Comparison of experimental setups for the production of milk concentrates and subsequent characterization

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ABSTRACT

Vacuum concentration is an intermediary step in the dairy powder production and an essential step in the process scheme for the production of sweetened concentrated milk, a product widely popular and produced in Brazil. As any food product, it is subject to research and development in order to improve the original recipe and propose new derivatives, whether in academic or industrial environments. The tests required for the development of products may be costly in relation to the supply in raw material and cleaning agents, energy and water consumptions as well as water treatment after the cleaning of the equipment. As part of the reduction of production costs and environmental impact, the present work aims to evaluate the possibility of using smaller equipment for concentration as well as to produce relevant data on the processing of SCM. Two types of vacuum evaporators were used for this work: a pilot scale falling film evaporator and a lab scale rotary evaporator. Based on the observed results, it was possible to evidence the similarity of the data obtained for the concentrates (with and without sucrose addition) in both equipment for the measurements of density, viscosity and surface tension with deviations less than 10%.

1. Introduction

Vacuum concentration is used mainly for the production of powders in the dairy industry in order to preconcentrate products before spray drying. But it is used too for the production of concentrates such as sweetened concentrated milk (SCM). SCM is a widely popular product used mainly as ingredient in desserts. Brazilian people are especially fond of this product and dairy manufacturers in Brazil are then the main worldwide producers of SCM. The production is about 608 000 tons per year (Zacarchenco, Van Dender, & Rego, 2017). The industrial manufacturing steps for the production of SCM are successively fat standardization of skim milk, heat treatment of standardized milk, sucrose addition, fat homogenization, concentration by vacuum evaporation up to a dry matter content equal to 700–720 g/kg, seeding and lactose crystallization, cooling and packaging (Nieuwenhuijse, 2016; Renhe et al., 2017). Regarding to the Regulatory Instruction No. 47, October 26, 2018 – MAPA (BRASIL, 2018), the final product must have a dairy dry matter content equal to at least 280 g/kg and a fat content ranging from 80 to 160 g/kg. The protein content in the non-fatty dairy dry matter content must be equal to at least 340 g/kg.

In the dairy industry, concentration by vacuum evaporation is mainly carried out in falling film evaporators. This type of evaporator has a shell and tube arrangement. A thin film of liquid falls down on the internal surface of high vertical tubes whereas live steam circulates on the shell side and provides energy to the liquid for water evaporation. These evaporators are well adapted to heat sensitive products as they can work under vacuum and operate between 45 °C and 70 °C for the concentration of dairy products. Moreover, they have large heat transfer areas and short residence times. Lastly, even if evaporation is known to be a highly energy-intensive phenomenon, many efforts are made to reduce energy consumption through the reuse of the vapor coming from the product as a heating medium and the implementation of mechanical and thermal vapor recompression systems. Falling-film evaporators are nowadays more energy efficient than spray-dryers in the process scheme for the manufacture of powder. Concentration in evaporators is conducted to a concentrate dry matter content as high as possible in order to

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reduce the overall energy consumption of the process scheme, the limit of the operation being determined by the concentrate viscosity (Anema, 2009; Anema & McKenna, 1996).

Conversely, the implementation of different energy recovery systems makes tricky the configuration of evaporators and it is quite challenging to know the operating conditions applied to the product when it passes through the evaporator. In the meantime, the progressive concentration of all the components induces changes that may be irreversible, such as denaturation of proteins and mineral destabilization, into the dairy concentrates and affects their biochemical and physical properties. As an example, concentration leads to a pH decrease, an ionic strength increase, the transfer of some calcium phosphate from the continuous phase to the dispersed phase as well as more protein instability. These biochemical changes affect the physical properties of product such as viscosity, surface tension and density. These properties influence in turn film flow through the film thickness δ (Equation (1)) and the minimum wetting rate Γ_{min} of the evaporation tubes (Equation (2)). Moreover, these values are required for modelling and simulation of the evaporation process (Madoumier, Azzaro-Pantel, Tanguy, & Gésan-Guiziou, 2015).

$$\delta = \sqrt[3]{\frac{3\,\eta\,\Gamma}{\rho^2\ g}} \tag{1}$$

where δ is the film thickness (m), η is the product viscosity (Pa.s), ρ is the density (kg/m^3), g is the gravitational constant equal to 9.81 m/s^2 and Γ the tube wetting rate of evaporation tubes (kg/(s.m))

$$\Gamma_{min} = 1.69\left(\frac{\eta\rho}{g}\right)^{\frac{1}{3}}(\sigma(1-\cos\theta))^{\frac{3}{5}} \tag{2}$$

where Γ_{min} is the minimum wetting rate (kg/(m.s)), η is the product viscosity (Pa.s), ρ is the density (kg/m^3), σ is the surface tension (N/m) and θ is the advancing contact angle of the liquid on the tube (rad). This equation was defined by Hartley and Murgatroyd (1964).

To improve the control of the concentration process and better understand the behavior of concentrates during concentration, experiments in falling-film evaporators are required. However, the configuration of these equipment and their working under vacuum makes this task tricky. It explains why there are few experimental studies on falling-film evaporators and most of them are conducted at industrial scale (Bienvenue, Jiménez-Flores, & Singh, 2003; Jeurnink, Walstra, & de Krutt, 1996; Jeurnink & Brinkman, 1994). Some lab and pilot scale set up were developed but they were dedicated for the study of a specific feature of the operation. Gordon, Bolisetti, Ting, and Reitsma (2017) used of a vertical tube for studying the formation of the liquid film depending on the heat flux through the wall, the viscosity of the product and the velocity of the film. Morison and Tie (2002) studied the behavior of proteins and minerals depending of the heat flux and the flow velocity of the product in a set up without vacuum and concentration. Kessler (1986) studied the fouling of falling-film evaporators using a pilot scale set-up made with a 2-m high tube. Tanguy et al. (2019) also used a single effect pilot-scale falling-film evaporator for studying the behavior of acid whey during vacuum concentration. However, the carrying out of experiments at pilot scale raises the questions of the initial volume of product needed for the trials, the duration of the experiments and the consumption of cleaning solutions. For example, the trials carrying out in (Tanguy et al., 2019) to produce whey concentrate at 400 g/kg DM require at least 100 L of liquid whey at 60 g/kg DM, last about 8 h (including more than 3 h for the cleaning before and after experiments). It consumes about 40 L of both alkaline and acid cleaning solutions. But, only 2 L of concentrated acid whey are eventually sampled for further analytical characterization. Therefore, it would be interesting to carry out experiments at lab scale in order to get samples from a lower initial volume of raw material, to reduce the duration of experiments and the quantities of cleaning solutions. The rotary evaporator is a suitable alternative to the pilot scale falling film evaporator as the working principle of both equipment are quite similar. Both are working under vacuum and the vaporization energy is provided through an indirect heat transfer between the product and the heat transfer medium (water and condensing steam in rotary and falling-film evaporators respectively). The main difference is the product does not flow in the form of a thin film in a rotary evaporator as it is held in a flask. Even if the experimental conditions are not exactly the same, it could be interesting when the outputs of the studies are to get data about the composition and the functional properties of the concentrates.

