during storage. Before reaching the use of medicinal plants, several steps are necessary. Care begins at planting and goes through harvesting and drying, before being used medicinally. Its total use as a green plant is practically impossible, due to its availability in short periods, so it is necessary to harvest, dry, store and make it available for a longer period of time.

In view of the above, the objective of this work study was to evaluate the effect of different drying systems on the colour and compounds of *B. forficata* and *P. aduncum* leaves.

1 THEORETICAL REFERENCE

1.1. Medicinal plants

The use of medicinal plants in the treatment of diseases has been used since past generations, which characterizes the decades-long use of these products juxtaposed with knowledge and popular knowledge passed on from generation to generation (FEIJÓ *et al*., 2012). The use gradually made the beliefs for curing diseases of that time to be limited, and the fact that plants can help in cure became accepted (JAMSHIDI-KIA *et al*., 2018).

The prospect of studying medicinal plants is great. There are about half a million plants around the world, most of them have not yet been studied in medical practice, and future studies of medical activities may be effective in curing diseases. However, there are species that change the synthesis of compounds in different climates, develop synergistic compounds that lead to adverse effects of antagonists, or other unexpected changes in bioactivity (PASSARI *et al*., 2015).

B. forficata is an example of a medicinal plant widely used in the production of pharmaceutical drugs. The species is widely used as a natural remedy to cure blood circulation diseases (SALGUEIRO *et al.*, 2016). Its consumption is basically through tea to control diabetes. Another species that has shown a growing increase in its studies is the *P. aduncum* L. The leaves, in long baths, have been used in cases of fall of the uterus. With the leaves, a tea is prepared, good for combating hemorrhages. The same tea is also indicated against diarrhea (MARCHESE *et al.*, 2006). Being important to remove water to ensure conservation, as well as obtaining various products and for an extended period.

1.2. Drying system

Drying is one of the main processes carried out to ensure the maintenance of the quality of most agricultural products. Among the benefits are the preservation of the product, stability of the aromatic components at room temperature, protection against enzymatic degradation and oxidation, in addition to contributing to more adequate storage conditions, making the product available at any time of the year (QUEQUETO *et al.*, 2019; SIQUEIRA *et al.*, 2020). Since each product presents a behavior during the drying process (SIQUEIRA *et al.*, 2020), it is important that the process is conducted in a controlled manner, in order to guarantee the preservation of quality (MABASSO *et al.*, 2023).

There are two general methods used for drying the leaves of medicinal plants, one of them is natural drying, where the product is exposed to the sun or to the shade. Although it has no energy cost, it is disadvantageous because of the drying time and uneven final moisture content. In addition, some compounds may be degraded or oxidized due to the long drying period or high temperature. (MASHKANI *et al.*, 2018). Silva *et al.* (2016), when working with the drying of *Plectranthus barbatus*, found reduction in the flavonoid content of leaves dried in a solar dryer, when compared to oven-dried leaves.

The other method is the artificial drying, in this case the product is subjected to hot air drying. Traditionally, drying is carried out on a fixed bed, which consists in forcing the drying air flow through a layer of product that remains static inside the dryer. Another technique is fluidised-bed drying, which has been increasingly used by industries. In this case, the product is subjected to high air flows to the point of causing mass fluctuation, that is, there is no contact of the product with the plate or bottom of the dryer. Its advantages include high heat and mass transfer and drying uniformity, with a consequent reduction in drying time (SIVAKUMAR *et al.*, 2016).

Moreover, regardless of the hot air-drying method used, the drying of leaves of medicinal plants is a very delicate process, because the substances present can be degraded during water removal, mainly due to the excess temperature or to the exposure time required by the process, because of the drying rate (GÜMÜŞAY *et al.*, 2015).

Along with the type and concentration of the compounds present in the leaves, colour is a parameter of great importance, as it is considered a good indicator of product quality and even of the presence of bioactive compounds (LU *et al.*, 2014).

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2 MATERIAL AND METHODS

2.1. Preparation of samples and drying

Fresh leaves of *B. forficata* and *P. aduncum* were acquired from the Garden of Medicinal Plants of the Federal University of Grande Dourados, Dourados/MS, Brazil. The leaves were collected in the morning, avoiding days when there were remnants of dew and/or irrigation. The leaves were selected and those exhibiting any type of injury were discarded. The average initial water contents were 63.77% for *B. forficata* and 78% for *P. aduncum*, determined by the gravimetric method, using an oven with forced air circulation at 103 ± 1 ºC for 24 h, in triplicate (ASABE, 2007).

Then, the leaves (leaf blade) were subjected to drying at 3 temperatures, 40, 50 and 60 °C and in two drying systems, fixed bed and fluidised bed. Air speed was equal to 0.4 m s^{-1} for fixed-bed drying 2.0 m s^{-1} for fluidised-bed drying. A set of two trays was used, supported in the drying chamber, each containing approximately 25 grams of product.

Drying tests were conducted using a fixed layer dryer, equipped with a system that precisely controls the drying air speed and temperature and has a series of sensors connected to a control panel, aiming to obtain a fine adjustment and monitor the drying air conditions.

After the drying process began, the mass of the samples was measured at pre-established times on an analytical scale with resolution of 0.01 g. Water content during drying was determined by mass difference, based on the known initial water content, until reaching the final water content of approximately 10%.

The moisture reduction rate during drying was determined according to Eq. 1:

$$MRR = \frac{Mw_0 - Mw_i}{DM\,(t_i - t_0)} \quad (1)$$

MRR: moisture reduction rate, kg kg^{-1} h^{-1}; Mw_0: previous total mass of water, kg; Mw_i: current total mass of water, kg; DM: Dry mass, kg; t_0: previous total drying time, h; and t_i: current total drying time, h.

