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# Modeling mango ripening during shelf life based on pulp color nondestructively measured by time-resolved reflectance spectroscopy

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# ABSTRACT

Non-destructive techniques could help mango growers to pick fruit at the proper maturity degree and to monitor fruit quality during storage and marketing to satisfy the consumer expectations. The absorption coefficient nondestructively measured at 540 nm (*μ*a540) by time-resolved reflectance spectroscopy (TRS) was shown to be a maturity index for mango fruit and was highly correlated with pulp color parameters and carotenoids content. The aim of this work was to model *μ*a540 using the biological shift factor theory to verify if *μ*a540 is able to assess the maturity degree of individual mango fruit and then to use *μ*a540 to model pulp color in order to relate *μ*a540 to an important index of mango ripening.

Mango fruit (*Mangifera indica* L. cv Tommy Atkins) at commercial maturity for ship and air transport maturities, were measured by TRS, ranked according to decreasing *μ*a540 (decreasing maturity), randomized into six batches per transport maturity and analyzed for *μ*a540 and pulp color parameters after 0, 1, 2, 5, 6 and 7 days of shelf life at 20 °C.

The *μ*a540 as a function of biological shift factor increased during ripening following a logistic/exponential model (R<sub>adj</sub>=99%) with a faster rate in less mature fruit than in more mature ones. The changes in pulp color during mango ripening depended on fruit maturity (i.e., *μ*a540) and on time of shelf life at 20 ◦C. By converting the  $\mu$ <sub>a</sub>540 into the biological shift factor it was possible to model the increasing trend of  $a^*$ ,  $b^*$ ,  $C^*$ , and  $I_Y$  and the decreasing trend of *L*\* and *h*◦ during the shelf life period explaining 91.2–99.8% of the variation and to differentiate mango fruit according to their biological age. Similarly to *μ*a540, color changes occurred earlier in more mature fruit and later in less mature ones with the same pattern in time.

There is a synchronization between changes of *μ*a540 and changes of *a*\*, *b*\*, *C*\* and yellowness during ripening in mango fruit which allows to use *μa*540 to sort fruit according to their maturity degree and then to optimize fruit management along the supply chain.

### **1. Introduction**

Mango (*Mangifera indica* L.) is a tropical/subtropical climacteric fruit that is typically harvested at hard green stage, providing time for transport to distant markets and marketing.

Firmness, pulp color, total soluble solids content, dry matter, titratable acidity and aroma volatiles are the main attributes related to ripening in mango fruit. Flesh color is an important indicator of maturity

and ripeness as all mango cultivars develop yellow and orange pigments due to carotenoids accumulation in the flesh during maturity and ripening, while changes in skin color are not always correlated with maturity, ripeness or internal eating quality ([Padda et al., 2011](#page-16-0)). During ripening, mesocarp tissue color changes exhibiting a progressive decrease in L\* and h $^{\circ}$  values while flesh a\*, b\* and C\* values increased ([Padda et al., 2011\)](#page-16-0). The best color attributes to follow changes during mango ripening are a\* and  $b*$  of the flesh with  $b*$  showing the highest

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**Fig. 1.** Scheme of the setup used for TRS measurements on mango fruit. GRIN: graded-index; PMT: photomultiplier tube; SYNC: synchronization signal; CFD: constant fraction discriminator signal; TCSPC: time-correlated single-photon counting board.

correlation with maturity score and dry matter ([Subedi et al., 2007](#page-16-0); [Kienzle et al., 2011](#page-15-0); [Padda et al., 2011\)](#page-16-0).Unfortunately, mesocarp color can be measured only in a destructive way; therefore, the development of non-destructive techniques could help growers to pick fruit at the proper maturity degree for each market destination (local, distant) especially in the view of minimizing quantitative and qualitative losses during the supply chain. Among optical methods, continuous wave near infrared (NIR) or visible (Vis) -NIR spectroscopy has been used for predicting pulp color ([Subedi et al., 2007;](#page-16-0) Cortés et al., 2016). This approach, however, predicts maturity parameters indirectly and the robustness of the prediction model is linked to the extent and variation of the population used for the calibration. An improvement with respect to the standard Vis-NIR approach could be the direct non-destructive measure of the relevant parameters in the fruit mesocarp, which can be achieved by the time-resolved reflectance spectroscopy (TRS) or space- resolved reflectance spectroscopy (SRS) techniques.

TRS and SRS, differently from continuous wave methods, can simultaneously measure the effects of light absorption, due to chemical compounds, and of light scattering, mainly due to microscopic changes in refractive index caused by membranes, organelles, vacuoles, starch granules and air [\(Lu et al., 2020\)](#page-15-0). In TRS, a short pulse of monochromatic light is injected into the fruit: whenever a photon strikes a scattering center, it changes its trajectory and keeps on propagating in the tissue, until it is eventually reemitted across the boundary, or it is captured by an absorbing center. Usually, the laser light is injected into and collected from the fruit by using two optical fibres placed in contact with the surface at a distance of 1–2 cm. The laser light probes a banana-shaped volume of tissue to a depth of 1–2 cm, in contrast to continuous-wave Vis-NIR spectrophotometers, which have a useful penetration depth of a few millimeters, depending on the wavelength ([Lammertyn et al., 2000\)](#page-15-0). By measuring the photon distribution of time-of-flight both the absorption  $(\mu_a)$  and reduced scattering  $(\mu_s')$  coefficients in the Vis–NIR spectrum region are estimated ([Cubeddu et al.,](#page-15-0)  [2001;](#page-15-0) [Torricelli et al., 2008](#page-16-0)). TRS relies on the ability to measure the optical properties of the fruit mesocarp with no or limited influence from the skin as the spectra acquired on whole fruit for both absorption and scattering were shown to be very similar to the spectra of the same fruit when peeled ([Cubeddu et al., 2001](#page-15-0); Saeys et al., 2008; [Torricelli et al.,](#page-16-0)  [2008; Spinelli et al., 2012](#page-16-0); [Rizzolo and Vanoli, 2016](#page-16-0)).

TRS has been used to study the internal fruit attributes related to maturity ([Rizzolo and Vanoli, 2016](#page-16-0)). The *μ*a measured in the chlorophyll-*a* absorption region near 670 nm is considered an index of the biological age of the fruit and have been used to define harvest maturity and to model fruit ripening on an individual fruit basis. Fruit maturity on the tree is not homogeneous but depends on flowering time, position of fruit on the tree relative to leaves and other fruit, microclimate and

hormonal and nutritional effects. This variability can be expressed as biological shift factor (BSF, [Tijskens et al., 2005](#page-16-0)). In nectarines [\(Tijskens](#page-16-0)  [et al., 2007b](#page-16-0)) and in apples ([Rizzolo et al., 2021](#page-16-0)), *μ*a670 declines during fruit ripening and was successfully used to predict the softening rate in nectarines and to select fruit for different market destinations ([Eccher](#page-15-0)  [Zerbini et al., 2009](#page-15-0); [Rizzolo et al., 2021\)](#page-16-0) when it was converted into the biological shift factor In mangoes, [Pereira et al. \(2010\)](#page-16-0) showed that  $\mu$ <sub>a</sub>630 could be used to predict softening rate of 'Tommy Atkins' mangoes, even if the model explained only 70% of the variation in firmness decay, whereas [Eccher Zerbini et al. \(2015\)](#page-15-0) by using both *μ*a670 and *μ*a540 (carotenoid tail) in 'Haden' mangoes was able to assess fruit maturity and to predict the ripening of individual fruit with 80% of variability explained for the ethylene and firmness logistic models. [Rizzolo et al. \(2016\)](#page-16-0) and [Vanoli et al. \(2018\)](#page-16-0) reported that *μ*a540 can be used as a non-destructive maturity index for mangoes, as it was able to classify intact mangoes of different cultivars according to pulp color and to the contents of total and individual carotenoids.

