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1 **Modelling mastication and aroma release from white rice during**

2 **FOP**

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15 **Abstract**

16

17 A mathematical model describing the aroma release from white rice during food oral
18 processing (FOP) was developed based of a coupled selection-breakage and mass transport
19 models. An integrated selection and breakage model was able to predict the changes of bolus
20 surface area over time, assuming that the pasted portion of masticated rice particles was the
21 dominant mechanism when aroma was released to the liquid bolus during chewing. Model
22 predictions were validated against experimental data for all subjects when the input
23 parameters were directly obtained from the coupled chewing-aroma release model. Adjusting
24 the input parameters from one of the validated coupled model showed that the portion size,

25 initial concentration of the studied aroma compound, initial liquid volume and the rice pasted
26 fraction were the most sensitive product-related parameters. The oral cavity volume, pharynx
27 volume, nasal cavity volume and the breathing frequency were the most sensitive
28 physiological parameters. The physico-chemical parameter which had the most significant
29 effect was the mass transfer coefficient in the saliva phase.

30 Keywords;

31 *Particle Size Distribution; Paste; Bolus surface area; Mass transfer; Mastication; Ordinary*
32 *Differential Equations*

33

34 **1. Introduction**

35 Mathematical models have been developed in the literature to predict the aroma release of
36 various food matrix types during FOP. Harrison et al. (1998) provided the first model for
37 predicting the aroma release of solid and semisolid foods. However, the model lacked
38 experimental validation. Strong simplifying assumptions were made for the suggested model
39 such as constant breath airflow and the model application was limited to the type of solid and
40 semi-solid foods that retained their shape and did not further disintegrate into the surrounding
41 saliva with increasing chewing cycles (Harrison et al., 1998; Trelea et al., 2007). Since
42 Harrison et al. (1998), improvements have been made by incorporating a more realistic
43 description of physiological mechanism, such as for mastication of semi-solid products
44 (Wright & Hills, 2003; Wright et al., 2003; Trelea et al., 2007; Doyennette et al., 2014;
45 Harrison et al., 2014) and for liquid products (Rabe et al., 2004; Doyennette et al., 2011; Le
46 Révérend et al., 2013).

47 The most comprehensive model to predict aroma release during oral processing of
48 semi-solid foods to date was developed by Doyennette et al. (2014). The model was
49 constructed using one-dimensional differential equations for mass transfer and flavour release
50 and was validated against experimental *in vivo* data using cheese as the food system. A
51 sensitivity analysis performed on the mathematical model showed that the air-bolus contact
52 surface area could affect aroma release; however, the model did not include the coupling to
53 any food breakdown model to predict the air-bolus contact surface area. Due to fragmentation
54 caused by mastication, where food particles are reduced in size, the model assumes that the
55 contact area between the solid product and the liquid bolus during mastication evolves
56 linearly with time. The authors acknowledged that this was a simplified assumption, as the
57 exact rate of change of contact area is not known for products such as cheese and rice, where
58 when ingested, a fraction of the food product can form very fine particles that can dissolve in

59 the liquid phase of the bolus due to the combined action of mastication, saliva incorporation
60 and the warming of the product in the mouth, known as the pasted fraction (Doyennette et al.,
61 2014; How et al., 2021). The limitation of the model was acknowledged by the authors as the
62 lack of coupling to a dynamic food breakdown model to predict the particle size distribution
63 with respect to chew number stating that validating the model would require complex
64 experimental protocols which are challenging to execute (e.g. bolus spitting after a variable
65 number of bites).

66 The aim of the study was to develop a model that could accurately describe how the
67 concentration of aroma release changes over time during mastication of cooked white rice.
68 The work carried out here is limited to predicting the volatile components of released aroma
69 in the nasal cavity, rather than those related to taste, which is a subject of ongoing work. This
70 work is an extension to our previous study investigating the role of oral processing on *in vivo*
71 aroma release of rice by comparing experimental results with a conceptual model (How et al.,
72 2021). The main novelty of the model developed in this study compared to previous aroma
73 release models developed in the literature is the coupling of a food breakdown model based
74 of an integrated mechanistic selection and breakage functions (How et al., 2022) to predict
75 the air-bolus contact area in the mouth compartment. Selection is the probability per chew
76 that food particles are damaged or broken and has been shown to depend on particle size and
77 number through the one-way and two-way competition models (van der Glas et al., 1992,
78 2018). Breakage is the distribution function of the daughter particles that originate from each
79 selected particle which could be described using mechanistic and empirical functions (Austin,
80 1971; van der Bilt et al., 1987; van der Glas et al., 1987; Gray-Stuart, 2016). To ensure
81 conservation of volume after breakage of particles, a discretised population balance method is
82 commonly employed to track selection and breakage of individual particles (Gray-Stuart,
83 2016; How et al., 2022).

84 In addition to the food breakdown aspect, the model would have to consider the
85 mechanisms for mass transfer between the solid particles and liquid content of the bolus and
86 how the interfacial area between the bolus and air phases change during mastication. In this
87 study, the model was validated by comparing the model simulations to *in vivo* aroma release
88 data of cooked white rice flavoured with two food grade aroma compounds (2-nonanone and
89 ethyl propanoate), measured by Proton Transfer Reaction-Mass Spectrometry (PTR-MS) in
90 real time on five panellists.

91 **2. Conceptual model development**

92 Figure 1 shows the conceptual diagram for the interconnected compartments that are involved
93 in aroma release during the consumption of cooked white rice. Refer to Appendix A1 for the
94 nomenclature table. The compartments that were involved in flavour release during food
95 consumption were denoted as follows: the oral cavity (index O), the pharynx (index F), the
96 nasal cavity (index N) and the trachea (index T). The model used here is an adaptation from a
97 chemical engineering approach where the various parts of the upper respiratory tract are
98 viewed as interconnected reactors, containing an air phase (index A) and the saliva phase,
99 index (S). To take the retention effect of lubricated mucosa (index M) into account, the
100 lubricated mucosa layers were also included in each compartment (oral cavity, pharynx and
101 nasal cavity). The compartments were included as one of the aroma compounds used to spike
102 the cooked white rice, 2-nonanone, was known to interact with the lubricated mucosa
103 (Doyennette et al., 2014; Déléris et al., 2016). The airflow rates (Q_{Na} , Q_{Ta} , Q_{Oa}), were
104 considered to be positive if their direction is the one indicated by the arrows in Figure 1
105 (inhalation) and negative when in the opposite case (exhalation).

106 Aroma concentrations in all compartments (oral cavity, pharynx and nasal cavity) were
107 calculated using mass transfer equations and mass balances. The mass balances include the

108 flavour release at the saliva – product (rice) interface, air-saliva interface and the air and
109 lubricated mucosa layer interface. In general, when two phases are in contact (e.g. air and
110 saliva), volatile transfer occurs across the interfacial layers. At each side of the interface, the
111 driving force is the concentration difference between the bulk phase and the interface. At the
112 infinitely thin interface, local equilibrium is expressed via the partition coefficient between
113 phases. The released volatile flux will depend on the contact area between phases and the
114 transfer resistance in each phase, expressed via mass transfer coefficients. Other than the
115 interfacial release, bulk flow may also occur between the various compartments. The volatile
116 mass balances for this case involve the bulk concentrations and the bulk flow rates.

117 *2.1 Assumption of the contact area between the rice and saliva phase*

118 We assumed that the breakdown of a cooked rice particle during mastication follows a
119 ‘cleave and paste’ mechanism. In this mechanism, rice kernel when occluded between
120 opposing molars are assumed to produce one or a several large particles, and a fraction of the
121 original kernel is pasted into very fine particles (paste) which become effectively part of the
122 liquid phase of the bolus. As described in our previous work in How et al. (2021), it was
123 found that during the mastication of rice, the bolus forms a bimodal particle size distribution
124 (PSD) when measured using a laser diffraction method. It follows that particles with a size
125 less than 0.355 mm were not broken down in subsequent chewing cycles (i.e. pastes), which
126 are assumed to be individual separated swollen starch granules. Pasting involves swelling of
127 granules and it is known that native rice starches have granule sizes in the range of 1.9–26
128 μm . During cooking, the granule can swell 2 to 48 times of their initial size when heated to
129 various temperatures (How et al. 2021). Therefore the threshold of 0.355 mm used as an
130 assumption when particles become pastes simplifies the model.

131 Therefore, during mastication of rice, two simultaneous phenomena can occur:

- 132 • the transfer of aroma compounds from the non-pasted daughter particles into the
133 liquid phase of the bolus, particularly from newly exposed surface area generated
134 during a chewing cycle, and
- 135 • the release of the aroma compounds contained in the pasted particles into the liquid
136 phase of the bolus, where it was assumed because of the small size, that once
137 transferred, the concentration of the aroma compounds in the pasted and liquid phases
138 reach equilibrium instantaneously.

139 The direct transfer of aroma compounds from the solid surfaces into the air phase was
140 assumed negligible as after ingestion there will be at least a thin layer of saliva/moisture
141 present between these two phases and therefore aroma transfer always takes place through the
142 liquid phase.

143 Because of the challenge to distinguish between the relative contributions of each
144 mechanism, it was assumed that the release of the aroma compounds was dominated by the
145 transfer of the aroma compounds from the pasted particles into the liquid phase of the bolus.
146 This was a reasonable assumption as the surface area to volume ratio of the pasted particles
147 will be significantly larger than the surface area to volume ratio of the particles that were
148 greater than 0.355 mm. Similarly the distances for diffusion in the pasted particles was very
149 small. Thus, it is expected that the transfer of the aroma compounds from the pasted particles
150 to the liquid phase of the bolus is significantly faster compared to the transfer from the larger
151 particles.

152 Once in the liquid phase of the bolus, the concentration of the aroma compounds will be
153 diluted by saliva flow into the oral cavity (Harrison et al., 1998). At the same time, volatiles
154 partition from the saliva into the air phase which then transports them to the pharyngeal
155 compartment. During mastication, some aroma release to the pharynx and further to the nasal
156 cavity is possible for panellists with imperfect velopharyngeal closure (Trelea et al., 2007). In

157 the model, it is assumed that all subjects possess an imperfect velopharyngeal closure if the
158 concentration of aroma compounds increases after food ingestion and continues to do so
159 during the mastication period (i.e. the period before swallowing).

160 *2.2 Other model assumptions*

161 Further to the assumptions described above, the following additional assumptions were
162 made to develop the model equations.

- 163 • The assumptions for the aroma release model from oral, pharyngeal and nasal cavities
164 during consumption of solid (chewed) products were described in detail in
165 Doyennette et al., 2014.
- 166 • The assumptions for the integrated selection and breakage model to predict PSD of
167 rice particles during FOP have been described in detail in How et al., 2022.
- 168 • The assumptions concerning the interaction of the two models were as follows:
 - 169 ○ Food oral processing took place in isothermal conditions. That is, the
170 temperature of the cooked rice which was served at 37°C, was constant
171 throughout the duration of mastication. This is a reasonable assumption,
172 considering the body temperature is at 37°C. This assumption also ensures the
173 partition coefficient of aroma compounds will not change with temperature
174 and avoids the need to include a heat transfer model.
 - 175 ○ Food particles in the mouth were immediately coated with saliva at time zero
176 and after each chew when new surface area of particles were generated by
177 food breakage.
 - 178 ○ It was assumed that due to a relatively short mastication time, α -amylase had
179 zero or minimal contribution to the mass transfer coefficient of aroma
180 compounds by decreasing the bolus viscosity.

181 ○ The properties of the aroma compounds, such as the partition coefficient, were
182 not affected by the variations in saliva compositions between individuals and
183 by interaction with food components.

184 **3. Formulation of model equations**

185 The aroma release model equations used in this study are based on the one described in detail
186 in Doyennette et al. 2014 and the integrated selection and breakage models are based on How
187 et al. 2022. The main equations related to the interaction of the two models are given below
188 and the full set of equations are reported in appendices A2 and A3 respectively.

189 *3.1 Liquid bolus in the oral cavity*

190 The liquid bolus compartment is initially composed of pure saliva and is progressively
191 flavoured by the addition of rice particles which were pasted. The volume increases by the
192 addition of saliva (salivary flow rate) and with the addition of pasted particles.

193 The volume of the liquid bolus $V_{Ol(t)}$ can then be divided into two parts:

$$V_{Ol(t)} = V_{Os(t)} + V_{Opasted(t)} \#(1)$$

194 where

$$\frac{dV_{Os(t)}}{dt} = Q_{Os} \#(2)$$

195

196 *3.2 Concentration of aroma compounds in the saliva phase*

197 After each chewing cycle, a new volume of pasted particles are formed. These were then
198 added to saliva, which form the liquid bolus. The concentration of the aroma compounds in
199 the saliva phase and pasted particles were assumed to reach equilibrium instantaneously.
200 Therefore, after each chewing cycle, the mass of the aroma compounds in the saliva phase is

201 a combination of the initial mass (before chewing) and the new mass of aroma being added
 202 from the pasted particles. Hence,

$$V_{Os_i}C_{Os_i} + V_{Opasted_i}C_{Opasted_i} + (V_{Opasted_{i+1}} - V_{Opasted_i})C_{Op} = V_{Os_{i+1}}C_{Os_{i+1}} + V_{Opasted_{i+1}}C_{Opasted_{i+1}} \quad \#$$

203 ... (3)

204 The volatile concentration on the pasted particles side, using the partition conditions at the
 205 interface, is given by

$$C_{Opasted} = \frac{C_{Os}}{K_{Osp}} \quad \#(4)$$

206 After rearranging, the concentration of the aroma compound in the saliva after a single chew
 207 can be described as

$$C_{Os_{i+1}} = \frac{(V_{Os} + \frac{V_{Opasted}}{K_{Osp}})_i C_{Os_i} + (V_{Opasted_{i+1}} - V_{Opasted_i})C_{Op}}{(V_{Os} + \frac{V_{Opasted}}{K_{Osp}})_{i+1}} \quad \#(5)$$

208 This step change in aroma concentration occurs instantaneously with each chew and then
 209 changes dynamically due to dilution with the addition of saliva or losses to the oral airspace.
 210 Thus,

$$\frac{dC_{Os}(t)}{dt} = \frac{-\left(Q_{Os}C_{Os}(t) + A_{Ob}(t)k_{Os}\left(C_{Os}(t) - \frac{C_{Oa}(t)}{K_{Oas}}\right)\right)}{\left(V_{Os}(t) + \frac{V_{Opasted}(t)}{K_{Osp}}\right)} \quad \#(6)$$

211 3.3 Bolus surface area

212 For clarity to the reader, only the main equations used to predict the bolus surface area are
 213 described in this section. Full explanation is given in Appendix A3.