2.2. Colour evaluation

Colour was evaluated in fresh leaves of *B. forficata* and *P. aduncum* and after drying, by direct reading of reflectance of the coordinates L*, a* and b*, using a portable high-precision colorimeter, Konica Minolta, CR-400 model, with the HunterLab colour system, with L* relative to white and black, a* to red and green, and b* to yellow and blue.

The coordinates L*, a* and b* were used to calculate chroma (C*) (Eq. 2) and Hue angle (H*) (Eq. 3). Chroma (C*) indicates the saturation level or colour intensity, while Hue angle (H*) indicates the observable colour (tone) considering the coordinates a* and b*.

$$C^* = \sqrt{(a^*)^2 + (b^*)^2} \quad (2)$$

$$H^* = \arctan\left(\frac{b^*}{a^*}\right) \quad (3)$$

C*: chroma; H*: Hue angle (º); a*: chromatic coordinate with variation from green to red; b*: chromatic coordinate with variation from blue to yellow.

2.3. Preparation of extracts

The dry material was crushed in the Laboratory of Bromatological Analyses of UFGD. Subsequently, 40 mg of the plant material received the mixture of ethanol:water (7:3 v/v) and was extracted for 15 minutes in ultrasound. Subsequently, the extracted sample was centrifuged at 3000 rpm for 10 minutes and the supernatant was filtered (1 mL) using Millex PTFE 0.22 µm filter.

2.4. Analyses of *B. forficata* and *P. aduncum* leaves by LC-DAD-MS

The analyses were performed on a UFLC (Shimadzu) coupled to a diode array detector and a mass spectrometer (MicrOTOF-Q III, Bruker Daltonics) with electrospray ionization source and analysers quadrupole and Time-of-Flight (TOF). The chromatographic column Kinetex C-18 (Phenomenex, 150 × 2.1 mm, 2.6 µm) was used in the analyses. The mobile phase was composed of ultrapure water (A) and acetonitrile (B) both mixed with 0.1% formic acid (v/v), the flow rate was 0.3 mL/min and the elution profile was the following: 0-2 min - 3% B, 2-25 min - 3-25% B and 25-40 min - 25-80% B. *B. forficata* and *P. aduncum* leaves (10 mg) were extracted by methanol and water (7:3 v/v, mixed with 0.1% formic acid), maintained in ultrasonic bath for 10 min, centrifuged (12000 rcf for 10 min), and the supernatants were filtered (Millex, PTFE, 0.22µm) and 2 µL were injected in the system. The samples were analysed in triplicates. The pool (quality control for metabolomic analyses) was produced from the mixture of 50 µL each sample and injected after 10 sample analyses. Nitrogen was applied as dry (9 L/min), nebulizer (4 Bar) and collision gas in the mass spectrometer. The capillary voltage was 2.500 kVv. The analyses were performed in negative and positive ion modes.

2.5. Metabolomics analyses: data processing and statistical analyses

Initially, the data were aligned by Metalign software and the inputs were reduced using MSclust Software. The replicates and the sample pool were compared to assess the reproducibility of the device and the exported data were analysed by the Metaboanalyst 4.0 platform using the Principal Component Analysis (PCA) tool, which aimed to evaluate the variations between treatments. The PCA was generated with log transformation, transformed and self-dimensioned for the dispersion profile of compounds and hierarchical grouping. The data were organized using Pearson's distance, the complete clustering algorithm and random forest analysis ($p \leq 0.001$).

3 RESULTS AND DISCUSSION

3.1. Drying and colour of *B. forficata* leaves

The times required for *B. forficata* leaves to reach the water content of approximately 10% in fixed-bed drying were 5.67, 2.33 and 1.25 h at temperatures of 40, 50 and 60 °C, respectively. For drying in a fluidised bed, the times were 3.58, 1.50 and 0.75 h, for the same temperatures.

The increase in temperature resulted in a shorter drying time, which occurs because the increment in temperature causes the increase in the vapor pressure difference between the product and the drying air, causing the water to migrate from the inside of the leaves to their extremities and then to the drying air, in a shorter time. Note that the drying time was reduced in the fluidised bed system, since it has higher drying air speed (Figure 1A). These behaviours have been observed by Gasparin *et al*. (2017) when drying *Mentha piperita* leaves, Silva *et al*. (2017) when studying the drying of *B. forficata* leaves and Sousa *et al*. (2015) in leaves of *Ziziphus joazeiro*.

There was greater water removal from *B. forficata* leaves at the beginning of the drying process, compared to the end, under all conditions (Figure 1A), since at first the product had a high moisture content on its surface, so this water will be removed faster. According to Babalis *et al*. (2006), after the evaporation of water on the surface of the product, which occurs in a more marked way, the water present inside will move to the surface and, at this time, the speed of the drying air exerts little effect on water removal, giving room for liquid diffusion, which is more influenced by temperature.

The drying systems showed greater interferences at the temperature of 40 °C. According to Martins *et al*. (2018), the effect of drying air speed is more evident at low temperatures, as it has greater contribution to the removal of water on the surface of the product.

For the coordinate a*, only the temperatures of 50 and 60 °C in the fluidised-bed drying showed significant differences ($p < 0.01$) compared to the fresh plant. Considering that the coordinate a* varies from green to red, the values increased as the temperature increased, indicating that *B. forficata* leaves showed a tendency to green colour for low temperatures (Figure 1B).

