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Article

In Vivo Aroma Release and Dynamic Sensory Perception of Composite Foods

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ABSTRACT: Condiments such as spreads, dressings, or sauces are usually consumed together with carrier foods such as breads or vegetables. Dynamic interactions between condiments and carriers occur during consumption, which can influence aroma release and perception. This study investigated *in vivo* aroma release (PTR-MS) and dynamic sensory perception (time−intensity) of mayonnaises spiked with lemon aroma (limonene, citral). Mayonnaises were assessed without and with carrier foods (bread, potato). When different mayonnaises were consumed and assessed alone, aroma release and intensity perception were positively correlated. Interestingly, when mayonnaises were combined with carriers, aroma release and perception were no longer positively correlated. Addition of carriers increased release of limonene and citral into the nasal cavity during consumption but decreased perceived aroma intensity of condiments. The increase in aroma release induced by the carriers can be explained by differences in oral processing behaviors and by the increased surface area of mayonnaise-carrier combinations. Carrier addition is likely to modulate aroma perception of composite foods by cross-modal texture−aroma interactions. This work demonstrates that not only physicochemical characteristics of foods but also cross-modal interactions play a role in influencing flavor perception of composite foods.

KEYWORDS: *aroma release, aroma perception, food texture, food flavor, PTR-MS, time-intensity*

INTRODUCTION

In addition to consumption context, physiological and social-economic status, availability, and many other factors, food sensory properties have a functional role in food choice, food acceptance, and energy intake regulation.[1] Understanding which food properties contribute to sensory perception of foods is therefore of utmost importance to influence consumer and food intake behavior. Sensory perception is multidimensional and encompasses aroma, taste, and texture perception, which are well known to interact with each other through cross-modal interactions.[2−4] Food structural transitions during consumption influence *in vivo* aroma release and thereby sensory perception.[5,6] Aroma compounds are released from the food matrix and reach the olfactory receptors located in the human nasal cavity (retro-nasal pathway) throughout consumption.[−11] Aroma release is a rather complex process, which is influenced by food composition, food structure, and dynamic changes during oral processing.[7−9] To better understand aroma release and perception, it is essential to couple dynamic sensory methods (i.e., time−intensity profiling (TI)) simultaneously with instrumental methods to follow in *in vivo* real-time aroma release from the food during consumption (i.e., in-nose proton transfer reaction mass spectrometry (PTR-MS)).

In many meals, different foods are combined. Condiments such as spreads, dressings, or sauces are often used to complement or enhance the flavor of bland carrier foods to increase sensory pleasure.[10] Here, we refer to the combination of condiments with carriers as composite food. When different foods are consumed together, dynamic interactions between the foods occur during consumption, as they are mixed in the mouth and continuously broken down by mastication. Yet, factors contributing to aroma release and sensory perception of composite foods received surprisingly little attention in the field of food science.

Addition of carrier foods to condiments is generally known to decrease overall perceived flavor intensity of condiments.[11−14] Addition of bread or carrots to mayonnaises has been shown to reduce perceived intensities of several mayonnaise-related flavor attributes.[14] The mechanisms underlying the reduction in flavor intensity upon carrier addition are not known. To the best of our knowledge, it has not been investigated to what extent the decreased flavor perception of composite foods is due to physicochemical interactions between carrier and condiment leading to a lower release of aroma compounds into the nasal cavity due to differences in oral processing behaviors caused by the addition of solid carriers leading to changes of aroma release kinetics or due to unconscious, perceptual cross-modal interactions.[15]

Several studies investigated the effects of food properties (composition, rheological properties) on *in vivo* aroma release and perception. While in many cases, aroma release and perception were positively correlated, i.e., an increase of *in vivo* aroma release led to an increase in aroma perception,[16,17] other

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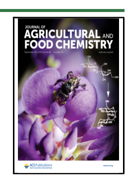

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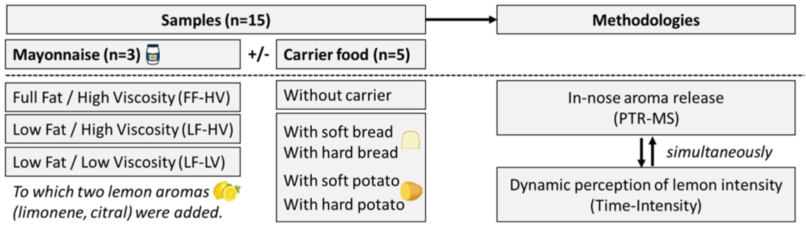

Figure 1. Experimental design outlining the approach. Mayonnaises varying in fat content (high/low) and viscosity (high/low) were tested without carrier food (n = 3) and together with carrier foods differing in moisture absorption capacity (bread, potato) and hardness (hard/soft). Mayonnaises were spiked with two lemon aroma compounds (limonene, citral), which allowed characterization of in-nose aroma release and dynamic lemon intensity perception.

studies observed discrepancies between aroma release and perception, i.e., an increase of *in vivo* aroma release was accompanied by a decrease in aroma perception.[16,18−21] In the case of chewing gum, mint aroma perception followed sucrose release rather than menthone release.[22] Food texture has also been shown to influence aroma perception by cross-modal interactions.[23,24] Cross-modal interactions between food texture, taste, and aroma play a key role in multisensory flavor perception.[3,4,15] Currently, it is not well understood how addition of accompanying foods changes aroma release and perception of single foods. In this work, we refer to composite foods as foods that are consumed together within one bite and are composed of one solid carrier food and one condiment.

This study aimed at understanding the relationships between *in vivo* in-nose aroma release and dynamic aroma perception of composite foods. As condiment, we used mayonnaises differing in fat content (high, low) and viscosity (high, low) to understand the effect of different physicochemical properties on in-nose aroma release and perception. As carriers, we used bread and potato, as examples of different carriers varying in moisture absorption capacity. Carrier texture (soft, hard) was varied to investigate the role of carrier texture in aroma release and perception of condiments (Figure 1). We monitored in-nose release of two aroma compounds differing in hydrophobicity (citral and limonene) using *in-vivo* nose-space PTR-MS coupled with time−intensity profiling (TI) for capturing dynamic flavor perception.

MATERIALS AND METHODS

Samples. Three different mayonnaises varying in fat content and viscosity were prepared, namely, full fat/high viscosity (FF-HV; 69% w/w oil; Calvé De echte, Unilever, The Netherlands), low fat/high viscosity (LF-HV), and low fat/low viscosity (LF-LV) mayonnaises. For the low fat mayonnaises, 2.5 or 1.0% xanthan in water solution (E415, Pit&Pit bvba, Belgium) was gradually spooned into the FF-HV mayonnaise following a 1.6:1.0 weight ratio to create the LF-HV or LF-LV mayonnaises (26.5% w/w oil), respectively. To limit differences between the samples in the diffusion of hydrophobic aroma compounds through the oil droplet interface, we prepared emulsions with the same oil droplet sizes ($D_{3,2}$ = 10.9 ± 2, 7.9 ± 1 and 7.1 ± 1 μm (mean ± SD) for FF-HV, LF-HV, LF-LV, respectively). Two lemon aroma compounds varying in hydrophobicity, citral (M_w = 152 g/mol , log P = 2.76, 1 mg/g mayonnaise) and limonene (M_w = 136 g/mol , log P = 4.2, 1 mg/g mayonnaise), were gently mixed into the mayonnaises using a spatula. The addition of these compounds made the mayonnaise easier to track during the proton transfer reaction mass spectrometry (PTR-MS) analysis and easier to be perceived by the participants. The two compounds were chosen based on their aroma, their different physical/chemical properties and their masses after some preliminary measurements on volatile organic compounds (VOCs) emissions on both mayonnaises and carriers to verify interferences. Mayonnaises were served at a weight of 2 g.