However, until now no model was developed combining *μ*a540 and the BSF for assessing ripening in mangoes, as achieved for  $\mu_a$ 670 in nectarines and in apples. Hence, the aim of this work was to model *μ*a540 using the biological shift factor theory to verify if *μ*a540 is able to assess the maturity degree of individual mango fruit and then to use *μ*a540 to model pulp color in order to relate *μ*a540 to an important index of mango ripening.

# **2. Materials and methods**

# *2.1. TRS instrumentation*

A scheme of the portable TRS setup developed at Politecnico di Milano and used for measurements is reported in Fig. 1. It is based on a supercontinuum fiber laser (SC450–6 W, Fianium, UK) as a source, that provides white-light picosecond pulses, adjustable in power by means of a variable neutral-density attenuator. The spectral selection of the injected light is performed through a filter wheel loaded with 14 bandpass interference filters with characteristic wavelengths in the range 540 - 940 nm. Light is, then, delivered to the sample exploiting a 100 μmdiameter multimode graded-index fiber.

As for the detection side, diffuse re-emitted light by the sample is collected by a 1 mm-diameter step-index fiber 1.5 cm apart from the injection point, and delivered to a photomultiplier (HPM-100–50, Becker&Hickl, Germany) for detection. Finally, by means of a timecorrelated single-photon counting board (SPC-130, Becker&Hickl, Germany), distributions of time-of-flight of re-emitted photons at different wavelengths are recorded. Further details of this instrumentation can be found in [Spinelli et al. \(2012\).](#page-16-0)



**Fig. 2.** Exemplum of the best fit of the model (red line), describing photon diffusion in a turbid medium, on measured DTOF (blue dots) after convolution with instrument IRF (pink line). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

In order to assess the optical properties of the sample, recorded DTOFs (distribution of photon time-of-flight) have been analyzed with a model describing the photon diffusion in turbid media based on the solution of the diffusion equation in a homogeneous semi-infinite medium [\(Martelli et al., 2009](#page-15-0)). In particular, after a convolution with the instrument response function (IRF), the model was best fitted to the recorded DTOF considering the absorption and reduced scattering coefficients as free parameters. This analysis procedure is depicted in Fig. 2.

### *2.2. Fruit*

Mango fruit (*Mangifera indica* L., cv 'Tommy Atkins') were harvested in commercial orchards in Pernambuco (Brazil) at commercial maturities for ship- and air- transport, which had been defined according to the importer's protocol for each type of shipping (less mature for ship – longer transport time; more mature for air – shorter transport time), and carried by plane to Milano (Italy) immediately after harvest. According to the diagram shown in Fig. 3, on arrival at CREA-IT lab (about 5–7 days from harvest), 180 fruits per type of transport were selected, measured by TRS at 540 nm on two opposite sides in the fruit equatorial region and ranked within each type of transport by decreasing *μ*a540 averaged over the two sides, that is from more (high  $\mu_a$ ) to less mature fruit (low  $\mu_a$ ). The ranked fruit were grouped by 6, for a total of 30 groups, corresponding to 30 levels of *μ*a540. Each fruit from each group was randomly assigned to a different sample. In this way, 6 samples per type of shipping were obtained, each one containing 30 fruits from the whole range of  $\mu$ <sub>a</sub>540 and used for a single time of shelf life at 20 °C: 0, 1, 2, 5, 6, and 7 days (labelled d0, d1, d2, d5, d6 and d7, respectively).

Then at each shelf-life time, *μ*a540 and pulp color was measured in correspondence of the positions of TRS measurements with a spectrophotometer (CM-2600d, Minolta Co., Japan), using the primary illuminant D65 and 2◦ observer in the *L*\*, *a*\*, *b*\* color space. From the *L*\*, *a*\* and  $b^*$  values, hue ( $h^{\circ}$ ), chroma ( $C^*$ ) and Yellowness Index ( $I_Y$ ) were computed according to the following equations:

$$
h^* = \arctan(b^* / a^*) \times 360 / (2 \times 3.14)
$$
 (1)

$$
C* = (a^{*2} + b^{*2})^{-2}
$$
 (2)

$$
I_Y = [(81.2746X - -1.0574Z) / Y] \times 100
$$
\n(3)

after converting *L\*a\*b\** parameters into the *XYZ* color space [\(Jha et al.,](#page-15-0)  [2006\)](#page-15-0). Data were averaged per fruit.



### **FOR EACH TRANSPORT MATURITY**

**Fig. 3.** Diagram of the procedure used for the formation of the six equivalent samples of 30 fruit.

<span id="page-3-0"></span>The sample d7 was measured by TRS at 540 nm also at times 0, 1, 2, 5 and 6 days of shelf-life.

### *2.3. The μa540 model*

Basing on the fact that several biological phenomena such as color development, fruit softening and chlorophyll degradation, develop or decay following a sigmoid curve, in order to model the *μ*a540 behavior with the fruit biological age, it was assumed that the increase of  $\mu_a$ 540, mainly due to carotenoids development, follows a sigmoid curve increasing with the biological age of the fruit, which can be described using the logistic model [\(Penchaiya et al., 2020](#page-16-0)) of Eq. (4):

$$
y = (y_{ref} - y_{min}) \times e^{-km \times (t + \Delta t)} + y_{min}
$$
 (4)

where: *y* is *μ*a540; *t* is the time of shelf life; *km* is the rate constant and Δ*t*  the biological shift factor. The *ymin* is the minimum *μ*a540 value ever possible at minus infinite time,  $y_{ref}$  is the chosen reference value of  $\mu_a$ 540 at which the BSF equals zero, i.e., the midpoint of the overall range of change in  $\mu$ <sub>a</sub>540.

The BSF Δ*t* is a stochastic variable containing all the information related to maturity for each individual fruit in the whole batch, and is the shift of individual maturity in relation to the intermediate maturity  $(\Delta t = 0)$  corresponding to  $\mu_a$ 540 equal to half of the maximum and can be computed according to Eq. (5) ([Tijskens et al., 2010\)](#page-16-0):

$$
\Delta t = -\left(\frac{\log\left(\frac{y_0 - y_{min}}{y_{ref} - y_{min}}\right)}{km}\right) \tag{5}
$$

where  $y_0$  is the  $\mu_a$ 540 value of the fruit used for fruit ranking at the start of the experiment.