The drying treatments for fixed bed did not negatively affect the coordinate a*, suggesting that the chlorophylls present in the leaves were not degraded under high temperatures as in fluidised-bed. For the coordinate b*, there was a statistically significant difference ($p< 0.01$) for all conditions in comparison to the fresh plant. This coordinate corresponds to the variation from blue to yellow and, as its values were positive, *B. forficata* leaves showed the presence of yellow colour (Figure 1C).

The greater presence of yellow colour after drying may be related to the release of carotenoids, due to the probable rupture of the internal structures of the leaves. However, the excess heat in the process was not enough to degrade this pigment, so the yellow colour was maintained.

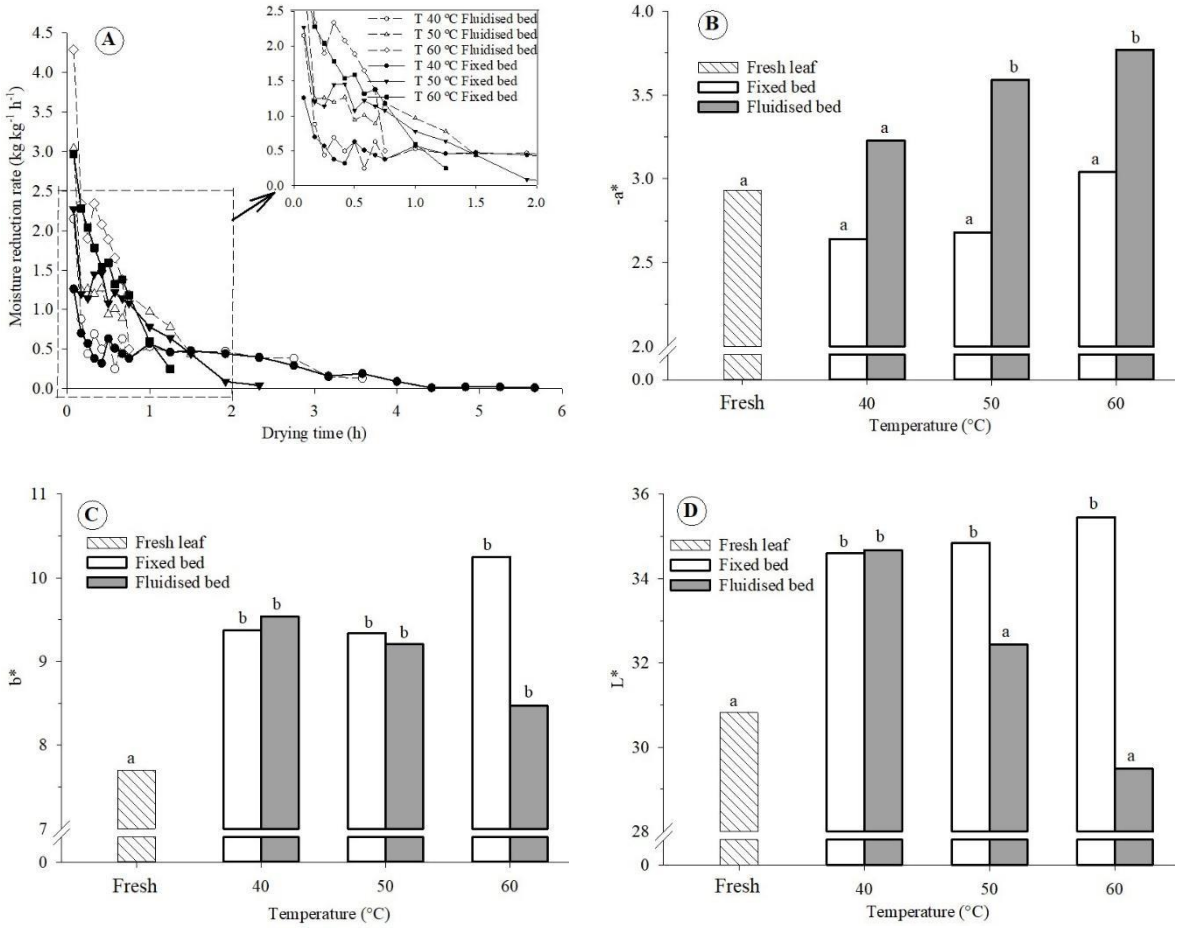
For the coordinate L*, only the temperatures of 50 and 60 °C in the fluidised-bed drying did not cause significant differences ($p < 0.01$) in comparison to the fresh plant. For fixed-bed drying, the colour of the leaves tended to become lighter with the increase in temperature, whereas for fluidised-bed drying, the values decreased with increasing temperature, resulting in darker leaves (Figure 1D).

