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ORIGINAL PAPER

Improved prediction of wheat baking quality by three novel approaches involving spectroscopic, rheological and analytical measurements and an optimized baking test

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Abstract

Baking quality, defined as loaf volume, is one of the most important quality attributes of wheat. An accurate and rapid determination is of great interest for the wheat supply chain. However, this remains difficult to date, because reported predictions based on other wheat characteristics (e.g. protein content) or flour spectroscopy are poor. This study investigates three novel approaches to improve the prediction of specific loaf volume determined by an optimized mini-baking test. The predictions are based on a large variety of rheological and analytical data as well as fluorescence, near-infrared (NIR) and Raman spectroscopy of flour and flour fractions. Furthermore, the influence of data fusion on the predictions is investigated. All three approaches presented promising results and showed great potential for practical application with $R^2_{CV} > 0.90$ for various regression models. For example, the combination of farinograph data with solvent retention capacity data or NIR flour spectra yielded R^2_{CV} of 0.91 in both cases. Combining Raman spectra of the <32 µm and 75–100 µm fractions as well as NIR spectra of gluten, flour and starch both also yielded R^2_{CV} of 0.91. The results underline that loaf volume is a complex quality characteristic that can be better predicted when different data types are combined. Different rheological and analytical tests and different spectroscopic methods capture specific wheat quality characteristics that have different relations to baking volume and can therefore provide complementary information for improved predictions. Furthermore, the importance of rheological tests (especially farinograph, extensograph, alveograph) and the baking procedure for the prediction of baking quality are emphasized.

Keywords Baking quality · Chemometrics · Loaf volume · Rheology · Spectroscopy · Wheat quality

Introduction

Wheat products are among the most important staple foods in the world. Determining wheat quality is therefore of utmost importance for the entire wheat supply chain, because it determines the wheat price and the flours suitability for a given purpose [1, 2].

The prominent quality characteristic of a wheat sample is its baking quality that is usually defined as its ability to produce a bread loaf of a certain volume. Baking quality is assumed to be mainly influenced by protein quantity and protein quality. A prerequisite to carry out an accurate determination of baking quality is the ability to perform baking tests. This is often impractical for mills and breeders in particular, as baking tests are time consuming, expensive and usually require a large amount of flour. Consequently, it has

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become common practice to predict baking quality based on other wheat quality characteristics that can be determined more easily and rapidly [1–4]. In the industry, protein content has been used for this purpose for many years, because it is an analytical parameter that can be predicted well within a couple of minutes using near-infrared (NIR) spectroscopy and it has been shown to have a positive correlation to baking quality. However, the strength of this correlation is being questioned, since correlations reported in literature show high variability with $r^2_{Pearson}$ ranging from values close to 0 to values above 0.80. Nevertheless, the protein content is still the most relevant price building attribute for wheat. This is not only problematic because it appears not to be a good single indicator for baking quality, but also because this leads farmers to use high nitrogen fertilization to achieve high protein contents for economic reasons, accompanied by environmental risks like ground water pollution [1, 5]. Besides protein content, many different empirical rheological tests have been established that try to bridge this remaining gap between analytical characteristics and actual baking tests. They produce useful information simulating the behavior of dough during kneading and proofing processes and therefore reflect protein quality in some way that is assumed to also strongly influence baking quality [2, 3]. Still, correlations of individual parameters of these rheological tests and baking quality reported in literature are usually rather low [4, 6–8]. This is why researchers have focused on finding new methods that allow an improved prediction of baking quality instead of using simple correlations to protein content or to other wheat quality characteristics. Many attempts have been made to predict loaf volume using spectroscopy of flour or grains. Besides NIR spectroscopy, also fluorescence and Raman spectroscopy have been tested for this purpose. This approach can provide an improved prediction, but prediction accuracies are usually only accurate enough for screening purposes, as the obtained coefficients of determination R^2 are often below 0.70 [1, 5]. Other researchers have focused on combining specific parameters of analytical and rheological measurements, often using multilinear regression models, to achieve an improved prediction of baking quality [4, 7, 9]. While some studies reported promising results [4], others do not show great potential of this method for the prediction of baking quality [9]. However, these studies often have limitations, because they tend to use a limited number of analytical and rheological measurements and usually only establish models for specific wheat varieties. Consequently, more research is necessary to find novel methods to achieve an even more accurate prediction of baking quality.

In general, not only the correlations of protein content and baking quality reported in literature show high variability, but also the correlations to rheological parameters as well as the coefficients of determination obtained by predictions using spectroscopy or combinations of analytical and rheological parameters [1, 4–7, 9–12]. Many different reasons are discussed, mainly focusing on reference analyses of predictor variables (e.g. high measurement errors, number of repetitions), environmental factors (e.g. weather, fertilization) and properties of samples used in a study (e.g. number of samples, varieties, variability, protein content range) [1, 4, 5]. The influence of the baking test itself used for the characterization of baking quality is often ignored. There is no single standard baking test used, but rather many different baking tests are reported, which differ not only in size, but also in the used ingredients, their amounts as well as the complete baking procedure (e.g. kneading time, proofing time, method of shaping dough). A few studies have focused on the influence of the baking test on correlation results and have shown that the baking test methodology can be the decisive factor for the fact that there is either a low or a high correlation of baking volume and other wheat quality characteristics [6, 13, 14]. Furthermore, it is known that, for a baking test to reflect the true bread-making potential of a flour, there must not be any limiting factor. Often, studies use baking tests with a fixed baking procedure, e.g. a fixed kneading time, applied to every sample. This can be seen as a limiting factor, because an optimal dough development should be achieved for every flour sample to utilize its full bread-making potential [3]. Therefore, when relating flour quality characteristics to baking quality, special attention should be paid to the choice of the baking test and optimized baking procedures may be preferred.

This study proposes three novel approaches to improve the prediction of baking quality by extending the existing analytical, rheological and spectroscopic methods and relating them to the result of an optimized baking test. Fifty international, commercially available flour samples of different qualities are used, representing mixtures of many different wheat cultivars. Data fusion methods are key features of all three approaches presented in this study, because individual wheat quality characteristics or spectra have not achieved accurate predictions of baking quality in the past. As baking quality is a complex trait that is influenced by many different factors, using complementary information of different analytical, rheological and spectroscopic methods may be beneficial to achieve improved predictions. They provide not only information about protein quantity and quality, but also on other sample components and how all of them interact in processes like kneading or proofing [2, 15, 16]. However, data fusion is normally associated with a higher effort, as more measurements have to be carried out. Consequently, data fusion should be limited by keeping practical applicability and cost-benefit efficiency in mind.

In the first approach (a), many different analytical and rheological methods are used to calculate regression models for the prediction of specific loaf volume. For this, either individual quality characteristics or combinations of different quality characteristics are used. In the second approach (b), a preprocessing of flour samples into air classified fractions, sieve fractions as well as gluten, starch and dough is followed by spectroscopic analyses (fluorescence, NIR, Raman) and regression modeling. This preprocessing approach may improve predictions, since studies have shown that different components are enriched or depleted by the fractionation procedures [17, 18] and additional rheological information may be contained in spectra of gluten and dough [19, 20]. This could lead to an increased information content in the spectra relevant for the prediction of baking quality and may reduce the strong superimposition of spectral signals compared to spectra of flour samples. In this approach, regression models are not only based on single spectra, but also on combinations of different spectra. In the third approach (c), regression models are calculated using combinations of flour spectroscopic analyses (fluorescence, NIR, Raman) and different analytical and rheological quality characteristics.

The aims of this study are to test

(1) the potential of three novel approaches to improve the prediction of baking quality, determined by an optimized baking test, compared to the simple correlations to analytical and rheological parameters (e.g. to protein content) as well as compared to predictions from flour spectroscopy. For this, calculated regression models are

   (a) based on a large variety of analytical and rheological data.
   (b) based on fluorescence, NIR and Raman spectra of flour, air classified fractions, sieve fractions, gluten, starch and dough.
   (c) based on fluorescence, NIR and Raman spectra of flour combined with analytical and rheological data.

(2) the influence of data fusion methods on the prediction accuracy of regression models for all three approaches.

