



Modelling water and salt diffusion of cold-smoked Atlantic salmon initially immersed in refrigerated seawater versus on ice

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ABSTRACT

The effect of dry salting during the cold-smoking process was evaluated on Atlantic salmon initially stored in refrigerated seawater (RSW) or ice. A 2D mathematical model was developed from first principles, simulating the heat and mass transfer process during dry salting at increasing salting duration. This model was validated using experimental values and compared with the empirical model of Zugarramurdi and Lupin. The predicted values were used for water activity prediction and validated. It was found that salting duration influenced drip loss, redness and yellowness, texture, water activity, salt uptake and water loss. Smoked salmon from RSW fish were more reddish with a lower water activity than iced fish after vacuum storage. Drip loss and colour were significantly influenced by the processing step (salting, smoking and storage). Overall, the model presented reasonable predictions for temperature, salt and water content, water activity and was in close agreement with the empirical model.

1. Introduction

As the population becomes more mindful of their health, “The Keyhole” is a voluntary Nordic label on food packages that encourages consumers to choose healthier products. In Norway, to obtain this label for cold-smoked salmon, the regulation states that the final salt (sodium chloride, NaCl) content must be less than 3 g NaCl/100 g product (Ministry of Health and Care Services, 2015). Cold-smoked salmon is a lightly preserved fish product with a salt content of 1.7–5.1 % in the water phase. Its water content is between 65 and 70 % (Cardinal et al., 2004) and pH between 5.8 and 6.3 (Hansen et al., 1995).

The recent advancement in the salmon processing industry introduces a novel fish slaughter method that effectively circumvents several steps in the value chain. This method slaughters fish directly by the net pens onboard a slaughter vessel followed by superchilling (<0 °C) them in refrigerated seawater (RSW) tanks during transportation. The study of Chan et al. (2020a) showed that immersing fish in RSW leads to slightly higher water and salt uptake than the traditional method of storing on ice. Nevertheless, there were minimal differences in quality when both groups were cold-smoked, and the primary

determinant was storage duration. This gives an insightful notion that RSW fish can also produce high-quality cold-smoked salmon like those on ice. Thus, in addition to the successive benefits the new slaughter method brings (for instance, reduced environmental impact, shortened lead time and increased fish welfare), there presents a possibility that this slaughter method could revolutionize the fish processing industry.

Water activity (a_w) is a dimensionless physical parameter that measures the availability of water and is related to microbial growth. Various standard salting procedures are used in the salmon processing industry that can change the product's functionality, such as organoleptic properties, dehydration, solubilizing proteins and changing osmotic pressure to prevent microbial growth. Dry salting is one of the oldest food preservation methods by depressing a_w . It enhances shelf life by spreading crystalline NaCl on the product's surface until it diffuses into the product and equilibrates. The primary process in salting is diffusion, causing counter current water and salt transport between the salt and muscle and resulting in a high salt concentration in the fillet surface during the smoking process (Lerfall et al., 2011). The conformation of muscle protein is affected, causing changes in water holding capacity (WHC) and potentially, protein denaturation. At low salt

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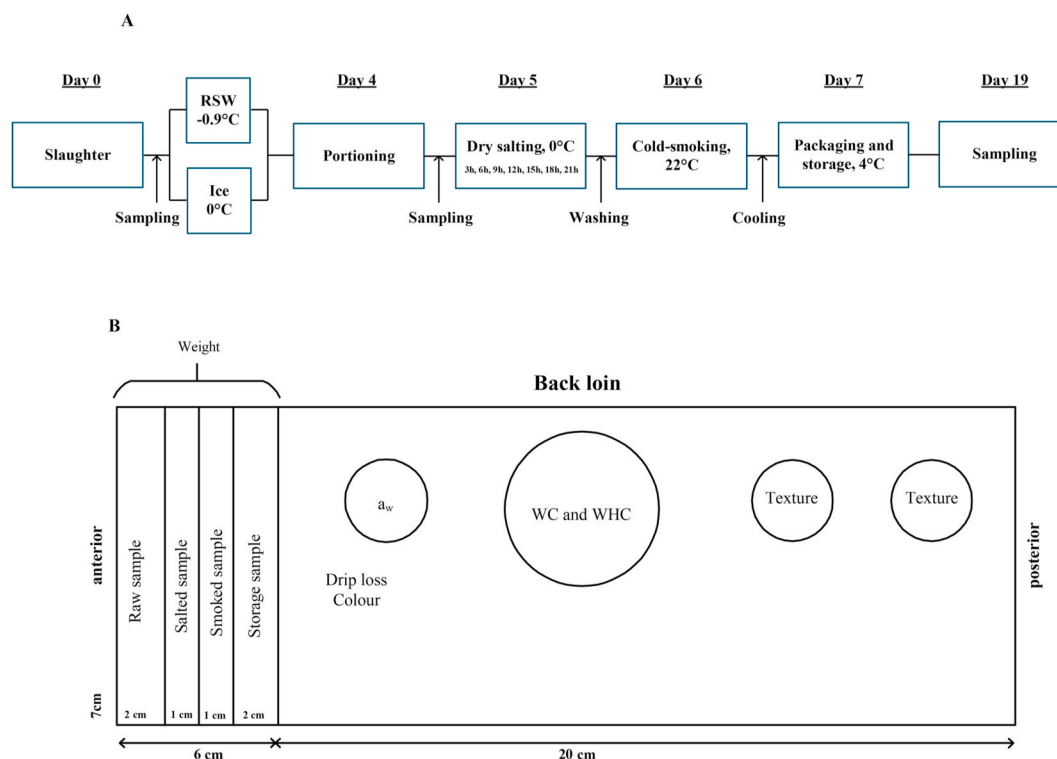


Fig. 1. (a) Graphical illustration of the experimental timeline. (b) Graphical illustration where sampling was done on the fillet portion after 12 days of storage, on day 19 post mortem. Small pieces were cut off (total of 6 cm) at every processing step and deep-frozen as backup analysis.

concentrations (<5–6%), a lower degree of protein denaturation occurs and causes swelling from the electrostatic repulsion of chloride ions weakly attached to the myofibrillar and sarcoplasmic proteins (Larsen and Elvevoll, 2008; Offer and Trinick, 1983). This expands the filament lattice and entraps free water, leading to an increase in WHC. Therefore, salting influences several quality parameters like WHC, taste, shelf life, texture, colour and fillet yield in the final smoked product (Birkeland and Bjerkeng, 2005; Birkeland et al., 2004; Løje, 2007).