### *2.4. Color parameters models*

It has been shown that color parameters with ripening follows a sigmoidal curve ([Penchaiya et al., 2015, 2020](#page-16-0)). For  $b^*$ ,  $C^*$  and  $I_Y$  it was assumed that these *y* color parameters could follow a sigmoid curve increasing with the biological age of the fruit reaching a maximum value (*y*max):

$$
y = \frac{y_{max}}{1 + e^{\left(-k_m \times \left(t + \Delta t_y^*\right)\right)}}\tag{6}
$$

while for  $a^*$  the model (7), in which  $y_{\text{min}}$ , the minimum value ever possible, is included and the range of values replaces *y*max, best fitted *a*\* data ranging from negative to positive values:

$$
y = y_{min} + \frac{(y_{max} - y_{min})}{1 + e^{(-k_m \times (t + \Delta t_y^*))}}
$$
(7)

As for *L*\* and *h*◦, it was assumed that these *y* color parameters decrease with ripening to minimum values without reaching the zero value:

$$
y = y_{min} + \frac{y_{max} - y_{min}}{1 + e^{k_m \times (t + \Delta t_y^*)}}
$$
(8)

In these models, *ymax* and *ymin* are the maximum and minimum values ever possible for the *y* color parameter (at minus and plus infinite time),  $k<sub>m</sub>$  is the rate constant for the color parameter *y*, *t* is the normalized time which is zero when the value of the *y* color parameter is halfway between its maximum and minimum values within each set, and Δ*t*<sup>\*</sup><sub>y</sub> is the BSF in time relative to time  $t = 0$  for the *y* color parameter, which accounts for the different age of individual fruit in regard to *a\*, b\*, C\** and yellowness index increases and *L*\* and *h*◦ decreases.

By these models moreover it was assumed that all fruit, grown in the

same orchard and edaphic conditions, had the same behavior as regard to  $L^*$ ,  $a^*$ ,  $b^*$ ,  $C^*$ ,  $h^{\circ}$  and  $I_Y$  while ripening, and each fruit, with its BSF, represents a different step of the same process. As described for  $\mu_a$ 540,  $\Delta t_\mathrm{y}^*$  is a stochastic variable containing all the information related to maturity for each individual fruit in the whole batch, expressed in standardized dimensionless time [Tjiskens et al., 2007a](#page-16-0)). Since the increase of  $a^*$ ,  $b^*$ ,  $C^*$  and  $I_Y$  was positively related to the exponential increase of carotenoids during ripening ([Ornelas-Paz et al., 2008](#page-15-0)), and that  $\mu$ <sub>a</sub>540 was positively related to total carotenoids in mango (Vanoli et al., [2016; Vanoli et al., 2018](#page-16-0)) and negatively related to *L*\* and *h*◦ [\(Spinelli](#page-16-0)  [et al., 2012](#page-16-0); [Vanoli et al., 2016\)](#page-16-0), it can be assumed that the BSF of each color parameter ( $\Delta t_y^*$  in the Eqs. (6), ((7) and (8)) could be expressed as a function of the measured  $\mu_a$ 540 ( $\mu_a$ 540<sub>0</sub>) relatively to its range

$$
\Delta t_y^* = \alpha_{540} \times \delta + \beta \text{ with}
$$
  
\n
$$
\delta = \log \frac{\mu a 540_{\text{max}} - \mu a 540_0}{\mu a 540_0 - \mu a 540_{\text{min}}}
$$
\n(9)

where  $\alpha_{540}$  and  $\beta$  are parameters to be estimated. The subscript 0 indicates the  $\mu$ <sub>a</sub>540 value measured in each fruit by TRS on the same day as color parameters measurement, while the indices max and min indicate the maximum (0.84 cm<sup>-1</sup>) and minimum (0.05 cm<sup>-1</sup>) values ever possible found in experiments on mango fruit, where extreme values were found [Eccher Zerbini et al., 2015](#page-15-0)). To express Δ*t*<sup>\*</sup><sub>y</sub> in time dimension, in Eqs.  $(6)$ ,  $((7)$  and  $(8)$ , this variable was divided by the range of the *y* color parameter and the rate constant *km*.

## *2.5. Statistics*

Modeling of *μ*a540 in shelf life was carried out using Statgraphics ver. 5.1 Plus (Manugistic Inc., Rockville, MD, USA). From model (4) combined with Eq.  $(5)$ ,  $k_m$  was estimated using the non-linear regression procedure, in which *y*<sub>min</sub> was fixed to 0.08 cm<sup>-1</sup> on the basis of *μ*<sub>a</sub>540 data obtained in this or other experiments with mango fruit, *y*ref was equal for each data set to the mean of  $\mu$ <sub>a</sub>540 values of all the fruit of the set at the start of the experiment (sorting),  $y_0$  is the  $\mu_a$ 540 value of the fruit used for fruit ranking at the start of the experiment, and *t* was expressed in days of shelf life. The initial *km* was set at 0.0001 and the estimation was carried out by the Marquardt method and stopped when the convergence of residual sum of squares was achieved. In the nonlinear regression procedure  $y_0$  and *t* were considered independent variables and the fruit  $\mu_a$ 540 value was the dependent variable. The rate constant  $k_m$  was estimated considering the  $\mu_a$ 540 value of the 30 fruits of sample d7 during the whole period of shelf-life. Then, both for sample d7 in shelf life and for d0, d1, d2, d5, d6 and d7 samples, the Δt of each individual fruit was computed according to  $Eq. (4)$  using the estimated rate constant for the specific data set (ship-transport maturity, airtransport maturity, ship+air set). Then, using the regression analysis procedure, the models of  $\mu_a$ 540 vs.  $t + \Delta t$ , with *t* being the normalized time, with 0 value when  $\mu_a$ 540 is in the midpoint between the minimum and the maximum value, were studied.

Color data were analyzed by non-linear regression (Statgraphics ver. 5.1 Plus, Manugistic Inc., Rockville, MD, USA) to be represented as functions of fruit maturity at the time of measure, as assessed by  $\mu_a$ 540 optical property. From model (6) for  $b^*$ ,  $C^*$  and  $I_Y$ , model (7) for  $a^*$  and model (8) for *L*<sup>\*</sup> and *h*<sup>○</sup>, all combined with Eq. (9),  $k_m$ , α540 and *β* were estimated, after having computed for each fruit the  $\delta$  value. Then,  $\Delta t_y^*$  of each individual fruit for every color parameter was computed according to Eq. (9), and expressed in the time dimension by dividing it by the range and  $k_m$ . Then, using the regression analysis procedure, the models of each color parameter vs.  $\Delta t_y^*$  expressed in time dimension, were studied.

<span id="page-4-0"></span>

**Fig. 4.** Absorption coefficient *μ*a540 of fruit of d0, d1, d2, d5, d6 and d7 samples of ship (left) and air (right) transport maturity degree at harvest according to ranking order.



**Fig. 5.** Absorption coefficient at 540 nm (*μ*a540) of sample d7 of ship and air transport maturities in function of time of shelf life at 20 ◦C: fitted exponential model, confidence limits at 95% level (inner lines), prediction limits (outer lines) and experimental data (left) and estimates and standard error (SE) of intercept and slope, Rsquared adjusted for d.f. ( $R_{adj}^2$ ), standard error of the estimate (SEE) and mean absolute error (MAE) of the fitted model (right).

Estimates of the rate constant of *μ*a540 increase in time (*km*), asymptotic standard error and 95% confidence interval of *km* estimate, R2 adj, standard error of the estimate (SEE) and mean absolute error (MAE) of the [Eq. \(4\)](#page-3-0) model for ship, air and ship+air transport maturities data sets.



<span id="page-5-0"></span>



**Table 3** 

BSF for  $\mu$ <sub>a</sub>540 (Δt<sub>μa540</sub>) for samples d0, d1, d2, d5, d6 and d7 of ship and air data sets: minimum, maximum, mean, standard deviation values of distributions and results of tests for normality ( $n = 30$ ). For frequency histograms and density traces see [Figs. A.1](#page-11-0) (ship set) and [A.2](#page-12-0) (air set).