Materials and methods

Flour samples

A total of 50 commercially available wheat flour samples were used in this study, representing mixtures of many different cultivars. The samples were harvested in the years 2019, 2020, 2021 and 2022. Twenty-seven samples originated from Germany while others were grown in Australia, USA, Latvia, Lithuania, Mexico, India, Poland, Romania and Ukraine. Wheat classes were mostly unknown, but the sample set contained samples of many different qualities. The flour samples were provided by Mühlenchemie GmbH & Co. KG (Ahrensburg, Germany). The ash content had been adjusted to approximately 0.60% after milling.

Mini-baking test

An optimized mini-baking procedure was carried out to assess the baking quality of the 50 flour samples. The following ingredients were used: 160 g flour (14% moisture), 1.6 g dry yeast, 1.6 g sugar, 1.6 g peanut fat, 2.4 g salt and 0.0032 g ascorbic acid that was dissolved in a variable amount of water for every sample. The amount of water for every sample was equivalent to the unique water absorption (WAM) determined prior to the baking test using ICC 115. Doughs were prepared in a 300 g farinograph (27 °C, 63 rpm). After 1 min of mixing the dry ingredients, the ascorbic acid solution was added. Starting from this point, dough was kneaded according to the dough development time (DDT) determined prior to the baking test using ICC 115. In case the DDT was shorter than 1.5 min, the kneading time was set to 1.5 min to ensure proper kneading. After kneading, the dough temperature was 27.5 ± 0.5 °C (room temperature was 20–25 °C). Afterwards, the dough was placed in the proofing chamber at 32 °C and 80% rel. humidity (SCH-80, STAMMKÄLTE Heinz Stamm GmbH, Wegberg, Germany) for 30 min minus the individual kneading time. Afterwards, the dough was divided into 4 × 60 g pieces that were rounded to balls in a rounding device (Brabender Type 844000, Brabender GmbH & Co. KG, Duisburg, Germany) for 10 s each. The rounded dough pieces were placed in baking rings (H: 4.35 cm; Ø 7.65 cm) on a baking tray that was then placed back in the proofing chamber for 70 min minus the time it took to round the dough pieces. Then, the dough pieces on the tray were placed in the oven (PICCOLO 1–4 STIR, Wachtel GmbH, Hilden, Germany) and baked for 20 min (230 °C, 8 s steam). After 1 h of cooling at room temperature, bread weights and volumes (VolScan Profiler 600, Stable Micro Systems, Surrey, UK) were determined and the specific loaf volume was calculated for every bread in mL/g bread.

This baking procedure was designed in such a way that it tries to reflect the true bread-making potential of every flour sample. This was ensured by an optimized water addition and kneading time to achieve optimal hydration and dough development for every sample. Furthermore, by subtracting the kneading time and the time for rounding from the resting times, the total elapsed time from the addition of water to the end of the baking test was the same for every sample and

thus also the time in which the yeast was able to produce gas. Additionally, operator-specific influences were minimized by mechanical rounding of dough pieces.

As the amount of flour was limited, baking tests were mostly performed as single replicates. For 13 of the flour samples, a double determination was carried out to test the reproducibility. For those samples, the mean percentage standard deviation of the specific loaf volume was 3.7%. Specific loaf volumes of all 50 samples ranged from 2.76 to 4.84 mL/g bread with a mean of 3.45 mL/g bread. The squared Pearson correlation coefficient r^2_{Pearson} of specific loaf volumes and protein contents was 0.80.

Rheological and analytical measurements

A large number of reference analyses were conducted to obtain analytical and rheological characteristics of the 50 flour samples. The used analyses and methods are specified in Table 1. For Osborne and SDSS-GMP (sodium dodecyl sulfate soluble proteins - glutenin macropolymer) fractionation, method specifications are provided in the subsequent sections. Furthermore, subset abbreviations are introduced in Table 1. Their use is further explained in section "Chemometric analysis". A total of 104 parameters were determined by the different methods. For a few flour samples, some measurements were missing. The total number of samples

Table 1 List of reference analyses with used methods and measured parameters as well as subset abbreviations used

Reference analyses	Subset abbreviation	Method	Number of replicates	Measured parameters
Osborne fractionation	OS	see section Rheological and analytical measurements "Osborne fractionation"	3	osborne total extractable proteins; albumins and globulins; gliadins; glutenins; gliadin/glutenin-ratio
SDSS-GMP fractionation (Sodium dodecyl sulfate soluble proteins - glutenin macropolymer)	SDSS-GMP	see section Rheological and analytical measurements "SDSS-GMP fractionation"	3	SDSS-GMP total proteins; SDSS; GMP; GMP-HMW; GMP-LMW
Wet gluten; Gluten Index	P/F	ICC 155	2	wet gluten content; Gluten Index
Flour protein	P/F	ICC 159	2	protein content
Falling number	P/F	ICC 107/1	2	falling number
Starch damage	S	ICC 172 & AACC 76−31	1	starch damage (Ai%); starch damage (UCD)
Solvent retention capacity	SRC	Method based on AACC 56-11.02 using an automated system	1	water; sucrose; lactic acid; sodium carbonate
Farinograph	FA	ICC 115	2	dough development time DDT; consistency C; water absorption (WZ, WAC, WAM); stability S; dough softening DS (10 min after start, 12 min after DDT); quality number FQN
Extensograph	EX	ICC 114/1	2	water absorption; energy (45, 90, 135 min); resistance (45, 90, 135 min); extensibility (45, 90, 135 min); maximum (45, 90, 135 min); ratio number (45, 90, 135 min); ratio number max. (45, 90, 135 min)
Micro-Visco Amylograph	MVA	Standard method of Brabender	2	point A (time, temperature, viscosity); point B (time, temperature, viscosity); start of holding (temperature, viscosity); start of cooling (temperature, viscosity); point E (temperature, viscosity); point F (temperature, viscosity); viscosity B-D; viscosity E-D
Alveograph	AL	ICC 121	1	maximum pressure Cmax; tenacity P; extensibility L; swelling index G; deformation energy W; ratio P/L; elasticity index Ie; strength coefficient K; strain hardening index SH; minimum of first derivative Dmin; maximum of first derivative Dmax
Mixolab	ML	ICC 173	1	water absorption; dough development time; stability; torques (C1; CS; C2; C3; C4; C5)
ViscoQuick	VQ	Standard method of Brabender	2	beginning of gelatinization (time, temperature, viscosity); maximum (time, viscosity); minimum viscosity; viscosity at 50, 30, 20 °C; breakdown viscosity; setback viscosity
GlutoPeak	GP	Standard method of Brabender	3	peak maximum (time, torque); torque before maximum; torque after maximum; areas (0–1, 1–2, 2–3, 3–4, 4–5)

for every measurement, their units as well as corresponding descriptive statistics are presented in Supplementary Material S1.

Osborne fractionation

Flour (100 mg) was extracted in three stages according to Wieser et al. [21]. First, the albumin and globulin fraction was obtained using salt solution (400 mmol/L NaCl and 67 mmol/L Na_2PO_4/KH_2PO_4 (pH 7.6)). One mL was added to the flour, vortexed for 2 min and stirred for 10 min at 22 °C. After centrifugation (25 min, 22 °C, 3550 rcf), the supernatant was collected in a 2 mL volumetric flask and the procedure was repeated. For the gliadin fraction, 60% aqueous ethanol was used. The residue from the albumin and globulin fraction was extracted three times using 0.5 mL of 60% ehanol each time using the same procedure as for the albumin and globulin fraction. Lastly, the glutenin fraction was obtained by extracting the residue from the gliadin fraction under reducing conditions using 10 mg/mL DTT in buffer solution (0.1 mol/L TRIS-HCl (pH 7.5)/1-propanol (50%, v/v) with 12 g urea). One mL of the buffer solution was added to the residue, vortexed for 2 min and stirred for 30 min in a water bath at 60 °C. After centrifugation (30 min, 22 °C, 3550 rcf), the supernatant was collected in a 2 mL volumetric flask and the procedure was repeated one more time. The collected supernatants in the three volumetric flasks of all three extraction steps were filled up to 2 mL with the respective solvents, filtered (0.45 µm) and separated by UHPLC. Specifications on UHPLC separation are provided in Supplementary Material S1.