Mayonnaises were assessed alone and in combination with different carrier foods. Two commercial carrier foods were used, namely, bread (Plaisir de mie toastbrood, Jacquet, France) and potatoes (Waxy potatoes, Albert Heijn, The Netherlands). Bread and potatoes were selected based on their difference in water absorption capability. Bread has been shown to absorb moisture during consumption, whereas potatoes hardly absorb moisture during mastication.[64] Mayonnaise on bread represents a simplified model food for sandwiches and mayonnaise on potato represents a simplified model food for salads. Bread cubes without crust (35 × 35 × 8 mm) were served fresh and oven-dried for 40 min at 100 °C (Venti-line, VWR) to obtain two bread samples with varying properties. Peeled potato cubes (30 × 12 × 12 mm) were cooked sous-vide at 90 °C for 15 and 45 min to obtain two potato samples with varying properties. Carrier-mayonnaise combinations were prepared just before serving in order to minimize moisture transfer of the mayonnaises into the carriers before consumption.

Table 1 presents an overview of the composition and product properties of the mayonnaises (fat content, viscosity, oil droplet size)

Table 1. Product Properties of Mayonnaises Varying in Fat Content and Viscosity (a) and the Carrier Foods Bread (B) and Potato (C) Varying in Preparation Methods (Mean ± SD)

(A) mayonnaise properties	full fat/high viscosity (FF-HV)	low fat/high viscosity (LF-HV)	low fat/low viscosity (LF-LV)
fat content (w/w %)	70	27	27
viscosity at 1 s^{-1} (Pa·s)	84 ± 19	73 ± 12	11 ± 3
viscosity at 10 s^{-1} (Pa·s)	13 ± 3	10 ± 1	2 ± 1
viscosity at 100 s^{-1} (Pa·s)	2.1 ± 0.4	1.3 ± 0.2	0.3 ± 0.1
$D_{3,2}$ (μm)	10.9 ± 2	7.9 ± 1	7.1 ± 1
(B) bread properties	fresh bread	dried bread	
compression force (N)	5 ± 2	100 ± 38	
A_w	0.92 ± 0.02	0.20 ± 0.16	
(C) potato properties	soft potato	semi-hard potato	
fracture stress (kPa)	40 ± 17	198 ± 132	
A_w	0.98 ± 0.01	0.99 ± 0.00	

and the carrier foods (firmness, water activity). The mayonnaises' properties were measured each morning before data collection (n = 10 days of data collection), and carrier properties were measured for each new preparation batch (n = 4 batches) to ensure that samples were stable over the data collection period. To determine the viscosity of the mayonnaises, mayonnaises were sheared at shear rates ranging from 1 to 1000 s^{-1} after a resting period of 5 min using a rheometer (MCR 301 Rheometer, Anton Paar Benelux BVBA, Belgium) equipped with an Inset I-PP50/SS plate and a CP50-1 cone. The oil droplet size of the mayonnaises (D3,2) was measured by light scattering (Mastersizer 2000, Malvern Instruments) in triplicate using the refractive index of sunflower oil (1.469). To determine the

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firmness of the carrier foods, uniaxial compression tests were performed with a Texture Analyzer (TA.XT Plus, Stable Micro Systems, United Kingdom) fitted with a 50 kg load cell, a cylindrical plate with a diameter of 100 mm, and a constant speed of 1 mm/s. Bread samples were compressed until 20% strain, and the mean force needed to compress the bread samples was calculated. Potato samples were compressed until 50% strain, and the mean fracture stress of the potatoes was calculated. The water activity of the carrier foods was measured using a LabMaster aw (Novasina).

Participants. A group of 14 Caucasian, European females (23 ± 3 years) participated in the study. As the focus of the study was to understand aroma release and perception of composite foods varying in properties, a relatively homogeneous group of participants was selected to minimize inter-individual variation. Participants were selected based on their mechanically stimulated saliva flow rate (1.4 ± 0.6 g/min, mean ± SD), size of the oral cavity (73.5 ± 10.4 g water, mean ± SD), and natural eating time of the samples (16 ± 5 s, mean ± SD),[25] which were assessed during one selection session of 1 h. In addition, they had non-smoking habits (self-reported), good dental health (self-reported), and were consumers of mayonnaise, bread, and potato on a regular basis. All participants gave written informed consent, completed the study, and received financial compensation for participation.

Chewing Protocol. Participants were instructed to follow a chewing protocol to minimize the influence of individual differences in mastication behavior on aroma release and perception throughout consumption. Participants were instructed to consume each sample within one bite and to swallow after 20 s of consumption (timer was shown on the screen). In the case of mayonnaises alone, they were instructed to swirl samples in their mouth. In the case of mayonnaise-carrier combinations, they were instructed to chew the sample with a frequency of 1 chew/s (i.e., approximately 20 chews) using a metronome and the timer on the screen. Furthermore, participants were asked to raise their hand each time they swallowed, which was recorded by the researcher. In addition, they were asked to keep their mouth closed during all the evaluations.

Nose-Space Analysis, Data Extraction, and Peak Selection. *In vivo* aroma release was measured using a commercial PTR-MS instrument (Ionicon Analytik GmbH, Innsbruck, Austria) equipped with a time of flight and quadrupole ion guide (PTR-QiTOF). H_3O^+ was used as the precursor ion, and the ionization conditions were the following: 1000 V drift voltage, 60.0 °C drift temperature, 3.8 mbar drift pressure, resulting in an E/N ratio of 133 Td. Acquisition was set to 1 spectrum per second. Sampling was carried out via a heated (95 °C) inlet tube with an inlet flow of 45.02 sccm. The mass resolution (m/Δm) was at least 5000.

The nose-space experimental setup was adapted from previous PTR-MS in-nose studies.[26] For each measurement, laboratory air was sampled for 20 s. After that, participants were asked to insert two Teflon tubes (diameter: 6.8 mm, length: 6.4 cm, connected to the heated inlet tubes) in the nose. They were asked to breathe normally through their nose, and participants' breath was sampled for 60 s. Then they consumed the samples for 20 s. After swallowing the sample, participants kept on breathing for 90 s. This led to a total sampling time of 190 s. Samples were assessed in triplicate by each participant.

PTR-MS data were treated with TOFO office software (Department of Food Quality and Nutrition, Edmund Mach Foundation) as described in Cappellin *et al.* (2011). A total of 247 mass peaks were extracted from 20 to 250 *m/z*, and in-nose concentration was calculated. From that, 73 peaks were selected for the further analysis based on pilot experiment reports, the literature, and the high concentration of the release curve for the relevant aroma compounds of citral, limonene, mayonnaise, food carriers, and the exhaled gases from participants. In the work, only mass peaks corresponding to the two lemon aroma compound are considered: *m/z* 138.139 and 153.131 tentatively identified as the limonene isotope ($^{13}CC_9H_{16}H^+$) and citral ($C_{10}H_{16}OH^+$) were chosen as representative examples, respectively. The isotope was chosen due to the high concentration of *m/z* 137.132 that in some measurements was saturating the detector. *m/z* 135.119 ($C_{10}H_{15}^+$) was chosen as the main fragments of citral.