The objective of this work is to establish if the data obtained at lab scale in a rotary evaporator are representative to those obtained at pilot scale in a falling film evaporator. The comparison was made for the production of sweetened concentrate milk. Even if it is a widely consumed product, there are few data in the literature about the physical properties of this product. Moreover, the behavior during concentration is unknown, especially the evolution of density, viscosity and surface tension, some physical properties useful to characterize film flow in evaporators.

2. Materials and methods

2.1. Experimental procedure

Concentrated milks were prepared at pilot scale in the Dairy Platform (STLO - Science and Technology of Milk and Egg research unit, Rennes, France) following the processing scheme described in Fig. 1. Raw materials were cream and thermised skim milk provided by a local commercial dairy manufacturer. A standardized milk at 320 g/kg DM was prepared with 36 kg of cream and 464 kg of thermised skim milk. This mixture was heated to 40 °C using a heat exchanger (S14A, SONDEX, Saint-Genis-Laval, France) and then passed through the homogenizer (LAB 16/50, RANNIE-APV, Evreux, France) with pressure of 24 MPa (4 MPa in the 2nd stage and 20 MPa in the 1st stage). Supplementary step was added to the processing scheme related to preparation of sweetened milk that is the addition of powdered sucrose at 200 g/kg of standardized milk. The composition of both homogenized milks is given in Table 1.

A small part of the milks was kept for further concentration at lab scale whereas most of milks was concentrated using a pilot scale falling-film evaporator (GEA Process Engineering, Montigny-Le-Bretonneux, France). This equipment is composed of three evaporation tubes in series that are connected to the same indirect condenser (coil-type heat-exchanger). It was well described and characterized by Silveira et al. (2013) and Silveira et al. (2015). The evaporation rate of the equipment is about 27 kg/h at a feed mass flowrate of 70 kg/h and a heating power of 25.2 kW.

The experimental setup was adapted from Tanguy et al. (2019). The experiments were carried out at an absolute pressure of 0.02 MPa corresponding to an evaporation temperature of 60 °C. The products were first preheated from 20 °C to 60 °C using a tubular heat-exchanger and they were concentrated by passing through the pilot scale falling-film evaporator at a mass feed flowrate of 70 kg/h. Several successive runs were carried out to produce milk concentrates at increasing concentration factors (Fig. 2). The concentration factor is the ratio of the dry matter content of the concentrate at the outlet of the evaporator over the dry matter content of the homogenized milk. Evaporation temperature and feed mass flowrate were kept constant whatever the run whereas the heating power was adjusted to modify the evaporation rate and produce concentrates at specific dry matter contents.

The concentrates were prepared at lab-scale using a rotary evaporator (Hei-VAP Value Digital, Heidolph Instruments, Schwabach, Germany) at the same absolute pressure and evaporation temperature used in the pilot (i.e. 0.02 MPa and 60 °C respectively). The rotation speed of the 1-L flask was 80 rpm. For each concentrate, the starting material was always 500 mL of homogenized milk and the vacuum concentration was

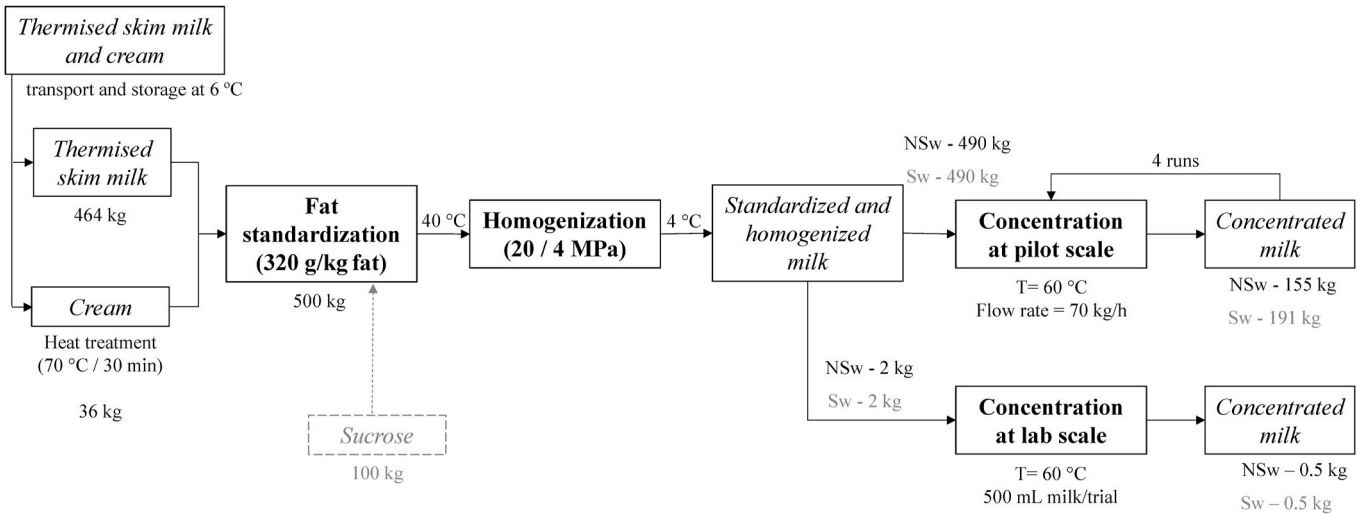

Fig. 1. Process scheme for the preparation of non-sweetened (NSw) and sweetened (Sw) milk concentrates. For the production of Sw milk concentrates, there is a supplementary step (in gray) i.e. addition of 200 g/kg of powdered sucrose to homogenized milk.

Table 1
Composition of non-sweetened homogenized milks AB_0 and sweetened homogenized milk CD_0.