SDSS-GMP fractionation

Flour (100 mg) was extracted using 1 mL of SDS solution (1% (w/v) SDS in 0.05 mol/L NaH_2PO_4 (pH 6.9)). After vortexing for 2 min and stirring for 30 min at 22 °C, the sample was centrifuged (25 min, 22 °C, 3550 rcf). The supernatant was collected in a 5 mL volumetric flask and the procedure was repeated one more time. The GMP fraction was further extracted using GMP extraction solution (50% (v/v) propan-1-ol, 0.05 mol/L Na_2HPO_4/KH_2PO_4 (pH 7.5) and 1% (w/v) DTT). One mL of GMP extraction solution was added to the residue, vortexed for 2 min and stirred for 30 min in a water bath at 60 °C before centrifugation (25 min, 22 °C, 3550 rcf). The supernatant was collected in a 2 mL volumetric flask and the extraction was repeated one more time. The collected supernatants in the two volumetric flasks were filled up with the respective solvents, filtered (0.45 µm) and separated by GP-UHPLC. Specifications on GP-UHPLC separation are provided in Supplementary Material S1.

Flour fractionation and dough preparation

Air classification of flour samples into a fine and coarse fraction was performed using the Hosokawa Alpine AG (Augsburg, Germany) air classifier 1213/25 D. Flour (3.5 kg) was processed using a classifier wheel speed of 15,000 rpm and a feed rate of 30 g flour/min. The coarse fraction was classified for a second time. In the end, the proportion of fine fraction was about 6–8%. Particle size distributions of fine fractions were characterized by D[4,3] below 10 µm and Dv (90) below 15 µm. For three samples, no coarse fraction was available for analyses.

Air jet sieving was performed using the 200LS-N Hosokawa Alpine AG (Augsburg, Germany) air jet sieve machine at 2500–2600 Pa. Flour (70 g) was processed using the following sieves from Haver & Boecker OHG (Oelde, Germany): 32 µm, 50 µm, 75 µm, 100 µm. In this way, the following five sieve fractions were obtained: < 32 µm, 32–50 µm, 50–75 µm, 75–100 µm, > 100 µm. Each fraction was sieved for 10 min before the flour remaining on the sieve was transferred to the next larger sieve.

Dough was prepared in a 50 g farinograph using 30 g of flour (14% moisture) and 0.60 g of salt according to ICC 115 (30 °C, 63 rpm, optimum water absorption WAM) until the dough development time was reached. Afterwards, two times 15 g of dough were washed-out with a Glutomatic 2202 (Ing. Stefan Kastenmüller GmbH, Martinsried, Germany) according to ICC 155 using 80 µm metal sieves. The remaining dough and the washed-out wet gluten were covered and rested in a temperature-controlled chamber for 10 min at 25 °C before they were frozen at -28 °C. This process was repeated three times for each flour sample. Afterwards, the collected starch slurry was centrifuged at 3046 rcf for 1 min using a VWR Mega Star 600R centrifuge (VWR international GmbH, Darmstadt, Germany). The precipitate was frozen at -28 °C and the supernatant discarded. The next day, frozen dough, gluten and starch samples were freeze-dried for 24 h in a Christ Alpha 1–4 (Martin Christ Gefriertrocknungsanlagen GmbH, Osterode, Germany). The freeze-dried samples were milled using the Ultra Centrifugal Mill ZM 200 (0.5 mm sieve, 12,000 rpm) (Retsch GmbH, Haan, Germany).

Flours and fractions were stored in airtight containers in the dark at around 15 °C to minimize sample changes during storage. Freeze-dried gluten, starch and dough samples were additionally stored in aluminum containers to prevent permeation of water vapor. 24 h prior to spectroscopic analyses, they were stored at room temperature (21 ± 1 °C).

Spectroscopic analyses

The BioView® Sensor (Delta Light & Optics, Hørsholm, Denmark) was used to record 2D fluorescence spectra by scanning the excitation between 270 and 550 nm and the emission between 310 and 590 nm using 20 nm increments. The used settings were: three repetitions per wavelength combination, high sensitivity for all wavelength combinations, 1350 gain for all wavelength combinations of the excitation wavelengths 270 and 290 nm and 1050 gain for the remaining wavelength combinations. Each sample was compacted in a petri dish (Ø 6 cm) using a 600 g weight. Six spectra were recorded for every sample at different positions.

Near-infrared spectra were recorded using the MPA (Bruker Optik GmbH, Ettlingen, Germany) in diffuse reflectance mode as absorbance spectra. Samples were measured in a rotating cup (quartz glass, Ø 5.1 cm). For settings, a resolution of 8 cm^{-1}, a scanner velocity of 10 kHz and 64 scans per measurement were used. Six measurements were performed for every sample. In between measurements, samples were thoroughly mixed. Before every measurement, a slight compaction was carried out by lightly tapping the filled cup on the table for ten times.

Raman measurements were performed with the Raman 785 (inno-spec GmbH, Nuremberg, Germany) using a fiber optic probe (RamanProbe™ RPP 785/15 − 5 025411, InPhotonics Inc., Norwood, USA). The laser wavelength was 785 nm and the maximum power output was 500 mW. Integration time and number of scans per measurement for every sample type were chosen so that the signal intensity was in the linear detector range and sufficiently high to provide a good signal-to-noise ratio. As baselines differed considerably for various sample types due to laser-induced autofluorescence, the following integration times and number of scans were chosen: 9 s and 12 scans for starch, 2.5 s and 50 scans for gluten, 6.5 s and 15 scans for all other sample types. Each sample was compacted in a petri dish (Ø 6 cm) using a 600 g weight. Then, six spectra were recorded at six different positions. For every measurement, the sample was positioned in the focal point of the fiber optic probe.

Chemometric analysis

All analyses were performed using MATLAB (R2021b, The MathWorks, Inc).

Prediction models for the specific loaf volume were calculated based on three different approaches using spectroscopy of flour, flour fractions and dough, rheological and analytical data of flour as well as a combination thereof. A visualization is presented in Fig. 1 and details on the specific methods are further explained in the subsequent sections.

For every model, a leave-one-out cross-validation was performed and the predictive ability was evaluated by calculating the following metrics according to Eq. 1 to 3.

$$R^2_{\mathrm{CV}} = 1 - \frac{\sum_{i=1}^{n} (y_i - \hat{y}_i)^2}{\sum_{i=1}^{n} (y_i - \bar{y})^2} \tag{1}$$

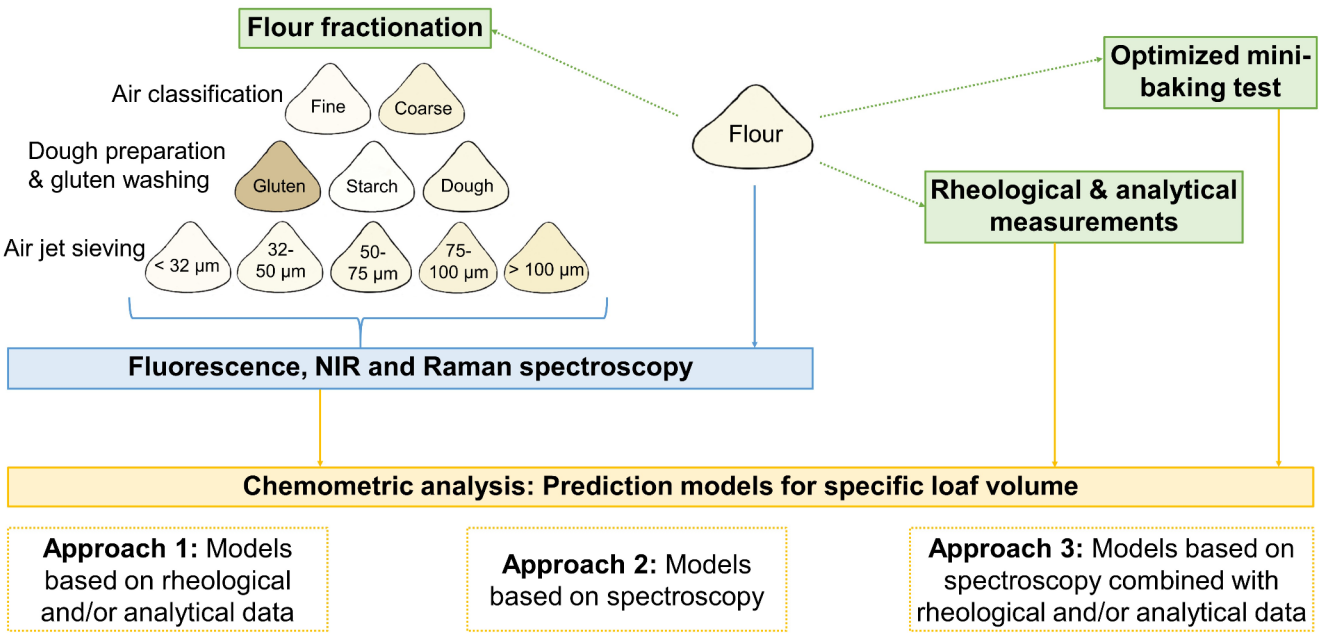

Fig. 1 Overview of the process for generating prediction models for specific loaf volume based on three different novel approaches involving fluorescence, NIR and Raman spectroscopy of flour, flour fractions and dough, rheological and analytical measurements that characterize flour quality and a combination thereof