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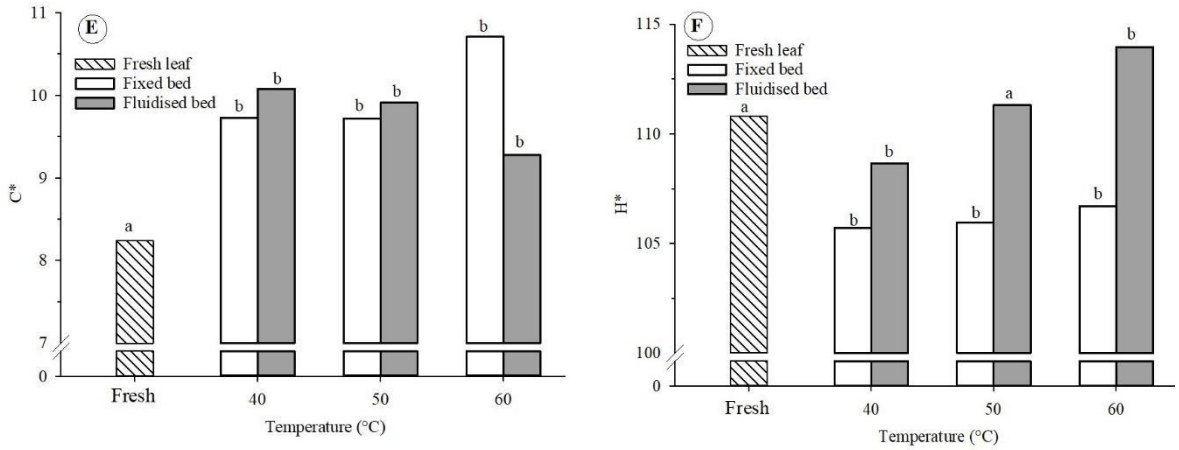
It is noted that the drying process intensified the colour of *B. forficata* leaves, since the values of chroma (C*) increased. A statistically significant difference was observed ($p < 0.01$) for all conditions in comparison to the fresh plant (Figure 1E).

The hue angle (h°) ranges from 0 to 360° and indicates the observable colour (tone) of the sample. It was observed that only the temperature of 50 °C in the fluidised-bed drying did not show statistically significant differences ($p < 0.01$) in comparison to the fresh plant. In both drying systems, H* values increased as temperature increased, indicating that the leaves lost their colour tone (Figure 1F).

Figure 1 - Moisture reduction rate for *B. forficata* leaves along the drying time as a function of temperature and drying system (**A**). Mean values of a* (**B**), b* (**C**), L* (**D**), C* (**E**) and H* (**F**) for fresh and dried leaves of *B. forficata* as a function of temperature and drying system.



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Means with the letter "a" do not differ at 1% probability level by Student's t-test, compared to the fresh plant.
Source: Own authorship.

3.2. Metabolomics analyses of *B. forficata* leaves

From statistical analyses, the main compounds for differences between sample groups were identified (Table 1). The compounds were identified based on UV, accurate MS and MS/MS profile data compared to published data

Table 1 - Compounds identified in *B. forficata* leaves by LD-DAD-MS

Peak	Compound	RT (min)	MF	UV (nm)	MS (*m/z*) $[M-H]^-$	MS/MS (*m/z*)	MS (*m/z*) $[M+H]^+$
1	NI	11.4	$C_{20}H_{36}O_{11}$	324	451.2199	167/149	-
2	5-*O*-E-caffeoylquinic acid	12.5	$C_{16}H_{18}O_9$	299/326	353.0890	191	-
3	*O*-hexosyl tuberonic acid	12.6	$C_{18}H_{28}O_9$	-	387.1647	163	389.1808
4	*O*-hexosyl tuberonic acid	13.5	$C_{18}H_{28}O_9$	-	387.1674	207/163	389.1808
5	3-*O*-*E*-coumaroylquinic acid	13.5	$C_{16}H_{18}O_8$	299/310	337.0929	191/163	339.1074
6	Phenylmethyl O-deoxyhexosyl-hexoside	14.1	$C_{19}H_{28}O_{10}$	-	415.1624	269	417.1735
7	5-*O*-*E*-coumaroyquinic acid	15.0	$C_{16}H_{18}O_8$	290/310	337.0930	191	339.1074
8	di-*O*-deoxyhexosyl hexosyl quercetin	17.4	$C_{33}H_{40}O_{20}$	262/352	755.2077	300/271/255/179/151	757.2186
9	*O*-deoxyhexosyl-hexosyl quercetin	18.8	$C_{27}H_{30}O_{19}$	265/348	609.1461	300/271/255/179/151	611.1607
10	di-*O*-deoxyhexosyl-hexosyl kaempferol	18.8	$C_{33}H_{40}O_{19}$	265/348	739.2130	284/255/179/151	741.2237
11	di-*O*-deoxyhexosyl-hexosylmethyl-quercetin	19.3	$C_{34}H_{42}O_{20}$	268/354	769.2236	314/299/286/271/179	771.2342

RT: retention time; MF: molecular formula; NI: non-identified; UV: ultraviolet spectra
Source: Own authorship.

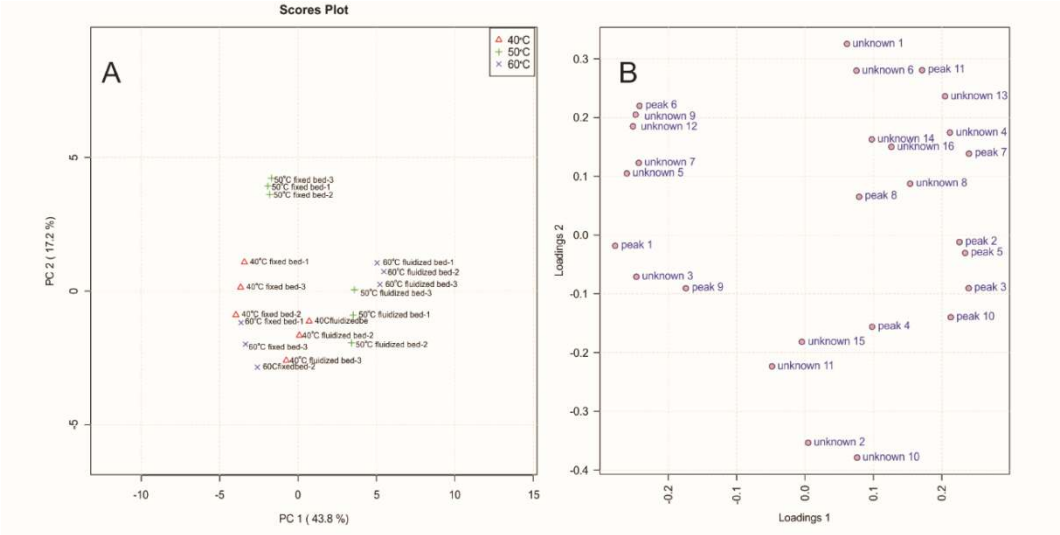
The principal component analysis (PCA) showed data variability of 43.8% and 17.2% for PC1 and PC2, respectively, so these data explained 61.0% data variability (Figures 2A and 2B). The PC1 is relative to the component of higher data variability and the samples from the treatment at 50 and 60 °C by fluidised-bed dryer were closer, thus indicating higher significance.