For each mass peak, a release curve was obtained by plotting between peak concentration (ppbV) and time (s). Each release curve was divided into four time separate windows: lab air session (1−20 s), breathing (21−80 s), mastication session (81 s to first swallowing point), and post-swallowing session (first swallowing point until 195 s). Each part of the curve was averaged for the entire panel and superposed to create an average release curve for each sample. For comparing the different mayonnaise aromas released and the food carrier interactions, the baseline (signal before the sample was ingested) was then subtracted, and three main parameters were extracted from each individual release curve: the area under the curve (AUC_R), the maximum concentration (I_{max}_R), and the time to reach the maximum concentration (T_{max}_R).

Time-Intensity (TI) Sensory Methodology. Dynamic lemon aroma intensity of mayonnaises was determined using the time−intensity (TI) methodology. Participants were instructed to place the sample in the mouth and simultaneously click the start button. Then, they continuously scored the lemon intensity over time by moving the cursor horizontally on a 100 mm unstructured line scale anchored from not at all to very (Eye Question software, version 4.11.19). The total duration of the evaluation was set at 110 s, meaning that participants evaluated lemon intensity during chewing (approximately 20 s) and after the sample had been swallowed (approximately 90 s). Intensity scores were recorded with an interval time of 500 ms. From the time-intensity profiling, the total area under the curve (AUC_S), the maximum perceived intensity (I_{max}_S), and the time to maximum intensity (T_{max}_S) were obtained. In the present study, Liu&MacFie standardization was applied to correct for individual signature curves.

Experimental Approach. Participants participated in 10 sessions over a time period of 1 month. Participants were first trained over four sessions of 1 h, after which dynamic aroma perception and *in vivo* aroma release were determined simultaneously by using TI and PTR-MS during the subsequent six sessions of maximum 1.5 h.

During the four training sessions, participants were acquainted with the chewing protocol (training 1 and 2) and the TI methodology (training 3 and 4). The first training started with an introduction to the chewing protocol, after which the participants practiced the protocol. During the second training session, participants were familiarized with the nose tubes used to connect the participants' nasal cavity with the PTR-MS. During this session, participants continued practicing the chewing protocol while having the nose tubes in their nose. The third session was used to introduce the TI methodology to the participants, after which they practiced with the tasting protocol, nose tubes, and TI methodology using mayonnaises spiked with lemon aroma. The fourth session was a pilot experiment, during which they practiced with the tasting protocol, nose tube, and TI methodology for all samples included in the present study.

During the six data collection sessions, participants were requested to not eat, drink, or brush their teeth 2 h before the experiment and to not wear perfume or lotion. All samples were assessed following a 3 × 5 design: three mayonnaises with five carrier conditions (without carrier, with fresh bread, with oven-dried bread, with shortly cooked potato, with long cooked potato). Samples were assessed in triplicate leading to a total of 45 nose-space measurements and sensory analyses for each participant. Each replicate was assessed over two sessions. Within each replicate, samples were presented in a random order following a completely randomized design. Samples were presented with three-digit codes, served on a spoon to facilitate easy intake. Between each sample, participants cleansed their palate for at least 6 min using cold water, hot water, and tongue scrapers to aid the removal of oil from their tongue. No other palate cleansers were used since they might affect the volatile release of follow-up samples.

Statistical Data Analyses. Results were reported as mean values with standard error (n = 14 participants, in triplicate). Outliers (Z score > 3.29 or Z score < −3.29) were removed from the data. The averaged release curves together with their standard error were plotted for each sample for *m/z* 138.139 and 153.131. Average and standard error were chosen instead of median and standard deviation

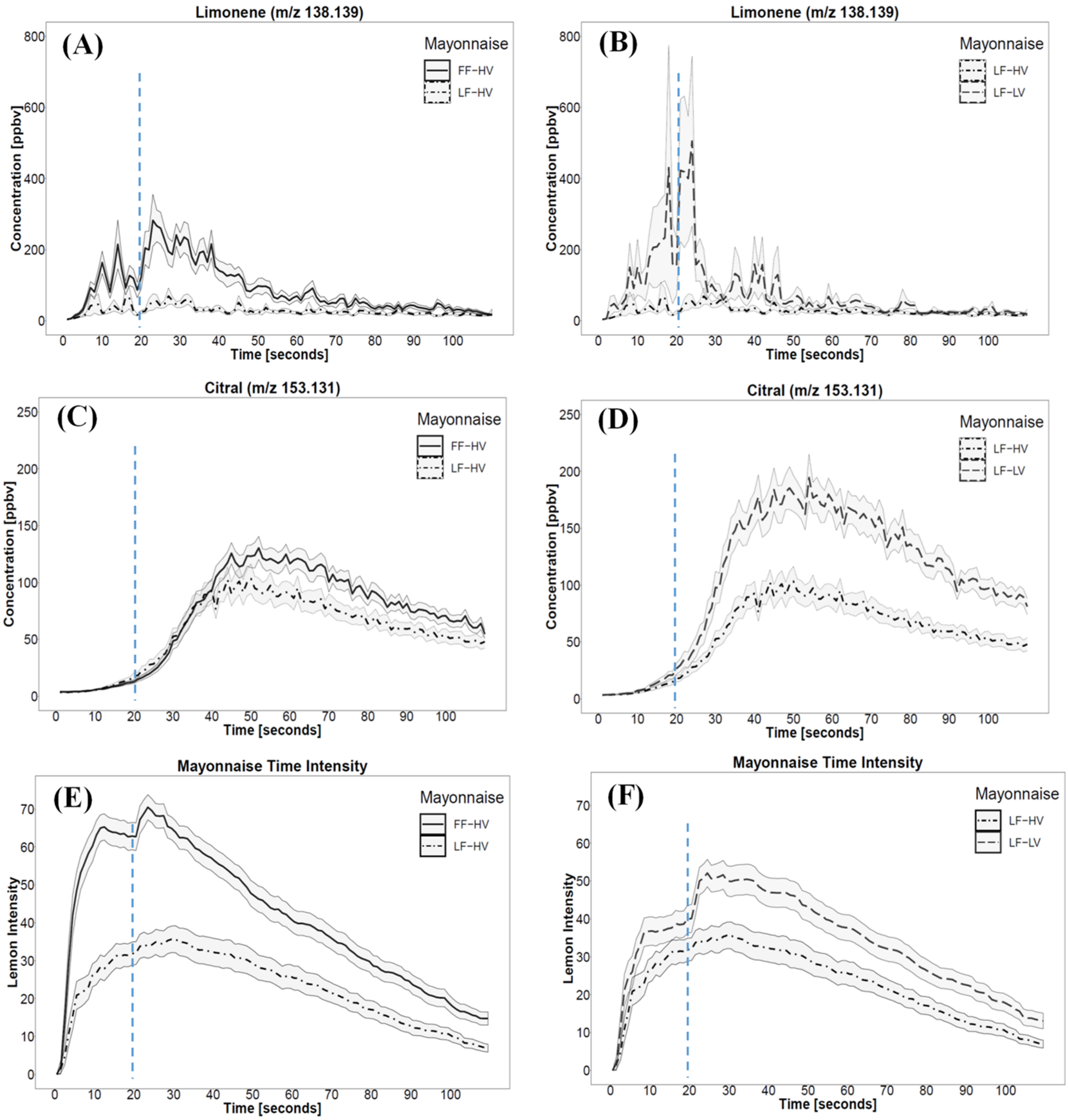

Figure 2. Averaged in-nose limonene release (m/z = 138.139) (A,B), in-nose citral release (m/z = 153.131) (C,D), and lemon intensity perception (E,F) during mastication and after swallowing for mayonnaise varying in viscosity and fat content (n = 14 participants, in triplicate). Mayonnaises differing in viscosity (LF-HV and LF-LV) are presented on the right (B,D,F), and mayonnaises varying in fat content (FF-HV and LF-HV) are presented on the left (A,C,E). The shaded bars represent the standard error of the mean. The moment of swallowing is indicated as dashed lines at 20 s.