The salting process can be simulated with a modelling approach. Quality measurements for food can often be time-consuming and dependent on laboratory-based premises. Some of the bottlenecks in laboratory analysis include cost, rate and labour intensity. Mathematical modelling is a powerful tool that can be used as an alternative for a wide variety of purposes in the food industry. For the past decades, it has received much attention in food science, technology and engineering, significantly reducing the experimentation process (Banga et al., 2008; Datta, 2008). The heat and mass transport phenomena in food can be described with coupled partial differential equations (PDEs) under various assumptions (including appropriate initial and boundary conditions) and solved numerical method to predict the state variables (e.g., temperature, concentration) as a function of space and time. The model predictions are validated with experimental data. The established model can aid in predictions, process designing and optimization in the industry concerning food quality and safety. Several researchers have used numerical methods based on mechanistic principles to study the prediction of heat and mass transfer in a convection oven heating process on fish and meat products like cod (Blikra et al., 2019), chicken (Rabeler and Feyissa, 2018), chicken patties (Chen et al., 1999) and pork (Feyissa et al., 2009, 2013). Modelling the mass transfer phenomena has also been done in salting of cod (Andrés et al., 2002; Blikra et al., 2020), brining on herring (Laub-Ekgreen et al., 2019) and brining (Wang et al., 1998, 2000) and dry salting on salmon (Martínez-López et al., 2019).

On the other hand, empirical models are built solely on mathematical equations developed from experimental data. The application of empirical models, like the Zugarramurdi and Lupín (1980) model (Z&L)

for fish salting, is also used in predicting the development of average salt and water concentration over time. This uses an exponential approach to the equilibrium values and has been verified on several fish species like catfish (Corzo et al., 2015), sardines (Bellagha et al., 2007) and anchovies (Bellagha et al., 2007).

A better understanding of salting kinetics can help industries evaluate existing conditions and develop new products. Therefore, this study aims to compare the quality parameters of cold-smoked salmon produced from fish initially chilled in RSW and on ice subjected to different salting times. A mathematical model was developed to predict the temperature during salting, water and salt profiles, and water activity at increasing salting duration with a fixed cold-smoking procedure. In addition, to demonstrate the robustness of the model for RSW and ice stored fish, it was compared to the empirical model of Zugarramurdi and Lupín (1980).

2. Materials and methods

2.1. Experimental design

Before the experiment, an 800-L polyethylene tank was obtained and thoroughly washed with lye before filling in refrigerated seawater (RSW, 3.5 % salinity) kept at a temperature between -0.5 and -1 °C. After that, 54 farmed Atlantic salmon (*Salmo salar*) were obtained from a local slaughterhouse (November 2020) with an average weight of 3.6 kg (starved for 9 days, core temperature ~ 1.6 °C). The fish were electrically stunned, thoroughly bled, and gutted before being weighed and packed in either the RSW tank ($n = 24$) or expanded polystyrene (EPS) boxes containing ice ($n = 24$). Six fish were used to sample for raw material determination of pH, water content (WC), water holding capacity (WHC), salt, protein, and fat content. In each group, two TrackSense Pro temperature loggers (Ellab A/S, Denmark) were inserted into the mid-abdomen of two random fishes while one logger was placed in the surrounding environment. The tank and boxes were transported to the laboratory within 2 h and kept in a 0 °C storage room for 4 days. The

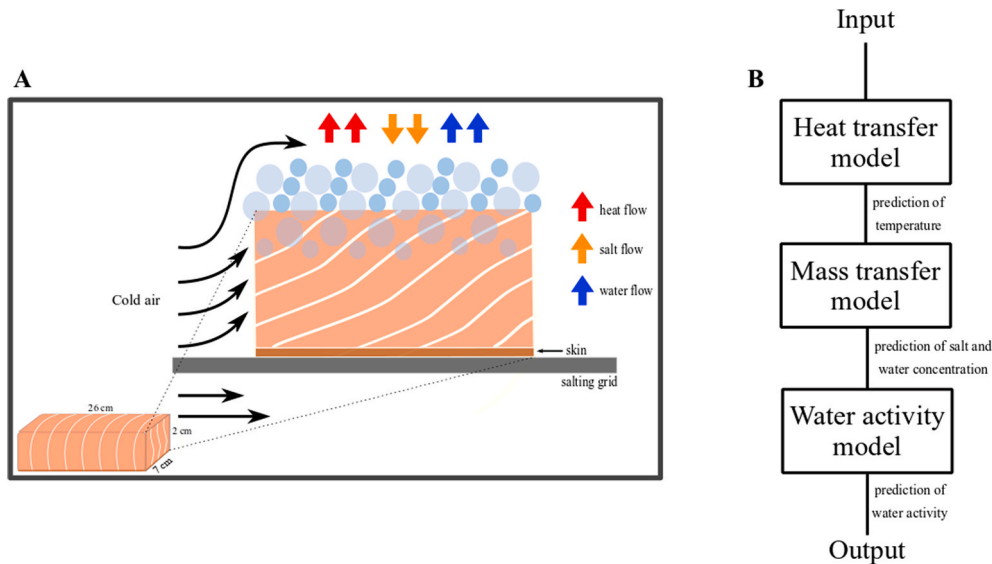


Fig. 2. (a) Transport process during dry salting in a cold room. (b) Input and output parameters used for prediction.

tank's temperature was monitored continuously and kept below 0 °C by adding clean seawater ice, similar to the study of Chan et al. (2020a). A graphical illustration of the experimental timeline is shown in Fig. 1a.

2.2. Processing and quality analysis

On day 4, the RSW tank was drained and the fish were gently wiped with paper. All fish from both groups were individually weighed and manually filleted, and five fish from each group were used for sampling. The remaining fillets were portioned on the back loins into rectangular parallelepiped-shaped samples (26 cm × 7 cm × 2 cm). Weights and colour measurements were taken prior to dry salting the samples' surface with excess refined salt (99.8 % NaCl, GC Rieber, Norway) in randomized grids at 0 °C for every 3 h interval until 21 h. Thermocouples type K (PR Electronics Inc., USA) was inserted at the center of one random salmon portion. The temperature was logged in an Eval Flex recorder every second (Eval Flex, Denmark). After salting, the portions were washed with cold tap water (6–8 °C), gently dried and weighed before cold-smoking using the alternating drying and smoking protocol of Birkeland and Skåra (2008). After smoking, the portions were cooled overnight at 1 °C, then vacuum-packaged (99.9 % vacuum) and stored for 12 days at 4 °C. Sampling was done after storage, as seen in Fig. 1b. Colour analyses were also taken after smoking and after storage. The weights of the portions were recorded during every processing step before cutting a small piece of sample on the anterior part and frozen at –80 °C for further analysis. The weight change is calculated as the % difference with respect to the initial weight before processing.