214 The bolus surface area was predicted by adapting the concept of a sintering mechanism which
 215 is used in forming metal, ceramic, polymer and composite components from particles
 216 (German, 2016). In a sintering mechanism, the total surface area of the particles is reduced by
 217 growing bonds (bridges) between contacting particles during a heating process. The same
 218 concept was applied to model the bolus surface area. As the number of chews increases, the
 219 amount of saliva incorporated in the bolus also increases. This also increases the number of
 220 saliva bridges among the particles in the bolus, which promotes the merge of particles. For
 221 the sintering mechanism, a linear relationship was proposed between the surface area and the
 222 packing density where the surface area declines as the density increases (German, 2016).
 223 Thus, the surface area of a bolus can be described as
 224

$$A_{Ob} = \frac{SA_{sphere} - SA_E}{1 - \phi_E} (\phi - 1) + SA_{sphere} \#(7)$$

225 where SA_E is the initial surface area of the bolus, which is the summation of all surface area
 226 of individual rice particles predicted using the integrated selection and breakage functions
 227 (How et al., 2022). As individual particles merge with the help of liquid saliva bridges to
 228 form a bolus, the total surface area of the bolus decreases. The bolus saturation, S decides
 229 when Eq. 7 is used to estimate the bolus surface area where

$$S = \frac{V_{opasted} + V_s}{V_{ptotal}} \cdot \left(\frac{1 - \varepsilon}{\varepsilon} \right) \#(8)$$

230 For any saturation value less than 1, Eq. 8 will be used to estimate the bolus surface area. For
 231 the bolus when the saturation surpasses the value of 1, the bolus surface area was estimated
 232 by assuming the bolus is of a spherical shape where

$$SA_{sphere} = 4 \pi R^2 \#(9)$$

233 3.3.1 Selection and Breakage model

234 The chewing model used to predict A_{Ob} in Eq. 7 consists of selection and breakage sub-
235 models. The mechanistic one-way and two-way competition selection models (van der Glas
236 et al., 1992, 2018; How et al., 2022) were initially applied and compared to identify which
237 model that best fits the experimental data. The breakage function used in the chewing model
238 was as described in Eq. 10 below which was previously applied on brown rice (Gray-Stuart,
239 2016). The equation assumes that the pasted fraction is constant.

240

$$241 \quad B(X, X_0) = (1 - P) \cdot \left[1 - \left(1 + r \cdot \frac{X}{X_0} \right) \cdot \left(1 - \frac{X}{X_0} \right)^r \right] \#(10)$$

242 The PSD was predicted using the discretised population balance model and is described in
243 detail in How et al. (2022).

244 3.4 Initial conditions

245 The initial concentration of aroma compounds in all compartments for the when the product
246 was introduced in the mouth up to the first chewing cycle (chew 0 to chew 1), is zero. The
247 initial volume of saliva to solve Eq. 2 was set as the volume of saliva at rest. Thus, the initial
248 conditions from chew 0 to chew 1 are:

$$V_{Os}(t_0) = V_{Osrest}$$

$$C_{Os}(t_0) = C_{Oa}(t_0) = C_{Om}(t_0) = C_{Fs}(t_0) = C_{Fm}(t_0) = C_{Fa}(t_0) = C_{Nm}(t_0) = C_{Na}(t_0) = 0$$

249 After the 1st chew, rice will break into smaller particles, and some will be pasted which
250 dissolves in the liquid bolus. The volatiles from the pasted particles are then transferred into
251 the liquid bolus, and an instant equilibrium was assumed (see section 2.1).

252 Thus, the initial conditions of the model following the first chew are:

253

$$C_{Os}(t_0) = C_{Os}(t_{chew=1}) \text{ calculated from Eq. 5}$$

$$V_{Os}(t_0) = V_{Os}(t_{chew=1})$$

$$C_{Oa}(t_0) = C_{Oa}(t_{chew=1})$$

$$C_{Om}(t_0) = C_{Om}(t_{chew=1})$$

$$C_{Fs}(t_0) = C_{Fs}(t_{chew=1})$$

$$C_{Fm}(t_0) = C_{Fm}(t_{chew=1})$$

$$C_{Fa}(t_0) = C_{Fa}(t_{chew=1})$$

$$C_{Nm}(t_0) = C_{Nm}(t_{chew=1})$$

$$C_{Na}(t_0) = C_{Na}(t_{chew=1})$$

254

255 *3.5 Model numerical solution*

256 The model was solved numerically using MATLAB program version R2019a using ode45
257 solver with a default relative error tolerance value of 0.001 which was shown to produce
258 negligible numerical error.

259 **4. Experimental methods**

260 *4.1 Food system*

261 White Jasmine rice (*Oryza sativa* L.) was used as the test food system of the model. Cooked
262 rice is aromatic and forms a particulate bolus with high bolus recovery (80-95%) which
263 allows the coupled selection-breakage and aroma models to be used (How et al., 2021). The
264 cooking method followed the procedure described in How et al. 2021 where it was cooked
265 using a 1:2 ratio, cooked in a microwave rice steamer. The rice was also cooked in three
266 batches to check the repeatability of the cooking method, where the moisture content (61

267 g/100 g of sample, average of three samples) showed $\pm 0.9\%$ variation between batches.
268 Approximately 5 g of cooked rice samples were transferred into small containers and were
269 kept warm at 60 °C. The rice was served to the subjects after cooling down to approximately
270 50 °C, which is the temperature at which rice is usually consumed (How et al., 2021).

271 *4.2 Subject's physiological characteristics*

272 The physiological characteristics are critical input parameters required to predict aroma
273 release. The input parameters were determined from the experimental measurements made in
274 How et al. (2021) or from the information found in the literature. Appendix A6 summarises
275 the physiological parameters required for the model, the values used and their source.

276 The oral cavity volume, $V_{O_{amean}}$, the volume of air in the pharynx, V_{Fa} and the volume
277 of air in the nasal cavity, V_{Na} were measured using the rhinopharyngometer as described in
278 Doyennette et al. (2014). The volume of saliva at rest, V_{Osrest} and the saliva flow rate, Q_{Os}
279 were obtained from the y-intercept and the slope of the subject's bolus saliva content and
280 chew number relationship based on the method previously discussed in detail in Motoi et al.
281 (2013). The number of chews required to reach swallow point, n_{chews} , the chewing frequency,
282 fr_{chew} and the time taken to swallow, $t_{swallow}$ for each subject during the *in vivo* aroma release
283 experiment were determined according to the protocol as previously described in How et al.
284 (2021).

285 The breathing frequency of the panellists during the consumption of the cooked rice
286 was estimated from the acetone signal (m/z 59) measured in the nasal cavity which was
287 recorded synchronously with the concentration of the target aroma compounds (Trelea et al.,
288 2007). The breathing frequency was determined by measuring the time it takes to complete
289 one breathing cycle. This was determined by identifying the point when the signal decreases
290 and increases, as it symbolises when the subject inhales and exhales. The breathing frequency

291 used in the model was the average of five replicates of breathing frequencies. Other
292 physiological variables including the area of the mucosa in all compartments (oral cavity,
293 pharynx and nasal cavity), the thickness of the mucosa layer, and the breath volume/tidal
294 volume were determined from the literature.

295 *4.3 Physico-chemical model input parameters*

296 Other input parameters required for the model include the physico-chemical properties of the
297 aroma compound that was flavoured in the cooked rice, such as the partition coefficient
298 between different phases. The air/rice partition coefficient at 37 °C and the initial
299 concentration of the studied aroma compounds were determined using the phase ratio
300 variation (PRV) method by headspace chromatography (Atlan et al., 2006; Doyennette et al.,
301 2011). Further explanation are given in Mohd Firdaus How (2021), Chapter 9. The mass
302 transfer coefficient was also required to predict aroma release, which was obtained from the
303 literature. Appendix A7 summarises the remaining parameters required for the model.

304 *4.4 Chewing model input parameters*

305 The initial PSD of 5 grams of rice (How et al., 2021) was used as the starting distribution in
306 the model. The size of particles were between 5-6.5 mm of diameter (Mohd Firdaus How,
307 2021). The input parameters required for the one-way and two-way selection functions are
308 the number of particles, the number of breakage sites and the affinity factor. When
309 considering the possibility of particle piling between antagonistic posterior teeth during jaw-
310 closing for the two-way interaction model, particle height, i.e. thickness or intermediate
311 diameter is relevant because the length axis of the rice grains will approximately parallel to
312 the occlusal plane between antagonistic teeth. It was found in van der Bilt et al. (1991) and
313 van der Glas et al. (2018) that particles can only pile when their height are smaller than 4
314 mm. In this study, assuming that the rice grains followed a similar aspect ratio of 3.3 ± 0.3 as
315 reported in Yu et al. (2019), the thickness (smallest diameter) or intermediate diameter of the

316 grains will be about 1.7 mm. Therefore, the piling conditions are completely favourable for
317 the two-way interaction model due to a small initial particle height of about 1.7 mm of rice
318 grains. As such, for efficiency, the two-way interaction model was used to describe the
319 selection process in the integrated selection-breakage model.

320 The degree of fragmentation variable and the pasted fraction are required for the breakage
321 function (section 3.3.1).

322 4.4.1 Selection-Breakage model fitting using Particle Swarm Optimisation 323 (PSO)

324 Using the initial PSD as described above as the input to the model, the rice bolus PSD after 1
325 chew, 2 chew, 25%, 50%, 75% and 100% of swallow point was determined for each subject.
326 The PSD of the bolus samples was analysed by image analysis following the procedure
327 described in How et al. (2021), which provided the projected area of individual particles in
328 mm². To transform the projected area (2D) to volume (3D), it was assumed that each rice
329 particle was of cylindrical shape, where the volume was determined by multiplying the
330 projected area with an assumed height. The height was obtained by multiplying the
331 characteristic dimension of the projected area with a factor. A circular shape was assumed to
332 determine the characteristic dimension of the projected area (diameter in mm). Thus,

$$V = h.A \#(11)$$

333
334 where V is the volume of a particle in mm³, h is the assumed height in mm, and A is the
335 projected area of a particle in mm². The constant h is determined by the following equation

$$h = f. \left(\frac{4.A}{\pi} \right)^{\frac{1}{2}} \#(12)$$

336
337 where f is a factor. The factor f was obtained by minimising the residual sum of squares
338 between the total predicted volume of particles calculated using equation 11 and the

339 experimental recovered volume of bolus (Gray-Stuart, 2016; Zheng et al., 2021). While the
340 assumption of a cylindrical shape rice particles provided a simple starting point in this study,
341 a more realistic shape for rice such as ellipsoidal (Yu et al., 2019) could be used in the future
342 where the ratios between the major and minor axes could also be assessed on the basis of
343 experimental data.

344 Due to the large number of model parameters to be solved, PSO algorithm was deployed to
345 solve the model input parameters by minimising the normalised sum of squares residuals
346 between the model and the experimental data. PSO has been used in food-related
347 optimisations such as modelling spray drying of coconut milk, mastication of peanuts and
348 more recently in the optimisation of restaurant atmosphere by coupling with Artificial Neural
349 Network (Ming et al., 2021; How et al., 2022; Kantono, How & Wang., 2022). The residual
350 was calculated from 10 d-values that were the intercepts for 10% (d10), 20% (d20) 30%
351 (d30), 40% (d40), 50% (d50), 60% (d60), 70% (d70), 80% (d80), 90% (d90) of the
352 cumulative volume distribution of the model and the experimental data. The algorithm was
353 also solved to minimise the normalised residual of the pasted fraction (defined as the volume
354 fraction for particles less than 0.354 mm) of the model and experimental data. Due to the
355 probabilistic nature of the way the selection and breakage models were implemented, the
356 model was repeated for 50 times and the average was determined to calculate the residual for
357 the model fitting (How et al., 2022).

358 4.5 *In vivo* aroma release data for model validation

359 Aroma release of five panellists' were measured using PTR-MS (Ionicon Analytik,
360 Innsbruck, Austria). These was the same data collected in our previous study in How et al.
361 (2021) where a minimum of five replicates were performed for all *in vivo* measurements. In
362 previous studies involving the modelling of aroma release, the model prediction is compared
363 against experimental measurements by representing the data as a peak line (Doyennette et al.,

2011; Doyennette et al., 2014). This is done by smoothing the breath-by-breath aroma release profiles by plotting a curve linking the maxima of the sinusoids (Doyennette et al., 2011). However, due to a longer sampling time nature of the experimental data and lack in consistency to when sampling was initiated relative to breathing (further explanation in Chapter 9 of Mohd Firdaus How. (2021)), the cumulative area under the curves of both model simulations and experimental data (normalised concentrations by dividing concentration against maximum concentration) were compared for validation.

5. Results and Discussions

5.1 Selection-Breakage model fitting

Table 1 below shows the comparison of the best-fit input parameter results when the breakage function of Eq. 10 and when the two-way competition selection model were applied (How et al., 2022). The R-squared was calculated as described in How et al. 2022. Figure 2 shows the model fits for Subject A4. For clarity to readers, comparisons between the fitted d -values and pasted fraction against experimental data for all of the five subjects are shown in Appendix A4.

For most of the subjects, a better fit was achieved when the two-way competition model was used as the selection model. This is supported by the higher R-squared values when the two-way competition model was used across most subjects in Table 1. Subject A1 however had a better fit when the one-way competition model was used as it had a greater number of larger particles (particles between 4-5.7 mm) compared to other subjects even at the later stages of mastication (Appendix A4).

5.2 Validation of the coupled selection breakage-mass transport models

The PSD outputs from the selection-breakage model were then used to calculate the volume of the pasted particles and the total surface area of particles as they were the variables

388 required to couple with the mass transport models.. Similarly, the physiological input
389 parameters of each subjects and the physicochemical parameters as described in the
390 experimental methods were applied. The cumulative area under the curve were then obtained
391 for the normalised concentrations of 2-nonanone (m/z 143) and ethyl propanoate (m/z 103) in
392 the nasal cavity.