to improve the figures' readability. To investigate the effect of mayonnaise properties, linear mixed models were performed on a subset of the data including the data of the single mayonnaises (i.e., without carriers) only. For this analysis, mayonnaise was set as a fixed effect and participant, replicate, serving order, and session were set as random effects using the Lmer package. To investigate the interplay between mayonnaise properties and carrier properties, linear mixed models were performed with mayonnaise, carrier, and mayonnaise:-carrier interaction as fixed effects and participant, replicate, serving order, and session as random effects. This analysis was performed for bread and potato carriers separately. In addition, multiple factor analysis (MFA) was performed on the selected mass peaks from PTR-MS analysis and on the time−intensity data using the FactoMineR package. Only the AUC was used in this case, and data were scaled to unit variance before performing the analysis. R language (RStudio, version 1.0.143) was used to perform all statistical tests. A significance level of $p < 0.05$ was chosen.

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Table 2. Summary of Parameters (Mean ± SE) Describing *In Vivo* Limonene Release, *In Vivo* Citral Release, and Dynamic Lemon Intensity Perception for Mayonnaises Varying in Fat Content (FF = Full Fat, LF = Low Fat) and Viscosity (HV = High Viscosity, LV = Low Viscosity)[a,b]

		mayonnaise		FF-HV				LF-HV				LF-LV			
AUC		*F*	*p*	mean	±	SE		mean	±	SE		mean	±	SE	
AUC_R (ppbV·s)															
limonene	*m/z* 138.139	10.3	<0.001	9617	±	701	*a*	2695	±	407	*b*	8571	±	2003	*a*
AUC_R (ppbV·s)	*m/z* 135.119	25.8	<0.001	4210	±	277	*b*	3228	±	321	*b*	6226	±	424	*a*
citral	*m/z* 153.131	23.9	<0.001	7550	±	538	*b*	6007	±	608	*b*	11,767	±	850	*a*
AUC_S (mm·s)		29.0	<0.001	9198	±	506	*a*	5096	±	489	*c*	7362	±	566	*b*
$\mathbf{I_{max}}$		*F*	*p*	mean	±	SE		mean	±	SE		mean	±	SE	
I_{max}_R (ppbV)															
limonene	*m/z* 138.139	5.9	<0.01	857	±	80	*ab*	269	±	50	*b*	1030	±	271	*a*
I_{max}_R (ppbV)	*m/z* 135.119	26.3	<0.001	88	±	5	*b*	72	±	7	*b*	134	±	8	*a*
citral	*m/z* 153.131	24.8	<0.001	173	±	12	*b*	145	±	14	*b*	267	±	16	*a*
I_{max}_S (mm)		30.6	<0.001	77	±	3	*a*	48	±	4	*c*	62	±	4	*b*
$\mathbf{T_{max}}$		*F*	*p*	mean	±	SE		mean	±	SE		mean	±	SE	
T_{max}_R (s)															
limonene	*m/z* 138.139	3.8	<0.05	28	±	2	*b*	38	±	3	*a*	33	±	3	*ab*
T_{max}_R (s)	*m/z* 135.119	0.0	NS	51	±	2		49	±	2		50	±	3	
citral	*m/z* 153.131	0.3	NS	56	±	2		52	±	2		52	±	2	
T_{max}_S (s)		4.8	<0.05	33	±	3	*b*	39	±	3	*ab*	43	±	3	*a*

[a]Lower case letters: significant differences between mayonnaises varying in fat content and viscosity ($p < 0.05$). [b]The release parameters AUC_R, I_{max}_R, T_{max}_R correspond to the area under the curve, the maximum concentration and time to reach the maximum concentration. The sensory parameters AUC_S, I_{max}_S and T_{max}_S correspond to the total area under the curve, the maximum perceived intensity and the time to reach the maximum perceived intensity.

RESULTS

In-Nose Aroma Release and Dynamic Lemon Perception of Mayonnaises without Carriers: Effect of Viscosity and Fat Content. Dynamic *in vivo* aroma release and dynamic lemon intensity perception of mayonnaises without carrier foods are shown in Figure 2. Table 2 provides a summary of all aroma release (AUC_R, I_{max}_R, and T_{max}_R) and perception (AUC_S, I_{max}_S, and T_{max}_S) parameters. As can be seen from Figure 2A–D, limonene and citral display different release profiles. While limonene was released fast, resulting in a sharp peak (Figure 2A,B), citral was released slowly during consumption and mainly after swallowing (later T_{max}_R, see Table 2) resulting in a later and broader peak (Figure 2C,D). Citral has a higher boiling temperature and lower vapor pressure due to its higher molecular weight and its molecular structure, resulting in a lower volatility than limonene.

Mayonnaise viscosity (LF-HV vs LF-LV) clearly affected in-nose aroma concentrations (Figure 2A,C) and dynamic lemon intensity perception (Figure 2E). *In vivo* limonene release, *in vivo* citral release, and lemon intensity perception decreased with increasing mayonnaise viscosity. For example, in the case of limonene (*m/z* 138.139), AUC_R decreased by 69% and I_{max}_R decreased by 74% with increasing viscosity (Table 2). Congruently, with respect to sensory perception, AUC_S decreased by 31% and I_{max}_S decreased by 23%. The times to reach the maximum concentration and intensity (T_{max}_R, T_{max}_S) were not significantly affected by mayonnaise viscosity.

Mayonnaise fat content (FF-HV vs LF-HV) also affected in-nose aroma concentration (Figure 2B,D) and dynamic lemon intensity perception (Figure 2F). *In vivo* limonene release, *in vivo* citral release, and lemon intensity perception decreased upon fat reduction of mayonnaises from 70 to 27 wt %. For example, AUC_R of limonene (*m/z* 138.139) decreased by 72% and AUC_S decreased by 45% with decreasing fat content. Similar trends were found for the I_{max} values (Table 2). A reduction of fat content slowed down the release of limonene ($p < 0.05$, T_{max}_R), but no significant effect was observed for citral release.