For quality analysis, WHC was measured based on the centrifugation method of Skipnes et al. (2007), which also calculates WC. Acidity was measured using a portable pH meter (Mettler Toledo SevenGo pro, Mettler Toledo Inc, USA). Water activity (a_w) analysis was done using the NMKL (2001) method no. 168 with an a_w meter (AquaLab Series 3, METER Group, USA). Fat and protein content were extracted using the Bligh and Dyer (1959) and the Kjeldahl method (AOAC 928.08). Salt analysis was done using the automatic titration method with 0.1 M AgNO₃ (VWR International, Norway) using SI Analytics Titroline 7000 (Xylem Analytics, Norway) (Chan et al., 2020a). Colour analysis was implemented using the DigiEye complete system (VeriVide Ltd, UK) connected to a Nikon D80 camera (Nikon Corp, Japan). The $L^*a^*b^*$ values were calculated using the DigiPix software (VeriVide Ltd, UK), where L^* represents lightness, a^* redness and b^* yellowness (CIE, 1994). The ΔE value was further calculated by

$$\Delta E = \sqrt{(L_2^* - L_1^*)^2 + (a_2^* - a_1^*)^2 + (b_2^* - b_1^*)^2} \quad (1)$$

Texture analysis was performed in replicates with the puncture test using a Texture Analyzer TA-XT plus (SMS Ltd, UK) connected to a 12.5 mm flat-ended cylinder at a constant speed of 2 mm/s. The force-time graph was recorded using the Texture Exponent software, and the force to press the cylinder down to 80 % of the fillet height represents the firmness.

2.3. Statistical analysis

Statistical analysis was performed using Minitab® 19. The association of treatment (RSW, ice), salting time (3 h–21 h), processing step (raw, smoked, stored), and response variables were analyzed using the general linear model (GLM) at a 95 % confidence interval. Fillet height was added as a covariate for texture analysis. Normality assumptions were tested before the analyses. The root mean square error (RMSE) was used to evaluate the performance between the experimental data and model predictions. All results are presented as mean ± standard deviation and statistical significance set at $p \leq 0.05$.

3. Model formulation

Dry salting in cold storage is a cooling process involving heat transfer from the product to the surrounding air. The primary mechanism during salting of salmon is molecular diffusion through the aqueous phase of the muscle, which induces water from the muscle fibers to migrate out (Gómez-Salazar et al., 2015) (Fig. 2a). During dry salting and cold-smoking, cold air is transferred by convection to the product surface and conduction within the product. The temperature at the centre of the salmon portion in cold storage during salting was first predicted using the input parameters, including the estimated heat transfer coefficient, coupled with the moisture and salt transport. Afterwards, the obtained moisture and salt concentrations were used to predict the water activity (Fig. 2b).

The model focused mainly on water and salt transport. Certain assumptions were made for model simplification to formulate the coupled heat and mass transfer for a rectangular parallelepiped shape of fish meat, as follows. There was no internal heat generation. The shrinkage during salting was also neglected. Furthermore, the skin was present only on the bottom side, so it did not hinder water and salt transport. Since the length of the sample was much larger than the thickness of the

Table 1
Model input parameters.

Parameter	Symbol	Value	Unit	Source
Initial temperature	T_0	279.35	K	measured
Initial composition				
protein	y_p	0.22	kg/kg	measured
fat	y_f	0.09	kg/kg	measured
water	y_w	0.661	kg/kg	measured
ash	y_a	0.018	kg/kg	Aas et al. (2019)
Initial concentration				
salt				
RSW	C_{s0}	0.002	kg/kg	measured
ice		0.0015	kg/kg	measured
water				
RSW	C_{w0}	0.668	kg/kg	measured
ice		0.66	kg/kg	measured
Surrounding concentration				
salt	C_{s1}	0.15	kg/kg	estimated
water vapour in ambient air	C_{w1}	0.05	kg/kg	Rabeler and Feyissa (2018)
Density				
water	ρ_w	998	kg/m ³	Rao et al. (2014)
protein	ρ_p	1330	kg/m ³	Rao et al. (2014)
fat	ρ_f	926	kg/m ³	Rao et al. (2014)
ash	ρ_a	2424	kg/m ³	Rao et al. (2014)
fish	ρ_s	1071	kg/m ³	calculated
Moisture diffusion coefficient	D_w	3.98×10^{-10}	m ² /s	Martínez-López et al. (2019)
Salt diffusion coefficient	D_s	6.64×10^{-10}	m ² /s	Akköse and Aktaş (2016)
Heat transfer coefficient	h_a	15.0	W/(m ² K)	estimated
Thermal conductivity of fish	k_s	0.47	W/(m K)	Rao et al. (2014)
Mass transfer coefficient between muscle and salt	$k_{m,s}$	3.54×10^{-7}	m/s	Martínez-López et al. (2019)
Mass transfer coefficient between muscle and water	$k_{m,w}$	1.73×10^{-8}	m/s	estimated
Specific heat capacity salmon	c_p	3436	J/(kg K)	calculated

sample, 2D geometry was used for the modelling.

3.1. Calculation of heat transfer coefficient

The heat transfer coefficient of convective heat transfer during the salting and cooling process was determined by comparing the experimental and predicted temperature profiles. Thermocouples type K were also inserted in the center of three Teflon cylinders ($d = 2$ cm, $x = 20$ cm) hanged at the bottom, middle and top positions of the salting grid concurrent to the salmon salting process in the cold room. The temperature was also logged in an Eval Flex every second until it stabilized, and the average temperature was calculated. The lumped system analysis was first tried to calculate the heat transfer coefficient using Eqs. (2a) and (2b) (Isleroglu and Kaymak-Ertekin, 2016).

$$\ln\left(\frac{T_\infty - T(t)}{T_\infty - T_0}\right) = -bt \quad (2a)$$

$$b = \frac{h_a A}{\rho C_p V} \quad (2b)$$

where h_a is the combined heat transfer coefficient (W/(m² K)), T_∞ is the surrounding temperature (K), T_0 is the initial temperature (K), A is the surface area (m²), V is the volume (m³), ρ is the density (2200 kg/m³), c_p

is the specific heat capacity (1172 J/(kg K)) of the material, and t is time (s). The obtained h_a was 13.2 W/(m² K) with an RMSE value of 1.50 W/(m² K). However, as the Biot number of the Teflon cylinder was slightly higher than 0.1, this obtained value may be inaccurate. Hence, a heat transfer model of the Teflon cylinder was predicted in COMSOL Multiphysics as described in Section 3.3.1 and 3.4 using the reverse estimation method, replacing the thermophysical properties with those of Teflon. The h_a value was adjusted until the predicted and measured temperature profiles showed close agreement. The final value obtained was 15.0 W/(m² K) and RMSE value 0.15 W/(m² K), which was a better fit than the Lumped capacity method. This value was therefore used as the input parameter for modelling water and salt transport.

3.2. Thermophysical properties

The model input parameters are presented in Table 1. The density of salmon and its heat capacity was estimated from its composition using Eqs. (3) and (4), respectively (Choi and Okos, 1986).

Density of salmon:

$$\rho = \frac{1}{\sum \frac{y_i}{\rho_i}} \quad (3)$$

Specific heat capacity of salmon:

$$c_p = (2y_p + 2y_f + 4.2y_w + 2.4y_a) \cdot 10^3 \quad (4)$$

where ρ , ρ_w , ρ_p , ρ_f and ρ_a are the densities (kg/m³) of fish, water, protein, fat and ash respectively, y_i is the mass fraction of each component and c_p is the specific heat capacity of fish (J/(kg K)). Thermal conductivity of 0.47 W/(m K) was used (Rao et al., 2014).