$$\mathrm{RMSECV} = \sqrt{\frac{\sum_{i=1}^{n}(y_i - \hat{y}_i)^2}{n}} \quad (2)$$

$$\mathrm{NRMSECV}_{\mathrm{range}}\,[\%] = \frac{\mathrm{RMSECV}}{y_{\max} - y_{\min}} \cdot 100\,\% \quad (3)$$

with:

R^2_{CV} = Coefficient of determination of cross-validation
RMSECV = Root mean squared error of cross-validation
$\mathrm{NRMSECV}_{\mathrm{range}}$ = Range normalized RMSECV
n = total number of samples
y_i = measured value of i-th sample
$\hat{y}_i$ = predicted value of i-th sample
$\bar{y}$ = mean value of sample set
$y_{\max}$ = maximum value of sample set
$y_{\min}$ = minimum value of sample set

It should be emphasized that in this study the R^2_{CV} was specified as the coefficient of determination according to Eq. 1. In contrast to the squared Pearson correlation coefficient, which is also frequently reported in literature, this metric can be negative. Negative values indicate that the predictions are very poor, even worse than if the mean value of the sample set was simply used instead of the model for the prediction.

One major focus of this study was to test the influence of data fusion on the predictive quality using the three approaches visualized in Fig. 1. The exact data fusion procedures for each approach are explained further in the following three sections. To assess the effect of data fusion on the prediction, the percentage improvement or worsening of the RMSECV of the fused data model compared to the models based on the individual data was calculated according to Eq. 4. For the $\mathrm{RMSECV}_{\mathrm{reference}}$, the minimum RMSECV of the two individual models was used. For example, model 1 was based on data 1 and yielded RMSECV 1 and model 2 was based on data 2 and yielded RMSECV 2. Model 3 was based on the concatenated data 1 and 2 and yielded RMSECV 3. Then, RMSECV 3 would be used for $\mathrm{RMSECV}_{\mathrm{fused\ data}}$ and the minimum value of RMSECV 1 and RMSECV 2 would be used for $\mathrm{RMSECV}_{\mathrm{reference}}$. Positive values of I indicate that data fusion improves the RMSECV, while negative values indicate that data fusion worsens the RMSECV.

$$I\,[\%] = 100\,\% - \frac{\mathrm{RMSECV}_{\mathrm{fused\ data}}}{\mathrm{RMSECV}_{\mathrm{reference}}} \cdot 100\,\% \quad (4)$$

with:

I = Improvement of RMSECV by data fusion
$\mathrm{RMSECV}_{\mathrm{fused\ data}}$ = RMSECV of model based on fused data
$\mathrm{RMSECV}_{\mathrm{reference}}$ = minimum RMSECV of models based on individual data

Models based on rheological and analytical data

First, a simple linear regression was carried out using every single of the 104 parameters obtained by the various rheological and analytical measurements shown in Table 1.

Second, subsets of data were used to build more complex regression models. For this, parameters, mostly of an individual method, were concatenated to obtain a subset. The abbreviations of the twelve subsets generated are shown in Table 1. For example, all farinograph parameters obtained in a measurement were concatenated to obtain the "FA" subset. For the "P/F" subset, protein related characteristics and the falling number were combined, as those measurements are often readily and easily performed to get a first estimation of flour quality. Furthermore, this data fusion approach was extended by concatenating different subsets before the regression analysis. For example, "FA + P/F" was generated by concatenating all parameters included in the "FA" and the "P/F" subsets. For this subset approach in general, autoscaling of variables was performed before regression, since the units of individual parameters differed. After data fusion and autoscaling, principal component regression (PCR) and partial least squares regression (PLSR) were carried out using individual subsets or concatenated subsets. A maximum of ten components was used for modeling. The best models were determined by the minimum RMSECV. The focus of this data fusion approach using subsets of rheological and/or analytical data was on fusing one to three subsets. Combining more than three subsets would significantly worsen the cost-benefit efficiency of the method, as a lot of flour and equipment would be needed and a lot of working time and manpower would be required to perform the necessary measurements prior to predicting baking quality.

Models based on fluorescence, NIR and Raman spectroscopy

Because every flour was fractionated into nine fractions and dough, a total number of 547 samples (three coarse fractions were not available) were analyzed using three different spectrometers. As six spectra were recorded for every spectrometer and sample, nearly 10,000 spectra were available for analysis.

Spectra were preprocessed using a number of different preprocessing routines including Savitzky-Golay smoothing (order: 2, frame length: 25) and differentiation (order: 2, frame length: 13 for NIR, 9 and 15 for Raman) filters, detrending, highpass filter, standard normal variate transformation (SNV), multiplicative scatter correction (MSC),

extended multiplicative scatter correction (EMSC), min-max normalization, 1. norm and 2. norm normalization and autoscaling of variables. For Raman spectra, baselines were removed by a custom baseline removal algorithm that was based on iterative polynomial baseline fitting (IPBF), but instead of fitting a polynomial baseline in every iteration, the spectrum was excessively smoothed using the Savitzky-Golay method until only the baseline remained. After preprocessing, mean spectra were calculated. Then, data fusion was performed for flour and sample types of a particular fractionation method and spectrometer. For example, for NIR spectra of flour and two air classified fractions, all possible combinations of the three sample types were generated by concatenating spectral intensities. In this case, a total of seven combinations (three single spectra, three combinations of two spectra and one combination of three spectra) were tested for modeling. The same method was applied to NIR spectra of sieve fractions and gluten, starch and dough as well as to fluorescence and Raman spectra of the three mentioned fractionation methods. After preprocessing and data fusion, PCR and PLSR were carried out. One to ten components were tested for modeling. The best models were determined by the minimum RMSECV. No data fusion across fractionation methods and/or across spectrometers was tested in this study. The reason is that this would drastically increase the amount of work and equipment needed to obtain all necessary spectra for prediction, making the procedure irrelevant for practical application.