393 The main assumption of the model is that during chewing, aroma compounds are transferred
394 to the saliva phase from the pasted rice particles instantaneously. The size threshold for when
395 rice particles are pasted was set as 0.354 mm by Gray-Stuart (2016) in his work. Here, the
396 model was predicted using different pasted size thresholds (0.2 mm, 0.354 mm, 0.5 mm and
397 when all particles are pasted) and compared against the experimental data to see if the
398 difference in the threshold has a pronounced effect on the prediction. The five replicates of
399 the experimental data were compared against the model prediction. It should be considered
400 that for the particle breakdown model the threshold was set to describe particles that become
401 too small to be actively broken down by occlusion. In terms of aroma release, the pasted
402 particle threshold corresponds to particles that are assumed to instantaneously equilibrate
403 their aroma compound with the liquid portion of the bolus. Figure 3 shows the comparison of
404 the model predictions against the experimental data for one of the subjects tested in the study
405 (subject A4). The results for the rest of the subjects are shown in Appendix A5. A common
406 trend can be seen from all figures is that the model predictions satisfactorily agreed with the
407 experimental data well for ethyl propanoate (m/z 103) compared to 2-nonanone (m/z 143).
408 When compared against different size thresholds,

409 *5.3 Using the integrated selection-breakage and mass transport models to* 410 *provide insights for food design* 411

412 Figure 4 shows the effects of some parameters related to the product and to the individual to
413 the aroma release (concentration of aroma compound in the nasal cavity, C_{na}) which could be

414 of interest to a food manufacturer. For clarity to readers, the results for subject A4 are used
415 here as a reference.

416 It can be observed when the portion size increased (Figure 4 a), C_{na} increased. This is
417 expected as the higher the portion size, the higher the volume of particles which will be
418 broken into pasted particles during mastication. This is consistent with the main model
419 assumption which assumes mass transfer of aroma compounds had only occurred from the
420 paste to the saliva phase. Therefore, the higher the volume of particles that are pasted, the
421 higher the C_{na} . The same trend can be observed when the initial particle size is varied (Figure
422 4 b). The larger the particle size, the higher the volume of a particle. Thus, the higher the
423 volume of pasted particles will be formed, which result to a higher C_{na} . A higher C_{na} can also
424 be observed when the initial concentration is increased as to be expected (Figure 4 c).

425 The fragmentation variable, r was also varied to test the effect of the breakage function of
426 foods on C_{na} . It can observed from Figure 4 d that the larger the r value, the higher the C_{na} ,
427 although the difference between the other r values is not obvious. A larger r value
428 corresponds to a higher degree of fragmentation (van der Glas et al., 1987). Therefore, a
429 larger r value will produce a higher number of smaller daughter particles. Theoretically, a
430 higher C_{na} should be observed with a larger r value as the increase in the number of smaller
431 particles will have created more surface area, resulting to a faster release and movement of
432 the aroma compounds from the rice matrix into the saliva and vapour phases (How et al.,
433 2021).

434 The reason why a less obvious trend (between the three r values used in the sensitivity
435 analysis) was observed could be due to the assumption of an entirely constant P value (Eq.
436 10), during chewing, which is a limitation of the study. When r and p values from Table 1 are
437 plotted (data not shown) against each other, it was shown that they are highly correlated (R^2

438 > 0.9), therefore any change in r must be accompanied by a subsequent change of p in the
439 same direction because of their causal relationship. By artificially breaking the causal
440 relationship, the effect of an increase of r is penalised by a constant value of p (which is too
441 small (Table 1) for the increased r), while a decrease in r is favoured by a constant, too large
442 value of p , which produces an artificial reverse effect of modifying r separately.

443 As expected, the magnitude of C_{na} is higher when p is larger, consistent with the main model
444 assumption (Figure 4 g). It is also expected that increasing p gives an expected increasing
445 effect on C_{na} because increasing p while keeping r constant also mimics the effect of
446 involving more particle sizes above the tiny ones (≤ 0.335 mm; sizes up to 1 mm) in aroma
447 release (Figure 2). Because of the limitation of Eq. 10, it is challenging to postulate the real
448 effect of r and p parameters on aroma release, without the need to develop a different model
449 that accounts for the strong causal relationship. The development of a new model will include
450 additional experiments to be able to better understand the specific nature and strength of the
451 causal relationship, which is subjected to future studies.

452 The higher the saliva flow rate (represented by the rate constant of the bolus saliva content
453 data), the smaller the magnitude of C_{na} (Figure 4 e), which is to be expected due to the
454 renewal of fresh saliva present in the mouth and pharynx (Doyennette et al., 2014). The
455 volume of saliva also determines the bolus saturation, which is a parameter required to
456 calculate the bolus surface area. A higher salivary flow rate will result in the bolus reaching
457 saturation faster, therefore may decrease surface area of the bolus. This results to a slower
458 rate of transfer of aroma from the bolus to the air phase of the mouth, hence, explains the
459 smaller C_{na} value. Thus, a food manufacturer may avoid adding additional
460 chemical/components (such as citric acid) that may increase the saliva flow rate during
461 mastication. The same trend can also be observed when the initial volume of liquid in the
462 mouth is varied, where a higher initial liquid volume results to a smaller magnitude of C_{na}

463 (Figure 4 f). This is to demonstrate when rice is served with liquid such as curries or soup
464 which will reach bolus saturation immediately and will therefore have smaller bolus surface
465 area. Finally, it is also interesting to test the effect of the chewing rate on C_{na} as it is
466 dependent on the food structure (e.g., soft vs hard foods). Comparing the magnitude of C_{na} in
467 the first 15 seconds in Figure 4 h, chewing faster has a higher magnitude as it takes a shorter
468 time to swallow for the same initial mass of aroma compounds.

469 Besides the product, aroma release is also influenced by the physiological parameters of
470 humans. This is a challenge for food technologists as humans show wide variation in these
471 parameters and in the way they consume food (Taylor, 2002). Mathematical modelling can
472 provide insights into understanding the role of individual physiological parameters as these
473 parameters are defined in the model to predict aroma release. In this way, the model can be a
474 tool to design food that can tailor to individual's physiological characteristics.

475 *5.3 Effect of physiological parameters on model predictions*

476 The physiological parameters that are used as the model inputs were manipulated to observe
477 its effects on the aroma release predictions. As can be seen from Figure 5, it can be observed
478 that aroma release was mostly impacted from the breathing frequency, the volume of
479 pharynx, the volume of oral cavity and the volume of nasal cavity while the rest of the
480 parameters seem to have a negligible effect. A higher breathing frequency seems to have
481 lower aroma concentration as it increases the removal of the aroma compound from the
482 mouth. A higher aroma concentration is also observed with a larger oral cavity and it
483 becomes more apparent towards the later stages of mastication. A larger volume of oral
484 cavity indicates a larger volume of aroma-rich air in the oral cavity. Due to this, a higher
485 concentration of aroma will be observed in the nasal cavity as it takes longer for the aroma-
486 rich air in the oral cavity to be depleted during breathing (for the same breath flow rate). The
487 variation of the volume of pharynx gives a higher C_{na} when the volume is smaller. The same

488 trend can be observed with the variation of the volume of nasal cavity. The effects can be
489 explained from the combination of an aroma-rich air from the oral cavity/pharynx with
490 aroma-free ambient air while breathing. A lower pharynx/nasal cavity volume implies higher
491 renewal rate, therefore it leads to a quicker increase and decrease of the aroma concentration
492 (Trelea et al., 2007). The remaining physiological parameters as seen in Figure 5 seem to
493 have negligible effect on the simulated nasal aroma concentration. This indicates that their
494 accurate knowledge of the parameters is not vital for running the model.

495 The substantial effects of some of the physiological parameters on aroma
496 concentration can provide knowledge to food manufacturers to design foods to a specific
497 class of consumer. For example, race and gender are known to be the important factors
498 affecting the oral and nasal structures (Xue & Hao, 2006). A study by Xue & Hao (2006)
499 compared the vocal tract dimensions of 120 healthy adult subjects with equal numbers of men
500 and women of three races (White American, African American and Chinese). The results
501 showed that the men have a larger vocal tract dimension (e.g. oral and pharynx volume)
502 compared to women. Chinese people seem to have the largest oral and pharynx volume,
503 followed by White American and African American. Thus, the physiological parameters of
504 the subjects need to be considered by a food manufacturer in the food designing process. If a
505 subject possesses a large oral volume, perhaps only a small initial concentration of aroma
506 compound is required in the food to be able to perceive the 'right amount of flavor'.
507 However, besides the physiological parameters, it is also known that other factors such as the
508 nature of the food matrix and physicochemical factors can affect aroma release. It can be
509 challenging to identify the parameters that have the most effect on a subject through a series
510 of lab experiments as these are time-consuming and financially expensive. Therefore,
511 development of mechanistic models linking oral processing and aroma release provides tools

512 to explore these interactions and can lead to the development of foods influencing sensorial
513 and digestive outcomes.

514 *5.4 Effect of physico-chemical parameters on model predictions*

515

516 Physico-chemical factors such as partitioning, interfacial mass transport and diffusion are
517 mechanisms that can affect aroma release (Taylor, 2002). Food technologist is interested in
518 this area as upon mastication, flavour components are released, and the overall sensory
519 appreciation is influenced by the way the components are distributed over the different
520 phases (that make up the food microstructure) and the diffusion kinetics of flavor release and
521 transport of the volatiles to the olfactory epithelium in the nasal cavity (Bruin, 1999).

522 As can be seen from Figure 6, almost all the physico-chemical parameters have negligible
523 effect on the simulated aroma concentration in the nasal cavity except the mass transfer
524 coefficient of saliva in the oral cavity. A higher aroma concentration is observed with higher
525 value of mass transfer coefficient. This parameter was also pointed out to be one of the key
526 factors governing the release of aroma compounds when a sensitivity analysis was carried out
527 in the aroma release mechanistic model developed for cheese (Doyennette et al., 2014). The
528 mass transfer coefficient could be influenced by the viscosity of the saliva and the stirring
529 rate (tongue and cheek movements), both of which determine the thickness of the stagnant
530 layer (Nahon et al., 2000). Increasing the viscosity of the surrounding fluid by addition of
531 thickeners or simply raising the concentration of aroma in the saliva will therefore decrease
532 the mass transfer coefficient and the rate of flavour release (Nahon et al., 2000). The rest of
533 the physico-chemical parameters such as the partition coefficients (saliva to rice, air to saliva)
534 and the mass transfer coefficients in all physiological compartments all seem to have a
535 negligible effect on the aroma concentration, which indicate that their accurate knowledge is
536 not essential to run the model.

537 **6. Conclusions**

538 An integrated selection-breakage model was coupled with mass transport models to predict
539 aroma release during the consumption of cooked white rice. The validity of the model was
540 tested by comparing model predictions against *in vivo* aroma release data of five subjects.
541 Among the product related parameters studied, the model showed that the portion size, initial
542 concentration of aroma, initial liquid volume and the pasted fraction have the most impact on
543 the aroma concentration. Physiologically, the model showed that the oral cavity volume,
544 pharynx volume, nasal cavity volume and the breathing frequency were the variables that
545 affect aroma concentration the most. The mass transfer coefficient of saliva has the most
546 significant effect on the aroma release among all physico-chemical parameters studied in the
547 model. The effects of the partition coefficient of different aroma compounds were also
548 explored where aromas that had the highest affinity in air showed the highest aroma release.
549 All in all, the incorporation of the mechanistic chewing model in this study provides the first
550 step upon the development of mechanistic models that can lead to the development of foods
551 to influence sensorial and digestive outcomes.

552 **CRedit authorship contribution statement**

553
554 **Syahmeer How:** Conceptualization, Methodology, Investigation, Visualization, Formal
555 analysis, Funding acquisition, Writing - original draft. **Jim R. Jones:** Supervision,
556 Visualization, Writing - review & editing. **Marco P. Morgenstern:** Supervision, Writing -
557 review & editing. **Eli Gray-Stuart:** Supervision. **John E. Bronlund:** Supervision, Funding
558 acquisition, Visualization, Formal analysis, Writing - review & editing. **Anne Saint-**
559 **Eve:** Formal analysis. **Ioan Cristian Trelea:** Visualization, Writing - review &
560 editing. **Isabelle Souchon:** Conceptualization, Methodology, Supervision, Formal analysis,
561 Funding acquisition.

562 **Declaration of competing interest**

563 The authors declare they have no competing interest.

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565

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670

List of Figure Captions

Figure 1. Conceptual diagram of the interconnected compartments and the mechanisms involved in flavour release during the consumption of cooked white rice.

Figure 2. Best-fit model against experimental data for Subject A4. Two-way interaction model was used as the selection model. The plot on the left of the image is the comparison of the d-values of the best fit model and the experimental data whereas the plot on the right shows the comparison of the pasted fraction values. The error-bar of the model is the standard deviation of 50 simulations. All 3 replicates of the measured data (three different markers) were plotted for comparison.

Figure 3. Model prediction against experimental data of 2-nonanone (m/z 143) and ethyl propanoate (m/z 103) for Subject A4. Rel. AUC refers to the relative cumulative area under the curve, where the cumulative area under the curve was normalised against the total area under the curve. The model was also predicted using different pasted size threshold (0.2 mm, 0.354 mm, 1 mm and when all particles were pasted) and compared against the five replicates of the experimental data.