3.3. Governing equations

3.3.1. Heat and mass transfer

The heat transfer within the salmon muscle is based on the heat diffusion equation, given by

$$\frac{\partial T}{\partial t} = \frac{k_s}{c_p \rho_s} \nabla^2 T \quad (5)$$

where c_p , ρ_s and k_s are the specific heat capacity (J/(kg K)), density (kg/m³) and thermal conductivity (W/(m K)). ∇ is the Nabla operator, i.e. partial derivative in x , y and z -direction ($\nabla = \frac{\partial}{\partial x} + \frac{\partial}{\partial y} + \frac{\partial}{\partial z}$). T is the temperature (K), and t is the time (s).

The governing equation for mass transfer of water (Eq. (6a)) and salt (Eq. (6b)) within the muscle is based on the conservation of mass (Fick's second law), given by

$$\frac{\partial C_w}{\partial t} = D_w \nabla^2 C_w \quad (6a)$$

$$\frac{\partial C_s}{\partial t} = D_s \nabla^2 C_s \quad (6b)$$

where C_w and C_s are the water and salt concentrations (kg/kg), respectively. D_w and D_s are the water and salt diffusion coefficients (m²/s), respectively.

3.4. Initial and boundary conditions

We assume a uniform initial temperature, moisture, and salt distribution throughout the whole sample.

$$T(x, y, z, 0) = T_0 \quad (7)$$

$$C_w(x, y, z, 0) = C_{w0} \quad (8a)$$

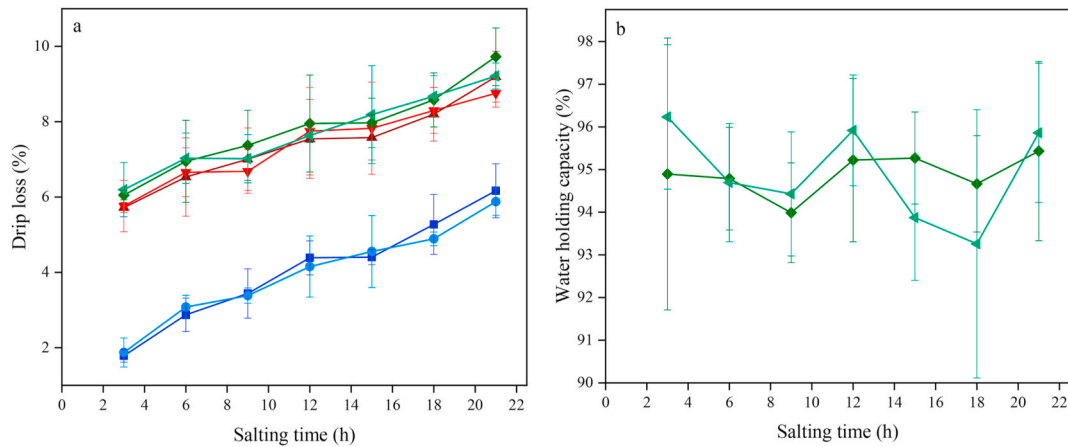


Fig. 3. (a) Drip loss for RSW fish after salting (■), ice fish after salting (●), RSW fish after smoking (▲), ice fish after smoking (▼), RSW fish after storage (◆), ice fish after storage (◀) (GLM; salting hour – after salting: $p < 0.001$, after smoking: $p < 0.001$, after storage: $p < 0.001$; treatment – after salting: $p = 0.596$, after smoking: $p = 0.951$, after storage: $p = 0.499$). (b) WHC for RSW fish after storage (◆), ice fish after storage (◀) (GLM; salting hour: $p = 0.895$, treatment: $p = 0.975$). All results are presented in mean \pm standard deviation.

$$C_s(x, y, z, 0) = C_{s0} \quad (8b)$$

where is T_0 , C_{w0} and C_{s0} are the initial temperature (K), water and salt concentration (kg/kg) respectively.

The boundaries that are exposed to the surrounding air and heat flux is given by:

$$-k_s \nabla T = h_a (T_\infty - T_s) \quad (9)$$

where k_s is the thermal conductivity of salmon (W/(m K)), h_a is the heat transfer coefficient (W/(m² K)), T_∞ is the surrounding temperature (K) and T_s is the surface temperature (K) of the salmon.

The boundary condition for the mass transfer is:

$$D_w \nabla C = k_{m,w} (C_{w1} - C_w) \quad (10a)$$

$$D_s \nabla C = k_{m,s} (C_{s1} - C_s) \quad (10b)$$

where $k_{m,w}$ and $k_{m,s}$ are the mass transfer coefficient (m/s) between muscle and water, and muscle and salt, respectively. C_{w1} and C_{s1} are the concentration of water vapour in the surrounding air and salt (kg/kg), respectively. C_w and C_s are the concentration of water vapour and salt at the surface of the fillet (kg/kg), respectively.

3.5. Calculation of water activity

Water activity can be calculated as a salt molality function that couples salt and water transport, given by Eq. (11) (Martínez-López et al., 2019; Pazuki, 2005).

$$\ln a_w = -M_w \cdot \varphi \cdot v \cdot m = -\frac{M_w}{M_s} \cdot \varphi \cdot v \cdot \frac{C_s}{C_w} \quad (11)$$

where M_w and M_s are the molecular weights of water (0.018 kg/mol) and salt (NaCl, 0.0583 kg/mol), φ is the osmotic coefficient, v is the number of solute ions (2), C_s and C_w are the water and salt concentrations on wet basis (kg/kg). The value of the osmotic coefficient as a function of molality at 0 °C was obtained from Pitzer et al. (1984). The value of molality was derived from the equation given by Fernández-Salguero et al. (1993).

3.6. Comparison with Zugarramurdi and Lupín model

As the Z&L model has been used previously for fish salting processes, this exponential approach to the equilibrium values of salt and water concentrations was included to compare with the numerical model:

$$\text{Salt uptake: } X_s = X_s^0 e^{-k_s t} + X_s^1 (1 - e^{-k_s t}) \quad (12)$$

$$\text{Water exudation: } X_w = X_w^0 e^{-k_w t} + X_w^1 (1 - e^{-k_w t}) \quad (13)$$

where X_s^0 and X_s^1 are initial and final salt contents (kg/kg), X_w^0 and X_w^1 are initial and final water contents (kg/kg). k_s and k_w are two theoretical coefficients (h⁻¹) calculated based on the experimental values obtained from 3 h salting time, while the equilibrium values used were obtained from 21 h salting time.

3.7. Model solution and validation

The mathematical model with coupled PDE of heat and mass transfer was solved in COMSOL Multiphysics v5.5 using the finite element method (FEM). A 2D rectangular geometry representing the salmon portion thickness with dimensions 7 cm \times 2 cm was created and meshed. The salt bulk concentration was estimated to be 15 % using trial and error to minimize the RMSE value between predicted and obtained value. For the determination of salt transport parameters, the data sets of 0 h and 21 h from the RSW fish were used, and the rest for validation. For the determination of water transport parameters, the data sets from 0 h to 18 h from RSW fish were used, and the rest for validation.