Table 2 Cross-validation results for prediction of specific loaf volume using linear regression of individual quality parameters

Quality parameter	R^2_{CV}	RMSECV [mL/g bread]
osborne total extractable proteins [g/100 g flour]	0.69	0.28
albumins and globulins [mg/g flour]	0.02	0.50
gliadins [mg/g flour]	0.71	0.27
glutenins [mg/g flour]	0.65	0.30
gliadin/glutenin-ratio [-]	-0.01	0.51
SDSS/GMP total proteins [g/100 g flour]	0.76	0.25
SDSS [mg/g flour]	0.70	0.28
GMP [mg/g flour]	0.44	0.38
GMP-HMW [mg/g flour]	0.36	0.40
GMP-LMW [mg/g flour]	0.43	0.38
wet gluten [%]	0.79	0.23
Gluten Index [-]	-0.10	0.53
protein content [% dm]	0.78	0.24
FA DDT [min]	0.68	0.28
FA S [min]	0.60	0.31
FA FQN [-]	0.56	0.33
EX energy 45 min [cm²]	0.58	0.33
EX extensibility 45 min [mm]	0.79	0.24
EX energy 90 min [cm²]	0.54	0.35
EX extensibility 90 min [mm]	0.80	0.23
EX energy 135 min [cm²]	0.58	0.33
EX extensibility 135 min [mm]	0.76	0.25
AL W [10E-4 J]	0.76	0.25
AL Ie [%]	0.51	0.35

Presented are all protein related parameters and all other parameters with an $R^2_{CV} > 0.50$. Abbreviations are explained in Table 1

Models based on spectroscopy combined with rheological and analytical data

Prediction models were also calculated based on a combination of flour spectra and subsets of rheological and analytical data, as shown in Table 1 and described in the previous section "Models based on rheological and analytical data". Preprocessing routines of flour spectra were equivalent to what is described in section "Models based on fluorescence, NIR and Raman spectroscopy". Because units of spectra and different rheological and analytical parameters differed, autoscaling of variables was performed prior to model building. After data fusion and autoscaling, PCR and PLSR were carried out using a concatenation of flour spectra of different spectrometers and different subsets of rheological and/or analytical data. One to ten components were tested for modeling and the best models were determined by the minimum RMSECV. As described in the other two sections, one focus of this study was on practical applicability and cost-benefit efficiency. Consequently, the focus of this data fusion approach was also on fusing flour spectra of individual spectrometers with one to two subsets of analytical and/or rheological measurements. Fusing spectra of multiple spectrometers, spectra of flour fractions or dough and more than two subsets would drastically increase the effort for the application of this method and would not be relevant in practice as a method to facilitate the prediction of baking quality.

Results and discussion

Prediction models based on rheological and analytical data of wheat

Linear regression of individual quality parameters

The results of linear regression analyses using 104 individual quality parameters of reference analyses described in Table 1 are shown in Table 2. All protein-related parameters are presented as well as all other parameters with an $R^2_{CV} > 0.50$. The results for all 104 parameters are provided in Supplementary Material S2.

It has been shown before that protein quantity and protein quality are the main drivers for the baking quality of wheat

flour, but the definition and measurement of protein quality remains a difficult task up to this day [2, 3]. One approach is to try and characterize protein quality by looking at amounts of individual protein fractions (e.g. gliadins or GMP). The prediction results using protein and wet gluten content, Osborne fractionation and SDSS-GMP fractionation presented in Table 2 emphasize the importance and differences of both protein quantity and quality for baking quality.

In this study, an R^2_{CV} of 0.78 was achieved just using protein content as the predictor variable. In literature, many different correlations for protein content and baking volume are reported, ranging from $r^2_{Pearson}$ close to 0 to $r^2_{Pearson} > 0.80$. Multiple factors are discussed as possible reasons for this high variation, focusing mainly on variety, growing location and growth conditions [1, 4, 6]. Often ignored is the strong influence of the baking test itself used to characterize baking quality. For a baking test to reflect the full bread-making potential of a flour, there must not be any limiting factor [3]. This is especially achieved by using an optimized kneading time to ensure optimal dough development for every sample, as it was also performed in this study. Studies using optimized baking tests are often the ones also reporting higher correlations for protein content and baking volume [4, 6, 7, 22], while studies using fixed kneading times usually report lower correlations [1, 6, 9, 23]. By using an optimized kneading time, the effect of protein quality on the baking result may be minimized, because the gluten network is optimally developed. In this way, a good correlation to protein quantity is achieved and the true bread-making potential of a flour is reflected by the baking result.

Regarding protein quality, as represented by individual protein fractions, prediction results were not as good compared to protein content. Gliadin and glutenin fractions showed R^2_{CV} values of 0.71 and 0.65, respectively, while the R^2_{CV} for albumins and globulins was only 0.02. This underlines that the gluten proteins are important for loaf volume, while non-gluten proteins do not play a major role [3, 24]. Also, the gliadin/glutenin-ratio, that has been discussed as an indicator for baking quality [25], did not show any predictive ability using linear regression with an R^2_{CV} of basically 0. Surprisingly, the GMP related fractions presented $R^2_{CV} < 0.50$ while the SDSS related fraction had an R^2_{CV} of 0.70. This was not expected, as GMP is meant to be an important protein fraction related to baking quality [6, 26]. Possible reasons could be that there is not a single key protein or protein fraction that defines protein quality related to baking quality. Also, the optimal amount of a certain fraction might not show a linear relationship with loaf volume. For example, HMW-glutenin subunits are known to be responsible for dough strength, but doughs can also be too strong, resulting in a small loaf volume [3]. There also might be an optimal amount of a certain protein fraction or an optimal gliadin/glutenin-ratio, but this amount or ratio could further be influenced by the characteristics and amounts of other protein fractions, like e.g. the molecular weight distribution of glutenins [27]. Consequently, protein quality is not easily captured by looking at amounts of individual protein fractions and also not easily related to baking quality just by looking at linear relationships of amounts of individual protein fractions and specific loaf volume.

Another way to try and characterize protein and flour quality is by the many empirical rheological tests established over the last decades. Many of them were also tested in this study, but only the most commonly established and used ones, namely farinograph, extensograph and alveograph measurements, presented prediction results of $R^2_{CV} > 0.50$ in linear regression. Interestingly, the extensibility at 90 min measured by the extensograph yielded an R^2_{CV} of 0.80, which is even better than the prediction using protein content. Farinograph measurements provide information about dough rheology in the mixing stage, while extensograph and alveograph measurements simulate dough properties during fermentation and oven rise by uniaxial and biaxial extension [15]. The results show that specific loaf volume is both related to dough properties during mixing and extension and that often multiple parameters determined by an empirical rheological test show a relationship to baking quality. It was also reported in literature before that some farinograph, extensograph and alveograph parameters are correlated to baking volume [4, 7, 12, 28]. Rheological tests that provide information about starch gelatinization and pasting behavior (e.g. Micro-Visco-Amylograph, ViscoQuick) and also novel methods like Mixolab and GlutoPeak did not show good predictive ability for baking volume in this study using simple linear regression, because all parameters had R^2_{CV} below 0.50.

Regression based on subsets of rheological and analytical data

The parameters of the individual measurements were combined into subsets, the abbreviations of which are listed in Table 1. Subsets were then used, either alone or in combination of two or three subsets, to calculate prediction models for specific loaf volume using PCR and PLSR. The results of the best models are presented in Tables 3, 4 and 5. As the influence of this data fusion approach should be investigated, the percentage improvement of the RMSECV *I* by data fusion is also presented in those tables. Further model specifications are provided in Supplementary Material S2.

For predictions based on single subsets, as shown in Table 3, the same protein and rheological measurements that presented the best results in linear regression shown in

Table 3 Cross-validation results for prediction of specific loaf volume using PCR or PLSR of single subsets of rheological and analytical data

Single subsets	R^2_{CV}	RMSECV [mL/g bread]	Improvement of RMSECV *I* [%] (compared to)
AL	0.82	0.22	12.0 (AL W)
P/F	0.80	0.23	0.0 (wet gluten)
EX	0.80	0.23	0.0 (EX extensibility 90 min)
OS	0.77	0.24	11.1 (gliadin)
SDSS-GMP	0.75	0.25	0.0 (SDSS/GMP total)
FA	0.73	0.26	7.1 (FA DDT)
MVA	0.58	0.33	28.3 (MVA max temp. or MVA visc. B-D)
GP	0.57	0.33	17.5 (GP area (3–4))
ML	0.53	0.35	14.6 (ML hydration or ML DDT)
SRC	0.44	0.38	5.0 (SRC sucrose)
VQ	0.29	0.42	4.6 (VQ breakdown)
S	-0.02	0.51	0.0 (starch damage)

Presented is also the improvement of the RMSECV *I* compared to the best linear regression result using individual quality parameters (see Table 2). Abbreviations are explained in Table 1

Table 4 Cross-validation results for prediction of specific loaf volume using PCR or PLSR and a combination of two subsets of rheological and analytical data