In-Nose Aroma Release and Dynamic Lemon Perception of Mayonnaise with Carrier Foods. Figure 3 shows averaged in-nose limonene release, in-nose citral release, and perceived lemon intensity curves for FF-HV mayonnaise without and with carrier foods. The release and perceived intensity curves of the other two mayonnaises (LF-HV and LF-LV) are provided as supplementary data (Figures S1 and S2), as addition of carriers affected release and perception of the different mayonnaises in a similar way. In-nose limonene and citral release parameters (AUC_R, I_{max}_R and T_{max}_R) and perceived lemon parameters (AUC_S, I_{max}_S and T_{max}_S) of mayonnaises without and with carriers are presented in Table 3. Overall, in-nose limonene and citral release increased with the addition of food carriers, whereas simultaneous lemon intensity perception of mayonnaises decreased. Bread and potato affected aroma release and perception of mayonnaises differently. The results of bread and potato addition are therefore reported separately in the following subsections.

Effect of Bread Addition on In-Nose Aroma Release and Aroma Perception of Mayonnaises. Addition of bread increased in-nose limonene and citral release of mayonnaises, regardless of bread texture (Figure 3A,C and Table 3A). For example, in the case of limonene (*m/z* 138.139), AUC_R increased by 136 and 144% after addition of soft and hard bread, respectively ($p < 0.001$; $p < 0.001$). For citral (*m/z* 135.199 and 153.131), AUC_R increased with addition of bread, ut this effect was only significant for LF-HV mayonnaise. Similar trends were observed for I_{max}_R values.

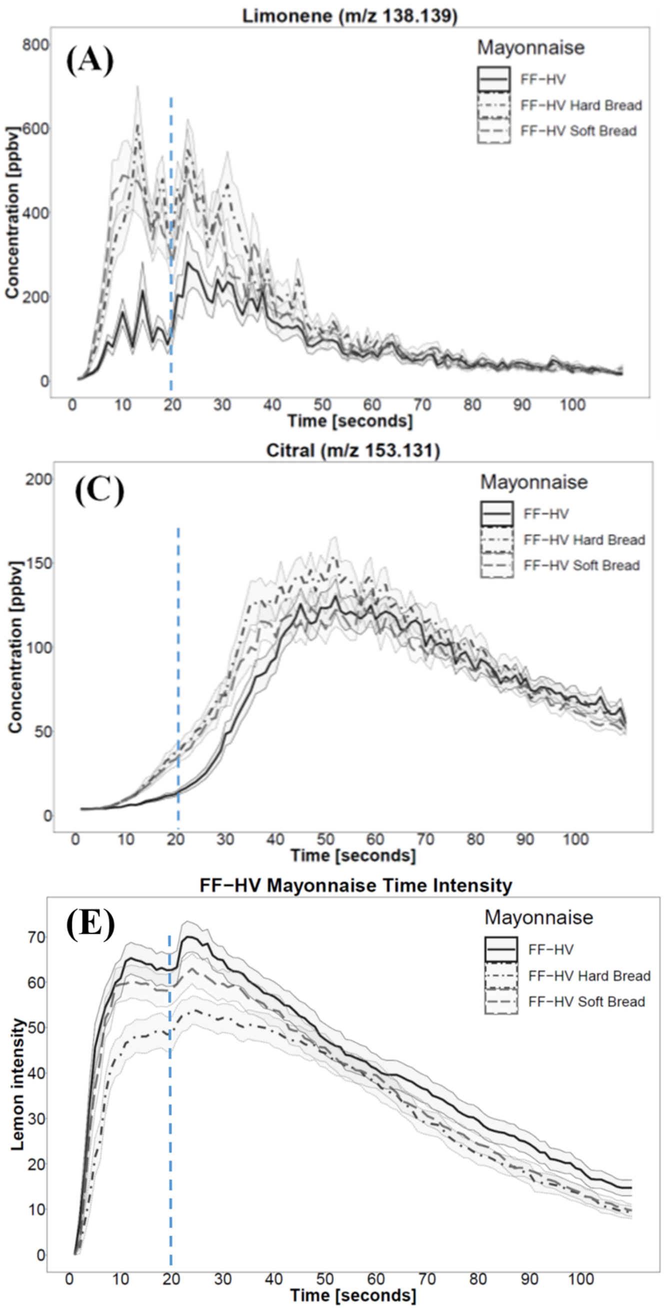

Figure 3. Averaged in-nose limonene release (m/z = 138.139) (A,B), in-nose citral release (m/z = 153.131) (C,D), and lemon intensity perception (E, F) during mastication and after swallowing for mayonnaise without and with different food carriers (n = 14 participants, in triplicate). Mayonnaise (i.e., FF-HV mayonnaise) with bread carriers (soft, hard) is presented on the left (A,C,E), and the mayonnaise with potato carriers (soft, hard) is presented on the right (B,D,F). The shaded bars represent the standard error of the mean. The moment of swallowing is shown as dashed line at 20 s.

Bread texture (soft vs hard) did not affect limonene and citral release concentrations significantly.

The time to reach maximum aroma concentration (T_{max}_R) was affected by the addition of bread, regardless of bread texture (Table 3A). Overall, T_{max}_R was reached earlier for mayonnaise-bread combinations than for mayonnaises consumed without bread. These differences in T_{max}_R were significant for LF-HV but not for FF-HV nor LF-LV.

Addition of bread decreased lemon intensity perception of mayonnaises (Figure 3E, Table 3A). However, the effect of bread on lemon intensity perception was not the same for each mayonnaise (significant mayonnaise:bread interaction). For FF-HV mayonnaise, AUC_S and I_{max}_S were lowered by 11 and 8% with soft bread ($p > 0.05$; $p > 0.05$) and by 21 and 22% with hard bread ($p = 0.003$; $p = 0.001$). For LF-LV mayonnaise, AUC_S and I_{max}_S decreased by 10 and 10% with soft bread ($p > 0.05$; $p > 0.05$) and by 22 and 15% with hard bread ($p = 0.030$; $p > 0.05$). No significant effect was observed for LF-HV. Hence, bread hardness partly affected lemon intensity perception of mayonnaises. T_{max}_S was not significantly affected by the addition of bread.

Effect of Potato Addition on In-Nose Aroma Release and Aroma Perception of Mayonnaises. Addition of potato to mayonnaises increased both limonene and citral

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Table 3. Summary of Parameters (Mean ± SE) Describing *In Vivo* Limonene Release, *In Vivo* Citral Release, and Dynamic Lemon Intensity Perception for Mayonnaises without and with Carrier Foods