Figure 4. Effect of parameters related to the product and individual which may be of interest to a food manufacturer. Using the physiological parameters of Subject A4, C_{na} of 2-nonanone was predicted. a. Aroma release (C_{na}) when portion size is varied (2.5 g, 5 g and 10 g of rice), b. Aroma release (C_{na}) when initial particle size is varied (halved, original and doubled size) for 5 g of rice c. Aroma release (C_{na}) when the initial concentration is varied for 5 g of rice, d. Aroma release (C_{na}) when the breakage function (represented by the fragmentation variable, r) is varied for 5 g of rice, e. Aroma release (C_{na}) when saliva flow rate is varied for 5 g of rice, f. Aroma release (C_{na}) when the initial liquid volume is varied for 5 g of rice, g. Aroma release (C_{na}) when the breakage function (represented by the pasted fraction, p) is varied for 5 g of rice, h. Aroma release (C_{na}) when the chewing rate (0.7 chew/s, 1.5 chew/s and 2 chew/s) is varied for 5 g of rice

Figure 5. Effect of physiological parameters on aroma release (C_{na}). Using the physiological parameters of Subject A4 as a reference value, C_{na} of 2-nonanone was predicted. a. Effect of breathing frequency on aroma release (C_{na}) (0.12 cycle/s, 0.25 cycle/s, 0.32 cycle/s), b. Effect of the oral cavity volume on aroma release (C_{na}) ($2 \times 10^{-5} \text{ m}^3$, $6 \times 10^{-5} \text{ m}^3$, $1 \times 10^{-4} \text{ m}^3$) c. Effect of the pharynx volume on aroma release (C_{na}) ($1.5 \times 10^{-5} \text{ m}^3$, $3.2 \times 10^{-5} \text{ m}^3$, $6 \times 10^{-5} \text{ m}^3 \text{ mm}^3$), d. Effect of the nasal cavity volume on aroma release (C_{na}) ($1.2 \times 10^{-5} \text{ m}^3$, $1.6 \times 10^{-5} \text{ m}^3$, $4 \times 10^{-5} \text{ m}^3$), e. Effect of the oral mucosa thickness on aroma release (C_{na}) ($5 \times 10^{-6} \text{ m}$, $5 \times 10^{-5} \text{ m}$, $5 \times 10^{-4} \text{ m}$), f. Effect of the pharynx mucosa thickness on aroma release (C_{na}) ($5 \times 10^{-6} \text{ m}$, $5 \times 10^{-5} \text{ m}$, $5 \times 10^{-4} \text{ m}$), g. Effect of the nasal mucosa thickness on aroma release (C_{na}) ($5 \times 10^{-6} \text{ m}$, $5 \times 10^{-5} \text{ m}$, $5 \times 10^{-4} \text{ m}$), h. Effect of tidal volume on aroma release (C_{na}) ($2.5 \times 10^{-4} \text{ m}^3$, $5 \times 10^{-4} \text{ m}^3$, $1 \times 10^{-3} \text{ m}^3$), i. Effect of oral cavity area on aroma release (C_{na}) ($5.8 \times 10^{-3} \text{ m}^2$, $1.6 \times 10^{-2} \text{ m}^2$, $2.32 \times 10^{-2} \text{ m}^2$), j. Effect of pharynx area on aroma release (C_{na}) ($3.3 \times 10^{-3} \text{ m}^2$, $6.5 \times 10^{-3} \text{ m}^2$, $1.3 \times 10^{-2} \text{ m}^2$), k. Effect of air/mucosa contact area in the nasal cavity on aroma release (C_{na}) ($7.5 \times 10^{-3} \text{ m}^2$, $1.6 \times 10^{-2} \text{ m}^2$, $3 \times 10^{-2} \text{ m}^2$), l. Effect of volume of saliva in pharynx (C_{na}) ($1 \times 10^{-7} \text{ m}^3$, $1 \times 10^{-7} \text{ m}^3$, $4 \times 10^{-7} \text{ m}^3$)

Figure 6. Effect of physico-chemical parameters on aroma release (C_{na}). Using the physiological parameters of Subject A4 as a reference value, C_{na} of 2-nonanone was predicted. a. Effect of saliva/rice partition coefficient on aroma release (C_{na}) (2.45×10^{-2} , 2.45×10^{-1} , 2.45), b. Effect of air/saliva partition coefficient on aroma release (C_{na}) (9.7×10^{-4} , 9.7×10^{-3} , 9.7×10^{-2}) c. Effect of air/mucosa partition coefficient in the oral cavity on aroma release (C_{na}) (1×10^{-5} , 1×10^{-3} , 1×10^{-1}), d. Effect of air/mucosa partition coefficient in the pharynx on aroma release (C_{na}) (1×10^{-5} , 1×10^{-3} , 1×10^{-1}), e. Effect of air/mucosa partition coefficient in the nasal cavity on aroma release (C_{na}) (1×10^{-5} , 1×10^{-3} , 1×10^{-1}), f. Effect of air/saliva partition coefficient in the pharynx on aroma release (C_{na}) (5×10^{-4} , 5×10^{-3} , 5×10^{-2}), g. Effect of mass transfer coefficient in saliva in oral cavity on aroma release (C_{na}) ($1 \times 10^{-8} \text{ m/s}$, $1 \times 10^{-6} \text{ m/s}$, $1 \times 10^{-4} \text{ m/s}$), h. Effect of mass transfer coefficient in saliva in pharynx on aroma release (C_{na}) ($1 \times 10^{-8} \text{ m/s}$, $1 \times 10^{-6} \text{ m/s}$, $1 \times 10^{-4} \text{ m/s}$), i. Effect of mass transfer coefficient in mucosa in oral cavity on aroma release (C_{na}) ($1 \times 10^{-8} \text{ m/s}$, $1 \times 10^{-6} \text{ m/s}$, $1 \times 10^{-4} \text{ m/s}$), j. Effect of mass transfer coefficient in mucosa in pharynx on aroma release (C_{na}) ($1 \times 10^{-8} \text{ m/s}$, $1 \times 10^{-6} \text{ m/s}$, $1 \times 10^{-4} \text{ m/s}$), k. Effect of mass transfer coefficient in mucosa in nasal cavity on aroma release (C_{na}) ($1 \times 10^{-8} \text{ m/s}$, $1 \times 10^{-6} \text{ m/s}$, $1 \times 10^{-4} \text{ m/s}$)

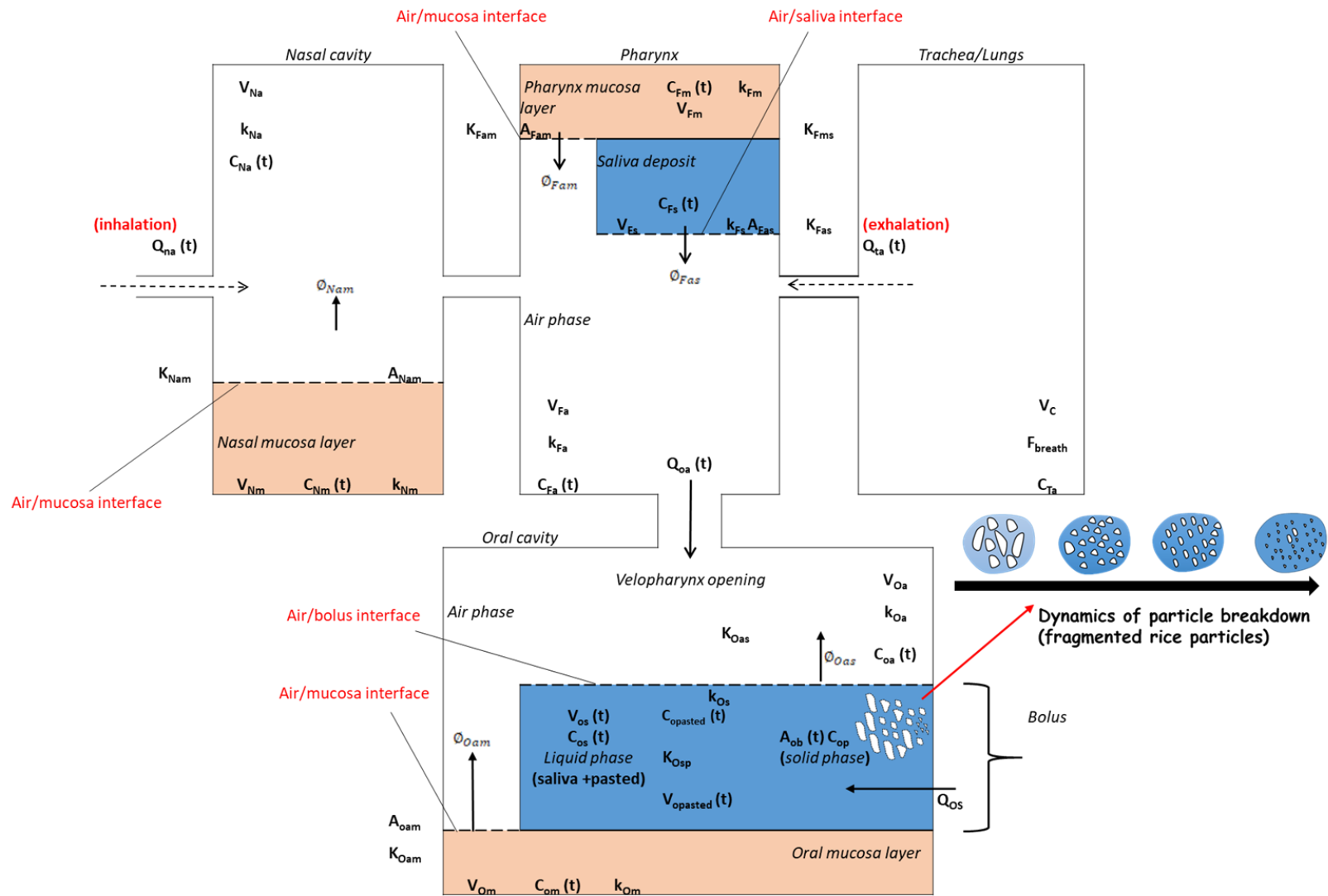


Figure 1. Conceptual diagram of the interconnected compartments and the mechanisms involved in flavour release during the consumption of cooked white rice.

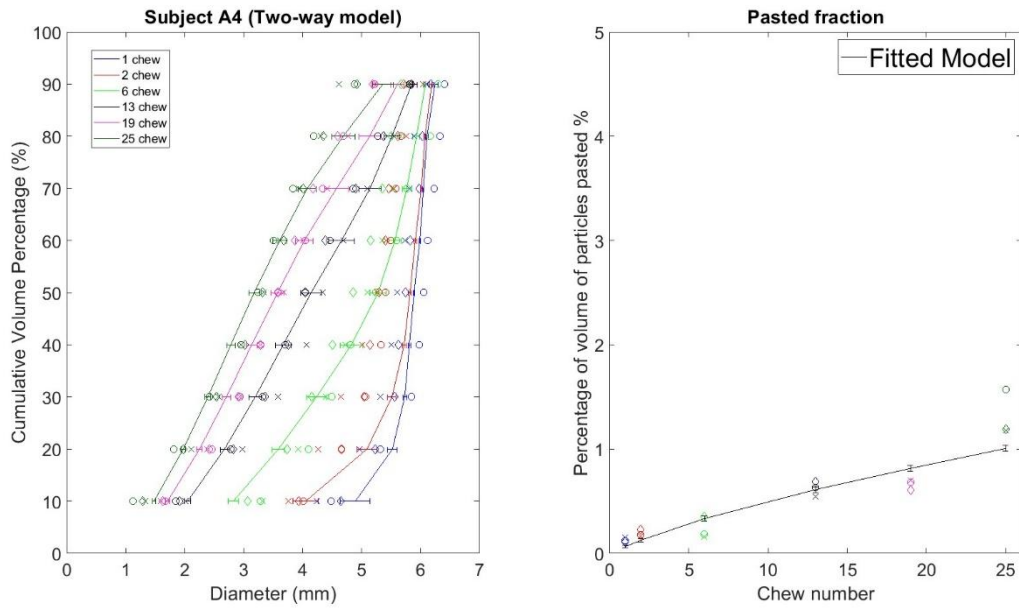
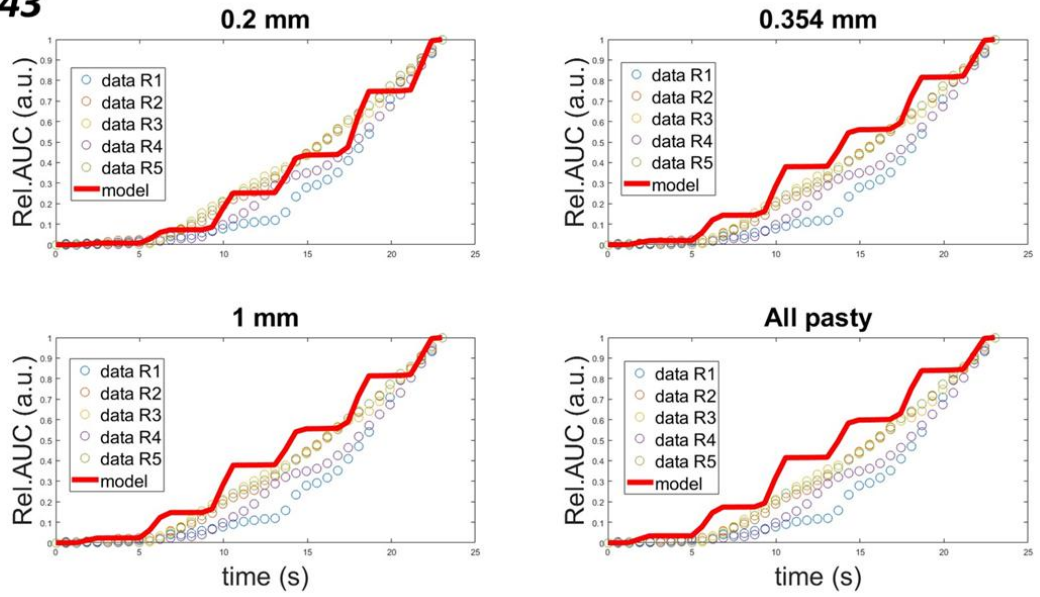


Figure 2. Best-fit model against experimental data for Subject A4. Two-way interaction model was used as the selection model. The plot on the left of the image is the comparison of the d-values of the best fit model and the experimental data whereas the plot on the right shows the comparison of the pasted fraction values. The error-bar of the model is the standard deviation of 50 simulations. All 3 replicates of the measured data (three different markers) were plotted for comparison.

a. m/z 143



b. m/z 103

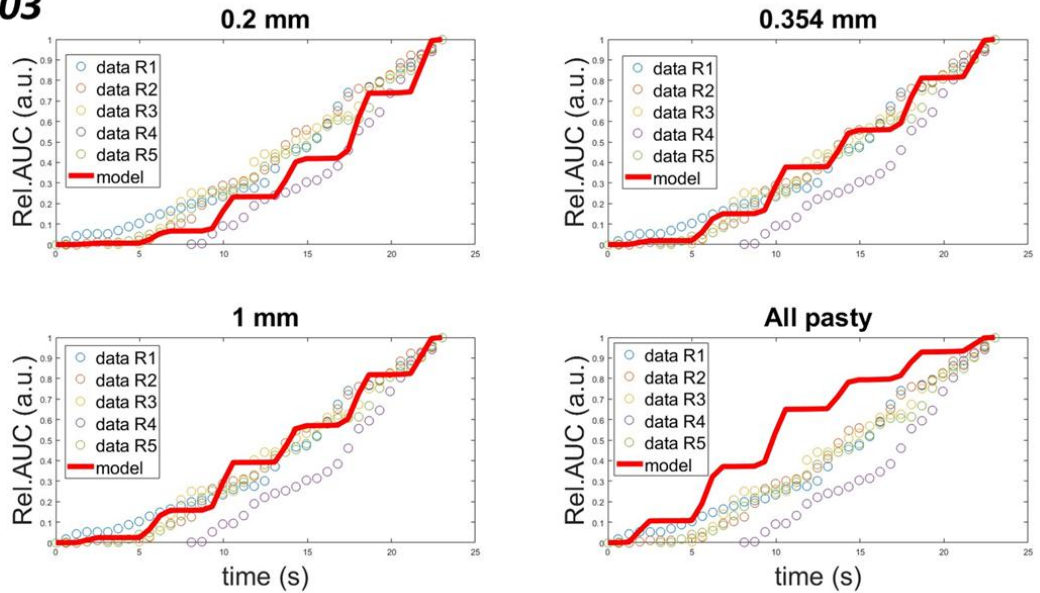


Figure 3. Model prediction against experimental data of 2-nonanone (m/z 143) and ethyl propanoate (m/z 103) for Subject A4. Rel. AUC refers to the relative cumulative area under the curve, where the cumulative area under the curve was normalised against the total area under the curve. The model was also predicted using different pasted size threshold (0.2 mm, 0.354 mm, 1 mm and when all particles were pasted) and compared against the five replicates of the experimental data.