Transport maturity		d <sub>0</sub>	d1	d2	d <sub>5</sub>	d6	d7
Ship minimum		$-4.79$	$-2.78$	$-1.76$	0.08	0.23	$-0.58$
maximum		3.95	6.17	3.92	4.71	5.15	5.62
mean		$-0.09$	0.51	1.16	2.27	2.68	2.85
Std dev		1.834	1.810	1.530	1.015	1.168	1.263
$\chi^2$ goodness-of-fit	statistic	10.0	9.0	7.0	22.4	12.0	13.9
	P-value	0.62	0.70	0.86	0.03	0.44	0.30
Shapiro Wilks W statistic	statistic	0.9852	0.9510	0.9767	0.9478	0.9851	0.9661
	P-value	0.94	0.21	0.76	0.20	0.94	0.50
Air minimum		$-3.49$	$-5.39$	$-2.10$	0.48	0.53	$-0.02$
maximum		7.31	4.86	5.95	5.91	9.15	6.60
mean		0.34	0.56	1.41	3.19	3.37	3.38
Std dev		2.331	2.318	1.974	1.320	1.699	1.560
$\chi^2$ goodness-of-fit	statistic	6.0	13.0	7.0	18.1	19.0	5.21
	P-value	0.92	0.37	0.86	0.11	0.09	0.95
Shapiro Wilks W statistic	statistic	0.9598	0.9820	0.9715	0.9775	0.9228	0.9721
	P-value	0.35	0.89	0.62	0.79	0.04	0.66

### **3. Results**

# *3.1. The μa540*

The *μ*a540 at fruit sorting did not differ between the two transport maturity stages, being (mean  $\pm$  standard error) 0.157  $\pm$  0.003 cm<sup>-1</sup> in fruit for ship transport and 0.166  $\pm$  0.004 cm<sup>-1</sup> for air transport; values were in the 0.103–0.403 cm<sup>-1</sup> (ship maturity) and 0.106–0.511 cm<sup>-1</sup> (air maturity) ranges, with a different distribution between the two transport maturities. In fact, considering the ship set, 51% of fruit had  $\mu_{\rm a}$ 540 values lower than 0.15 cm<sup>-1</sup>, 35% were in the 0.15–0.19 cm<sup>-1</sup> interval, 9.6% in the 0.19–0.23 cm<sup>-1</sup> and 6.2% had  $\mu$ <sub>a</sub>540 higher than 0.23 cm<sup>-1</sup>. Differently, in the air set 44% of fruit showed *μ*<sub>a</sub>540 lower than 0.15  $\text{cm}^{-1}$ , 13% were in the 0.19–0.23  $\text{cm}^{-1}$  range and 9% had  $\mu_{\text{a}}$ 540 values higher than 0.23 cm<sup>-1</sup>.

### *3.2. The μa540 model*

With shelf life  $\mu$ <sub>a</sub>540 increased in all fruit showing the largest vari-ation at the end of shelf life [\(Figs. 4 and 5\)](#page-4-0). Fitting the  $\mu_a$ 540 values of the 30 mangoes of the samples d7 in function of shelf life at 20 ◦C with the traditional approach of the exponential regression model without considering the BSF approach only about 36% and 28% of the variability, for ship and air transport maturity respectively, could be

explained with quite large standard errors of the estimate (*>*0.2).

To study the  $\mu_a$ 540 increase using the logistic model of [Eq. \(4\)](#page-3-0) and considering the individual information obtainable by the BSF approach, the rate constant  $k<sub>m</sub>$  was estimated using the nonlinear regression pro-cedure ([Table 1](#page-4-0)). The  $k_m$  was about 0.108 cm<sup>-1</sup> day<sup>-1</sup> for the ship set, 0.094 cm<sup>-1</sup> day<sup>-1</sup> for air set and 0.1 cm<sup>-1</sup> day<sup>-1</sup> for ship+air set with  $R_{\text{adj}}^2$  in the 78–83% range.

Considering the evolution of BSF of sample d7 fruit of the ship set with shelf life (Table 2), it ranged from −3.94 to 3.45 days at the beginning of shelf life to -0.58 to 5.62 days after 7 days, with the average value increasing from − 0.01 days at d0 to 2.85 days at d7 and the standard deviation decreasing from 1.77 at d0 to 1.26 at d7. The distributions of BSF were normal with 99% confidence at  $\chi^2$  goodnessof-fit statistic and Shapiro-Wills W statistic (Table 2).

As for the BSF of d7 sample fruit belonging to the air set (Table 2), the minimum values of BSF ranged from about − 5 days at the beginning of shelf life to − 0.02 days at d7, and the maximum values ranged from 4.83 days at d0 to 6.60 days at d7; the average value increased from 0.06 days at d0 to 3.38 days at d7, with the standard deviation decreasing from 2.39 at d0 to 1.56 at d7. The distributions of BSF were normal with 99% confidence at  $\chi^2$  goodness-of-fit statistic and Shapiro-Wills W statistic (Table 2).

Considering the BSF of the samples analyzed for pulp color parameters (Table 3), the minimum values were in the − 4.8 - − 0.6 days range

<span id="page-6-0"></span>Modeling of  $μ_3$ 540 vs *t*+Δ*t*<sub>ua540</sub> of sample d7 data during shelf life: estimates and standard error of intercept and slope of (A) logistic and (B) exponential regression models,  $R_{\text{adj}}^2$ , SEE and MAE for ship (d.f.=176), air (d.f.=173) and ship+air (d.f.=350) data sets.



#### **Table 5**

Modeling of  $μ_3$ 540 vs *t*+Δ*t*<sub>μa540</sub> using data of samples d0, d1, d2, d5, d6 and d7: estimates and standard error of intercept and slope of (A) logistic and (B) exponential regression models,  $R_{\text{adj}}^2$ , SEE and MAE for ship (d.f.=176), air (d.f.=176) and ship+air (d.f.=353) data sets.

	intercept		slope		$\mathrm{R}^2_{\mathrm{adj}}$	<b>SEE</b>	<b>MAE</b>	P-value model
Data set	estimate	SE	estimate	SE				
A - logistic								
Ship	$-1.6206***$	0.0008	$0.0669***$	0.0001	99.9	0.0091	0.0063	< 0.0001
Air	$-1.6035***$	0.0038	$0.0644***$	0.0005	98.8	0.0411	0.0140	< 0.0001
$Ship+air$	$-1.6960***$	0.0011	$0.0556***$	0.0001	99.8	0.0146	0.0099	< 0.0001
B - exponential								
ship	$-1.8016***$	0.0018	$0.0524***$	0.0003	99.4	0.0199	0.0134	< 0.0001
Air	$-1.7853***$	0.0013	$0.0490***$	0.0002	99.8	0.0136	0.0102	< 0.0001
$Ship+air$	$-1.8557***$	0.0025	$0.0424***$	0.0003	98.5	0.0340	0.0205	< 0.0001

for the ship set and in the  $-3.5 - -0.02$  days range for the air set, while the maximum values ranged from 3.9 days (samples d0 and d2) to 5.6 days (sample d7). The mean values increased from − 0.1 (sample d0) to 2.8 days (sample d7) for the ship set and from 0.3 (sample d0) to 3.4 days (sample d7) for the air set, with standard deviation decreasing with shelf life from 1.83 to 1.26 in the ship set and from 2.33 to 1.56 in the air set. All the distributions of BSF for the ship set were normal with 99% confidence at Shapiro-Wills W statistic, and, except for sample d5, at  $\chi^2$ goodness-of-fit statistic. In contrast, all the distributions of BSF for the air set were normal with 99% confidence at  $\chi^2$  goodness-of-fit statistic and, except for sample d6, at Shapiro-Wills W statistic [\(Table 3](#page-5-0)). The frequency histograms and density traces of BSF of ship set belonging to d7 sample and to samples analyzed for color parameters shown in [Fig. A.1](#page-11-0) compared with those of air set reported in [Fig. A.2](#page-12-0) confirm the similarity of the BSF evolution in shelf life of the six samples from both data sets, as well as underline the differences between the two transport maturities, with air set having a wider range of BSF than the ship set.