(A) Mayonnaise without and with bread

		bread		mayo:bread			no carrier				with soft bread				with hard bread			
		F	*p*	*F*	*p*		mean	±	SE		mean	±	SE		mean	±	SE	
						AUC												
AUC_R (ppbV·s) *limonene*	*m/z* 138.139	42.4	$p < 0.001$	1.3	NS		6995	±	770	*b*	16553	±	1278	*a*	17085	±	1173	*a*
AUC_R (ppbV·s)	*m/z* 135.119	13.1	$p < 0.001$	4.5	$p < 0.01$	*FF-HV*	4210	±	277	*a*	4532	±	248	*a*	5119	±	307	*a*
citral						*LF-HV*	3228	±	321	*b*	5970	±	400	*a*	5562	±	450	*a*
						LF-LV	6226	±	424	*a*	6766	±	423	*a*	6590	±	460	*a*
	m/z 153.131	6.2	$p < 0.01$	6.0	$p < 0.001$	*FF-HV*	7550	±	538	*a*	7833	±	477	*a*	8909	±	595	*a*
						LF-HV	6007	±	608	*b*	10828	±	775	*a*	9953	±	859	*a*
						LF-LV	11767	±	850	*a*	11571	±	736	*a*	10902	±	821	*a*
AUC_S (mm·s)		7.0	$p < 0.01$	4.0	$p < 0.01$	*FF-HV*	9198	±	506	*a*	8214	±	437	*ab*	7307	±	488	*b*
						LF-HV	5096	±	489	*a*	5844	±	490	*ab*	5515	±	481	*a*
						LF-LV	7362	±	566	*a*	6608	±	522	*ab*	5767	±	484	*b*
						Imax												
Imax_R (ppbV) *limonene*	*m/z* 138.139	17.9	$p < 0.001$	1.4	NS		720	±	99	*b*	1613	±	175	*a*	1533	±	147	*a*
Imax_R (ppbV)	*m/z* 135.119	11.2	$p < 0.001$	2.3	NS		97	±	5	*b*	121	±	5	*a*	128	±	7	*a*
citral	*m/z* 153.131	4.5	$p < 0.05$	4.8	$p < 0.001$	*FF-HV*	173	±	12	*a*	176	±	12	*a*	203	±	14	*a*
						LF-HV	145	±	14	*b*	248	±	19	*a*	223	±	20	*a*
						LF-LV	267	±	16	*a*	268	±	19	*a*	250	±	22	*a*
Imax_S (mm)		9.1	$p < 0.001$	3.5	$p < 0.01$	*FF-HV*	77	±	3	*a*	71		3	*ab*	61		3	*b*
						LF-HV	48	±	4	*a*	48		3	*a*	48		3	*a*
						LF-LV	62	±	4	*a*	56		3	*a*	52		4	*a*
						Tmax												
Tmax_R (s)	*m/z* 138.139	30.4	$p < 0.001$	4.0	$p < 0.01$	*FF-HV*	28	±	2	*a*	24	±	2	*a*	25	±	1	*a*
limonene						*LF-HV*	38	±	3	*a*	23	±	2	*b*	22	±	2	*b*
						LF-LV	33	±	3	*a*	19	±	1	*b*	24	±	2	*b*
Tmax_R (s)	*m/z* 135.119	6.3	$p < 0.01$	1.3	NS		50	±	1	*a*	43	±	1	*b*	46	±	1	*ab*
citral	*m/z* 153.131	1.4	NS	0.7	NS		54	±	1		52	±	1		55	±	1	
Tmax_S (s)		0.3	NS	1.4	NS		38	±	2		38	±	2		40	±	2	

(B) Mayonnaise without and with potato

		potato		mayo:potato			no carrier				with soft potato				with hard potato			
		F	*p*	*F*	*p*		mean	±	SE		mean	±	SE		mean	±	SE	
						AUC												
AUC_R (ppbV·s) *limonene*	*m/z* 138.139	21.8	$p < 0.001$	1.9	NS		6995	±	770	*b*	12697	±	1015	*a*	12350	±	803	*a*
AUC_R (ppbV·s)	*m/z* 135.119	9.0	$p < 0.001$	2.2	NS		4552	±	227	*b*	4957	±	195	*b*	5722	±	246	*a*
citral																		
	m/z 153.131	4.5	$p < 0.05$	3.1	$p < 0.05$	*FF-HV*	7550	±	538	*a*	7476	±	527	*a*	8091	±	605	*a*
						LF-HV	6007	±	608	*b*	8365	±	577	*ab*	9536	±	772	*a*
						LF-LV	11767	±	850	*a*	10706	±	634	*a*	12201	±	658	*a*
AUC_S (mm·s)		10.5	$p < 0.001$	2.1	NS		7236	±	335	*a*	6085	±	307	*b*	6200	±	300	*b*

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Table 3. continued

(B) Mayonnaise without and with potato

		potato		mayo:potato		no carrier				with soft potato				with hard potato			
		F	*p*	*F*	*p*	mean	±	SE		mean	±	SE		mean	±	SE	
Imax																	
Imax_R (ppbV) *limonene*	*m/z* 138.139	10.4	*p* < 0.001	1.7	NS	720	±	99	*b*	1373	±	151	*a*	1361	±	155	*a*
Imax_R (ppbV)	*m/z* 135.119	9.1	*p* < 0.001	2.3	NS	97	±	5	*b*	110	±	5	*b*	127	±	6	*a*
citral	*m/z* 153.131	5.4	*p* < 0.01	2.8	*p* < 0.05	*FF-HV* 173	±	12	*a*	172	±	14	*a*	182	±	14	*a*
						LF-HV 145	±	14	*b*	204	±	15	*ab*	233	±	20	*a*
						LF-LV 267	±	16	*a*	260	±	17	*a*	295	±	19	*a*
Imax_S (mm)		9.8	*p* < 0.001	1.4	NS	62	±	2	*a*	54	±	2	*b*	57	±	2	*ab*
Tmax																	
Tmax_R (s) *limonene*	*m/z* 138.139	16.1	*p* < 0.001	1.5	NS	33	±	1	*a*	24	±	1	*b*	27	±	1	*b*
Tmax_R (s)	*m/z* 135.119	5.8	*p* < 0.01	0.7	NS	50	±	1	*a*	45	±	1	*b*	46	±	1	*b*
citral	*m/z* 153.131	1.9	NS	0.2	NS	54	±	1		51	±	1		51	±	1	
Tmax_S (s)		0.2	NS	1.0	NS	38	±	2		38	±	2		37	±	2	

[a]Lowercase letters: significant differences between mayonnaises without/with bread carriers ($p < 0.05$). [b]The effects of bread (A) and potato (B) are presented separately. The release parameters AUC_R, I_{max}_R, and T_{max}_R correspond to the area under the curve, the maximum concentration, and time to reach the maximum concentration. The sensory parameters AUC_S, I_{max}_S, and T_{max}_S correspond to the total area under the curve, the maximum perceived intensity, and the time to reach the maximum perceived intensity

release (Figure 3B,D and Table 3B). For example, in the case of limonene release (m/z 138.139), AUC_R increased by 45 and 43% after addition of soft and hard potato ($p < 0.001$ and $p < 0.001$). In the case of citral release (m/z 135.119), AUC_R increased by 8% with soft potato ($p > 0.05$) and by 21% with hard potato ($p < 0.001$). Hence, potato texture (soft vs hard) affected citral release concentrations. Similar trends were observed for I_{max}_R.

The time to reach maximum aroma concentration of mayonnaises (T_{max}_R) was affected by the addition of potatoes, regardless of potato texture (Table 3B). On average, T_{max}_R was reached in shorter times after the addition of potato.

Regarding sensory perception, the presence of potato carriers decreased perceived lemon intensity. AUC_S decreased by 19% with addition of soft potato ($p < 0.001$) and by 17% with addition of hard potato ($p = 0.002$). Similar effects were observed for I_{max}_S, but the effect was significant for soft potato (reduction by 15% $p < 0.001$) but not for hard potato (reduction by 9%; $p > 0.05$). T_{max}_S was not significantly affected by addition of potato carriers.