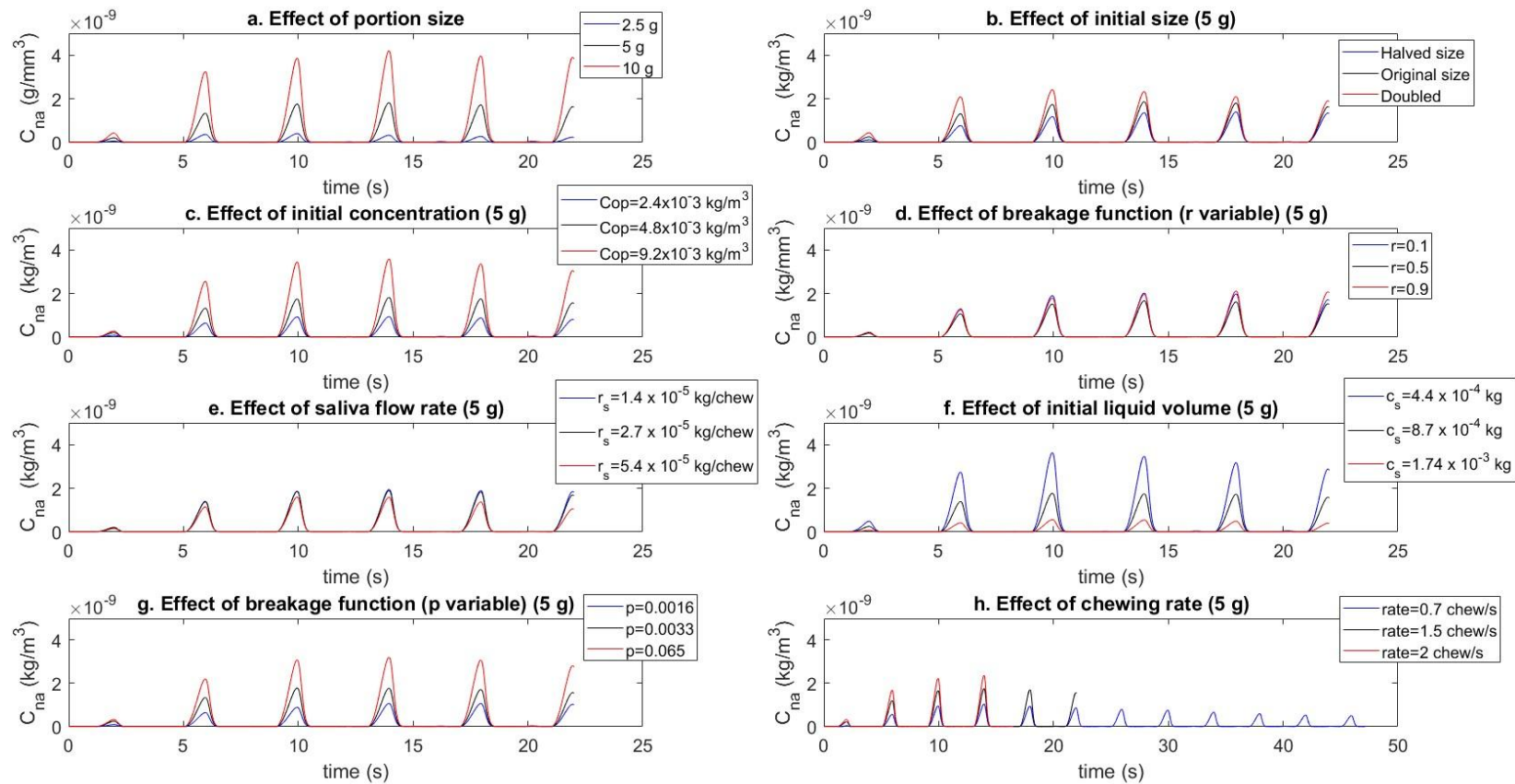


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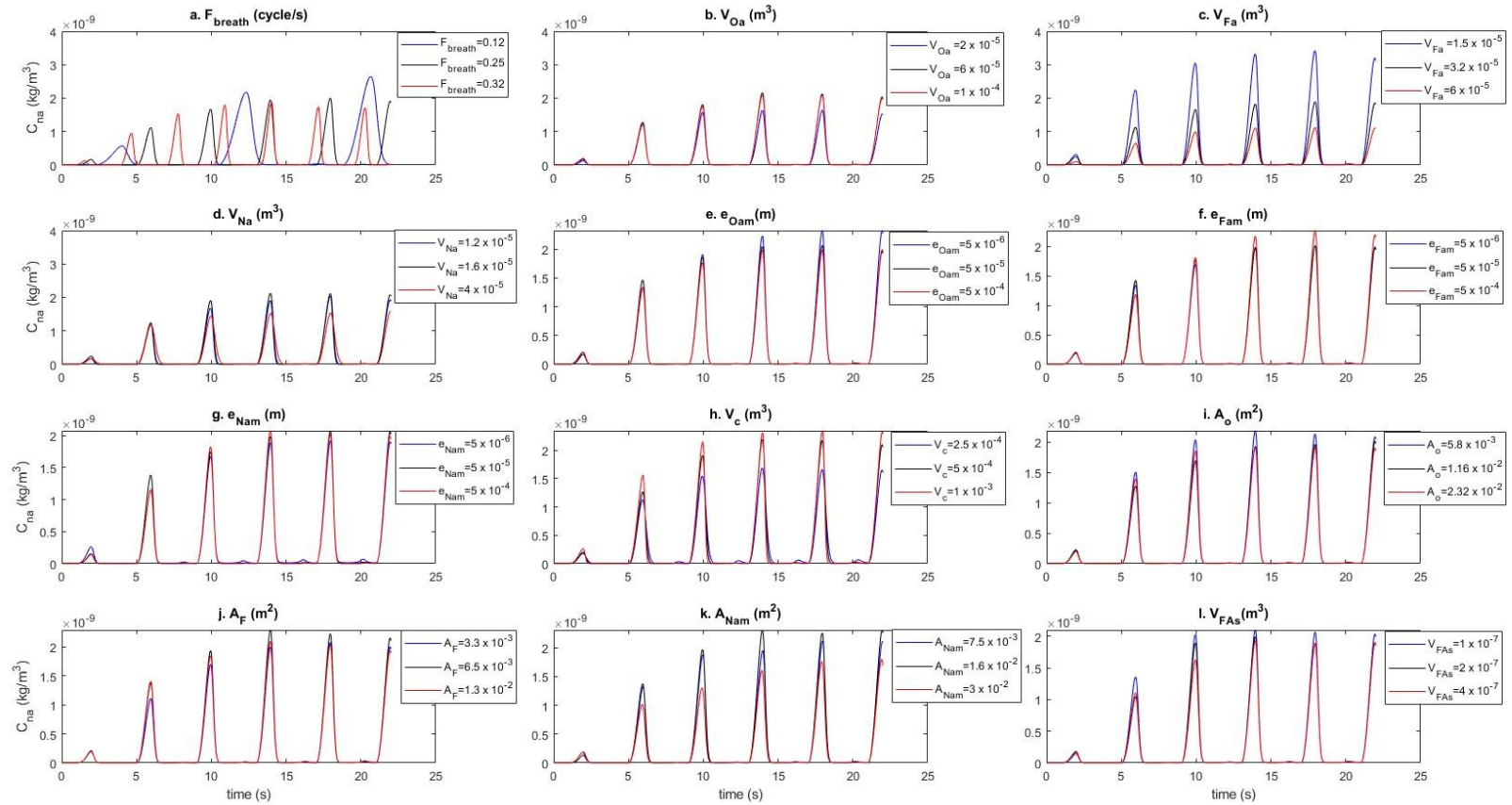


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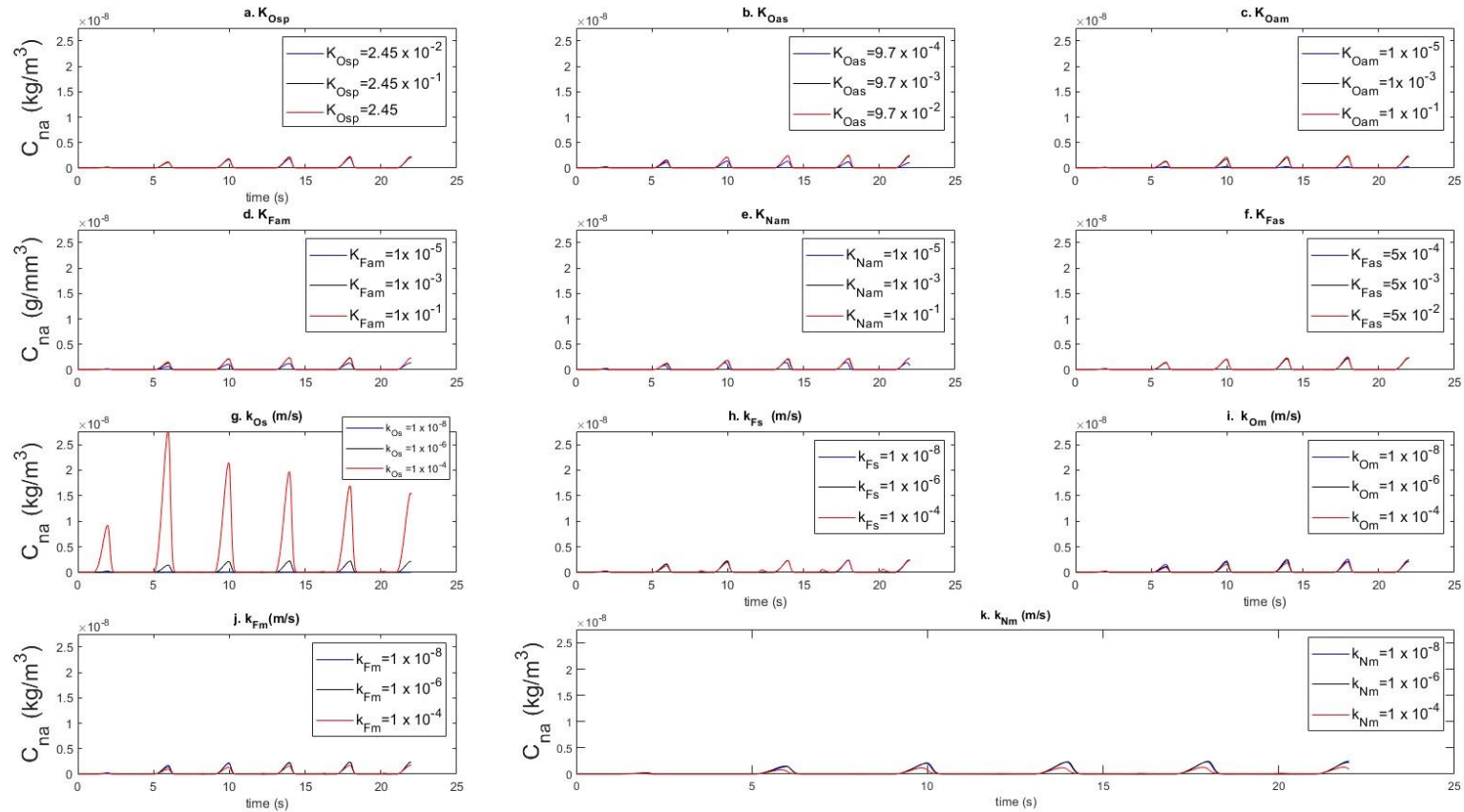


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List of Table Captions

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Table 1. Best fit input parameters of the PSD model evaluated from the PSO algorithm. The two-way competition selection model was applied integrated with a constant breakage model (Eq.10). The R-squared value was given to describe the goodness of fit.

Subject	Selection model	Selection model inputs				Breakage model input		Normalised		
		Number of breakage sites, n_b		Affinity factor, α_1		Fragmentation variable, r	Pasted fraction, p	SS residuals	SS	R-squared
		Multiplication factor, k	Power, m	Multiplication factor, p	Power, q					
A1	Two-way competition	199.83	1.19	0.003	1.58	0.48	0.003	3.69	33.35	0.79
A2	Two-way competition	448.14	1.68	0.001	1.98	0.65	0.006	2.54	23.53	0.84
A3	Two-way competition	341.5	2.11	0.002	1.69	0.61	0.004	5.07	48.96	0.73
A4	Two-way competition	212.42	1.34	0.002	1.96	0.59	0.003	0.95	4.22	0.98
A5	Two-way competition	210.22	1.08	0.002	1.35	0.66	0.005	0.83	10.86	0.94

Appendix A1 – List of Nomenclatures

Table A-1 defines the variables used in the model development used in this chapter.