As for the regression analysis of  $\mu_a$ 540 vs  $t + \Delta t_{\mu a}$ <sub>540</sub>, for each data set both the logistic and the exponential models were considered (Tables 4 and 5; [Fig. 6\)](#page-7-0) From [Fig. 6](#page-7-0) it can be seen that considering the individual differences in maturity by BSF, the  $\mu$ <sub>a</sub>540 values of [Fig. 5](#page-4-0) are shifted following the same curve, highlighting that each individual fruit represents a step in the same process.

The  $R_{\text{adj}}^2$  of all the models were higher than 99.0% for both logistic and exponential models.

#### *3.3. Mesocarp color*

Mesocarp color parameters during shelf life did not differ between the two transport maturity stages [\(Table 6](#page-8-0)). With shelf life, *L*\* and *h*◦ decreased, while  $a^*$ ,  $b^*$ ,  $C^*$  and  $I_Y$  increased, with the main changes occurring from d2 and d5 for both the transport maturities. Except for *L*\* of ship set, which further decreased till the end of shelf life, *L*\* of air set and  $a^*$ ,  $b^*$ ,  $C^*$ ,  $h^\circ$  and  $I_Y$  of both sets did not further change with time of shelf life.

#### *3.4. Color parameters models*

For each transport maturity stage, the *L*\*,  $a^*$ ,  $b^*$ ,  $C^*$ ,  $h^{\circ}$  and  $I_Y$  values of the 30 mangoes of each sample were plotted in function of shelf life at 20 ◦C, expressed as calendar time (days) and fitted with the regression model having the best performance (higher  $R_{\text{adj}}^2$ , lower SEE and MAE), without considering the BSF approach [\(Fig. B.1](#page-13-0)a and b). Raw data showed a large variation for all the color parameters, with a percentage variance explained ranging from 33 to 50%.

Aiming at modeling pulp color parameters in function of the BSF approach, the coefficient of the independent variable  $\mu$ <sub>a</sub>540 ( $\alpha$ <sub>540</sub>) and the *β* parameter of the [Eq. \(9\)](#page-3-0) were estimated, along with the rate constant  $k_m$ , by the nonlinear regression procedure, adopting model (6) for *b*\*, C\* and *I*Y, model (7) for *a*\*and model (8) for *L*\* and *h*◦ combined with [Eq. \(9\),](#page-3-0) in which the  $\delta$  value decreases with the increase of the shelf life time from  $0.773\pm0.024$  at d0 to  $0.488\pm0.021$  at d7, and increases with the increase of ranking order in the samples (i.e., decreasing maturity) from 0.126±0.072 for Rank 1 to 0.905±0.049 for Rank 30. The results of the estimates of the models' parameters for each data set are reported in [Table 7](#page-8-0); [Fig. 7](#page-9-0) shows for each color parameter the estimated response surfaces, i.e., the predicted values in function of time of shelf life expressed in normalized days and  $\delta$ , and the plots predicted vs. observed for the ship+air set.

To avoid over parameterization, the minimum and maximum values have been fixed at the minimum and maximum values ever measured in this and previous our experiments on mango fruit, and these values were used for all the data sets. Generally, the  $R_{\text{adj}}^2$  of the models was >90%, with higher values for *I*Y (96.5–99.1%) and *h*◦ (96.3–99.8%) than the R2 adj for *L*\* (94–97%), *a*\* (94.4–99.3%), *b\** (93–99%) and *C*\* (91.8–97.4%).

The rate constant  $k_m$  depended on the color parameter and/or the transport maturity [\(Table 7](#page-8-0)). Whatever the data set, the  $k_m$  for  $L^*$  and  $h^{\circ}$ showed higher values than the other color parameters, being, respectively, in the 0.9–1.1 and 0.4–0.8 ranges;  $b^*$  and  $I_Y$  showed the lowest  $k_m$ values, being in the 0.14–0.18 and 0.12–0.15 ranges, respectively. Comparing the two transport maturities, the  $k_m$  of  $L^*$  and  $b^*$  were significantly higher and those of  $I_Y$  and  $h$ <sup>°</sup> were significantly lower in the

<span id="page-7-0"></span>

**Fig. 6.** Measured (symbols) and predicted (lines) *μ*a540 in function of BSF (*t*+ Δt*μ*a540) for sample d7 data (panel a) for samples d0, d1, d2, d5, d6 and d7 data (panel b) of ship (left), air (center) and ship+air (right) sets by logistic (top) and exponential (bottom) models, Models' details are reported in [Tables 4](#page-6-0) and [5.](#page-6-0)

air set.

The BSFs, expressed in normalized time, differed among color parameters and between transport maturities ([Table 8](#page-9-0)). All the BSFs distributions were normal according to  $\chi^2$  goodness-of-fit and Shapiro Wilks W statistic tests (data not shown). The BSFs of ship transport maturity fruit related to  $L^*$ ,  $a^*$ ,  $h^\circ$  and  $I_Y$  [\(Table 8\)](#page-9-0) were lower than those observed for air transport maturity fruits, while the BSFs related to *b*\* and *C*\* were higher. Negative values from − 0.96 to − 0.20 normalized days (norm d) were observed for  $\Delta t_{\text{L}^*}$  for almost all the fruit of both ship and air transport maturities and for  $\Delta t_{a^*}$  for some fruit of the ship set. Low but positive values from 0.02 to 0.69 norm d were found for  $\Delta t_{\rm H}$ <sup>°</sup> for all the fruit of both the transport maturities. The ranges of Δ*t* values of the other color parameters were: 4.33–7.86 norm d for Δ*t*IY, 5.16–9.55 norm d for  $\Delta t_{b^*}$  and 9.17–15.74 norm d for  $\Delta t_{C^*}$  [\(Table 8\)](#page-9-0).

Considering the shelf-life time [\(Tables 10](#page-13-0)–[15\)](#page-14-0), in all the sets the minimum BSFs values were observed at the beginning of shelf life, then they increased significantly from d0 to d2 (only ship and ship+air sets) and from d2 to d5, then they did not change till the end of shelf life.

[Table 9](#page-10-0) and [Fig. 8](#page-10-0) (ship+air set) report the results of regression analysis of color parameters vs the specific biological shift factor (Δ*ty*) for ship, air, and ship+air sets.