The PCA graph can be divided into four quadrants, so, in general, it can be seen that the treatments located in the positive quadrant, in both axes, have higher intensities for a given compound, when compared to the treatments in the opposite quadrant (negative). For example, 5-*O*-*E*-coumaroyquinic acid (peak 5) had higher intensities in the treatments of 50 and 60 °C in fluidised bed.

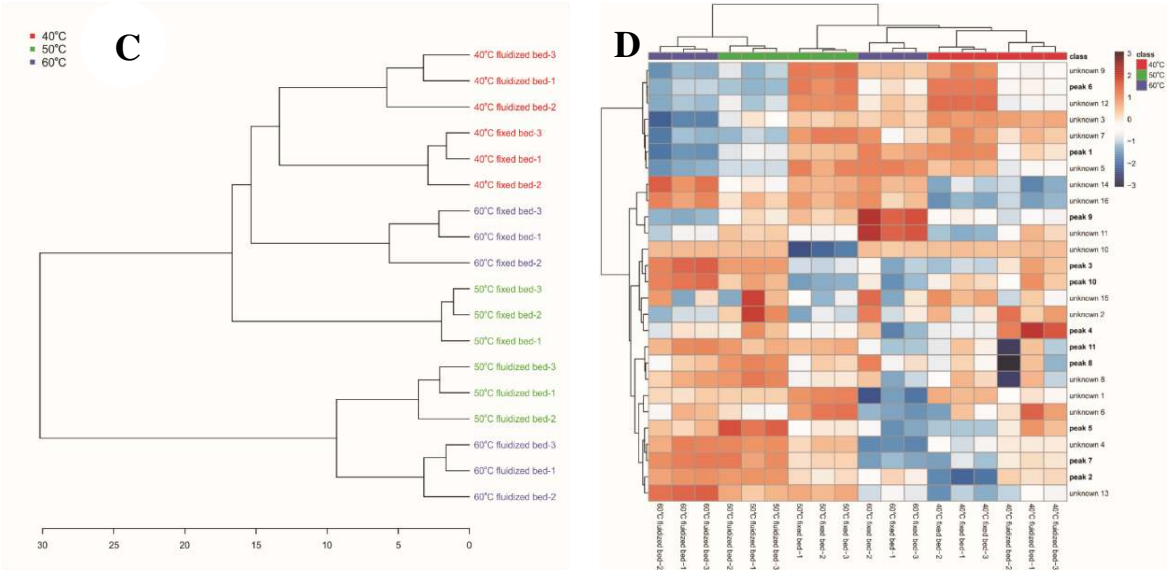
According to Reinato *et al*. (2012), the closer one treatment is to the other, the greater the similarity between the values of the variables studied. Thus, there are greater similarities between the chemical composition of samples (1) dried at temperatures of 60 °C and 50 °C in fluidised bed and (2) temperature 40 °C in fluidised bed and fixed bed, as demonstrated in the groups (Figure 2C).

The *heatmap*, constructed from the samples of *B. forficata* leaves shows the ion intensities for dryer treatments in each sample. The compound **2** (*O*-hexosyl tuberonic acid) shows higher ion intensities in the sample obtained with 40 °C by fluidised-bed dryer. For the most representative compounds in the division of the drying groups, the highest intensities were obtained under the following conditions: compound **1** at 40 °C in a fixed bed, compound **2** at 50 and 60 °C in a fluidised bed; compound **4** at 40 °C in a fluidised bed; compounds **5** and **7** at 50 and 60 °C in a fluidised bed; compounds **6**, at the temperature of 40 and 50 °C in a fixed bed; compound **8**, at the temperature of 50 °C in a fluidised bed; compound **11**, at the temperature of 50 and 60 °C in a fluidised bed; compound **9**, at the temperature of 60 °C in a fixed bed; and compounds **3** and **10**, at the temperature of 60 °C in a fluidised bed (Figure 2D).

Figure 2 - Principal component analysis (PCA) of samples from *B. forficata* leaves (**A**) and the loadings (**B**). Dendrogram of the samples from *B. forficata* (**C**). Heatmap and hierarchical grouping of the components of *B. forficata* (**D**).



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Source: Own authorship.

The compounds **8**, **9** and **11** of the flavonoid class had higher intensities under the highest temperature tested, 50 and 60 °C. According to Borgo *et al*. (2010), the thermal energy involved in the drying process causes the rupture of internal structures, promoting greater extraction of chemical compounds during the process of obtaining extracts.

The compounds **5** and **7** are chlorogenic acid and revealed higher intensities at temperatures of 50 and 60 °C. Similar results were found by López-Vidaña *et al*. (2016), who observed that when the material is dried at 60 °C, the chlorogenic acid content is more stable than when it is dried at lower temperatures. In this work we also found that the temperature of 40 °C favoured a high intensity of this class of compound when using fluidised bed for the compound (5) 3-*O*-coumaroylquinic acid.