	Non-sweetened milk AB_0	Sweetened milk CD_0	Analytical method described in section
pH	6.75 ± 0.02	6.74 ± 0.02	2.2.1
Sucrose (g/kg)	0	166.00	–
Dry Matter – DM (g/kg)	114.90 ± 0.10	255.99 ± 0.01	2.2.2
Dairy Dry Matter – DDM (g/kg)	114.90[a]	89.99[a]	–
Fat (g/kg)	31 ± 0	27 ± 0	2.2.3
Ash (g/kg)	6.89 ± 0.08	5.79 ± 0.04	2.2.4
TN (g/kg)	30.60 ± 0.03	26.83 ± 0.07	2.2.5
NCN (g/kg)	5.46 ± 0.42	5.35 ± 0.00	
NPN (g/kg)	1.47 ± 0.00	1.27 ± 0.00	
Calcium (mg/kg)	1176 ± 16	944 ± 1	2.2.6
Phosphate (mg/kg)	1880 ± 7	1545 ± 6	

[a]All the measurements were done in duplicate.
[a] Calculated: $(DM_{g/kg} = TDM_{g/kg} - Sucrose_{g/kg})$.

conducted up to the DM content reached at pilot-scale.

The DM content for the non-sweetened (AB_0) and sweetened (CD_0) milks were 114.9 and 256.0 g/kg. The method used for the determination of DM content is described in section 2.2.2. To reach the maximum DM of non-sweetened and sweetened concentrates usually achieved in industrial evaporators, it was necessary to carry out 4 and 3 runs in the pilot scale evaporator respectively. Four concentrates were produced from non-sweetened milk. The DM contents of the final concentrates produced at pilot scale and lab scale, A_4 and B_4 respectively, were 494.9 ± 0.1 and 491.3 ± 0.1 g/kg. Three concentrates were produced from sweetened milk. The DM contents of the final concentrates produced at pilot scale and lab scale, C_3 and D_3 respectively, were 708.0 ± 0.1 and 726.1 ± 0.1 g/kg (Fig. 2).

2.2. Physico-chemical characterization

In order to perform easily analysis and avoid age-thickening of more concentrated products, concentrates were diluted to the DM content of the corresponding homogenized milk for the determination of the following parameters: dry matter, nitrogen, ash and total ion contents.

The experimental values obtained for a biochemical parameter (nitrogen, ash and total ion contents) of a concentrate were compared to the theoretical value. It corresponds to the product of the experimental value in homogenized milk by the concentration factor.

2.2.1. pH measurement

The pH values of homogenized milks and concentrates were determined using a pH meter (HI 9024 microcomputer pH meter, Hanna Instruments, Lingolsheim, France) with a pH electrode (HI 1230B, Hanna Instruments, Lingolsheim, France).

2.2.2. Dry matter content

The DM contents were determined as described by IDF standard 21B (ISO-IDF, 1987). Five grams of each homogenized milk and re-diluted concentrate were mixed with sand in a capsule and were dried at 102 ± 2 °C in an oven for a period of 7 h. The weight loss of the capsule after drying was taken as the amount of water evaporated during vacuum concentration.

2.2.3. Fat content

The fat content of homogenized milks was determined using acid butyrometric method (FIL-IDF, 1997).

2.2.4. Ash content

The ash contents were determined by incineration of 10 g of each homogenized milk and re-diluted concentrate at 550 °C for 5 h and weight of the residue obtained (ISO-IDF, 1964).

2.2.5. Nitrogen contents

Total nitrogen (TN) contents of each homogenized milk and re-diluted concentrate were determined using the Kjeldahl method as described by IDF standard 20B (ISO-IDF, 1993). A factor of 6.38 was applied to convert nitrogen into milk protein content.

2.2.6. Ion contents

The mineral composition of homogenized milks and concentrates was determined using ion exchange chromatography (IEC-Dionex-500, Jouy-en-Josas, France) (Gaucheron, Le Graet, Piot, & Boyaval, 1996) for anions and atomic absorption spectrometry (AAS-220FS, Varian, Les Ulis, France) for cations (FIL-IDF, 1992). The experimental errors of both methods are ±0.5% and ±2.3%, respectively. The studied anions were inorganic phosphate, chloride and citrate and the studied cations were calcium, sodium, potassium and magnesium.

Total and soluble ion contents of milks and concentrates were determined according to Tanguy et al. (2019). The recovery of the soluble phase was carried out according to a 2-step procedure: (1)