The type and the performance of regression model of  $\Delta t$ <sub>y</sub> vs the *y* color parameter depends on color parameters and on two transport maturities, with  $R_{\text{adj}}^2$  ranging from 73 to 86%.

# **4. Discussion**

### *4.1. The μa540*

The  $\mu_a$ 540 was used as the non-destructive maturity index to sort fruit at arrival from the most mature (highest value in the batch) to the least mature (least value in the batch). The *μ*a540 was also used to obtain for each fruit its biological age expressed as biological shift factors specific for  $\mu_a$ 540 and for *L*\*,  $a^*$ ,  $b^*$ ,  $C^*$ ,  $h^{\circ}$  and  $I_Y$  color parameter, as each fruit at the time of examination had undergone a more or less advanced stage of the ripening process related to the specific property.

The  $\mu_a$ 540 showed similar values to those reported for the same cultivar [\(Vanoli et al., 2013\)](#page-16-0) and for other mango cultivars ([Vanoli et al.,](#page-16-0)  [2018\)](#page-16-0), and increased with the shelf-life time for both the transport maturities, in agreement with [Vanoli et al. \(2013\)](#page-16-0)'s findings even if the correlation was very low with day of shelf life. This low correlation could be due to the fact that fruit had been picked at a ripening degree

<span id="page-8-0"></span>Mesocarp color parameters  $L^*$ ,  $a^*$ ,  $b^*$ , chroma ( $C^*$ ), hue ( $h^{\circ}$ ) and Yellowness Index ( $I_Y$ ) of d0, d1, d2, d5, d6 and d7 samples of ship and air transport maturity degrees at harvest.



Means followed by different letters are statistically different (Tukey's test, *P<*0.001).

# **Table 7**

Parameters of the non-linear regression models for *L*\*,  $a^*$ ,  $b^*$ , chroma (*C*\*), hue (*h*<sup>◦</sup>) and yellowness index (*I*<sub>Y</sub>) pulp color parameters in function of  $\mu_a$ 540 for ship (d. f.=176), air (d.f.=176) and ship+air (d.f.=353) mango fruit sets,  $R_{\text{adj}}^2$ , SEE and MAE of the models.



<sup>a</sup> fixed values;  $\alpha_{540}$  is the coefficient of the independent variable  $\mu$ <sub>a</sub>540.

<span id="page-9-0"></span>

**Fig. 7.** Estimated response surface (top) and plot of predicted vs observed (bottom) by non-linear regression models for lightness (*L*\*), *a\** and *b\** pulp color parameters (panel a) and for chroma (*C*\*), hue and yellowness index (*I*<sub>Y</sub>) pulp color parameters (panel b) of ship+air set. See [Table 7](#page-8-0) for models' parameters.



more advanced than the preclimacteric mature-green stage, in order to have better quality at consumption, as shown by  $I_Y$  values greater than 100 at the beginning of shelf life This means that a certain amount of carotenoids was present in the pulp, which can further improve with shelf life day only in little quantities, as most likely both experiments started when carotenoid content fell in the last part of the logistic curve of carotenoid synthesis. Notwithstanding that on average the *μ*a540 did not differ between the two transport maturities, differences still exist

<span id="page-10-0"></span>Results of regression analysis between BSF and pulp color parameters for ship, air and ship+air sets: estimates and standard error of intercept and slope of regression models, R<sup>2</sup> adj, SEE and MAE. MT=model type (*L*=linear; *M*=multiplicative; RX=reciprocal-X; RY=reciprocal-Y; *Sc*=*S*-curve; SqrY=square root-Y).



<sup>a</sup> ln (H)=intercept – slope\*ln(BSF<sub>H°</sub>).



**Fig. 8.** Lightness ( $L^*$ ),  $a^*$ ,  $b^*$  (top), chroma ( $C^*$ ), hue and Yellowness Index ( $I_Y$ ) (bottom) pulp color parameters of ship+air set in function of the specific BSF  $\Delta t$ , fitted with the model having higher performance (for details see Table 9) (blue line), confidence limits at 95% level (inner red lines), prediction limits (outer red lines) and experimental data (symbols). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

between the two mango lots: the range of *μ*a540 values at sorting of ship transport maturity was narrower (0.3  $cm^{-1}$ ) than that of air transport maturity (0.405  $\rm cm^{-1}$ ), with 51% of fruit being less mature ( $\mu_{\rm a}$ 540  $<$ 0.15 cm<sup>−</sup> <sup>1</sup> ) and 15.8% of fruit being more mature (*μ*a540*>*0.19 cm<sup>−</sup> <sup>1</sup> ) vs 44% of fruit with  $\mu_a$ 540< 0.15 cm<sup>-1</sup> and 21% with  $\mu_a$ 540>0.19 cm<sup>-1</sup>.

# *4.2. Mesocarp color parameters*

The values of pulp color parameters and their behavior in shelf-life were like those reported for 'Tommy Atkins' mangoes from Brazil in

our previous works [\(Vanoli et al., 2013;](#page-16-0) [Vanoli et al., 2011](#page-16-0); [Rizzolo](#page-16-0)  [et al., 2011\)](#page-16-0) and in the literature ([De Morais et al., 2002](#page-15-0); [Sabato et al.,](#page-16-0)  [2009;](#page-16-0) [de Mello Vasconcelos et al., 2019\)](#page-15-0).

During ripening mesocarp color turned from greenish-yellow to orange, as  $a^*$ ,  $b^*$ ,  $C^*$  and yellowness index  $(I_Y)$  increased and  $h^{\circ}$  decreased with shelf life [\(De Morais et al., 2002;](#page-15-0) [Dang et al., 2008;](#page-15-0) [Ornelas-Paz](#page-15-0)  [et al., 2008](#page-15-0); Vásquez-Caicedo et al., 2006; [Vanoli et al., 2011](#page-16-0) and [2013](#page-16-0); [Rizzolo et al., 2011;](#page-16-0) [Spinelli et al., 2012;](#page-16-0) [Candelario-Rodríguez et al.,](#page-15-0)  [2014;](#page-15-0) [Gill et al., 2017\)](#page-15-0). Lightness (*L*\*) decreased with ripening due to changes in fruit texture rather than to pigment contents [\(De Morais](#page-15-0) 

<span id="page-11-0"></span>

**Fig. A.1.** Ship transport maturity set: Frequency histograms and density traces of BSF for *μ*a540 (Δt*μ*a540) of sample d7 data in shelf life (left) and of d0, d1, d2, d5, d6 and d7 samples (right).

<span id="page-12-0"></span>

**Fig. A.2.** Air transport maturity set: Frequency histograms and density traces of BSF for *μ*a540 (Δt*μ*a540) of sample d7 data in shelf life (left) and of d0, d1, d2, d5, d6 and d7 samples (right).

<span id="page-13-0"></span>

 $(b)$ 

Time of shelf life (days)

 $\overline{2}$ 

**Fig. B1.a.** Panel a: Lightness (*L*\*), **a**\* and *b\** pulp color parameters of ship (top) and air (bottom) transport maturities in function of time of shelf life at 20◦C, fitted with model having higher performance, confidence limits at 95% level (inner lines), prediction limits (outer lines) and experimental data (symbols), model type and R-squared adjusted for d.f. (R<sub>adj</sub>). MT=model type (L=linear; SqrX=square root-X). All models have P-value <0.0001. Panel b: Chroma (C\*), hue and Yellowness Index  $(I_Y)$  pulp color parameters of ship (top) and air (bottom) transport maturities in function of time of shelf life at 20°C, fitted with model having higher performance, confidence limits at 95% level (inner lines), prediction limits (outer lines) and experimental data (symbols), model type and R-squared adjusted for d.f. (R<sup>2</sup> adj). MT=model type (L=linear; RecY=reciprocal-Y; SqrX=square root-X; Sqr-Y=square root-Y). All models have P-value *<*0.0001.