In general, among all compounds, higher intensities are observed for 60 °C in both drying systems, and 50 °C in fixed bed.

3.3. Drying and colour of *P. aduncum* leaves

The times required for the drying of *P. aduncum* leaves to reach the water content of approximately 10% in a fixed bed were 6.67, 2.92 and 1.25 h at temperatures of 40, 50 and 60 °C, respectively. For the drying in fluidised bed, the times required were 5.83, 2.5 and 1.0 h, for the same temperatures.

It is observed that the drying time is reduced with the increase of temperature. The temperature influences air vapor pressure, so the higher the temperature, the greater the difference between the vapor pressure between the product and the drying air, triggering and accelerating the removal of water. It is also noted that the drying time was shorter in the fluidised bed system, which occurred because this system had higher air speed, which contributed to a higher rate of water removal (MARTINS *et al*., 2018).

For all treatments, the removal of water from *P. aduncum* leaves was more intense at the beginning of the drying process (Figure 3A), because in this initial period the leaves had high

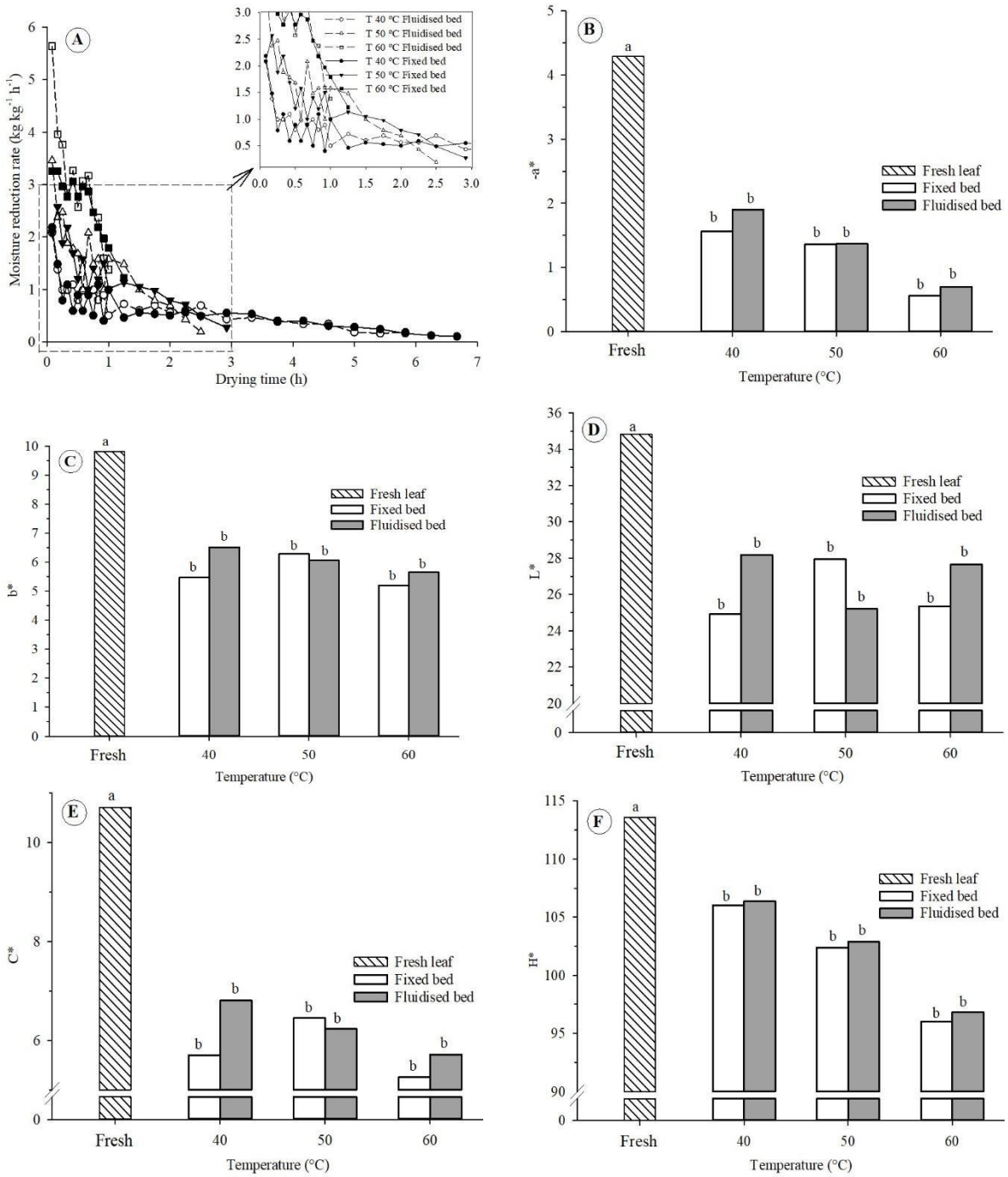
water content, so the water present at the extremities was rapidly removed and, after that, it took some time for the water present inside to move to the extremities and then be removed, which explains the fact that MRR is lower at the end of the process. When the reduction in MRR occurs, air speed no longer has as much influence on water removal, so there is a predominance of liquid diffusion, which is more influenced by temperature (BABALIS *et al*., 2006).

As in Figure 1A, the effect of the drying system can be observed at the lowest temperature, and the fluidised bed, for having higher drying air speed, contributes to a greater water removal, reducing the drying time.