4. Results and discussion

4.1. Initial composition, drip loss and water holding capacity

The pH and initial chemical composition of the sampled fish on day 0 after slaughter were 6.2 ± 0.0 , 22.0 ± 0.6 % (protein), 9.2 ± 0.9 % (fat), 66.1 ± 3.1 % (water) and 0.1 ± 0.1 % (salt). The temperature during RSW and ice storage was kept stable until day 4 at around -0.9 °C and 0 °C, respectively (data not shown). On day 4, the RSW fish gained 1.6 ± 0.5 % while iced fish remained stable at 0.1 ± 0.2 % in weight ($p < 0.001$), but there were no differences in salt content ($p = 0.173$; RSW: 0.2 ± 0.1 %, ice: 0.2 ± 0.0 %). The results were consistent with Chan et al. (2020a), where salt uptake was similar for both treatments while RSW fish gained 1 % weight after 4 days.

Weight gain during RSW storage is a common phenomenon, as observed in several studies on various eviscerated salmon species (Bronstein et al., 1985; Erikson et al., 2011). When slaughtered fish is kept in seawater, the flesh equilibrates with the surrounding solution and increases in weight (MacLeod et al., 1960). As Erikson et al. (2011) explained, the weight gain observed in the present study probably came

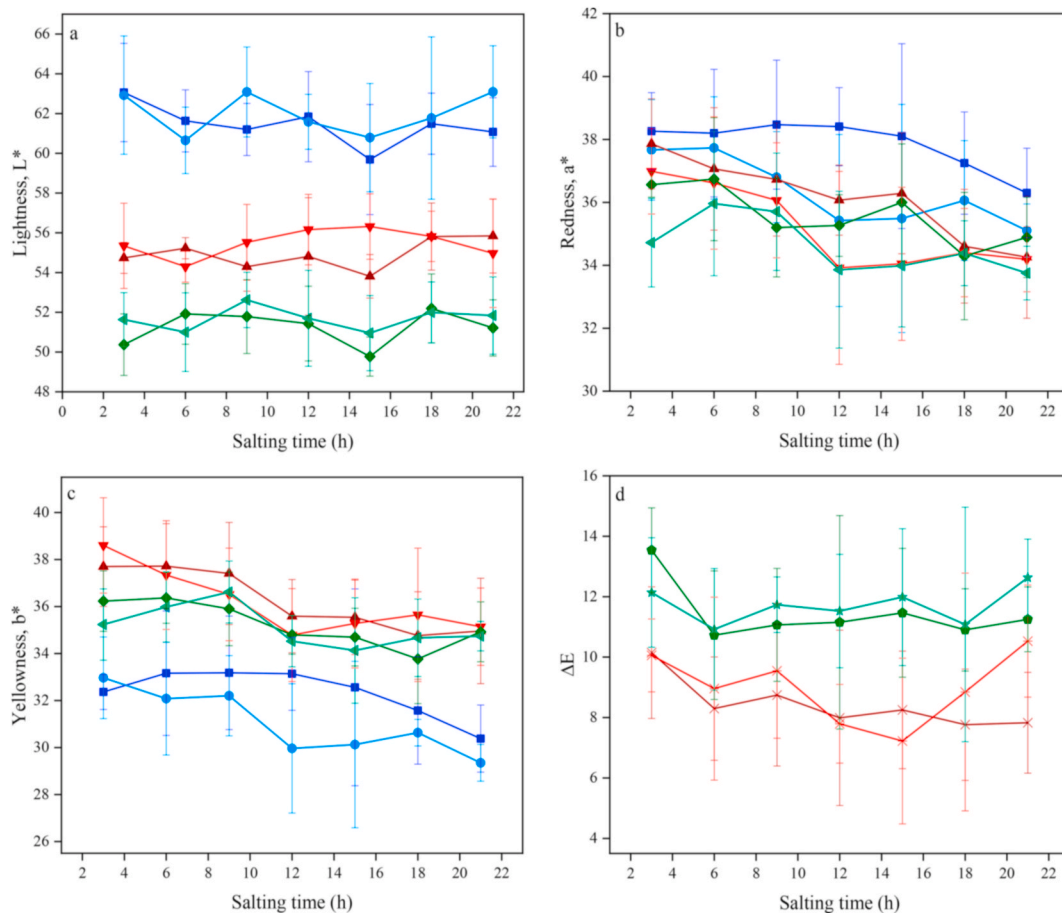


Fig. 4. Colour parameters of RSW raw fish (■), ice raw fish (●), RSW fish after smoking (▲), ice fish after smoking (▼), RSW fish after storage (◆), ice fish after storage (◀) (a) lightness, L^* (GLM; salting hour: $p = 0.947$, treatment: $p = 0.029$, processing step: $p < 0.001$). (b) Redness, a^* (GLM; salting hour: $p < 0.001$, treatment: $p < 0.001$, processing step: $p < 0.001$). (c) yellowness, b^* (GLM; salting hour: $p < 0.001$, treatment: $p = 0.052$, processing step: $p < 0.001$). (d) ΔE of RSW fish after smoking (×), ice fish after smoking (※), RSW fish after storage (●), ice fish after storage (★) (GLM; salting hour: $p = 0.219$; treatment: $p = 0.204$; processing step: $p < 0.001$). All results are presented in mean \pm standard deviation. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

mainly from the abdominal cavity since the salmon were bled and gutted. However, the % weight gain may vary according to species size and be unapparent, especially during the initial storage days when the differences are minor. Furthermore, Tomlinson et al. (1965) reported that the stress response before slaughter might influence the weight changes. As the initial pH observed in this study was lower than a typical pH of 7.5 for rested fish (Erikson and Misimi, 2008; Roth et al., 2012), this suggested that the fish used were possibly in a stressed condition during slaughter. Tomlinson et al. (1965) further found that the uptake of sodium ions significantly penetrated trout muscle only after adenosine triphosphate of the muscle was used. There was also a difference between the stress status, where unexercised fish initially lost weight while stressed fish gained weight after immersion. Hence, several factors could explain the rate of weight changes during RSW storage.

Heavy salting causes a significant weight reduction due to the consequential osmotic pressure of the salt on the moisture of muscle cells (Lauritzen et al., 2004). The drip loss for both treatments increased linearly through increasing salting time at every processing step after salting ($p < 0.001$), after smoking ($p < 0.001$) and after 2 weeks of storage ($p < 0.001$, Fig. 3a). This result was expected since salting is a diffusion process, and water is dragged out of the muscle until equilibrium is attained. The highest drip loss was observed after storage, where drip loss ranged from 6.1 % to 9.7 % for RSW fish and 6.2 %–9.2 % for iced fish salted for 3 h and 21 h, respectively. When both treatments were compared, no effect in drip loss during the different processing

steps was found. In line with previous studies (Chan et al., 2020a, 2020b), there were no differences in drip loss on smoked salmon from RSW treated or iced fish.