Table 10	
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BSF (norm d) for *L*<sup>\*</sup> (Δ*t*<sub>L<sup>\*</sup></sub>) of ship+air data set in function of shelf-life time at 20 ◦C: minimum, maximum, mean, and standard deviation values of distributions.

6

Time of shelf life (days)



[et al., 2002;](#page-15-0) [Vanoli et al., 2011, 2013](#page-16-0); [Gill et al., 2017](#page-15-0); [Penchaia et al.,](#page-16-0)  [2020\)](#page-16-0). [Rizzolo et al. \(2011\)](#page-16-0) found that the rate of color changes with shelf life in 'Tommy Atkins' mangoes was different in fruit classified as

# **Table 11**



 $\overline{2}$ 

Time of shelf life (days)

 $\mathbf{R}$ 



less, medium and more mature according to  $\mu_a$ 630: *h*<sup>∘</sup> decreased and *I*<sub>Y</sub> increased with the highest rate for less mature class and the least rate for more mature class, and *L*\* decreased faster in medium and more mature

<span id="page-14-0"></span>BSF (norm d) for  $b^*$  ( $\Delta t_{b*}$ ) of ship+air data set in function of shelf-life time at 20 ◦C: minimum, maximum, mean, and standard deviation values of distributions.

	d0	d1	d2	d5	d6	d7
N.	60	60	60	57	60	57
minimum	6.27	6.29	6.75	7.14	7.18	7.00
maximum	9.21	9.14	8.67	8.66	10.31	8.92
mean	7.19	7.29	7.45	7.80	7.90	7.92
Std dev	0.479	0.482	0.418	0.329	0.469	0.392

### **Table 13**

BSF (norm d) for  $C^*$  ( $\Delta t_{C^*}$ ) of ship+air data set in function of shelf-life time at 20 ◦C: minimum, maximum, mean, and standard deviation values of distributions.

	d0	d1	d2	d5	d6	d7
N.	60	60	60	57	60	57
minimum	10.04	10.08	10.92	11.66	11.73	11.39
maximum	15.49	15.36	14.49	14.47	17.54	14.94
mean	11.75	11.92	12.22	12.87	13.06	13.09
Std dev	0.890	0.894	0.775	0.610	0.869	0.728

#### **Table 14**

BSF (norm d) for  $h^\circ$  ( $\Delta t_{\text{H}^\circ}$ ) of ship+air data set in function of shelf-life time at 20 ◦C: minimum, maximum, mean, and standard deviation values of distributions.

	d0	d1	d2	d5	d6	d7
N.	60	60	60	57	60	57
minimum	$-0.34$	$-0.33$	$-0.03$	0.22	0.25	0.13
maximum	1.56	1.52	1.21	1.21	2.28	1.37
mean	0.26	0.32	0.42	0.65	0.72	0.72
Std dev	0.311	0.313	0.271	0.213	0.304	0.254

# **Table 15**

BSF (norm d) for  $I_Y (\Delta t_{IV})$  of ship+air data set in function of shelf-life time at 20 ◦C: minimum, maximum, mean, and standard deviation values of distributions.

	d0	d1	d2	d5	d6	d7
N.	60	60	60	57	60	57
minimum	4.73	4.76	5.38	5.92	5.98	5.73
maximum	8.76	8.65	8.02	8.00	10.26	8.35
mean	5.99	6.12	6.34	6.82	6.96	6.98
Std dev	0.657	0.659	0.572	0.450	0.641	0.537

classes than in less mature ones.

### *4.3. Modeling*

It has been previously shown that the maturity indices based on the absorption coefficients non-destructively measured by TRS are correlated to mesocarp color parameters ([Rizzolo et al., 2011](#page-16-0); [Vanoli et al.,](#page-16-0)  [2011,](#page-16-0) [2013,](#page-16-0) [2018](#page-16-0); [Spinelli et al., 2012](#page-16-0)), which depends on the main pigment concentrations, i.e., chlorophylls and carotenoids.

[Eccher Zerbini et al. \(2015\)](#page-15-0) reported that the optical absorptions showed a clear sequence in mango fruit: the increase of  $\mu_a$ 540 (linked to carotenoids) occurred only after the complete decrease of *μ*a670 (linked to chlorophyll). Similarly, upon ripening, carotenoids are accumulated in the mesocarp with an exponential behavior, only when chlorophyll is very low or undetectable [\(Kienzle et al., 2011,](#page-15-0) [2012](#page-15-0)). Previously, for 'Palmer', 'Haden' and 'Tommy Atkins' cultivars, positive correlations were found among  $\mu_a$ 540 and  $a^*$ ,  $b^*$  and  $I_Y$ , and negative correlation between *μ*a540 and *h*◦ and *L*\* ([Spinelli et al., 2012](#page-16-0); [Vanoli et al., 2013](#page-16-0), [2016, 2018](#page-16-0)).

Basing on the close relationship between *μ*a540 and carotenoid content, the production model reported in [Eq. \(4\)](#page-3-0) seems to be appropriate to describe the  $\mu$ <sub>a</sub>540 increase with ripening.

The  $\mu_a$ 670 alone or in combination with  $\mu_a$ 540 and scattering parameters have been so far used to model softening and ethylene production of mangoes ([Pereira et al., 2010;](#page-16-0) [Eccher Zerbini et al., 2015\)](#page-15-0), by integrating the BSF derived from the  $\mu_a$  with firmness decay or ethylene production. In these models, the BSF for *μ*a and the BSF for firmness and ethylene should be linearly related according to the  $Eq. (9)$  reported in [Section 2.3](#page-3-0), with  $\alpha_{540}$  being the synchronization factor between the ratio of  $\mu_a$  and firmness or ethylene and  $\beta$  representing the harvest date expressed as a shift factor relative to the midpoint of the curves [\(Tijsk](#page-16-0)[ens et al., 2007a\)](#page-16-0).

In this work also for color parameters it was assumed that  $\mu$ <sub>a</sub>540 increase and  $a^*$ ,  $b^*$ ,  $C^*$ ,  $I_Y$  increase or  $L^*$  and  $h^{\circ}$  decrease are synchronized, and, hence,  $\Delta t_{\mu a}$ 540<sup>\*</sup> and  $\Delta t_{v}$ <sup>\*</sup> should be related according to Eq. [\(9\)](#page-3-0). In order to estimate  $\alpha_{540}$  and the  $\beta$  parameter of the [Eq. \(9\)](#page-3-0), the increasing behavior of  $b^*$ ,  $C^*$  and  $I_Y$  has been approximated to the sigmoid curve model reported in Eq.  $(6)$ , that of  $a^*$  to the model reported in [Eq. \(7\)](#page-3-0), whilst the decreasing trends of *L*\* and *h*◦ have been approximated by the model reported in [Eq. \(8\).](#page-3-0)

## *4.3.1. The μa540 modeling*

Two models for  $\mu$ <sub>a</sub>540 as a function of biological time have been studied: logistic and exponential. The logistic model has already been adopted to describe the  $\mu_a$ 670 and the  $\mu_a$ 630 decays in nectarines ([Tijskens et al., 2006,](#page-16-0) [2007b\)](#page-16-0) and in mangoes ([Pereira et al., 2010](#page-16-0)), respectively. The exponential model was also studied, as *μ*a540 was linearly correlated with total carotenoids which accumulate in the mesocarp with an exponential behavior during ripening (V´ [asquez-Caseido et al., 2006](#page-16-0); [Ornelas-Paz et al., 2008](#page-15-0)).