For the coordinate a*, there were significant statistical differences ($p < 0.01$) between all drying conditions, in comparison to the fresh plant. It is observed that the increase in temperature indicated a tendency towards redder leaves (Figure 3B). The green colour of the leaves is attributed to the presence of chlorophyll, and in this case the increase in temperature may have caused Maillard reactions, oxidizing or degrading chlorophyll, causing the leaves to lose their initial characteristic colour (BUŠIĆ *et al*., 2014).

The coordinate b* showed a statistically significant difference ($p < 0.01$) for all conditions in comparison to the fresh plant. The values decreased with the increase in temperature, indicating loss of yellow colour and tendency to blue colour (Figure 3C).

Figure 3 - Moisture reduction rate of *P. aduncum* leaves along the drying time as a function of temperature and drying system (**A**). Mean values of a* (**B**), b* (**C**), L* (**D**), C* (**E**) and H* (**F**) for fresh and dried leaves of *P. aduncum* as a function of temperature and drying system.



Means with the letter "a" do not differ at 1% probability level by Student's t-test, compared to the fresh plant.

Source: Own authorship.

The yellow colour in the leaves is attributed to the presence of carotenoids (RODRIGUEZ-AMAYA, 2016), and the drying process may have degraded this pigment, causing the yellow tone to be reduced. Therefore, colour changes may be associated with the destructuring of cell membranes, thus affecting colour characteristics.

For the coordinate L*, a significant difference was noted ($p < 0.01$) between the fresh plant and the leaves subjected to all drying treatments. In general, the leaves become darker with drying (Figure 3D). These results were similar to those found by Reis *et al*. (2012), who dried leaves of *Ocimum basilicum* L. and concluded that drying resulted in their darkening.

C* and H* showed significant difference ($p < 0.01$) for all conditions in comparison to the fresh plant. For both drying systems, in general, the values of chroma and hue angle decreased with increasing temperature, resulting in leaves with lower colour intensity, i.e., opaquer and more yellowish with greater tone (Figures 3E and 3F).

3.4. Metabolomics analyses of *P. aduncum* leaves

Ten main compounds were identified, which were more representative in the division of the drying groups from *P. aduncum* leaves. Flavonoid (50%), carboxylic acid (30%) and chromene (20%) were identified in the compounds (Table 2).

Table 2 - Compounds identified in *P. aduncum* leaves by LD-DAD-MS

Peak	Compound	RT (min)	MF	UV (nm)	MS (*m/z*) $[M-H]^-$	MS/MS (*m/z*)	MS (*m/z*) $[M+H]^+$
1	*C*-hexosyl *O*-hexosyl kaempferol	11.9	$C_{27}H_{30}O_{16}$	268/350	609.1471	489/446/326/313/298	611.1607
2	*C*-hexosyl *O*-hexosyl-deoxyhexosyl kaempferol	12.1	$C_{33}H_{40}O_{20}$	269/352	755.2040	635/446/327/356/313	757.2186
3	Phenylmethyl *O*-pentosyl-hexoside	12.5	$C_{18}H_{26}O_{10}$	-	401.1462	269/161	403.1599
4	*O*-hexosyl dihydrovomifoliol	14.2	$C_{19}H_{32}O_8$	-	387.2027	225	389.2170
5	*C*-hexosyl *O*-hexosyl-deoxyhexosyl apigenin	18.0	$C_{27}H_{30}O_{15}$	270/334	593.1527	413/311/293	595.1639
6	Oxo-diprenyl hydroxyl benzoic acid	34.6	$C_{17}H_{20}O_4$	280	287.1289	231/217/191/173	289.1434
7	Dimethyl prenylchromene carboxylic acid	35.7	$C_{17}H_{20}O_3$	288	271.1340	227/204	273.1485
8	Di-prenyl hydroxyl benzoic acid	35.8	$C_{17}H_{22}O_3$	-	273.1496	229/206/173	275.1642
9	Oxo-diprenyl hydroxyl benzoic acid	38.3	$C_{17}H_{20}O_4$	272/340	287.1289	243/220/203/187/161	289.1434
10	Dimethyl-8-(3″-methyl-2″-butenyl)-2H -1-cromene-6-carboxylate	38.6	$C_{18}H_{22}O_3$	289	285.1496	229/189	287.1642