Table A-1 List of nomenclature

Symbol	Unit	Description
A_{Oam}	m^2	air/lubricated mucosa contact area in the oral cavity
A_{Ob}	m^2	air/bolus contact area in the oral cavity
A_{Fas}	m^2	air/saliva contact area in the pharynx
A_{Fam}	m^2	air/lubricated mucosa in the pharynx
A_{Nam}	m^2	air/lubricated mucosa in the nasal cavity
C_{Os}	kg/m^3	aroma concentration in the saliva in the oral cavity
C_{Oa}	kg/m^3	aroma concentration in the air in the oral cavity
C_{Om}	kg/m^3	aroma concentration in the lubricated mucosa in the oral cavity
C_{Op}	kg/m^3	aroma concentration in the product (rice) in the oral cavity
C_{Fs}	kg/m^3	aroma concentration in the saliva in the pharynx
C_{Fa}	kg/m^3	aroma concentration in the air in the pharynx
C_{Fm}	kg/m^3	aroma concentration in the lubricated mucosa in the pharynx
C_{Na}	kg/m^3	aroma concentration in the air in the nasal cavity
C_{Nm}	kg/m^3	aroma concentration in the lubricated mucosa in the nasal cavity
C_{Ta}	kg/m^3	aroma concentration in the trachea
F_{breath}	number of cycles/s	breathing frequency
$fr_{opening}$	occurrence number/s	opening frequency of the velopharynx
fr_{chew}	number of chews/s	chewing frequency
k_{Oa}	m/s	mass transfer coefficient in the air phase in the oral cavity
k_{Os}	m/s	mass transfer coefficient in the saliva phase in the oral cavity
k_{Om}	m/s	mass transfer coefficient in the lubricated mucosa in the oral cavity
k_{Fs}	m/s	mass transfer coefficient in the

k_{Fa}	m/s	saliva phase in the pharynx mass transfer coefficient in the air phase in the pharynx
k_{Fm}	m/s	mass transfer coefficient in the lubricated mucosa in the pharynx
k_{Na}	m/s	mass transfer coefficient in the air phase in the nasal cavity
k_{Nm}	m/s	mass transfer coefficient in the lubricated mucosa in the nasal cavity
K_{Oas}		air/saliva partition coefficient in the oral cavity
K_{Oam}		air/lubricated mucosa partition coefficient in the oral cavity
K_{Osp}		saliva/product(rice) partition coefficient in the oral cavity
K_{Fas}		air/saliva partition coefficient in the pharynx
K_{Fms}		lubricated mucosa/saliva partition coefficient in the pharynx
K_{Fam}		air/lubricated mucosa partition coefficient in the pharynx
K_{Nam}		air/lubricated mucosa partition coefficient in the nasal cavity
V_{bolus}	m^3	volume of bolus
V_c	m^3	current breath volume
V_{Os}	m^3	volume of saliva in the oral cavity
V_{Osrest}	m^3	volume of saliva at rest in the oral cavity
$V_{Opasted}$	m^3	volume of pasted rice particles in the oral cavity
V_{Om}	m^3	volume of mucosa in the oral cavity
V_{Oa}	m^3	volume of air in the oral cavity
V_{fluid}	m^3	volume of saliva surrounded on a single particle
V_{Fa}	m^3	volume of air in the pharynx
V_{Fs}	m^3	volume of saliva in the pharynx
V_{Fm}	m^3	volume of mucosa in the pharynx
V_{Na}	m^3	volume of air in the nasal cavity
V_{Nm}	m^3	volume of mucosa in the nasal cavity
V_p	m^3	volume of a single particle

$V_{p\ total}$	m^3	total volume of particles (non-pasted)
V_{tot}	m^3	volume of particle and surrounding saliva
Q_{Os}	m^3/s	saliva flow rate in the oral cavity
Q_{Oa}	m^3/s	air flow rate in the oral cavity
Q_{Na}	m^3/s	air flow rate in the nasal cavity
Q_{Ta}	m^3/s	air flow rate in the trachea
r	m	radius of a particle
R	m	radius of a bolus
S		saturation of the bolus
SA_E	m^2	surface area of expanded bolus
SA_{sphere}	m^2	surface area of a spherical bolus
t	s	time
ϕ		volume fraction of bolus
ϕ_E		volume fraction of expanded bolus
ϕ_{Oas}	kg/s	volatile mass flux between air and saliva in the oral cavity
ϕ_{Oam}	kg/s	volatile mass flux between air and lubricated mucosa in the oral cavity
ϕ_{Fam}	kg/s	volatile mass flux between air and lubricated mucosa in the pharynx
ϕ_{Fas}	kg/s	volatile mass flux between air and saliva in the pharynx
ϕ_{Nam}	kg/s	volatile mass flux between air and lubricated mucosa in the nasal cavity
ε	-	porosity

Appendix A2 – Other equations used to predict aroma release as described in Doyennette et al. (2014)

Air/bolus interfacial conditions in the oral cavity

$$C_{Os}(t) = \frac{C_{Oa}(t)}{K_{Oas}} \#(A2 - 1)$$

Aroma compound retention by the lubricated mucosa in the oral cavity, pharynx and nasal cavity

$$V_{Om} \frac{dO_m(t)}{dt} = -k_{Om} A_{Oam} \left(C_{Om}(t) - \frac{C_{Oa}(t)}{K_{Oam}} \right) \#(A2 - 2)$$

Air in the oral cavity

$$V_{Oa}(t) \frac{dC_{Oa}(t)}{dt} = A_{Ob}k_{Os} \left(C_{Os}(t) - \frac{C_{Oa}(t)}{K_{Oas}} \right) + k_{Om}A_{Oam} \left(C_{Om}(t) - \frac{C_{Oa}(t)}{K_{Oam}} \right) + \begin{cases} Q_{Oa}(t)(C_{Fa}(t) - C_{Oa}(t)) & \text{if } Q_{Oa}(t) \geq 0 \\ 0 & \text{if } Q_{Oa}(t) < 0 \end{cases} \quad \dots \quad (A2 - 3)$$

$$V_{Oa}(t) = V_{Oa\text{mean}} + \Delta V_{Oa} \sin(2\pi f r_{\text{opening}} t) \quad \#(A2 - 4)$$

$$Q_{Oa}(t) = \frac{dV_{Oa}(t)}{dt} = 2\pi f r_{\text{opening}} \Delta V_{Oa} \cos(2\pi f r_{\text{opening}} t) \quad (A2 - 5)$$

Bolus in the pharynx

$$V_{Fs} \frac{dC_{Fs}(t)}{dt} = -k_{Fs}A_{Fas} \left(C_{Fs}(t) - \frac{C_{Fa}(t)}{K_{Fas}} \right) \quad \#(A2 - 6)$$

Air in the pharynx

$$Q_{Na}(t) = -Q_{Ta}(t) + Q_{Oa}(t) \quad \#(A2 - 7)$$

$$V_{Fa} \frac{dC_{Fa}(t)}{dt} = k_{Fs}A_{Fas} \left(C_{Fs}(t) - \frac{C_{Fa}(t)}{K_{Fas}} \right) + k_{Fm}A_{Fam} \left(C_{Fm}(t) - \frac{C_{Fa}(t)}{K_{Fam}} \right) + \begin{cases} -Q_{Oa}(t)(C_{Oa}(t) - C_{Fa}(t)) & \text{if } Q_{Oa}(t) < 0 \\ Q_{Na}(t)(C_{Na}(t) - C_{Fa}(t)) & \text{if } Q_{Na}(t) \geq 0 \\ Q_{Ta}(t)(C_{Ta}(t) - C_{Fa}(t)) & \text{if } Q_{Ta}(t) \geq 0 \end{cases} \quad \dots \quad (A2 - 8)$$

Air in the nasal cavity

The mass balance of the aroma compound in the air phase in the nasal cavity is given by:

$$V_{Na} \frac{dC_{Na}(t)}{dt} = k_{Nm} A_{Nam} \left(C_{Nm}(t) - \frac{C_{Na}(t)}{K_{Nam}} \right) + \begin{cases} Q_{Na}(t) (0 - C_{Na}(t)) & \text{if } Q_{Na}(t) < 0 \\ Q_{Na}(t) (C_{Fa}(t) - C_{Na}(t)) & \text{if } Q_{Na}(t) \geq 0 \end{cases}$$

#... (A2 - 9)

Appendix A3 – Bolus Surface Area Full Derivation

The following derivation shows the steps required and assumptions made to calculate the bolus surface area.

For example, if each particle is assumed as a sphere (to provide clarity spherical shape is used here but the equations will be adaptable to other particle shape as well), the volume of a particle can be described as

$$V_p = \frac{4}{3}\pi r^3 \#(A3 - 1)$$

If it was assumed that each particle has an amount of fluid volume, V_{fluid} associated with it, the volume fraction of the fluid with respect to the volume of particle is

$$a = \frac{V_{fluid}}{V_p} \#(A3 - 2)$$

Therefore the V_{fluid} is

$$V_{fluid} = a \frac{4}{3}\pi r^3 \#(A3 - 3)$$

Assuming that each particle has an even and the same coating thickness, x (independent of size), the total volume of the particle and the fluid can be described as

$$V_{tot} = \frac{4}{3}\pi(r + x)^3 \#(A3 - 4)$$

where the V_{fluid} can also be calculated by subtracting V_{tot} with V_p . Hence,

$$V_{fluid} = \frac{4}{3}\pi(r + x)^3 - \frac{4}{3}\pi r^3 \#(A3 - 5)$$

Equating Eq.A3-3 and Eq. A3-5, x can be described as

$$x = r(1 + a)^3 - r \#(A3 - 6)$$

Therefore, the surface area of a particle with a coating thickness can be described as

$$A_p = 4 \pi (r + x)^2 \#(A3 - 7)$$

Substituting Eq. A3-6 into Eq. A3-7

$$A_p = 4 \pi r^2 (1 + a)^{\frac{2}{3}} \#(A3 - 8)$$

If n_p is the total number of particles, the total surface area of all particles (before they coalescence due to saliva bonding) can be described as

$$4\pi(1 + a)^{\frac{2}{3}} \sum_i^{n_p} r_i^2 \#(A3 - 9)$$

For the sintering mechanism, a linear relationship was proposed between the surface area and the packing density where the surface area declines as the density increases (German, 2016). Thus, the surface area of a bolus can be described as

$$A_{ob} = b\phi + c \#(A3 - 10)$$

Assuming that the initial packing density is ϕ_E , the initial surface area of the bolus is therefore

$$SA_E = b\phi_E + c \#(A3 - 11)$$

Assuming that the bolus forms a perfect sphere when the voidage between particles is 100% saturated with saliva ($\phi = 1$), the surface area of the bolus when at 100% saturation is

$$SA_{sphere} = b + c \#(A3 - 12)$$

Substituting Eq A3-12 into Eq. A3-11

$$SA_E = b(\phi_E - 1) + SA_{sphere} \#(A3 - 13)$$

Rearranging Eq. A3-13, the b constant can be described as

$$b = \frac{SA_{sphere} - SA_E}{1 - \phi_E} \#(A3 - 14)$$

Substituting Eq. A3-14 to Eq.A3-12, the c constant can be described as

$$c = SA_{sphere} - \frac{SA_{sphere} - SA_E}{1 - \phi_E} \#(A3 - 15)$$

Substituting Eq. A3-14 and Eq. A3-15 to Eq. A3-10, the surface area of the bolus can be described as

$$A_{Ob} = \frac{SA_{sphere} - SA_E}{1 - \phi_E} (\phi - 1) + SA_{sphere} \#(A3 - 16)$$

The initial surface area of the bolus, SA_E , can be estimated using equation A3-9. Thus,

$$SA_E = 4 \pi (1 + a)^{\frac{2}{3}} \sum_i^{n_p} r_i^2 \#(A3 - 17)$$

where

$$a = \frac{\varepsilon}{1 - \varepsilon} \cdot S \#(A3 - 18)$$

The total volume of particles can be described as below

$$V_{ptotal} = \frac{4}{3} \pi \sum_i^{n_p} r_i^3 \#(A3 - 19)$$

The total volume of the bolus is therefore

$$V_{bolus} = \frac{V_{ptotal}}{1 - \varepsilon} \#(A3 - 20)$$

If R is the radius of the bolus, the volume of the bolus can also be described as

$$V_{bolus} = \frac{4}{3} \pi R^3 \#(A3 - 21)$$

Equating Eq.A3-20 and Eq.A3-21, the radius of the bolus can be calculated as follows

$$R = \left(\frac{\sum_i^{n_p} r_i^3}{1 - \varepsilon} \right)^{\frac{1}{3}} \#(A3 - 22)$$

Therefore, the surface area of the bolus when at 100% saturation (assuming spherical shape) is

$$SA_{sphere} = 4 \pi R^2 \#(A3 - 23)$$

Finally, the packing density of the bolus in Equation A3-16 can be estimated as described below

$$\emptyset = (1 - \varepsilon) + S \cdot \varepsilon \#(A3 - 24)$$

where the saturation of the bolus, S is calculated using the equation below

$$S = \frac{V_{opasted} + V_s}{V_{ptotal}} \cdot \left(\frac{1 - \varepsilon}{\varepsilon} \right) \#(A3 - 25)$$

Appendix A4 – Comparison between fitted D-values and pasted fraction which represents the PSD against experimental data for the remaining four subjects (A1, A2, A3, A5)

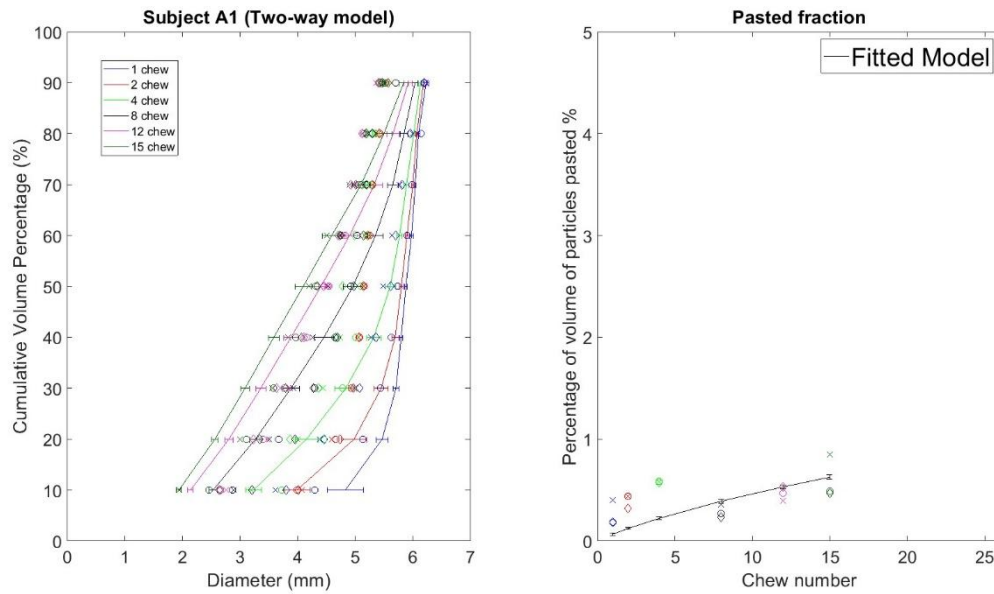


Figure A4-1. Best-fit model against experimental data for Subject A1. Two-way interaction model was used as the selection model. The plot on the left of the image is the comparison of the d-values of the best fit model and the experimental data whereas the plot on the right shows the comparison of the pasted fraction values. The error-bar of the model is the standard deviation of 50 simulations. All 3 replicates of the measured data (three different markers) were plotted for comparison.