The most significant loss occurred after the smoking process, with the highest loss for fillets salted for 21 h (RSW: 9.2 ± 0.7 %, ice: 8.8 ± 0.4 %). The process yield decreased with increasing salting time from 94 % to 91 % in both groups, which was close to the previously reported values of 86%–92 % (Birkeland et al., 2004; Lerfall and Rotabakk, 2016; Sigurgisladottir et al., 2000). This decrease in yield can be explained by the increasing salt content, which decreases the hydrophilic surface and enhances protein-protein interaction, causing water loss (Bjørnevik et al., 2018). As cold-smoking involves both drying and smoking operations, the drying process also causes heat or pressure to expel water from the product's interior and mechanical energy to remove water from the surface (Sebastian et al., 2005). In addition, the smoking process allows the product to absorb volatile components, which provides antioxidant and antimicrobial effects and the required taste (Sebastian et al., 2005). In this study, vacuum storage of the smoked salmon for a further 2 weeks only gave a 1 % loss in drip. Like the findings of Løje (2007), little liquid was lost during cold vacuum storage of smoked salmon for almost 3 weeks. The liquid loss during vacuum storage can be related to the fatty acid profile, where a high liquid loss is linked to high amounts of monounsaturated and n-6 fatty acids (Lerfall et al., 2016).

The initial WHC after slaughter was 93.9 ± 0.9 %. On day 4, the WHC for RSW and iced fish were 93.7 ± 2.1 % and 94.1 ± 2.5 %, respectively.

Table 2

Firmness (80 % compression force) of smoked fillets after storage through different salting time. ^aGeneral Linear Model (GLM) analysis of variance was done with treatment as a factor and salting time and fillet height as covariates. p_s , p_T and p_H are the significant levels for the effects of salting time, treatment, and fillet height, respectively. All results are presented in mean \pm standard deviation.

Firmness(N)		Salting time (h)						
		3	6	9	12	15	18	21
RSW		17.3 \pm 2.3	19.3 \pm 2.4	16.2 \pm 3.2	19.8 \pm 3.9	19.3 \pm 1.3	21.1 \pm 5.7	20.6 \pm 3.6
ice		17.8 \pm 3.4	18.1 \pm 2.6	17.9 \pm 4.1	20.5 \pm 3.3	20.6 \pm 5.4	19.9 \pm 2.6	18.0 \pm 3.5
GLM ^a	P_s				0.021*			
	P_T				0.986			
	P_H				<0.001*			

The WHC observed after vacuum storage for different salting times appears not to give a general trend. For iced fish, WHC for 3 h salted fillets were at 96.2 ± 1.7 % as compared to 94.9 ± 3.2 for RSW fish (Fig. 3b). This was reversed after 15 h (iced: 93.9 ± 1.5 %, RSW: 95.3 ± 1.1 %) and 18 h (iced: 93.3 ± 3.1 %, RSW: 94.7 ± 1.1 %) of salting. As observed in other studies (Chan et al., 2020b; Løje, 2007), chilled storage of smoked salmon decreases WHC due to changes in water distribution. A higher salt content would also lead to a higher WHC (Løje, 2007). However, these observations were not seen in the present study when comparing the WHC of smoked and raw fillets. This might be explained by the variations in lipid content which can also influence other factors like salt content.

4.2. Effects on colour and texture

The colour of cold-smoked salmon plays an important role in the purchasing decisions of consumers. In this study, statistical analysis showed that the processing step (raw, smoked, stored) affected the colour parameters (Fig. 4a–c; L^* : $p < 0.001$; a^* : $p < 0.001$; b^* : $p < 0.001$), and treatment affected L^* ($p = 0.029$) and a^* ($p < 0.001$). The dry salting and cold-smoking process significantly lowered lightness ($p < 0.001$) and redness ($p < 0.001$), and increased yellowness ($p < 0.001$) respective to the unprocessed fillets. This is a general trend for smoked salmon fillets, as confirmed in previous studies (Birkeland et al., 2004; Cardinal et al., 2001; Chan et al., 2020a, 2020b; Løje, 2007), and is due to the physical and chemical reactions that occur between the product and smoking compounds during smoking (Pittia and Antonello, 2016). Dry salting can affect colour and texture due to protein denaturation and precipitation in the muscle. Carotenoids, the pigments that give the red colouration in salmon, can be lost during processing due to their decomposition or extraction during the dry salting procedure (Lerfall et al., 2016).

Further vacuum storage for 2 weeks showed a greater reduction in colour ($L^*a^*b^*$) than the freshly smoked counterparts. This is in agreement with Chan et al. (2020a) and could be explained by the liquid leakages accumulated during vacuum storage that can negatively affect the product appearance (Birkeland et al., 2004). As colour measurements are sensitive to fillet surface changes, possibly influenced by water content or surface structure, light scattering properties could be affected and make the fillet appear darker (Bjørnevik et al., 2018). There was no significant difference in salting duration and treatment on L^* when individual processing steps were compared. As salting duration increases, there was a decreasing trend on a^* (smoked: $p < 0.001$; stored: $p = 0.002$) and b^* (smoked: $p < 0.001$, stored: $p = 0.001$), and smoked fillets initially stored in RSW appeared more reddish than those in ice (smoked: $p = 0.023$; stored: $p = 0.015$).

Total colour change (ΔE) helps determine the colour differences during storage (Fig. 4d). In this study, both smoking and storage altered the surface properties, and the ΔE was more significant after storage than after smoking ($p < 0.001$). Earlier studies reported a stepwise increase in ΔE from dry salting to cold-smoking (Birkeland and Bjerkeng, 2005). The smoking process influences colour through carbonyl-amino reactions of Maillard browning and protein and lipid oxidation (Hall, 2011). Besides, the observed colour differences in $L^*a^*b^*$ and ΔE may be

explained by how the smoke components reacted with the chemical compounds like fatty acids in the muscle (Lerfall et al., 2016). Lerfall et al. (2016) and Lerfall and Rotabakk (2016) reported that the colour of refrigerated vacuum stored smoked salmon was restored and more similar to the raw fillets. This was not observed in this study, possibly due to the denaturation of the surface or other mechanisms that affect surface properties as a function of time.

Before salting on day 4, the firmness of unprocessed fillets was measured to be 13.1 ± 1.8 N (RSW) and 13.6 ± 1.8 N (ice). After processing and storage, firmness generally increased through salting time ($p = 0.021$), while there was no effect on treatment (Table 2). Textural properties for processed fillets are usually higher than the unprocessed counterparts, causing the muscle to be denser and more elastic (Chan et al., 2020b; Løje, 2007). As texture firmness is negatively related to the water content of smoked salmon (Birkeland et al., 2004), this likely explains the observed increase in firmness as water content decreases through salting. The slight dip observed at 9 h and 21 h might be explained by various factors such as variations in lipid and collagen content (Løje, 2007). As also verified by Chan et al. (2020a), fillet height significantly influences the firmness of the final product ($p < 0.001$).