Results Overview: In-Nose Aroma Release and Sensory Perception of Mayonnaise-Carrier Combinations. Multiple factor analysis (MFA) was conducted to summarize the effects of mayonnaise viscosity, mayonnaise fat content, carrier addition, and carrier texture on aroma release and perception of mayonnaises (Figure 4). PC1 explained

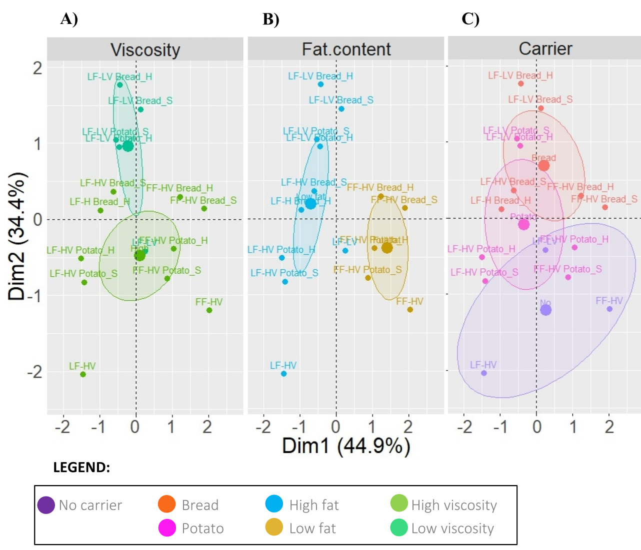

Figure 4. Scatter plot of multiple factor analysis (MFA) of in-nose aroma release and dynamic lemon perception data, in which the mayonnaise viscosity effect (A), the mayonnaise fat content effect (B), and the carrier effect (C) are highlighted using different colors.

44.9% of the total variance and accounted mainly for differences in mayonnaise fat content (Figure 4B). In this case, the 95% confidence ellipses highlight two clusters: one with full fat mayonnaise (FF-HV) and the other with the two low fat mayonnaises (LF-HV and LF-LV). PC2 explained 34.3% of the total variance and accounted for sample differences in viscosity (Figure 4A) and carrier addition (Figure 4C). In this case, the 95% confidence ellipses highlight two clusters: one with low viscosity mayonnaises (LF-LV) and one with high viscosity mayonnaises (LF-HV and FF-HV). Figure 4C highlights a difference between food carriers: bread

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samples are positioned further away from the single mayonnaises than potato samples, indicating that bread had a larger overall impact on lemon aroma release and intensity perception than potato.

To summarize, increasing mayonnaise viscosity or decreasing mayonnaise fat content reduced lemon aroma release and simultaneous lemon intensity perception. The two lemon aroma compounds (limonene, citral) had different release patterns, with limonene being released faster and with higher concentration due to its higher volatility than citral. When mayonnaises were combined with carriers, aroma release and perception were no longer consistent. Addition of bread and potato to mayonnaises enhanced lemon aroma release and decreased simultaneous lemon intensity perception. When comparing the different carrier foods, addition of bread increased lemon aroma release concentrations more than potato. Bread hardness did not influence lemon aroma release, but harder bread tended to decrease lemon intensity perception to a larger extent than soft bread. Potato hardness did not influence aroma release, but softer potato tended to decrease lemon intensity perception slightly more than harder potato.

■ DISCUSSION

The results showed that aroma release from condiments (mayonnaises) was enhanced when condiments were consumed together with carriers (bread or potatoes) compared to consumption without carriers. This was unexpected as we hypothesized that condiment aroma release would decrease with addition of carrier foods, as condiment aroma compounds might bind to the carriers. Although such binding might have occurred through physical, non-covalent bonds between carriers and condiments, a higher concentration of aroma compounds was released in the nose when carrier foods were added to mayonnaises. This indicates that for composite foods, other mechanisms than binding are more relevant and make a larger contribution to in-nose aroma release. We suggest that differences in food oral processing between mayonnaise and mayonnaise in combination with carriers explain the increase in aroma release. Mayonnaise-carrier combinations required chewing to safely break down the food before swallowing, whereas the mayonnaises without carriers were swirled around in the mouth without chewing following standardized consumption protocols (mayonnaise with carrier: chew with 1 chew/s for 20 s; mayonnaise without carrier: swirl in mouth for 20 s). The chewing required for the mayonnaise-carrier combinations apparently induced more aroma release. Moreover, as a result of chewing, the surface area of mayonnaise-carrier combinations might have increased since the carrier might have been broken down into multiple smaller bolus pieces. Consequently, the mayonnaise would be distributed over a larger area, which could have led to a higher transfer of aroma compounds from the mayonnaise to the vapor phase. This could explain why total aroma released increased upon addition of carriers to mayonnaises and this was also reflected in the time required to reach the maximum aroma concentration (T_{max}_R), which was faster in the case of the carrier-mayonnaise combinations than for single mayonnaises (Table 3). In addition, the velum-tongue border has been observed to open more frequently during consumption of solid foods than liquid foods,[22] which could increase the ability of aroma compounds to pass to the nasal cavity ahead of swallowing. Such an effect of oral processing behavior on *in vivo* aroma release is consistent with previous studies.[27−29] Addition of solid carrier foods to condiments thus increases oral movements, in-mouth food manipulations, and food's surface area and therefore favors an increase in in-nose aroma release of condiments throughout consumption.

Higher in-nose aroma release with the addition of carrier foods was still maintained after participants swallowed the foods. Such an effect might be explained by differences in bolus properties and oral retention. When mayonnaise is consumed on its own, it is mixed with saliva leading to a liquid-like bolus that is easily swallowed. We assume that little product remains in the mouth after swallowing.[30] When mayonnaise is consumed with a carrier, it is mixed with both the carrier and saliva leading to a relatively cohesive solid bolus that easily sticks to oral surfaces (teeth, tongue, palate) upon swallowing. We speculate that in this case more product remains in the mouth, which might lead to longer aroma release into the nasal cavity after swallowing.

The type of carrier food (bread versus potato) affected *in vivo* aroma release of mayonnaises since bread increased nasal aroma concentrations to a larger extent than potatoes (Figure 3 and Table 3). We suggest that this result could be explained by the properties of the starch in cooked potatoes. Potatoes contain starch granules, which are gelatinized upon cooking. The gelatinization leads to release of amylose from the granules into the continuous phase, whereas amylopectin resides mostly within the granules. Consequently, starch (mainly amylose) becomes available for interactions with hydrophobic aroma compounds after cooking through hydrophobic interactions. It is known that gelatinized starch retains hydrophobic aroma compounds including limonene to a larger extent than starch granules.[31] Such interactions can limit aroma release and could explain the lower release for potato. Together, these results show that mayonnaise aroma release depended on the properties of the carriers it is combined with.

The texture of carrier foods did not significantly influence mayonnaise aroma release. It is important to note that a standardized consumption protocol was used, meaning that both soft and hard carrier foods were chewed 20 times at the same chewing frequency. This did not allow participants to adapt oral behavior based on texture and presumably resulted in similar nasal air flows and release patterns, and this could be the reason why we see no effect of texture on release. In the case of free eating, differences in aroma release of mayonnaises depending on the texture of the carrier food might occur since softer foods generally require fewer chews than harder foods, likely to result in different nasal air flows which in turn can affect in-nose aroma release. For example, in the case of cheese, firmer cheeses were chewed for a longer time and broken down into more bolus pieces by which both the release rate and the total amount of released aroma were increased.[32,33]

Inter-individual variation between participants is known to affect oral behavior, aroma release, and perception.[26,34] To alleviate inter-individual variation, we selected a relatively homogeneous panel (young, female, Caucasian) and standardized their way of chewing by imposing a chewing protocol to ensure that differences in aroma release and perception could be attributed to varied product properties. This participant group is a segment of the entire population and is not representative of the population. Additional experiments should be performed to generalize the current findings toward the general population. A next step could involve studies investigating aroma release and perception of condiment-

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carrier combinations among participants with different physiological characteristics (*e.g.*, supertaster *vs* non-taster) and eating behaviors (slow *vs* fast eaters).