The explained parts are very high: 98.8–99.9% for the logistic models and around 98.5–99.8% for the exponential models. The biological shift factor (BSF) showed lower values for the ship set and higher values for the air set, indicating that the fruit of the ship set are less ripe than those of the air set with about a difference of 0.5–1 day. This pattern means that, notwithstanding the mean *μ*a540 value of the two fruit sets was not different at the start of the experiment, there was an influence of the shipping maturity on the behavior with shelf life.

Moreover, the rate constant was slightly lower in the air set respect to the ship set, and the standard deviations of the BSF were slightly higher in the air set. In apples, [Rizzolo et al. \(2021\)](#page-16-0) observed higher rate constant in less mature fruit than in more mature ones during maturation on the tree. Also, the standard deviation of the BSF was slightly higher in the air set than in the ship one, further confirming an influence of the shipping maturity on fruit ripeness. In fact, all the variation in the BSF is the result of the biochemical changes occurring during fruit growth and ripening at the tree, in the orchard conditions [\(Tijskens](#page-16-0)  [et al., 2007b](#page-16-0)).

#### *4.3.2. The modeling of color parameters*

With *μ*a540 and time as independent variables, the explained parts of the non-linear regression models are high for all the fruit sets, being the  $R<sub>adj</sub><sup>2</sup>$  in the 91.8–99.8% range for the ship set, from 97 to 99.6 for the air set and in the 94–97.1 interval for the ship+air set, indicating that almost all variation present in each data set was properly taken care of. The values of constant rates  $(k_m)$  for  $a^*$ ,  $b^*$ ,  $C^*$  and  $I_Y$  are not too different from the constant rates found for *μ*a540, whereas the estimates for  $h^\circ$  and  $L^*$  are about three-four times than the  $k_m$  values observed for the other color parameters and for  $\mu_a$ 540. The similar  $k_m$  values for  $a^*$ ,  $b^*$ ,  $C^*$  and  $I_Y$  could be due to their high correlation to carotenoid content, which in turn, is positively correlated to  $μ$ <sub>a</sub>540 [\(Vanoli et al., 2016](#page-16-0), [2018\)](#page-16-0). The higher  $k_m$  found for  $h^\circ$ , which was shown to be highly correlated in mangoes to  $\mu_a$ 630, linked to chlorophyll content (Rizzolo [et al., 2011\)](#page-16-0) and to *μ*a540 ([Vanoli et al., 2018\)](#page-16-0), could suggest that *h*◦ value depends on chlorophyll *b*reakdown and on carotenoid

<span id="page-15-0"></span>accumulation which are little overlapping and almost mutually exclusive, so that, depending on the fruit age, either one is prevalent (Eccher Zerbini et al., 2015).  $L^*$  showed the highest values of  $k_m$  as it is mainly influenced by changes in texture and not by pulp pigment dynamics. Changes in texture bound to mango softening have been previously modeled considering the BSF approach by using either *μ*a630 for 'Tommy Atkins' fruit during shelf life [\(Pereira et al., 2010](#page-16-0)) or *μ*a540, *μ*a670 and the equivalent size of the scattering centers (Mie's *B*  parameter) on 'Haden' fruit (Eccher Zerbini et al., 2015), obtaining in the first case a *k*m value for firmness decay of 0.004, far from those observed for *L*\* in this work. Hence, it can be envisaged that the texture changes influencing *L*\* regard other properties than softening measured by puncture test, such as the increase in juicy texture as suggested by [Penchaiya et al. \(2020\)](#page-16-0).

The estimated response surface plots obtained by the non-linear regression procedure well display the interplay of the time of shelf life, in this case expressed as normalized time for each color parameter, and the fruit maturity defined by  $\delta$  variable, bound to  $\mu$ <sub>a</sub>540 measured at the time of pulp color measurement, and to the BSF of each individual fruit. The decrease to minimum values of *L*\* and *h*◦, as well as the increase to maximum values of  $a^*$ ,  $b^*$ ,  $C^*$  and  $I_Y$  occurred earlier in more mature fruit (high  $\mu_a$ 540 value, low  $\delta$  value) and later in less mature ones (low  $\mu_a$ 540 value, high  $\delta$  value), with the same sigmoidal pattern in time.

For each color parameter, further information could be retrieved by the plot vs the specific BSF ( $\Delta t$ ). Considering lightness, the  $\Delta t_{L^*}$  values indicated that all fruit from both the transport maturities, having a negative BSF, were before the intermediate maturity in regard to *L*\* color parameter. In contrast, the  $\Delta t_{b^*}$ ,  $\Delta t_{C^*}$ ,  $\Delta t_{H^{\circ}}$  and  $\Delta t_{IY}$  values of fruit from both transport maturities and  $\Delta t_{a^*}$  values of air transport maturity fruit, having all a positive BSF, were beyond the intermediate maturity in regard to  $b^*$ ,  $C^*$ ,  $h^{\circ}$ ,  $I_Y$  and  $a^*$  color parameters. The negative  $\Delta t_{a^*}$ values of most of ship transport maturity fruit, on the other hand, indicated that for the ship set only few fruits were beyond the intermediate maturity in regard to *a*\*.

#### **5. Conclusion**

The non-destructive measurement of the absorption coefficient at 540 nm by means of TRS allowed to assess the biological age of each fruit in 'Tommy Atkins' mangoes, being *μ*a540 linked to carotenoid accumulation and to pulp color changes. The *μ*a540 as a function of the biological shift factor increased during ripening following a logistic/ exponential model with a faster rate in less mature fruit than in more mature ones.

The changes in pulp color during mango ripening depended on fruit maturity (i.e.  $\mu_3$ 540) and on time of shelf life at 20 °C. By converting the  $\mu$ <sub>a</sub>540 into the biological shift factor it was possible to model the increasing trend of  $a^*$ ,  $b^*$ ,  $C^*$ , and  $I_Y$  and the decreasing trend of  $L^*$  and *h*◦ during the shelf life period explaining 91.2–99.8% of the variation and to differentiate mango fruit according to their biological age. Similarly to  $\mu_a$ 540, color changes occurred earlier in more mature fruit and later in less mature ones with the same pattern in time.

There is a synchronization between changes of *μ*a540 and changes of a\*, b\*, C\* and yellowness during ripening in mango fruit which allows to use  $\mu_a$ 540 to sort fruit according to their maturity degree and then to optimize fruit management along the supply chain.

### **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.

# **Data Availability**

Data will be made available on request.

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# **Appendix A**

[Fig. A.1](#page-11-0). [Fig. A.2](#page-12-0). [Fig. B.1.](#page-13-0)

## **Appendix B**

[Table 10](#page-13-0), [Table 11,](#page-13-0) [Table 12](#page-14-0), [Table 13,](#page-14-0) [Table 14, Table 15](#page-14-0).

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