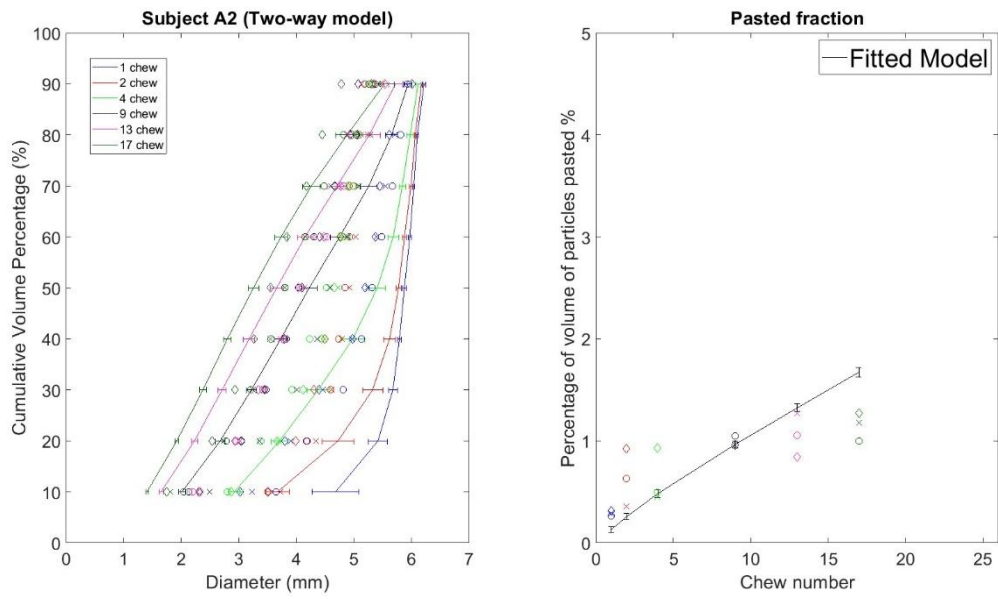


Figure A4-2.: Best-fit model against experimental data for Subject A2. Two-way interaction model was used as the selection model. The plot on the left of the image is the comparison of the d-values of the best fit model and the experimental data whereas the plot on the right shows the comparison of the pasted fraction values. The error-bar of the model is the standard deviation of 50 simulations. All 3 replicates of the measured data (three different markers) were plotted for comparison.

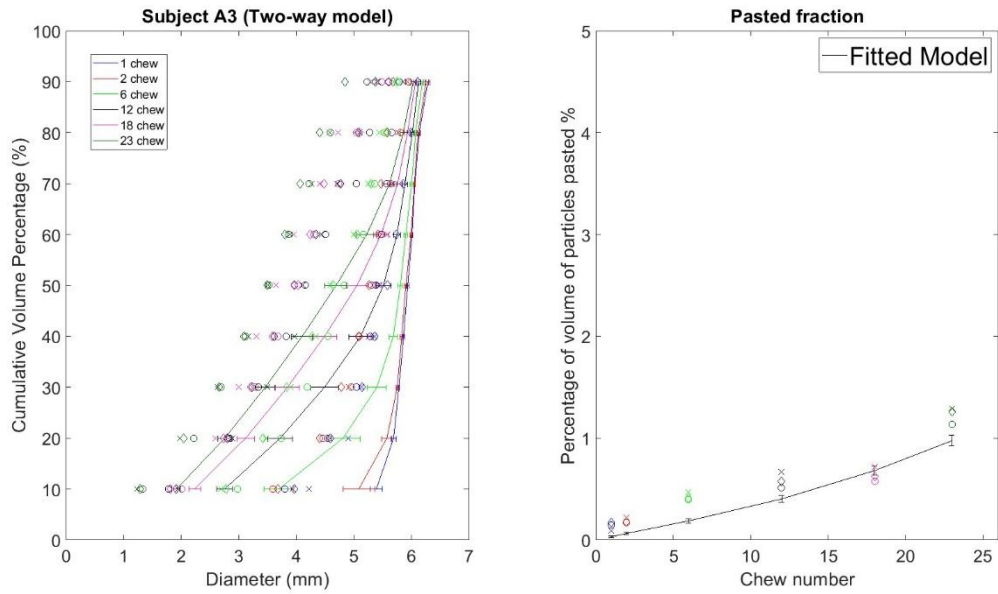


Figure A4-3. Best-fit model against experimental data for Subject A3. Two-way interaction model was used as the selection model. The plot on the left of the image is the comparison of the d-values of the best fit model and the experimental data whereas the plot on the right shows the comparison of the pasted fraction values. The error-bar of the model is the standard deviation of 50 simulations. All 3 replicates of the measured data (three different markers) were plotted for comparison.

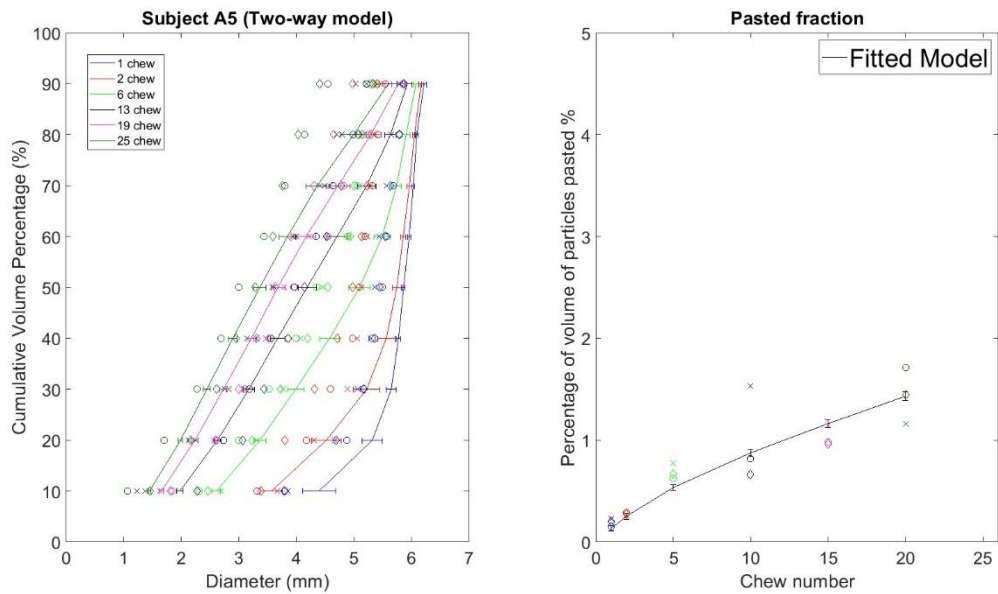
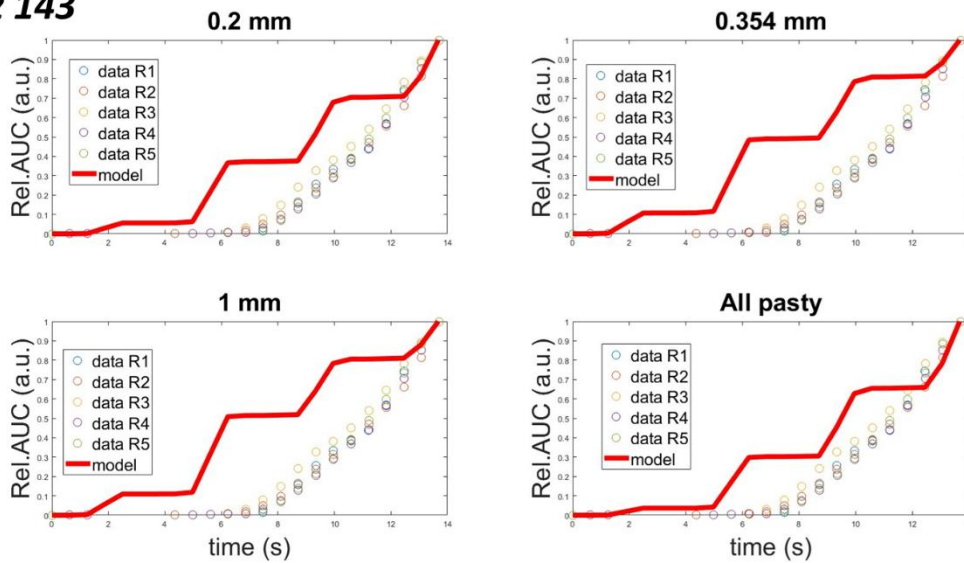


Figure A4-5. Best-fit model against experimental data for Subject A5. Two-way interaction model was used as the selection model. The plot on the left of the image is the comparison of the d-values of the best fit model and the experimental data whereas the plot on the right shows the comparison of the pasted fraction values. The error-bar of the model is the standard deviation of 50 simulations. All 3 replicates of the measured data (three different markers) were plotted for comparison.

Appendix A5 – Comparison between model predictions (coupled integrated selection –breakage and aroma release model) against experimental data for the remaining four subjects (A1, A2, A3, A5)

a. m/z 143



b. m/z 103

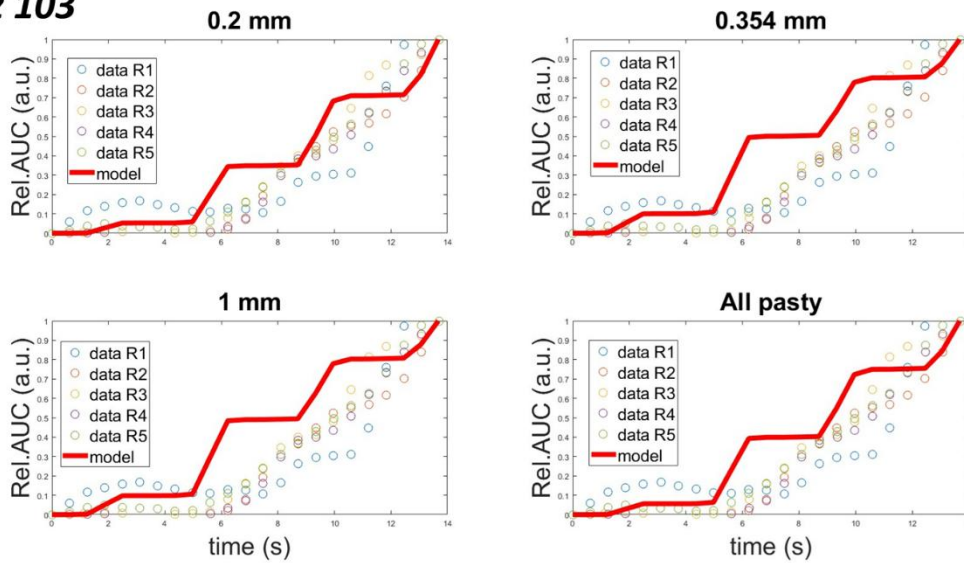
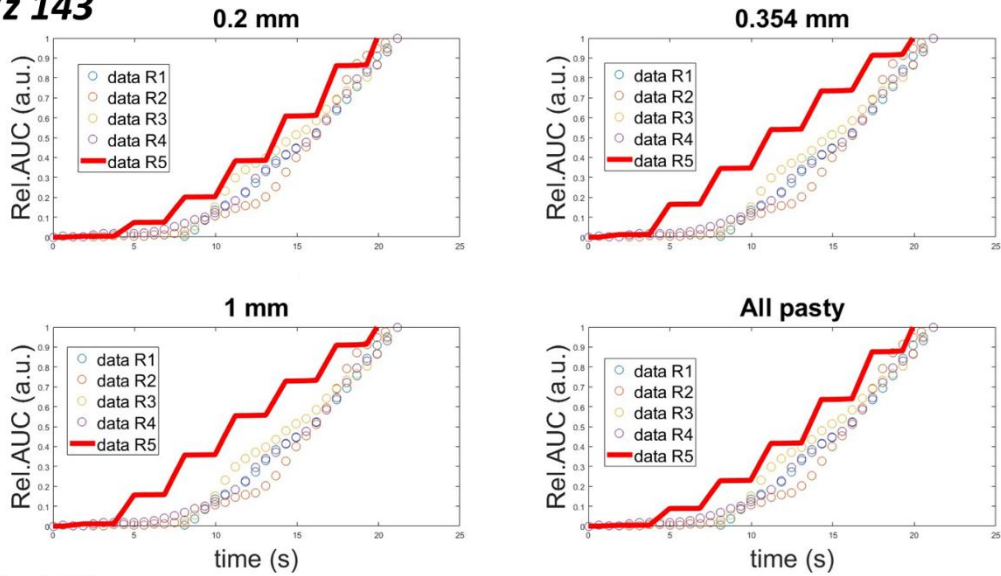


Figure A5-1. Model prediction against experimental data of 2-nonanone (m/z 143) and ethyl propanoate (m/z 103) for Subject A1. Rel. AUC refers to the relative cumulative area under the curve, where the cumulative area under the curve was normalised against the total area under the curve. The model was also predicted using different pasted size threshold (0.2 mm, 0.354 mm, 1 mm and when all particles were pasted) and compared against the five replicates of the experimental data.

a. m/z 143



b. m/z 103

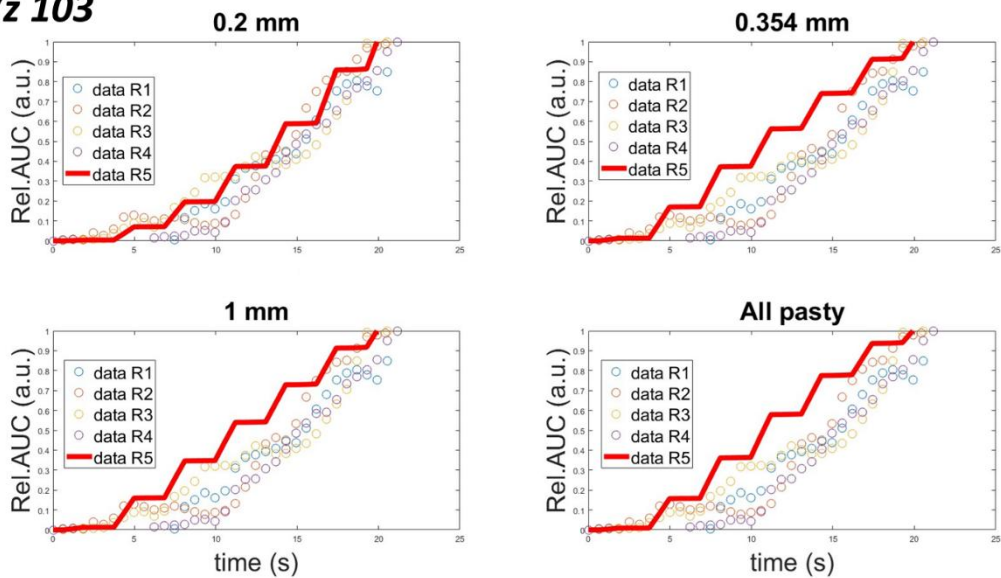
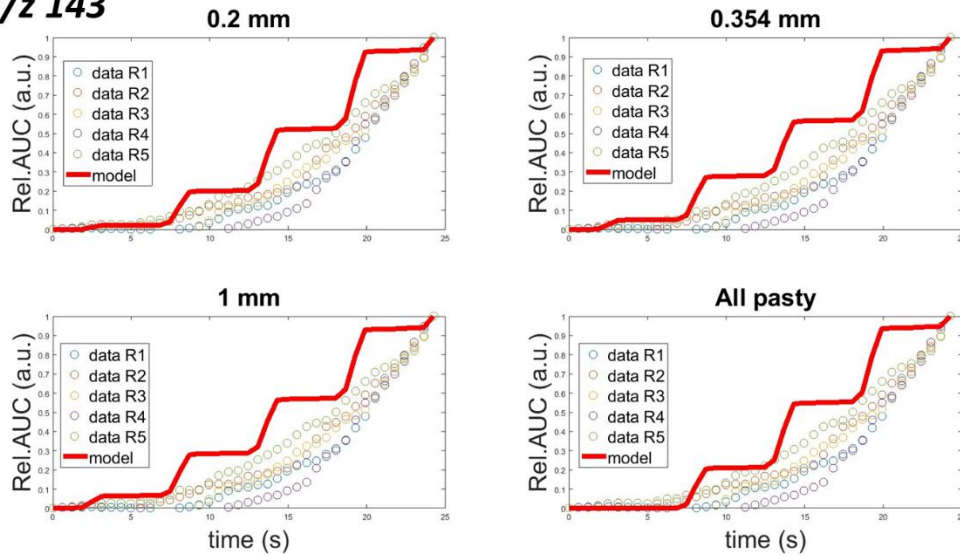