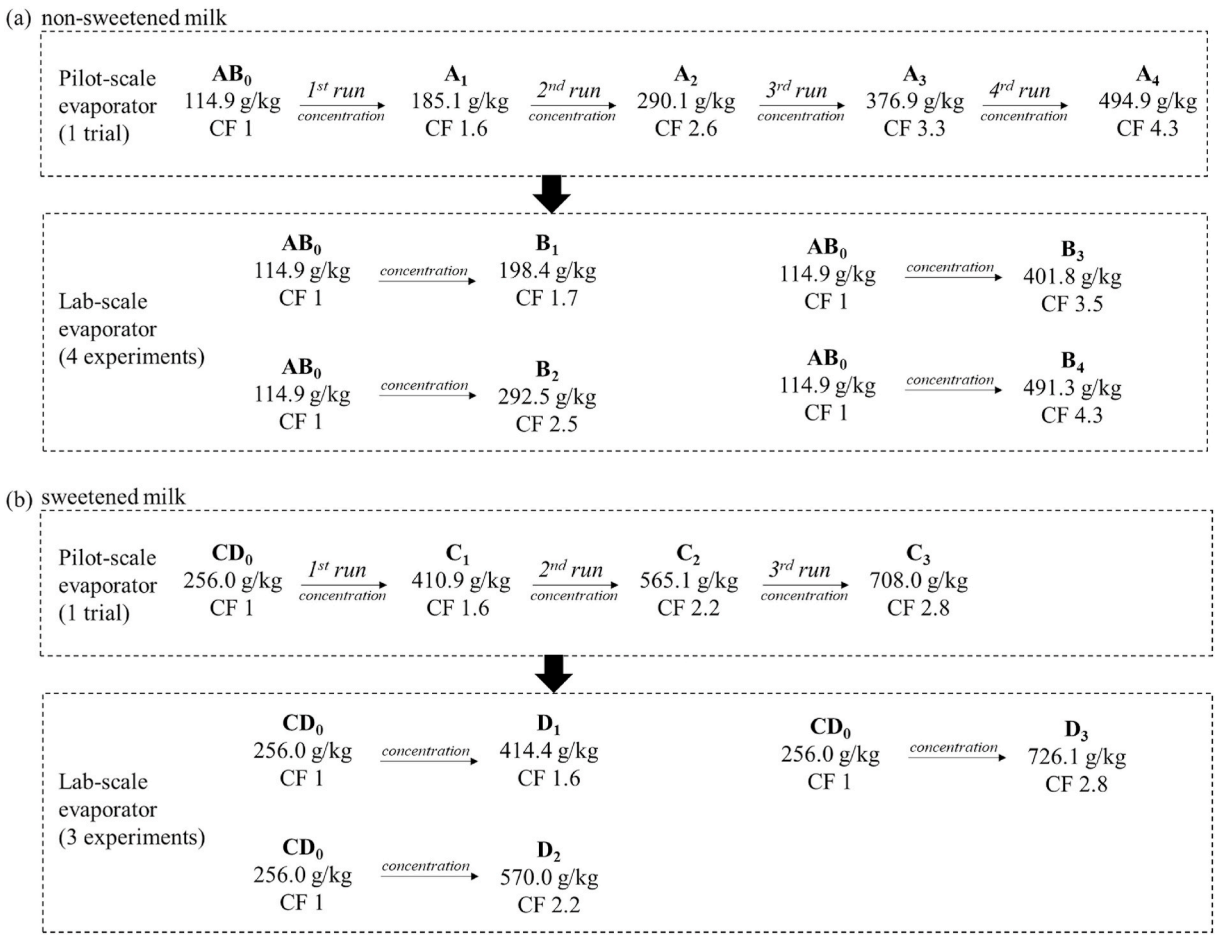

Fig. 2. Experimental procedure for the production of (a) non-sweetened milk concentrates and (b) sweetened milk concentrates at different DM contents using the pilot scale evaporator and the lab scale evaporator. The same milk was used for experiments at pilot scale and lab scale i.e. non-sweetened milk AB_0 (resp. sweetened milk CD_0) for the preparation of the non-sweetened milk concentrates (resp. sweetened milk concentrates). In both cases, the maximal DM contents was defined with respect to the maximal DM content achievable in industrial evaporators. The DM contents of the concentrates produced at lab-scale were defined in function of the DM contents of the concentrates produced at pilot-scale. DM = dry matter – CF = concentration factor.

ultracentrifugation of milks and concentrates for 1 h at 100,000 g and 40 °C in order to favor sedimentation of proteins (mainly caseins), (2) ultrafiltration of supernatant using analytical membrane (molecular weight cut-off of 10 kDa, Vivaspin, Palaiseau, France) and centrifugation for 1 h at 1800 g and at room temperature. This analytical procedure was performed in the same day that the concentration trials.

2.2.7. Viscosity measurement

The viscosity of concentrates was measured using a rotational viscometer type coaxial cylinder (RM 100 Plus, Lamy Rheology, Champagne-au-Mont-d'Or, France). Measurements were made at a controlled temperature of 60.0 ± 0.5 °C and at shear rates ranging from 100 to 500 s^{-1}. The experimental error was ±0.32 mPa s. After data collection, it is possible to make a graph and draw the trend line for each type of evaporator used (pilot and lab scale) and from them to predict the viscosity value in a certain desired dry matter value.

2.2.8. Density measurement

The density of milks and concentrates was measured using an oscillating density meter (DM48, Anton-Paar, Les Ulis, France) at a controlled temperature of 60.0 °C. The experimental error was ±0.1 kg/m^3. After data collection, it is possible to make a graph and draw the trend line for each type of evaporator used (pilot and lab scale) and from them to predict the density value in a certain desired dry matter value.

2.2.9. Surface tension measurement

The surface tension of milks and concentrates was measured at 60 °C using a pendant drop tensiometer ('Tracker', Teclis-Scientific, Civrieux-d'Azergues, France). To evaluate the impact of gravity on droplet shape, the Bond number Bo (Equation (3)) was calculated for all the samples.

$$\mathrm{Bo} = \frac{\rho g R^2}{\gamma} \tag{3}$$

where ρ is the droplet density (kg/m^3), g the acceleration of gravity (m/s^2), R the radius of the droplet (m) and γ stands for the droplet surface tension (N/m).

Drops with 8 μL and Bond number between 0.2 and 0.3 were formed at the tip of a syringe containing the samples and the measurements were made immediately after their formation. The drop profile was determined by image analysis using WDROP Software ('Tracker', Teclis-Scientific, Civrieux-d'Azergues, France) from which the surface tension was derived. Under mechanical equilibrium of capillary and gravity forces, the Laplace equation relates the pressure difference across the interface (liquid-air), the surface tension and the surface curvature. After data collection, it is possible to make a graph and draw the trend line for each type of evaporator (pilot and lab scale) used and from them to predict the surface tension value in a certain desired dry matter value.

2.2.10. Statistical analysis

The concentrations were done once and generated 4 products with

different concentration factors and analyzes made with each concentrate were performed in duplicate. The statistical analysis was done using R software version 3.5.3 (R Foundation for Statistical Computing, Vienna, Austria). With the results of density, viscosity and surface tension for the concentrates obtained in the rotary evaporator and in the pilot falling film evaporator an ANOVA was performed. If the F value indicated a difference between the means, the normality and homogeneity of the data was analyzed using the Shapiro-Wilk and Bartlett tests, respectively, and both tests at 5% significance. Finally, Tukey's analysis was used to identify group differences. Significance was defined as a P value < 0.05.

3. Results and discussion

3.1. Dry matter of concentrate

Four non-sweetened concentrates (A_1 to A_4) were prepared at pilot scale with DM contents ranging between 185.1 and 494.9 g/kg (Fig. 2). The DM content of the final concentrate A_4 is in the range of the maximum achievable values that can be obtained at the outlet of the industrial falling film evaporators for such a product (Schuck, Dolivet, & Jeantet, 2012).

The addition of powdered sucrose leads to an increase of the DM content of homogenized milk from 114.9 to 256.0 g/kg (AB_0 and CD_0 respectively). Three concentrates (C_1 to C_3) were prepared at pilot scale at 410.9, 565.1 and 708.0 g/kg (Fig. 2). The DM content of the final concentrate C_3 is the same that the one of a commercial product (BRASIL, 2018; Renhe et al., 2017).

The non-sweetened and sweetened concentrates have a variance less $\leq$1.8 when compared to the lab and pilot scales (Fig. 2).

3.2. Behavior of non-sweetened and sweetened concentrates during concentration

3.2.1. pH

Sucrose addition does not change the pH of homogenized milks (6.75 and 6.74, for AB_0 and CD_0 respectively). Then the pH of concentrates decreases during concentration in both cases probably due to (i) the increase in ionic strength that affects the activity coefficients and the pKa values of the protonated species, (ii) the transfer of some calcium and phosphate ions from the colloidal to the soluble phase, (iii) and the precipitation of calcium phosphate (Anema, 2009).