Mayonnaise properties (viscosity, fat content) were also observed to influence aroma release considerably. As both viscosity and fat content have been shown to influence aroma release of single foods in previous studies, these results are discussed only shortly here. Increasing mayonnaise viscosity by adding more xanthan resulted in lower aroma release and perception (LF-LV *vs* LF-HV). Viscosity is known to play a relevant role in aroma release, as the diffusion rate of aroma compounds is hindered by an increase in viscosity.[35,36] In addition, xanthan has been suggested to physically interact with hydrophobic aroma compounds by trapping them into a so-called "hydrophobic cavity".[37] Decreasing mayonnaise fat content while keeping the same viscosity resulted in decreased aroma release and perception (FF-HV and LF-HV). A similar observation was reported by Wendin *et al.*, who found that decreased fat content tended to decrease the perceived lemon intensity in mayonnaise.[38] However, these results do not support the general theory that partitioning of hydrophobic aromas into aqueous phases and air is greatly reduced with increasing fat/oil content.[39−41] This discrepancy may be due to different factors. First, aroma compounds may interact with xanthan in low fat emulsions (LF-HV), which was added to compensate for the difference in viscosity due to the reduction of fat. Thus, even though lowering oil content could provide the expected increase in aroma release, interactions with xanthan might have been more pronounced, eventually leading to a decrease in aroma release. Second, the FF-HV mayonnaise contains a higher number of fat droplets when compared to the LF-HV. This results in more interfacial area between oil and the continuous aqueous phase, and therefore interaction with saliva may be increased together with eventual transfer to the air phase. This may ultimately lead to a higher aroma release and an accompanying higher aroma concentration in the nose space.[42]

To summarize, our study highlights that aroma release from mayonnaises is enhanced when they are consumed together with carrier foods such as bread or potatoes. At this point, an unexpected result was found: the increase in aroma release with composite foods was not accompanied by an increase in aroma perception. Carrier addition decreased perceived aroma intensity of mayonnaises (Figure 3E,F, Table 3). This decrease in perceived intensity is in line with previous studies, which showed that flavor intensity of soy sauce and mayonnaise decreased with addition of solid carrier foods.[11,14] The present study revealed that the lower perceived intensity is not due to a lower delivery of aroma compounds into the nasal cavity, as aroma release was increased with addition of carriers (Figure 3A−D, Table 3). This discrepancy between aroma release and perception of mayonnaise-carrier combinations indicates that carriers modify condiment aroma perception not by physicochemical interactions between carrier and condiment but via other pathways independent of actual in-nose aroma concentrations. In this case, condiments were evaluated in combination with solid carrier foods, which introduced the texture perception of the solid carriers and the process of chewing solid foods. The perceived texture of the carrier probably induced perceptual, unconscious, cross-modal interaction effects between texture and aroma perception. Previous studies investigated texture-aroma cross-modal interactions in model foods including gels, liquid products, semisolid foods like yogurts, and model cheeses.[9,17,19,23,43−47] Increasing gel hardness resulted in decreased aroma intensity without affecting aroma release in two studies suggesting cross-modal correspondence between texture and aroma perception.[21,48] The addition of solid carriers like bread and potatoes to mayonnaise probably reduced perceived aroma intensity by similar perceptual cross-modal interaction effects.[9] While our study shows that more aroma is released through the retronasal pathway when the carrier is added, the reduction in aroma intensity could be due to a combination of perceptual cross-modal and physiological mechanisms similar to the ones reported by Gierczynski and colleagues.[49] These results demonstrate the complex and important role that texture plays in the multisensory perception of food flavor.[3]

While perceptual cross-modal interaction effects between carrier texture and condiment aroma are plausible, it is important to acknowledge a possible familiarity effect as well as a possible sensory dumping effect.[63,64] Regarding the familiarity effect, participants were more familiar with the consumption of mayonnaise-carrier combinations than mayonnaise, as they generally consume mayonnaise in combination with carrier foods. This difference in familiarly might have influenced their perception and aroma intensity ratings. To minimize a potential familiarity effect, both mayonnaise-carrier combinations and single mayonnaises were assessed during the training sessions to familiarize participants with the samples of the study. Regarding sensory dumping, a well-known limitation of the time−intensity methodology, carriers with different texture properties were added to mayonnaises with lemon aroma, and participants were asked to evaluate lemon intensity only. Participants probably perceived differences in texture and were asked to evaluate lemon aroma intensity only, which might have led to the projection of perceived differences and changes in texture into lemon intensity. To minimize the potential dumping effect, the perceived textural differences were carefully discussed during the multiple training sessions. Subsequently, the panel practiced with the evaluation of aroma intensity while being aware of the possible differences in texture. In this context, it is known that transfer of aroma compounds into the nasal cavity follows swallow breath.[50] Thus, aroma perception is known to increase just after swallowing. When looking at our time−intensity data (Figures 2E,F and 3E,F), we observe a consistent increase in perceived lemon intensity just after swallowing (20 s). This demonstrates that our panel functioned very well since they clearly perceived this increase in aroma intensity after swallowing, which strongly suggests that our panel was capable of properly evaluating aroma intensity and did not dump differences in texture perception into the assessment of aroma intensity. Furthermore, previous studies using a rate-all-that-apply (RATA) methodology for similar composite foods demonstrated similar results to that in our study where flavor intensity of mayonnaise decreased with addition of carrier foods.[11,14] In these studies, texture and flavor attributes were evaluated so dumping effects can be excluded. We therefore assume the sensory dumping effect in our study, if there was any, to be small.

The novelty of the present study is the fact that simultaneous aroma release and perception were assessed for condiment-carrier combinations and not only in model foods or single foods. Combining condiments and carrier foods increases complexity of the food consumed, which is more representative of the common consumption context. In

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summary, in-nose aroma release correlated positively with perceived aroma intensity when mayonnaise was consumed alone (i.e., when higher aroma concentrations were released in the nose, also higher perceived aroma intensity values were reported). This was not the case for more complex foods such as condiment-carrier combinations. Addition of carriers increased in-nose aroma release but decreased the perceived aroma intensity of mayonnaises. Since this decreased aroma perception was not due to a lower delivery of aroma compounds into the nasal cavity, we conclude that aroma release alone does not explain sensory perception of composite foods, but perceptual cross-modal interactions between carrier texture and condiment aroma influenced aroma perception. In the case of composite foods, cross-modal texture−aroma interactions are likely to modulate aroma perception of more complex food combinations. This provides further evidence that not only physicochemical food characteristics but also cross-modal interaction effects should be considered when investigating the mechanisms responsible for flavor perception.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.jafc.1c02649.

Figure S1 and S2: averaged in-nose limonene and citral release and lemon intensity perception during mastication and after swallowing for the LF-HV and LF-LV mayonnaises (PDF)

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Notes

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