Figure A5-2. Model prediction against experimental data of 2-nonanone (m/z 143) and ethyl propanoate (m/z 103) for Subject A2. Rel. AUC refers to the relative cumulative area under the curve, where the cumulative area under the curve was normalised against the total area under the curve. The model was also predicted using different pasted size threshold (0.2 mm, 0.354 mm, 1 mm and when all particles were pasted) and compared against the five replicates of the experimental data.

a. m/z 143



b. m/z 103

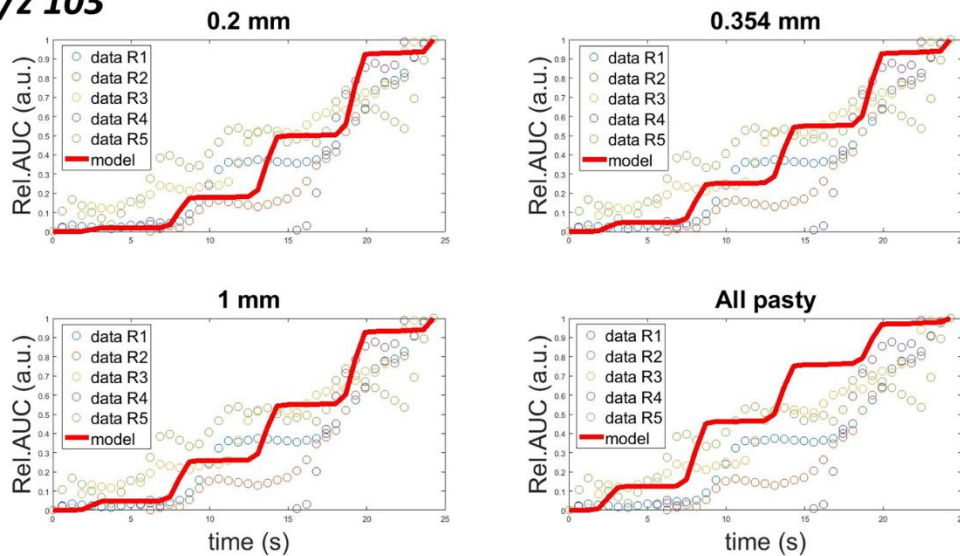


Figure A5-3. Model prediction against experimental data of 2-nonanone (m/z 143) and ethyl propanoate (m/z 103) for Subject A3. Rel. AUC refers to the relative cumulative area under the curve, where the cumulative area under the curve was normalised against the total area under the curve. The model was also predicted using different pasted size threshold (0.2 mm, 0.354 mm, 1 mm and when all particles were pasted) and compared against the five replicates of the experimental data.

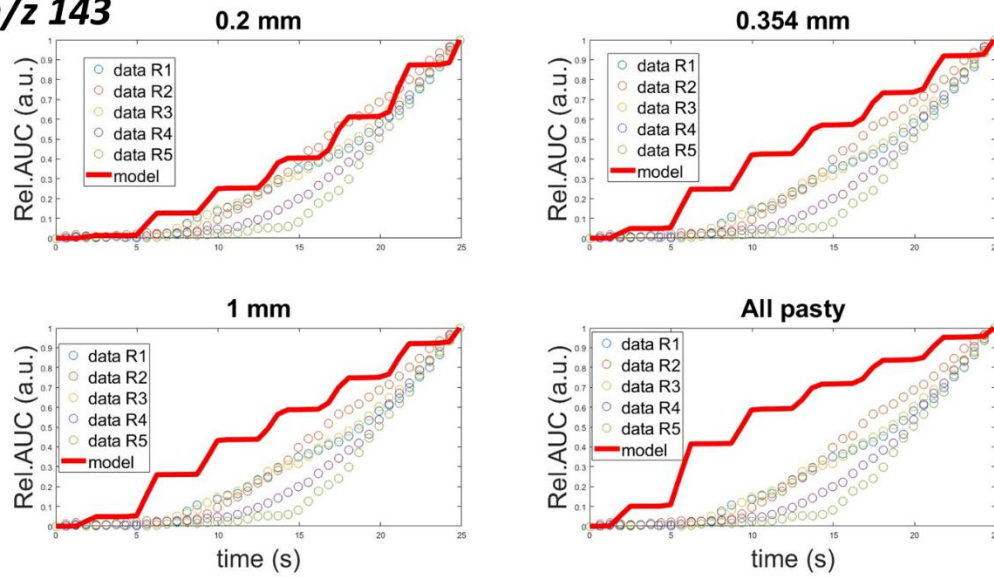
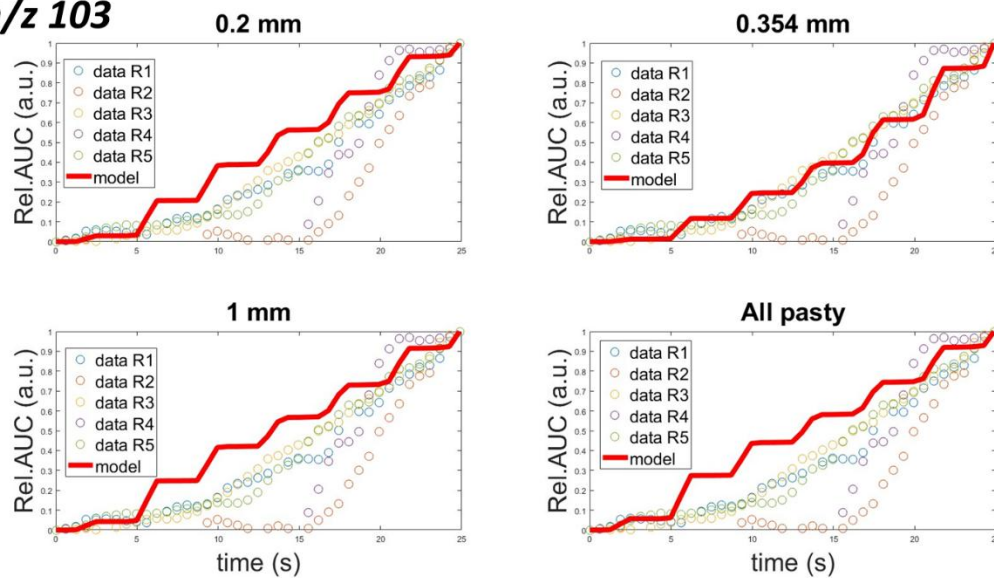
a. m/z 143**b. m/z 103**

Figure A5-4. Model prediction against experimental data of 2-nonanone (m/z 143) and ethyl propanoate (m/z 103) for Subject A5. Rel. AUC refers to the relative cumulative area under the curve, where the cumulative area under the curve was normalised against the total area under the curve. The model was also predicted using different pasted size threshold (0.2 mm, 0.354 mm, 1 mm and when all particles were pasted) and compared against the five replicates of the experimental data.

Appendix A6– Subject’s physiological input parameters

Table A6-1 defines the subject’s physiological input parameters used in the model.

Table A6-1 Subject’s physiological input values used in the model

Symbol	Unit	Description	Reference value	Range of variation	Source
A_O	m^2	Total area in oral cavity	1.2×10^{-2}	1.2×10^{-3} - 2.3×10^{-2}	(Déléris et al., 2016; Doyennette et al., 2014)
A_F	m^2	Total area of the pharynx	6.5×10^{-3}	3.3×10^{-3} - 1.3×10^{-2}	(Déléris et al., 2016; Doyennette et al., 2014)
A_{Oam}	m^2	air/lubricated mucosa contact area in the oral cavity	$= 10^{-1} \times A_O$	$10^{-1} \times A_O - A_O$	(Déléris et al., 2016)
A_{Fam}	m^2	air/lubricated mucosa in the pharynx	$= 10^{-1} \times A_F$	$= 10^{-1} \times A_F - A_F$	(Déléris et al., 2016)
A_{Fas}	m^2	air/saliva contact area in the pharynx	$= A_F - A_{Fam}$	/	(Déléris et al., 2016)
A_{Nam}	m^2	air/lubricated mucosa in the nasal cavity	1.5×10^{-2}	7.5×10^{-4} - 3×10^{-2}	(Déléris et al., 2016)
e_{Oam}	m	Thickness of wetted mucosa in the oral cavity	5×10^{-5}	5×10^{-6} - 5×10^{-4}	(Déléris et al., 2016)
e_{Fam}	m	Thickness of wetted mucosa in the pharynx	5×10^{-5}	5×10^{-6} - 5×10^{-4}	(Déléris et al., 2016)
e_{Nam}	mm	Thickness of mucosa in nasal cavity	5×10^{-5}	5×10^{-6} - 5×10^{-4}	(Déléris et al., 2016)
F_{breath}	number of cycles/s	breathing frequency	0.26	0.25-0.33	Experimental values
$f_{r_{chew}}$	number of chews/s	chewing frequency	1.3	1.2-1.4	Experimental values
$f_{r_{opening}}$	occurrence number/s	opening frequency of the velopharynx	$= F_{breath}$ or $f_{r_{chew}}$	/	(Doyennette et al., 2014)
V_c	m^3	current breath volume	8×10^{-4}	8×10^{-4} - 1.6×10^{-3}	(Déléris et al., 2016; Doyennette et al., 2014)
V_{Osrest}	m^3	volume of saliva at rest in the oral cavity	3.6×10^{-7}	1×10^{-7} - 8.7×10^{-7}	Experimental values

V_{Om}	m^3	volume of mucosa in the oral cavity	$= e_{Oam} \times A_{Oam}$	/	(Délérís et al., 2016)
V_{Oamean}	m^3	volume of air in the oral cavity	6.4×10^{-5}	5.7×10^{-5} - 6.8×10^{-5}	Experimental values
V_{Fa}	m^3	volume of air in the pharynx	3.4×10^{-5}	2.9×10^{-5} - 4.2×10^{-5}	Experimental values
V_{Fs}	m^3	volume of saliva in the pharynx	2×10^{-7}	2×10^{-7} – 4×10^{-7}	(Délérís et al., 2016)
V_{Fm}	m^3	volume of mucosa in the pharynx	$= e_{Fam} \times A_{Fam}$	/	(Délérís et al., 2016)
V_{Na}	m^3	volume of air in the nasal cavity	1.45×10^{-5}	1.03×10^{-5} - 1.95×10^{-5}	Experimental values
V_{Nm}	m^3	volume of mucosa in the nasal cavity	$= e_{Nam} \times A_{Nam}$	/	(Délérís et al., 2016)
Q_{Os}	m^3/s	saliva flow rate in the oral cavity	4×10^{-8}	1.8×10^{-8} - 1.8×10^{-8}	Experimental values
$t_{swallow}$	s	time taken to swallow	20	12-20	Experimental values
n_{chews}		number of chews required to reach swallow point	28	15-38	Experimental values

Appendix A7– Physico-chemical parameters

Table A7-1 defines the physico-chemical parameters used in the model.

Table A7-1: Physico-chemical parameters used in the model.

Symbol	Unit	Description	Reference value	Range of variation	Source
k_{Os}	m/s	mass transfer coefficient in the saliva phase in the oral cavity	10^{-6}	10^{-8} - 10^{-4}	(Déléris et al., 2016)
k_{Fs}	m/s	mass transfer coefficient in the saliva phase in the pharynx	10^{-6}	10^{-8} - 10^{-4}	(Déléris et al., 2016)
k_{Om}	m/s	mass transfer coefficient in the lubricated mucosa in the oral cavity	10^{-6}	10^{-8} - 10^{-4}	(Déléris et al., 2016)
k_{Fm}	m/s	mass transfer coefficient in the lubricated mucosa in the pharynx	10^{-6}	10^{-8} - 10^{-4}	(Déléris et al., 2016)
k_{Nm}	m/s	mass transfer coefficient in the lubricated mucosa in the nasal cavity	10^{-6}	10^{-8} - 10^{-4}	(Déléris et al., 2016)
K_{Oas}		air/saliva partition coefficient in the oral cavity at 37°C	12.9×10^{-3} (Ethyl propanoate) 9.7×10^{-3} (2-nonanone)	/	(Déléris et al., 2016)
K_{Osp}		saliva/product(rice) partition coefficient in the oral cavity at 37°C	2.14 (Ethyl propanoate) 2.5×10^{-1} (2-nonanone)	/	Experimental values
K_{Oam}		air/lubricated mucosa partition coefficient in the oral cavity	10^{-3}	10^{-5} - 10^{-1}	(Déléris et al., 2016)
K_{Fas}		air/saliva partition coefficient in the pharynx	5×10^{-3}	5×10^{-4} - 5×10^{-2}	(Déléris et al., 2016)
K_{Fam}		air/lubricated mucosa partition coefficient in the pharynx	10^{-3}	10^{-5} - 10^{-1}	(Déléris et al., 2016)
K_{Nam}		air/lubricated mucosa partition coefficient in the	10^{-3}	10^{-5} - 10^{-1}	(Déléris et al., 2016)

C_{Op}	kg/m ³	nasal cavity aroma concentration in the product (rice) in the oral cavity	1.5 x 10 ⁻⁴ (ethyl propanoate) 4.8 x 10 ⁻³ (2- nonanone)	/	Experimental values
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