The pH variation between the non-sweetened milk AB_0 and the concentrate A_4 is 0.53 $\pm$ 0.01. Likewise, it is equal to 0.55 between the sweetened milk CD_0 and the concentrate C_3. However, the pH decreases at a given concentration factor is sharper for the sweetened concentrates than for the non-sweetened concentrates (Fig. 3a). For example, at a concentration factor of 2.2, the pH of the sweetened concentrate is 6.31 whereas it is about 6.53 for the non-sweetened concentrate. This sharper decrease in pH in the sweetened concentrates may be due to the addition of sucrose to milk that induces a decrease in water activity and promotes the Maillard reaction. Indeed, the Maillard reaction is strongly favored when the water activity decreases from 1 to 0.6–0.7. Even if the Maillard reaction is not intensified during vacuum concentration, the first steps of this complex reaction may occur, leading to the formation of formic acid and acetic acid which tends to lower the pH (Martins, Jongen, & van Boekel, 2001).

3.2.2. Total nitrogen and ash contents

The total nitrogen and ash contents are equal to 30.60 and 6.89 g/kg in non-sweetened milk whereas they are equal to 26.83 and 5.79 g/kg in sweetened milk. These lower nitrogen and ash contents in sweetened milk are related to a dilution effect induced by the addition of powdered sucrose.

As shown on Fig. 3b and c, total nitrogen and ash contents increase linearly with increasing concentration factors. It indicated that there are no losses of both compounds in the equipment.

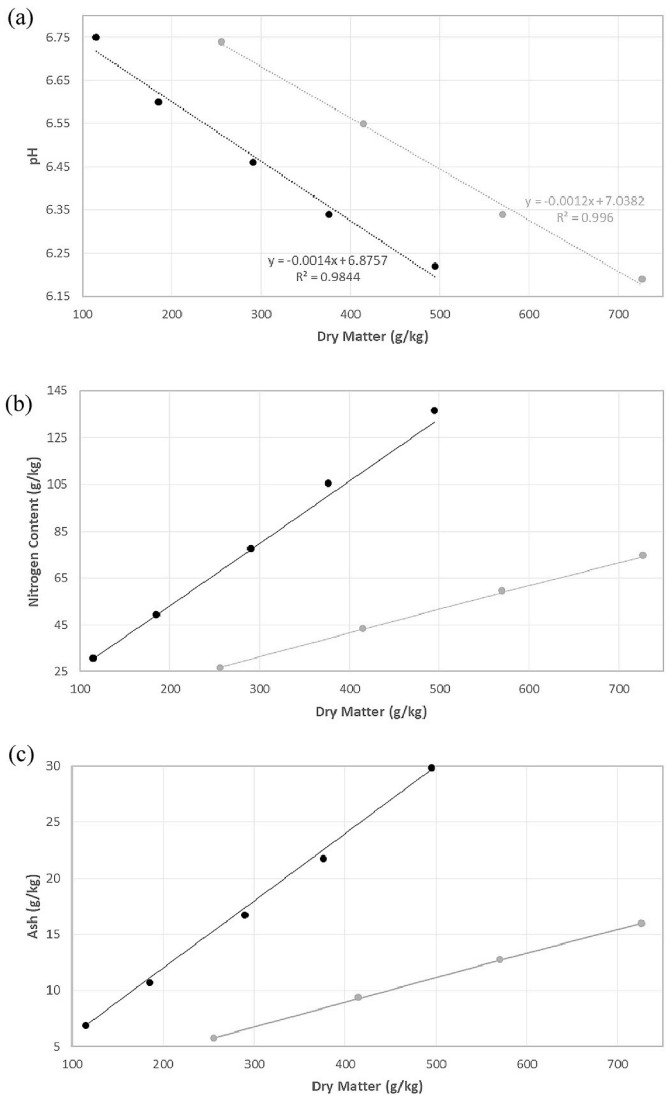

Fig. 3. Evolution of (a) pH, (b) total nitrogen and (c) ash contents of concentrates during concentration of non-sweetened (NSw) and sweetened (Sw) homogenized milks at pilot scale. Legend: Experimental data (●) NSw and (●) Sw; Theorical data (—) NSw and (—) Sw; Tendency lines (·····) NSw and (·····) Sw.

3.2.3. Minerals

Concerning the mineral composition of products, Fig. 4 highlights the experimental values of total calcium, phosphate, citrate and magnesium in the non-sweetened and sweetened concentrates are similar to the theoretical values, leading to the belief that there were no losses of these ions during the concentration process, more especially no ion deposit on the surface of evaporation tubes during concentration. Based on the visual analysis of the tubes and, mainly, on the analysis of minerals and proteins made in milks before concentration and throughout the concentration in the concentrated products. In the meantime, it is possible to observe that there is a difference between total and soluble ion contents and it increased with concentration. This difference corresponds in one hand to the ions into the colloidal phase that were mainly removed with caseins during the ultracentrifugation step, and in the other hand to the insoluble salts that may have precipitated in the concentrate during concentration. This latter part of insoluble ions was mainly removed during the analytical ultrafiltration and centrifugation steps. The difference between the total and soluble ion contents are all the more important with increasing concentration that there is a transfer