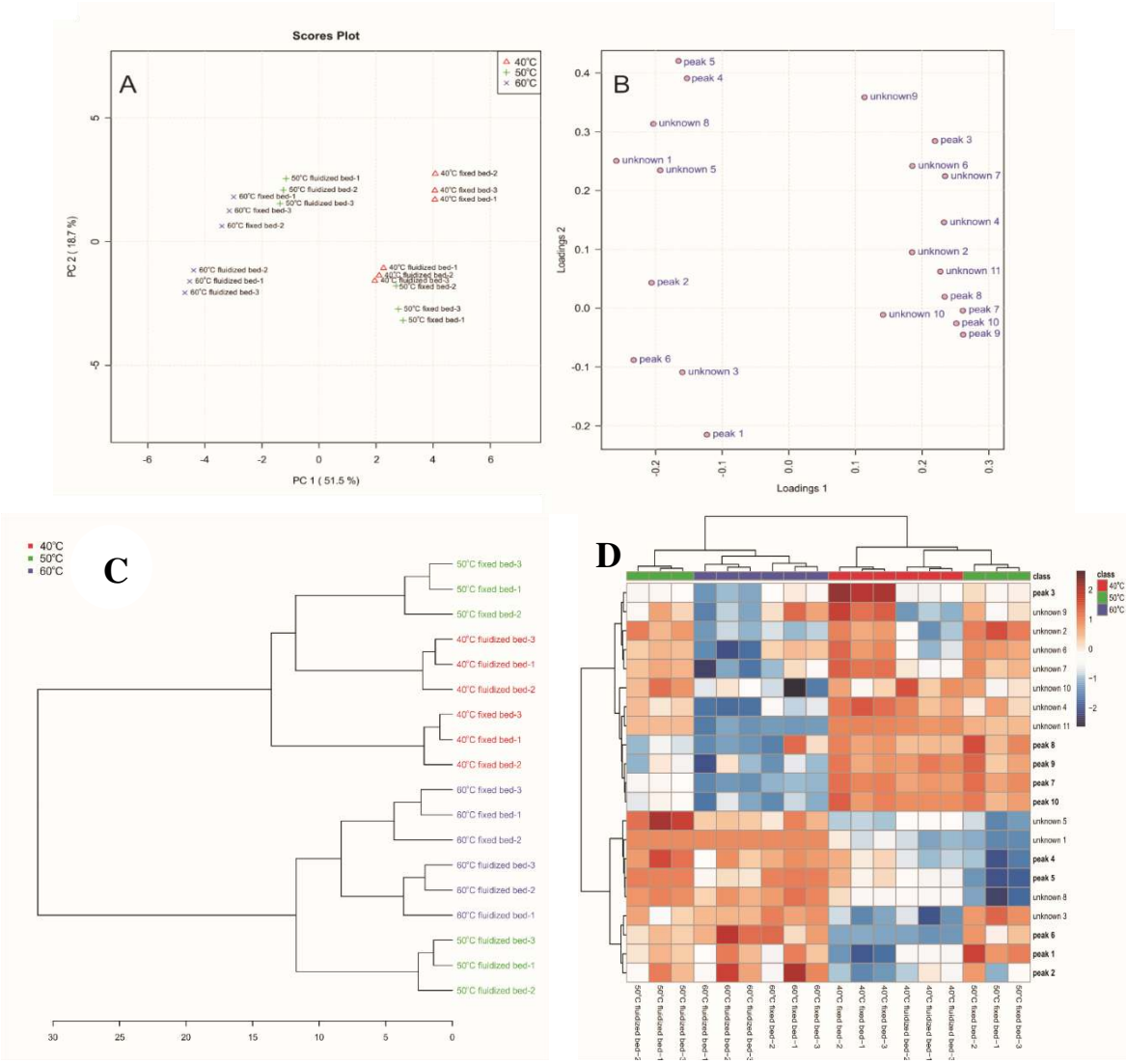
RT: retention time; MF: molecular formula; UV: ultraviolet spectra
Source: Own authorship.

The PCA analysis of samples from *P. aduncum* submitted to different drying processes explained 70.2% of the data variability. Components 1 and 2 explained 51.5% and 18.7%, respectively (Figures 4A and 4B).

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It is observed that the 40 °C treatment in both drying systems had higher contributions to the variance, when it was related to the intensities of the compounds. Since the intensities of the compounds are influenced by the position in the PCA quadrant, it is noted that the compound *O*-hexosyl diidrovomifoliol (4) had higher intensities at a temperature of 60 °C in fixed bed and 50 °C in fluidised bed, while it had lower intensities in the opposite quadrant, temperature of 40 °C in both drying systems and 50 °C in fixed bed.

Figure 4 - Principal component analysis (PCA) of the samples from *P. aduncum* (**A**) and the loadings (**B**). Dendrogram of the samples from *P. aduncum* (**C**). Heatmap and hierarchical grouping of the components of *P. aduncum* (**D**).



Source: Own authorship.

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The treatments closer to each other have few variations in their ion intensities, so the greatest similarities between the groups are observed: 40 °C in fluidised bed and 50 °C in fixed bed forming the first group; 40 °C in fixed bed forming the second group; 60 °C in both drying systems forming the third group; and 50 °C in fluidised bed forming the fourth group (Figure 4C).

For the most representative compounds in the division of drying groups, the highest intensities of the compounds are present in the following treatments: compound **1** at 50 °C in fixed bed and 60 °C in both drying systems; compound **2** at 60 °C in both drying systems; compound **3** at 40 °C in fixed bed; compound **4** at 50 °C in fluidised bed and 60 °C in fixed bed; compound **5** at a temperature of 50 °C in fluidised bed and 60 °C in fixed bed; compound **6** at a temperature of 60 °C in both drying systems; compounds **7**, **8**, **9** and **10** at a temperature of 40 °C in both drying systems and 50 °C in fixed bed (Figure 4D).

The results for compounds **3**, a phenyl methyl, were more intense at a temperature of 40 °C, showing that it could be thermally unstable. For the other compounds **1**, **2**, **4** and **5**, their highest intensities occurred at temperatures of 50 and 60 °C, and it is suggested that under this condition there was rupture of cell membranes, probably causing greater release of these compounds.

In general, higher ion intensities are observed at 50 °C in both drying systems. Allied to the temperature factor, the speed of the drying air also assumes great importance, since rapid drying interrupts the enzymatic and microbial action, preserving the chemical constituents in the dried plant.

CONCLUSION

The drying systems influenced the intensities of the compounds present in the extracts of *B. forficata* and *P. aduncum* leaves. The conditions of 60 and 50 °C in both drying systems resulted in higher intensities of the compounds derived from *B. forficata* leaves. For *P. aduncum*, the highest intensities were observed at 50 °C in both drying systems. After drying, *B. forficata* leaves became more greenish, yellower, and lighter, exhibiting more intense colours, but with less shade, while *P. aduncum* leaves became more reddish, bluer, and darker, exhibiting less intense colours, but with more shade.

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