4.3. Prediction of temperature

Fig. 5 presents the predicted temperature at the centre position of the salmon portion as a function of time in the cold room during salting. The sample temperature profile decreased rapidly to the surrounding temperature during the first 2 h and remained relatively stable afterwards. The model showed good agreement between the measured and predicted values (RMSE = 0.28 °C).

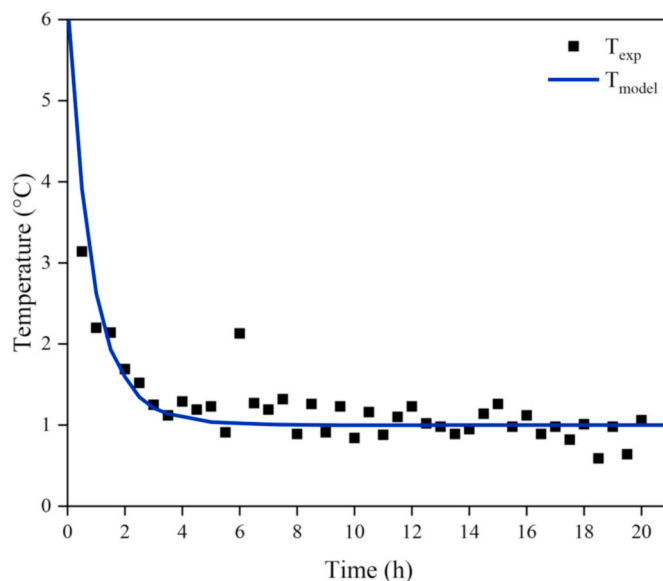


Fig. 5. Measured and predicted temperature profile at the middle position of the salmon portion during dry salting in a cold room (RMSE = 0.28 °C).

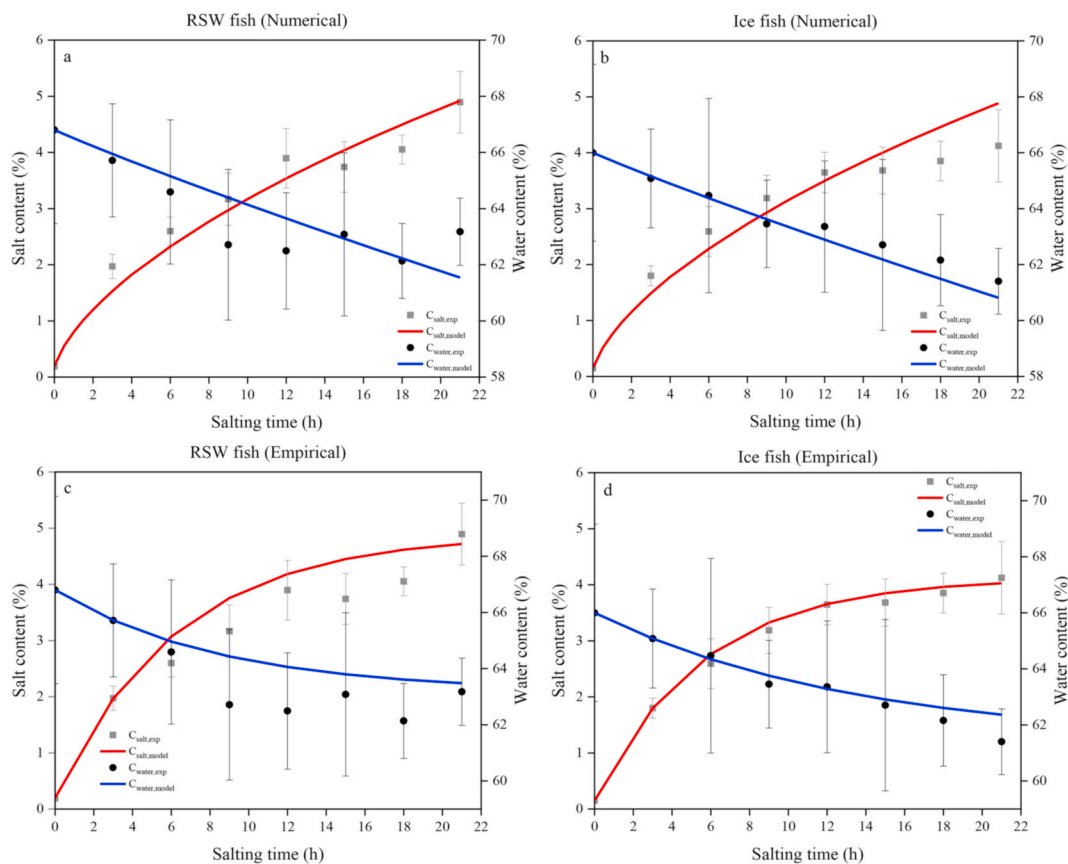


Fig. 6. Measured and predicted salt and water content for smoked salmon at different salting times from numerical modelling of (a) RSW (salt: RMSE = 0.30 %, water: RMSE = 0.95 %) and (b) ice fish (salt: RMSE = 0.41 %, water: RMSE = 0.41 %). Empirical modelling of (c) RSW (salt: RMSE = 0.43 %, water: RMSE = 1.02 %) and (d) ice fish (salt: RMSE = 0.11 %, water: RMSE = 0.40 %). Statistical analysis for measured salt content (GLM; salting time: $p < 0.001$, treatment: $p = 0.207$). Statistical analysis for measured water content (GLM; salting time: $p < 0.001$, treatment: $p = 0.511$). All results are presented in mean \pm standard deviation.

4.4. Prediction of salt and water transport

Statistical analysis revealed that there was an effect of salting time on salt (NaCl, $p < 0.001$) and water content ($p < 0.001$), but no differences was observed between treatments. Similar to Chan et al. (2020b), minimal differences were detected after salmon initially stored in RSW and ice were dry salted and cold-smoked. In this study, the water content obtained before salting was 66.8 ± 3.3 % and 66.0 ± 3.2 % for the RSW and ice fish, respectively. An inverse relationship was observed between water loss and salt uptake at increasing salting time for both treatments. The measured salt content at 21 h reached up to 4.9 ± 0.6 % (RSW) and 4.1 ± 0.7 % (ice), while water content to 63.2 ± 1.2 % (RSW) and 61.4 ± 1.2 % (ice).