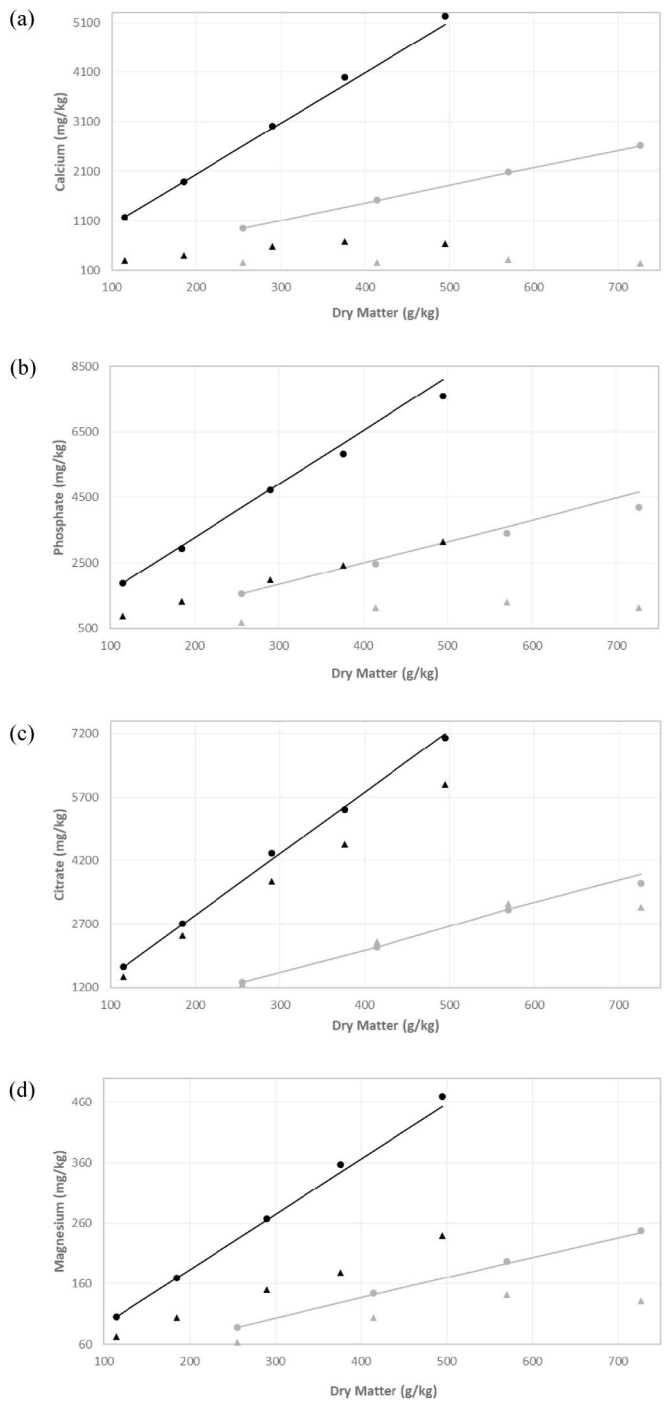

Fig. 4. Evolution of total and soluble ion contents during concentration of non-sweetened (NSw) and sweetened (Sw) homogenized milks: (a) calcium, (b) phosphate, (c) citrate and (d) magnesium. Legend: Theorical Total (—) NSw and (—) Sw; Experimental Total (●) NSw and (●) Sw; Soluble Experimental (▲) NSw and (▲) Sw.

of some salts from the soluble to the colloidal phase due to concentration.

3.2.4. Physical properties

Fig. 5 shows the evolution of some physical properties (density, viscosity and surface tension) of the concentrates throughout the concentration. In all cases, the experimental variance is plotted, but it is too small to see on graphics.

3.2.4.1. Density. The addition of 200 g/kg of powdered sucrose to milk induces an increase of milk density from 1.0111 ± 0.0002 for AB_0 to 1.0785 ± 0.0001 g/cm for CD_0[3]. The density increases linearly with the concentration factor for both types of products. Density of non-sweetened products increases from 1.0111 ± 0.0002 g/cm^3 for AB_0 to 1.1137 ± 0.0014 g/cm^3 for A_4 and B_4 (average between the two concentrates). In the meantime, density increases from 1.0785 ± 0.000 g/cm^3 for CD_0 to 1.28 ± 0.0001 g/cm^3 for C_3 and D_3 (average between the two concentrates).

3.2.4.2. Viscosity. The addition of 200 g/kg of powdered sucrose to milk leads to an increase of milk viscosity. Milk viscosity measured at 60 °C and 500 s^{-1} increases from 1.33 ± 0.14 mPa s for AB_0 to 3.02 ± 0.02 mPa s for CD_0. This is due to the addition of supplementary dry matter to milk.

Viscosity increases exponentially with concentration. It is all the faster than the dry matter of homogenized milk is high. The viscosity of non-sweetened products increases from 1.33 ± 0.14 mPa s for AB_0 to 23.4 ± 1.24 for A_4 and B_4 (average between the two concentrates) whereas the viscosity of sweetened products increases from 3.02 mPa s for CD_0 to 100 mPa s for C_3 and D_3 (average between the two concentrates).

3.2.4.3. Surface tension. The addition of 200 g/kg of sucrose to milk induces an increase in surface tension from 39.19 ± 0.69 mN m^{-1} for AB_0 to 43.82 ± 0.20 N m^{-1} for CD_0. According to the literature, sucrose (co-solvent) has a strong bond with water and this type of co-solvent does not bind directly to the surface of the protein, leaving a space that is filled with water making the protein hydrated, thus, this type of co-solvent is said to be preferentially excluded. This hydration protects globular proteins and mainly favors the native form of the protein, leaving it in its globular shape (Baier & McClements, 2003; Desu & Narishetty, 2013; McClements, 2002; Timasheff, 1993). The increase in surface tension observed in the analysis may occur due to three factors when adding sucrose: (i) the increase in viscosity that increases the stability of the product (Docoslis, Giese, & van Oss, 2000), (ii) the decrease in lipid-protein interactions at the interface (concentrate-air), because the protein is folded, leaving fewer groups exposed, like sulfhydryl groups, for interaction, with that the two constituents are on the surface, but in their separate forms (Wilde, Niño, Clark, & Patino, 1997) and (iii) the protein is preferably adsorbed at the interface (concentrate-air) (Wouters et al., 2017).

The surface tension of both products at 60 °C has the same behavior during concentration that is a decrease with concentration. The concentration of non-sweetened milk from AB_0 to A_4 and B_4 (average between the two concentrates) reduces the surface tension from 39.19 ± 0.69 to 34.32 ± 0.73 mN/m. In the meantime, the surface tension of sweetened products decreases from 43.82 ± 0.20 for CD_0 to 40.26 ± 0.33 mN/m for C_3 and D_3 (average between the two concentrates). Since proteins, fat and free fatty acids are the main surfactants found in milk, there is an increase in these constituents on the surface of the drop along the concentration, leading to the observed decrease (Williams, Jones, Paterson, & Pearce, 2005). With the dilution effect caused by sucrose addition to milk, there is also the dilution of these surfactants, thus leading to less variation in surface tension between non-sweetened and sweetened products.

3.2.5. Comparison of results at lab-scale and pilot-scale

As shown on Fig. 5, the trend curves of the physical properties of concentrates obtained at pilot and lab-scale are superimposed. However, some small deviations can be observed for properties that are sensitive to the variation of dry matter content, such as viscosity. In this case the variation of 18.2 g/kg DM between sweetened concentrates at pilot and lab scale (C_3 and D_3 respectively) showed a variance of 16.84 mPa s.