Combination of two subsets	R^2_{CV}	RMSECV [mL/g bread]	Improvement of RMSECV *I* [%] (compared to)
FA + SRC	0.91	0.15	42.3 (FA)
FA + AL	0.90	0.15	31.8 (AL)
FA + EX	0.89	0.16	30.4 (EX)
FA + P/F	0.89	0.17	26.1 (P/F)
EX + P/F	0.87	0.19	17.4 (P/F)
EX + VQ	0.87	0.19	17.4 (EX)
FA + OS	0.86	0.18	25.0 (OS)
AL + ML	0.86	0.19	13.6 (AL)
EX + SDSS-GMP	0.85	0.20	13.0 (EX)
EX + AL	0.85	0.20	9.1 (AL)
FA + GP	0.84	0.20	23.1 (FA)
AL + P/F	0.84	0.20	9.1 (AL)
ML + P/F	0.84	0.20	13.0 (P/F)
EX + ML	0.84	0.21	8.7 (EX)
EX + OS	0.83	0.21	8.7 (EX)
FA + SDSS-GMP	0.83	0.21	16.0 (SDSS-GMP)
ML + SRC	0.82	0.22	37.1 (ML)
GP + OS	0.81	0.22	8.3 (OS)

Presented is also the improvement of the RMSECV *I* compared to the corresponding best result using single subsets (see Table 3). Only results of subset combinations with an RMSECV improvement $I > 5.0\%$ are shown. Abbreviations are explained in Table 1

Table 2 also presented the best predictions using this subset method. The best prediction was achieved using the AL subset with an R^2_{CV} of 0.82 and an RMSECV improvement *I* of 12.0% compared to the linear regression using the AL W parameter alone. For the P/F, EX and SDSS-GMP subsets, no improvements of the RMSECV were achieved combining the respective individual parameters. For the OS and FA subsets on the other hand, RMSECV improvements *I* of 11.1% and 7.1% were achieved with R^2_{CV} of 0.77 and 0.73, respectively. For all other subsets, apart from S, the prediction models were also improved by combining the respective parameters of a particular measurement into a subset. For example, for the MVA and the GP subsets, RMSECV improvements *I* of 28.3% and 17.5% were achieved, respectively, and the R^2_{CV} were improved to values above 0.50, although the prediction models of those subsets still lack in accuracy.

Table 5 Cross-validation results for prediction of specific loaf volume using PCR or PLSR and a combination of three subsets of rheological and analytical data

Combination of three subsets	R^2_{CV}	RMSECV [mL/g bread]	Improvement of RMSECV *I* [%] (compared to)
FA + OS + SRC	0.94	0.13	13.3 (FA + SRC)
FA + EX + SRC	0.93	0.13	13.3 (FA + SRC)
FA + AL + SRC	0.93	0.13	13.3 (FA + AL or FA + SRC)
FA + EX + OS	0.92	0.14	12.5 (FA + EX)
FA + EX + P/F	0.91	0.15	6.6 (FA + EX)
FA + GP + OS	0.89	0.16	11.1 (FA + OS)
FA + GP + P/F	0.89	0.16	5.9 (FA + P/F)
EX + AL + P/F	0.89	0.17	10.5 (EX + P/F)
FA + OS + SDSS-GMP	0.88	0.17	5.6 (FA + OS)
EX + VQ + P/F	0.88	0.18	5.3 (EX + VQ or EX + P/F)
EX + P/F + SRC	0.88	0.18	5.3 (EX + P/F)
AL + ML + SRC	0.88	0.18	5.3 (AL + ML)
EX + ML + P/F	0.88	0.18	5.3 (EX + P/F)
EX + AL + ML	0.88	0.18	5.3 (AL + ML)
EX + VQ + OS	0.88	0.18	5.3 (EX + VQ)
EX + P/F + S	0.88	0.18	5.3 (EX + P/F)
AL + ML + P/F	0.88	0.18	5.3 (AL + ML)
EX + ML + SRC	0.88	0.18	14.3 (EX + ML)
AL + ML + OS	0.88	0.18	5.3 (AL + ML)
EX + VQ + SDSS-GMP	0.88	0.18	5.3 (EX + VQ)
EX + AL + OS	0.88	0.18	10.0 (EX + AL)
EX + VQ + SRC	0.87	0.18	5.3 (EX + VQ)
EX + P/F + SDSS-GMP	0.87	0.18	5.3 (EX + P/F)
EX + ML + OS	0.87	0.19	9.5 (EX + OS)

Presented is also the improvement of the RMSECV *I* compared to the corresponding best result using a combination of two subsets (see Table 4). Only results of combinations with an RMSECV improvement $I > 5.0\%$ are shown. Abbreviations are explained in Table 1

When this data fusion approach was extended by combining two subsets, as shown in Table 4, predictions were strongly improved for various combinations of subsets. Again here, especially FA, EX, AL and P/F subsets, either combined with each other or combined with other subsets, yielded the best predictions for specific loaf volume.

For many different combinations, R^2_{CV} of 0.85 or higher and RMSECV improvements *I* of > 15.0% were achieved. The best result was obtained when combining the FA and SRC subsets. Then, the R^2_{CV} was increased to 0.91 and the RMSECV was improved by 42.3% compared to just using the FA subset. SRC values have been shown to have good correlations with baking quality [13, 29, 30]. In this study, the SRC subset alone did not yield good predictions with an R^2_{CV} of only 0.44, but in combination, the SRC subset strongly improved predictions. Besides this, also the combinations of FA + AL, FA + EX and FA + P/F yielded R^2_{CV} of about 0.90 and RMSECV improvements *I* of > 26.0%.

Further improvements were achieved when combining three subsets. Results in Table 5 show that again, the most important subsets for prediction seem to be especially FA, EX, AL, P/F and also SRC. But here, those subsets were sometimes also combined with subsets that did not yield such good predictions when used as single subsets or in combinations of two subsets (e.g. GP, VQ, ML). The three best models were based on a combination of FA + SRC either with OS, EX or AL. In this way, R^2_{CV} of 0.94, 0.93 and 0.93 were achieved, respectively. For all three of those combinations, the RMSECV improvement *I* was 13.3%. Many other combinations of three subsets yielded R^2_{CV} of 0.88 or higher. Generally, the RMSECV improvements *I* for three subsets were often smaller than for two subsets but still > 5.0% for many subset combinations.

A combination of more than three subsets was tested, but compared to the labor and material costs associated with the additional measurements, this did not result in any significant improvements in the prediction accuracy compared to the already very good predictions achieved using two or three subsets. Also, combining many different rheological measurements may not be beneficial, as they often have relatively high measurement errors that might negatively influence the predictive quality of the corresponding models.

The promising results of this study are also supported by literature, because the approach of combining different parameters of rheological and/or analytical data to predict loaf volume has been tested before. Those studies mostly use a limited data set (limited varieties, countries of origin, harvest years), are usually based on a limited number of rheological and/or analytical reference analyses and often just use multilinear regression of a selected number of parameters to establish separate models for different wheat classes. For example, Dowell et al. [4] also used an optimized baking test and achieved model improvements by combining multiple parameters in multilinear regression. Protein content was the most important predictor for loaf volume, but the best models also included rheological measures (e.g. water absorption, farinograph stability). In this way, they achieved an $R^2_{Prediction\ set}$ of 0.90 for hard red winter wheat models and an $R^2_{Prediction\ set}$ of 0.85 for hard red spring wheat models. Selga et al. [9], on the other hand, were not able to accurately predict the baking volume of neither winter nor spring wheats using combinations of different rheological and analytical measures in PLSR with R^2_{CV} of only 0.50 and lower, but their models were also based on non-optimized baking tests. Nevertheless, their best models were also achieved using protein content, alveograph and farinograph parameters, but also other parameters, like damaged starch, were included. Addo et al. [12] reported an R^2 of 0.927 for an optimized baking test when combining protein content and different alveograph parameters in a multilinear regression to predict loaf volume, although it seems that this metric is only reported for calibration models and therefore has to be interpreted with caution.