Salt uptake and water loss simultaneously affect each other because of concentration and osmotic pressure difference, so water diffuses out while salt solubilizes in the water phase and diffuses into the muscle until equilibrium is attained, and the net rate of mass transfer is zero (Barat et al., 2003; Bellagha et al., 2007). In this experiment, the samples were analyzed after almost 2 weeks of vacuum storage. This is to ensure equilibrium as salt diffuses into the product at different speeds. During the dry salting process, a saturated layer of salt is first formed on the product's surface before salt migrates into the product (Pittia and Antonello, 2016). The evident increase in salt content during the first few hours of salting, as seen in this study, was likely due to the large concentration gradient between dry salt and muscle tissue. Thereafter, layer formation with high salt content close to the muscle surface acts as a barrier against further salt uptake (Akköse and Aktaş, 2016). This phenomenon was also observed in several studies (Akköse and Aktaş, 2016; Bellagha et al., 2007; Wang et al., 2000). The bulk salt

concentration was estimated to be 15 % (0.15 kg/kg) in the model. Since salt is used in excess during dry salting, this value can be influenced by the rate of salt diffusion and several factors such as surface-fillet thickness ratio and lipid content (Lerfall et al., 2016).

The calculated Z&L's specific constants k_s were 0.158 and 0.179 h^{-1} , and k_w were 0.118 and 0.074 h^{-1} for the RSW and iced fish. These values were similar to previous studies. For example, Bellagha et al. (2007) reported that k_s and k_w for dry salting of sardines were 0.139 and 0.191 h^{-1} . Corzo et al. (2015) also found that k_s and k_w salting of catfish sheets to be 1.125 and 1.489 d^{-1} .

The RMSE values obtained from the numerical model for RSW fish were 0.30 % (salt) and 0.95 % (water), while those of iced fish were 0.41 % (salt) and 0.41 % (water). In addition, the RMSE values from the Z&L model for RSW fish were 0.43 % (salt) and 1.02 % (water), while those of iced fish were 0.11 % (salt) and 0.40 % (water). Hence, the numerical and empirical model gave good agreements between the measured and predicted salt and water content for RSW and ice fish (Fig. 6).

Empirical models are developed by fitting the data with empirical correlations, relying on actual experiments. This can be beneficial and serve as a quick solution to observe the salting behaviour. However, such models cannot predict beyond the experimental range for a specific experiment, and biological variations like seasonal changes and raw material composition are omitted. In contrast, numerical models are based on the first principles and physical laws, which provides a better understanding of mechanisms. Food is a complex system with different physical properties such as shape, form, specific heat, thermal conductivity, density, and viscosity, changing with temperature. Therefore, numerical models can better manipulate process variables and adapt to

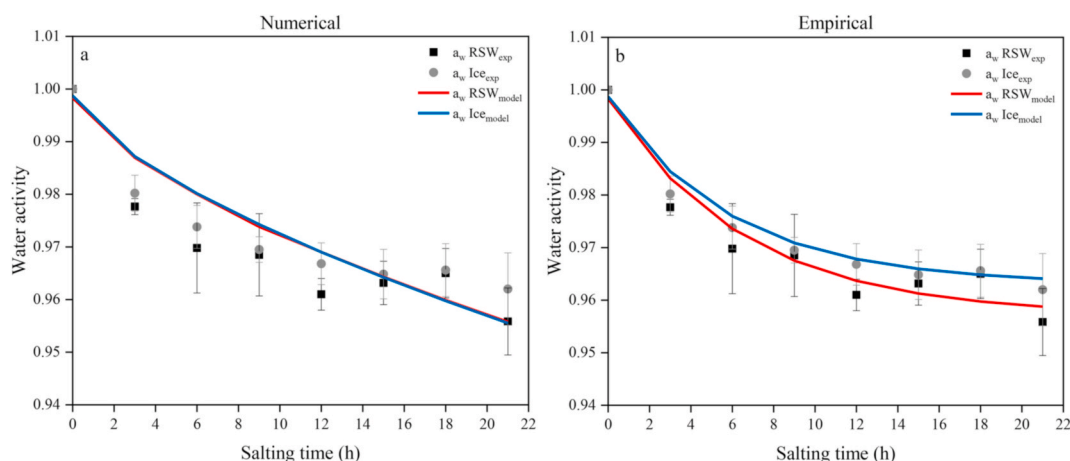


Fig. 7. Measured and predicted water activity for smoked salmon at different salting times from (a) numerical modelling of RSW (RMSE = 0.0063) and ice fish (RMSE = 0.0049); and (b) empirical modelling of RSW (RMSE = 0.0035) and ice fish (RMSE = 0.0021). Statistical analysis for measured water activity (GLM; salting time: $p < 0.001$, treatment: $p = 0.011$). All results are presented in mean \pm standard deviation.

changes in the conditions, providing a more flexible solution to include different input parameters and data extrapolation. It could be especially useful to observe spatial distribution and variations in temperature and water and salt concentration over time during salting to better understand the mechanism and control the salt uptake.

4.5. Prediction of water activity

In this study, the measured a_w values decreased during the whole salting process ($p < 0.001$), and the RSW fish generally had a lower a_w than the ice fish ($p = 0.011$). As water activity measures the amount of free water in the product, this could be the factor that explains the difference in a_w between the two treatments. The salt gain and water loss at increasing salting time explain the decrease in a_w , in agreement with other studies (Bellagha et al., 2007; Corzo et al., 2015).

The predictions of a_w using the predictions from salt and water content were validated against the experimental values. In general, the numerical (Fig. 7a) and empirical models (Fig. 7b) can accurately predict the a_w changes during dry salting for the two treatments. The RMSE values for RSW and ice fish predictions were 0.0063 and 0.0049 using the numerical model. Similarly, the empirical model gave RMSE values of 0.0035 and 0.0021 for the RSW and ice fish. Therefore, with the knowledge of water and salt content, the estimation of a_w is possible without needing to conduct laboratory analysis.

5. Conclusion

This study examined the quality parameters of dry salted and cold-smoked salmon that were initially immersed in RSW or ice, subjected to different salting times. According to the results, drip loss and colour were affected by the processing steps during salting, smoking and vacuum storage. Drip loss and salt (NaCl) content increased, while redness, yellowness and water content decreased with increasing salting duration. WHC was not affected by the salting time. In general, the smoked salmon from the RSW fish had redder and lower water activity values than the iced fish.

The heat transfer coefficient of 15 W/(m² K) gave a better fit using the reverse estimation method. The mathematical model of heat and mass transfer of salt and water during dry salting of salmon gave a reasonable agreement between the measured and simulated temperature, salt and water content. The predicted values of salt and water content were also able to simulate the water activity accurately. Comparison between numerical and empirical model showed good agreements from the low RMSE values obtained. Shrinkage occurs during dry salting due to moisture loss, which tightens the solid structure of the

product. Therefore, additional details may be included in the future to make the mathematical model more robust, such as measuring local concentrations, dimensional changes and fat distribution. Nevertheless, as more emphasis is given on the amount of salt added and salting time of the product, the model in this study can be a valuable tool for process optimization in the industry and understanding of the kinetics during dry salting of fish.

Author contributions

S.S.C. – conceptualization, methodology, investigation, writing original draft; A.H.F. – methodology, investigation, writing (review and editing); F.J. – supervision, writing (review and editing); B.R. – resources, supervision, writing (review and editing); A.N.J. – supervision, writing (review and editing); J.L. – conceptualization, investigation, supervision, writing (review and editing).

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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