Statistical analysis of data from the following groups: surface

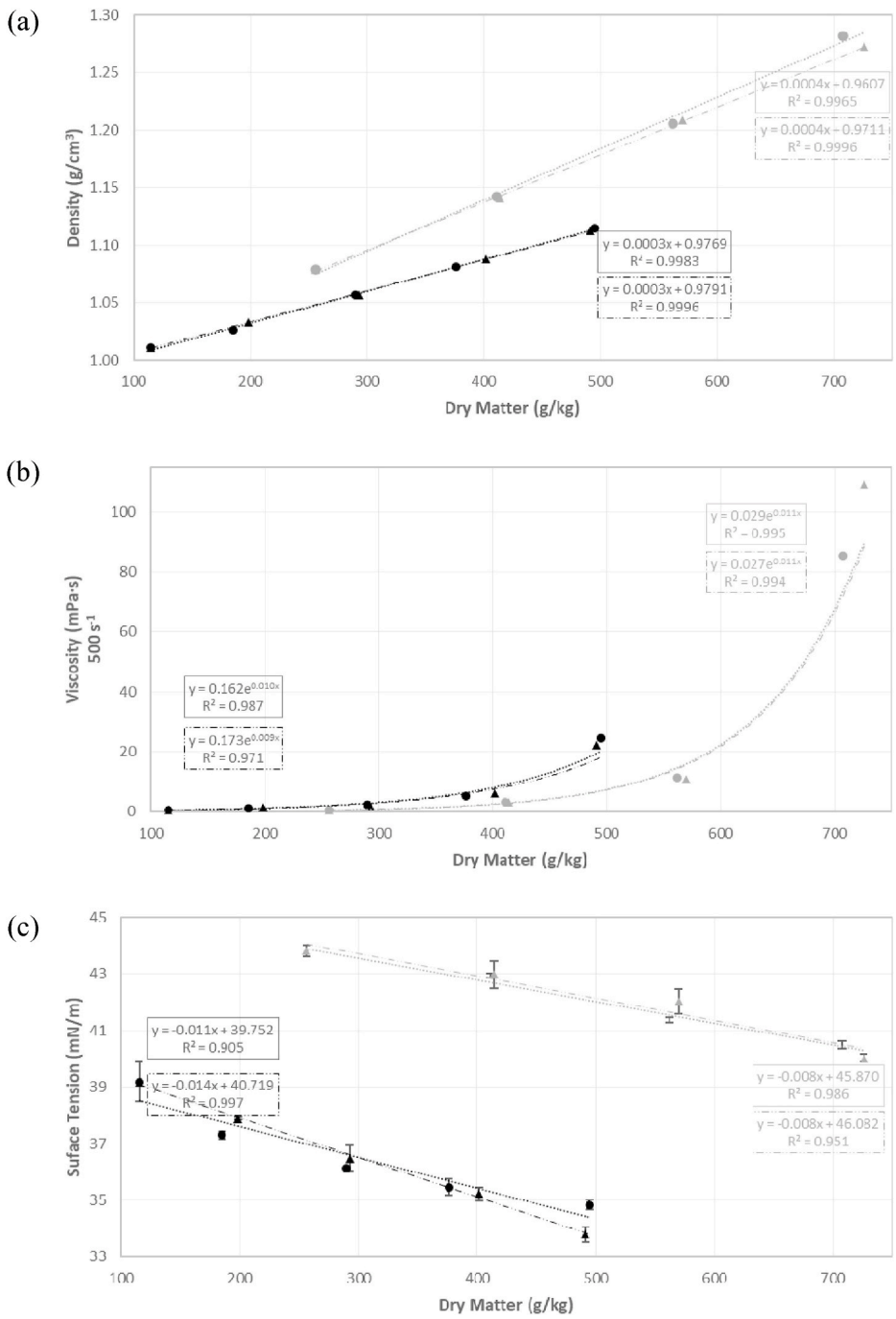

Fig. 5. Evolution of some physical properties of non-sweetened (**black**) and sweetened (**gray**) milk concentrates during concentration using a pilot scale evaporator (○) and a lab-scale evaporator (Δ): (a) density, (b) viscosity and (c) and surface tension.
Legend: (●) pilot scale-NSw, (▲) lab-scale-NSw, (●) pilot scale-NSw, (▲) lab-scale-NSw.

tension, density and viscosity detected through the F value of the ANOVA that there is some kind of significant difference within the data of each group at 5% significance. In order to identify what the differences would be, the Tukey test was carried out at 5% significance, which indicated that the differences were found at each concentration point and not between the samples of the two types of evaporators used. That is, when increasing the concentration of total solids, these concentrates had their properties significantly changed, but for the same concentration level in different equipment, a statistically significant difference was not detected. When modifying the concentration equipment there is no change in the product obtained at the same concentration level. Except, for the sweetened concentrates D_3 and C_3 (produced at lab scale and pilot scale respectively), which did not have similar averages for DM content (708.0 ± 0.0 and 726.1 ± 0.0 g/kg DM) and as a consequence, they have different viscosity values. But, when applying the trend line equations to 700 g/kg DM for example, similar viscosity values are obtained: 63.9 ± 0.2 and 68.7 ± 0.1 mPa s for concentrates prepared at lab-scale and pilot-scale respectively.

Density is a property highly dependent on the dry matter content and a variation of 18.2 g/kg in DM content between the sweetened concentrates produced at lab-scale and pilot-scale leads to a statistical differentiation between both concentrates. However, as for viscosity values, when comparing the results determined at a same DM content and using the trend lines, similar density values are obtained for

concentrates produced at lab-scale and pilot-scale.

These results showed that the values of density, viscosity and surface tension of concentrates produced at lab-scale are representative of values obtained for concentrates produced at pilot-scale. It is possible to change the falling film evaporator to the rotary evaporator in order to provide data on the physical properties of the concentrates.

3.2.6. Economic analysis

As both equipment used for this study allow producing concentrates at equivalent physical properties, it is interesting to evaluate the economic and environmental benefits of using smaller equipment.

The main difference between the rotavapor and the falling-film evaporator is the size of the equipment and their resulting evaporation rate, 0.5 kg/h of evaporated water and 27 kg/h respectively. It implies that the quantities of standardized milk required for carrying out experiments are greatly different. As shown in Table 2, when using the falling-film evaporator for the production of non-sweetened concentrate, at least 100 kg of standardized milk at 120 g/kg DM are necessary, which results in the production of about 24 kg of concentrate at 500 g/kg DM. If the objective of the trial is to recover some concentrate for further characterization such as biochemical composition and physical properties, a large part of the concentrate produced is thrown into drains and it generates unnecessary wastes.

We estimated that for our studies, a quantity of about 0.5 kg is enough to carry out microbiological and physico-chemical analysis on the concentrated products. The physico-chemical analysis included the measurement of viscosity, density, surface tension as well as the determination of the biochemical composition of the concentrates. Some samples can also be used to prepare the subsequent drying step in the production process of dairy powders using for example the desorption method (Schuck et al., 1998, 2009). This method allows to analyze the final product's moisture and water activity. Another possibility is to spray dry the concentrate obtained at lab scale in a small spray-dryer similar to the one used by Maury, Murphy, Kumar, Shi, and Lee (2005). From the combination of both equipment at lab-scale, it is possible to predict the behavior of the product during drying and identify the more relevant operating parameters for the drying stage at pilot and/or industrial scale. In addition, for both products, microbiological tests can be performed, to determine for example the growth rate and growth conditions of microorganisms and evaluate the evolution of the product during storage and their shelf life.