The results of the subset approach in this study show that a more complex consideration of rheological and analytical parameters is necessary to achieve a very good prediction of baking quality. This is firstly enabled by combining all parameters of a measurement into a subset. Rheological parameters are usually derived from a specific curve recorded during a measurement, e.g. from a farinograph curve, and therefore provide complementary information about the complex rheological behavior of a sample under a certain stress or strain [3, 31]. In other cases, e.g. for SRC values or Osborne fractionation, the obtained parameters characterize different components of the sample and consequently, the overall sample quality can be better understood by combining those parameters as well [16]. Secondly, it is known that also different rheological and analytical tests provide complementary information and have to be viewed in relation to each other in order to get a comprehensive understanding of the baking quality of a flour sample [2, 15]. This is strongly supported by the fact that in this study, many different combinations of two or three subsets show strong RMSECV improvements and yield very good predictions of baking quality when models are based on combined subsets. The importance of the SRC subset in this study for models based on two or three subset combinations also shows that parameters or subsets that do not seem to be as relevant for the characterization of baking quality when used on their own can be of great importance for the model improvement when combined with other data.

In this study, most important subsets and subset combinations were always based on rheological measurements (especially farinograph, extensograph and alveograph measurements), often in combination with SRC or analytical subsets. Rheological subsets showed higher importance than subsets related to protein analysis for the prediction of baking quality. This underlines the importance of rheological measurements for the characterization of flour quality in general. Many of them provide indirect measures for flour

protein quality. They measure the cumulative contributions and interactions of the individual flour components in measurements that attempt to characterize the actual behavior of a flour sample in a baking test by mimicking kneading and proofing processes [2, 15, 16]. Furthermore, it was shown here that many different rheological and analytical measurements are closely related to the specific loaf volume, which was determined using a baking test with an optimized kneading time. This emphasizes again the importance of an optimized kneading time in a baking test in order to adequately characterize the baking quality of a flour sample, because good correlations to many different rheological and analytical tests, which also claim to characterize the flour quality, were achieved with this method. It also demonstrates the strong potential of this subset approach for the prediction of baking quality, as many different instruments are applicable in practice and can be adapted depending on availability.

Prediction models based on fluorescence, NIR and Raman spectra of flour, flour fractions and dough

Example spectra for all sample types and spectrometers are shown in Fig. 2. Raw spectra are presented for fluorescence and NIR spectra, but Raman spectra were baseline-corrected to better visualize the peaks. Gluten Raman spectra are noisy and show some baseline-removal artefacts due to the really strong baselines present when measuring gluten samples with a laser wavelength of 785 nm, as used in this study. Spectral differences are visible for all sample types and every spectrometer, reflecting a change in sample composition caused by flour fractionation and dough preparation.

First, individual spectra of flour, flour fractions and dough were used to calculate prediction models for specific loaf volume using PCR and PLSR. Results of best models are presented in Table 6. Then, also models based on combinations of spectra were calculated. Corresponding results can be found in Table 7. Further model specifications are provided in Supplementary Material S2.

In this study, good predictions for specific loaf volume were already achieved just by using flour spectra. While an R^2_{CV} of 0.71 was obtained using fluorescence spectra of flour samples, R^2_{CV} of 0.85 and 0.81 were achieved by NIR and Raman spectroscopy of flour, respectively. Predictions of bread volume by flour spectroscopy found in literature are often worse than the results obtained in this study. For example, Gabriel et al. [1] obtained $R^2_{Validation}$ of 0.72 to 0.75 using NIR spectra of flour to predict bread volume. Jirsa et al. [10] achieved R^2_{CV} of about 0.60 for prediction of specific loaf volume from flour NIR spectra. Nagel-Held et al. [5] used fluorescence, NIR and Raman flour spectra to predict specific loaf volume and achieved R^2_{CV} of 0.57, 0.61 and 0.59, respectively. Neither Gabriel et al. [1] nor Nagel-Held et al. [5] used an optimized baking procedure and Jirsa et al. [10] did not make any specification regarding the kneading time.

The predictive ability of spectroscopic methods for bread volume is often attributed to the fact that protein content can be predicted very well, especially from NIR spectra, and that protein content is to some extent related to bread volume [11]. Consequently, the reason for the better predictions from flour spectra found in this study is likely due to the optimized baking tests used, as discussed before. Because of the optimized kneading time, the effect of protein quality may be minimized and then, a good correlation to protein content with an R^2_{CV} of 0.78 in linear regression was achieved. This is also supported by the fact that Dowell et al. [11] adapted the kneading time in his study, found a good correlation of specific loaf volume and protein content and achieved prediction accuracies of $R^2_{CV} \geq 0.80$ using NIR flour spectra. Nevertheless, the results of this study show that, compared to just using protein content in a linear regression, a better prediction is possible by using NIR and Raman spectra of flour. Nagel-Held et al. [5] and Gabriel et al. [1] also achieved improved predictions using spectroscopy compared to the correlation of baking volume and protein content, although their models still lacked in prediction accuracy, as mentioned above. This shows that some spectroscopic methods capture additional relevant information for the prediction of baking quality, besides protein content. It also emphasizes again the strong influence of the baking procedure on the predictive ability of regression models for prediction of baking quality.

When flour was fractionated or prepared to dough prior to spectroscopic analyses, prediction accuracies of models significantly changed compared to flour spectra. In many cases, using individual spectra of flour fractions or dough yielded equal or worse predictions compared to flour spectra. Especially spectra of fine fractions gave the worst results for every spectrometer with R^2_{CV} ranging from 0.38 to 0.49. This was not expected, since the fine fraction was tested to be protein-enriched compared to flour (results not shown) and spectra of fine fractions should therefore contain more relevant signals of protein constituents. In the literature, it is suggested that the proteins enriched in the fine fraction are mainly free "wedge" proteins. Those proteins are fragments of free protein that are formed when the protein matrix in the endosperm is broken up during milling. "Adherent" proteins on the other hand retain larger particle sizes, as those proteins are tightly connected to starch granules and covered by a lipid layer [18, 32]. Possibly, this indicates that free "wedge" proteins are not as important for the baking quality of wheat. For fluorescence and Raman spectroscopy, individual spectra of flour fractions or dough