Another drawback when using the pilot scale falling-film evaporator to produce samples for product characterization is the use of larger volumes of cleaning agents. Nitric acid and sodium hydroxide solutions are generally used for the cleaning of evaporators (Goode, Asteriadou, Robbins, & Fryer, 2013; Hagsten et al., 2016; Jeurnink & Brinkman, 1994). For our trials, we used about 20 L of alkaline solution (20 ml/L) and 20 L of acid solution (10 ml/L) to clean the equipment before experiment and again 20 L of alkaline solution (20 ml/L) and 20 L of acid solution (10 ml/L) after experiment. In comparison, the lab scale rotary evaporator requires only ~ 10 mL of biodegradable detergent. As a consequence, it reduces drastically the impact on the environment (Table 2).

The use of a rotary evaporator allows also reducing the experiment time. Considering only the concentration step of the product, the equipment use time do not differ too much, respectively 4.0 and 2.8 h for the rotary and the falling-film evaporators. However, if we consider the total duration of use of the equipment, the falling film evaporator is greatly longer than the one of the rotary evaporator. Indeed, the cleaning steps before and after concentration, the stabilization of operating parameters before concentration and the cooling of the equipment after concentration extend the duration of use of the equipment. A complete cleaning step lasts about 2 h, the stabilization of the operating parameters and the cooling about 30 min each (Table 2). At lab scale, these procedures are shorter (cleaning step and stabilization of operating parameters) or are not applied (no cooling of the equipment after concentration).

The energy consumption was calculated taking into account the electric power of each pump (manufacturer's manual data) of the equipment and their time use. Thus, for the pilot scale falling-film evaporator, 4 product pumps (0.75 kW each), 1 condensate pump (0.75 kW), 1 vacuum pump (1.1 kW) and 3 boilers (8.4 kW each) were used during 6.8 h, resulting in an electrical consumption equal to 204.3 kWh. For the rotavapor, 1 cooler (0.14 kW), 1 rotavapor (1.4 kW), 1 thermostatic bath (1.3 kW) and 1 vacuum pump (0.18 kW) were used during 3 h, resulting in a consumption of 9.1 kWh (Table 2). The pilot scale thus shows a saving of 95.5% in energy consumption. The analysis of energy consumption within the production of dairy products is very relevant, since evaporation alone consumes 12% of electrical energy and 39% of thermal energy for the manufacture of powdered milk (Finnegan, Goggins, Clifford, & Zhan, 2017). Therefore, it is extremely important when using an alternative equipment to do a test screening.

Longer duration uses of equipment, higher volumes of product and cleaning agents for the experiments at pilot scale impact greatly on the water and energy consumptions. We estimated that a trial on the falling film evaporator consumes about 2600 L (Table 2). It should be noted that about 92% of this volume is used at the condenser to ensure the production of vacuum inside the evaporator. The facility is not equipped with recovery systems and the installation of a closed-loop cooling system would reduce drastically water consumption. At lab scale, only an 8-L thermostatic bath and a cooler with recirculating water are used. Therefore, the water effectively consumed by this equipment is used to clean the evaporation flasks (approximately 2 l). Obviously, when using a recirculation method for the condenser water, this consumption value would be reduced and the impact would already be less in relation to the amount of water discharged. However, in the other parameters analyzed throughout the work, this would not influence when having equipment with similar characteristics such as evaporative capacity and flow.

Table 2
Comparison of the carrying out of experiments in a falling-film evaporator and a rotavapor – Application to the concentration of non-sweetened milk at 120 g/kg DM for the production of milk concentrate at 500 g/kg DM.

Parameters	Pilot scale *Falling-film evaporator*	Lab scale *Rotavapor*
Initial quantity of milk at 120 g/kg DM (kg)	100	2
Maximal evaporation rate of the equipment (kg/h)	27	0.5
Quantity of concentrate at 500 g/kg DM (kg)	24	0.5
Quantity of evaporated water (kg)	76	1.5
Duration of the concentration step (h)	4	2.8
Total duration of use of the equipment [a] (h)	6.8	3
Cleaning agents used	acid and alkaline solutions	Bio detergent
Volume of cleaning agents	40.44 l (alkaline solution) 40.16 l (acid solution)	~ 10 mL
Total water consumption [a] (L)	2600	2.0 + 8.0[b]
Total energy consumption [a] (kWh)	204.3	9.1

[a] Including the concentration and the cleaning steps.
[b] Reusable and/or recirculating water.

4. Conclusions

In the process scheme for the production of sweetened concentrated milk, it is known the addition of 200 g/kg of powdered sucrose to milk alters greatly the physico-chemical properties of milk and the resulting concentrates produced using vacuum concentration in falling-film evaporators. As an example, the maximum achievable dry matter content at the outlet of the evaporator is 500 and 720 g/kg for non-sweetened milk concentrates and sweetened milk concentrates. The

experimental results of this study provided more data on the physical properties of sweetened milk concentrates.

The non-sweetened and sweetened concentrates were prepared in parallel in a lab-scale rotary evaporator and in a pilot scale falling-film evaporator. They were characterized in particular in term of density, viscosity and surface tension. For these parameters, low variance was observed between the products obtained in both concentration equipment. Furthermore, the sensitivity of the viscosity and density parameters to a low variation in total solids between the last sweetened concentrate of both concentration equipment was reaffirmed. It makes then possible to change the concentration step for the production of sweetened and non-sweetened milk concentrates from pilot to lab scale by using a rotary evaporator. It allows to reduce the duration of trials, the volume of raw material, to save energy and water and preserve environment. However, the change can be used when the aims of the trials are to get samples for further characterization and when the volume required is low.

CRediT authorship contribution statement

Érica Felipe Maurício: Formal analysis, Writing – original draft, Data curation, Visualization. **Gaëlle Tanguy:** Methodology, Formal analysis, Writing – review & editing, Supervision, Project administration. **Cécile Le Floch-Fouéré:** Formal analysis, Resources, Supervision, Project administration. **Eric Beaucher:** Data curation. **Anne Dolivet:** Data curation. **Antonio Fernandes de Carvalho:** Resources, Funding acquisition. **Ítalo Tuler Perrone:** Methodology, Formal analysis, Writing – review & editing, Supervision, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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