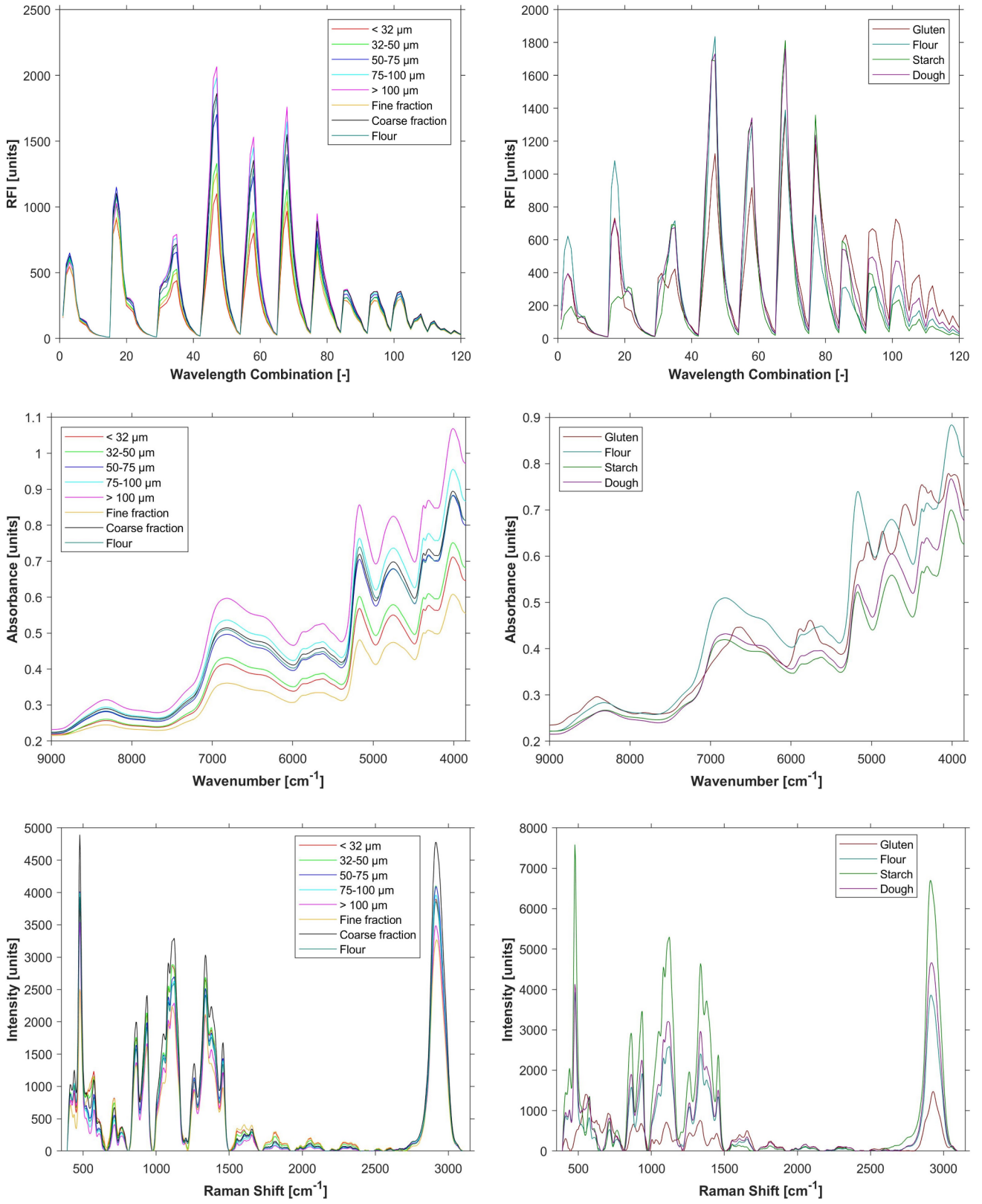

Fig. 2 Example spectra for fluorescence (top row), NIR (middle row) and Raman (bottom row) spectroscopy of flour, flour fractions and dough. 2D fluorescence spectra are presented in 1D by plotting the measured intensities over all scanned wavelength combinations. Raman spectra were baseline corrected

Table 6 Cross-validation results for prediction of specific loaf volume from individual fluorescence, NIR and Raman spectra using PCR or PLSR

Sample type	Fluorescence			NIR			Raman		
	R^2_{CV}	RMSECV [mL/g bread]	I [%][a]	R^2_{CV}	RMSECV [mL/g bread]	I [%][a]	R^2_{CV}	RMSECV [mL/g bread]	I [%][a]
Flour	0.71	0.27	-	0.85	0.20	-	0.81	0.22	-
Gluten	0.81	0.22	18.5	0.59	0.32	-60.0	0.58	0.33	-50.0
Starch	0.52	0.35	-29.6	0.69	0.28	-40.0	0.64	0.30	-36.4
Dough	0.71	0.27	0.0	0.84	0.20	0.0	0.84	0.21	4.6
Fine	0.47	0.37	-37.0	0.49	0.36	-80.0	0.38	0.40	-81.8
Coarse	0.64	0.29	-7.4	0.76	0.24	-20.0	0.81	0.22	0.0
< 32 µm	0.57	0.33	-22.2	0.72	0.27	-35.0	0.78	0.24	-9.1
32–50 µm	0.66	0.30	-11.1	0.81	0.23	-15.0	0.76	0.25	-13.6
50–75 µm	0.70	0.28	-3.7	0.82	0.21	-5.0	0.85	0.20	9.1
75–100 µm	0.74	0.26	3.7	0.85	0.20	0.0	0.85	0.20	9.1
> 100 µm	0.68	0.29	-7.4	0.83	0.21	-5.0	0.84	0.20	9.1

[a]Improvement of RMSECV I compared to flour spectra (≙reference data). Presented are the results using individual spectra as well as the improvement of the RMSECV I using dough and fractions spectra compared to flour spectra

Table 7 Cross-validation results for prediction of specific loaf volume from fluorescence, NIR and Raman spectra using PCR or PLSR of combined spectra

Combination		R^2_{CV}	RMSECV [mL/g bread]	Improvement of RMSECV I [%] (compared to)
			Fluorescence	
2 spectra	< 32 µm + 50–75 µm	0.75	0.26	7.1 (50–75 µm)
			NIR	
2 spectra	Gluten + Flour	0.88	0.18	10.0 (Flour)
	Flour + Starch	0.87	0.18	10.0 (Flour)
	32–50 µm + 75–100 µm	0.87	0.18	10.0 (75–100 µm)
	< 32 µm + > 100 µm	0.86	0.19	9.5 (> 100 µm)
3 spectra	Gluten + Flour + Starch	0.91	0.15	16.7 (Gluten + Flour or Flour + Starch)
			Raman	
2 spectra	< 32 µm + 75–100 µm	0.91	0.15	25.0 (75–100 µm)
	< 32 µm + 50–75 µm	0.88	0.18	10.0 (50–75 µm)
	< 32 µm + > 100 µm	0.87	0.18	10.0 (> 100 µm)
	50–75 µm + > 100 µm	0.87	0.18	10.0 (50–75 µm or > 100 µm)
	< 32 µm + Flour	0.86	0.19	13.6 (Flour)

Presented are the results using a combination of spectra as well as the RMSECV improvement I compared to the corresponding best result using a single spectrum (see Table 6) or a combination of spectra. Only results of combinations with a RMSECV improvement $I > 5.0\%$ are shown

also achieved improved predictions compared to flour spectra. The RMSECV was improved by 18.5% using gluten fluorescence spectra compared to flour fluorescence spectra. In this way, an R^2_{CV} of 0.81 was achieved. For Raman spectroscopy, the best models for individual spectra were based on the 50–75 µm or the 75–100 µm fractions, each with an RMSECV improvement I of 9.1% and an R^2_{CV} of 0.85. Using a combination of different spectra for modeling improved the prediction accuracies. Best predictions using this approach were achieved for NIR spectroscopy using a combination of gluten, flour and starch spectra with an R^2_{CV} of 0.91 and for Raman spectroscopy using a combination of < 32 µm and 75–100 µm spectra, also achieving an R^2_{CV} of 0.91.

The results suggest that air jet sieving as well as dough preparation and gluten washing are fractionation methods that have the potential to improve the prediction of baking quality compared to flour spectra, while air classified fractions did not improve the prediction. The exact reasoning for a particular combination of spectra obtained by

a particular spectroscopic technique to show an improved prediction is difficult to determine, as many different factors affect the result. This includes the changes induced by the fractionation methods that are apparent but their nature is mostly unknown, the signals captured by a particular spectroscopic technique and their unknown relation to baking quality, individual challenges of spectroscopic techniques and their unknown influence on prediction quality (e.g. strong baselines for Raman gluten spectra, sensitivity of NIR to differences in water content), used preprocessing and regression methods and many more. Nevertheless, the results support the hypothesis that flour fractionation alters the composition of samples and in this way, relevant spectral signals for prediction of baking quality can be enhanced, possibly by enriching components relevant for baking quality and/or depleting components that lower the spectral quality by superimposition of irrelevant signals in flour spectra. Dough and gluten spectra could also contain more relevant information, as their processing state is closer to that of the final bread compared to flour or sieve fractions, where components are still present in their native state. For example, Gabriel et al. [1] tested predictions based on dough NIR spectra compared to spectra of flour and also found an improved prediction. Additionally, the fact that a combination of spectra is necessary to achieve the best predictions in this study demonstrates that spectra of different fractions contain complementary information, possibly because components relevant for an overall estimation of baking quality have been separated into multiple fractions.

Prediction models based on spectroscopy combined with subsets of rheological and analytical data

Further prediction models were calculated based on a combination of flour spectra with either one or two subsets of rheological and analytical data for all three spectroscopic techniques. The results of the best models are presented in Table 8. Further model specifications are provided in Supplementary Material S2.

For fluorescence, NIR and Raman spectroscopy, best predictions for the combination of flour spectra and one subset were always achieved using either the FA or the EX subset. Especially for NIR spectroscopy, very good predictions with R^2_{CV} of 0.91 and RMSECV improvements $I \geq 20.0\%$ were obtained for both. Also, fluorescence and Raman spectroscopy showed good predictions with R^2_{CV} of 0.86 and 0.87, respectively, using a combination of flour spectra and the EX subset. When two subsets were included in the models, further improvements were observed. For fluorescence and Raman spectroscopy, R^2_{CV} close to 0.90 were obtained. When flour NIR spectra were combined with FA+SRC, FA+AL or EX+AL subsets, R^2_{CV} of 0.93, 0.92 and 0.91 and RMSECV improvements I of 6.7%, 6.7% and 6.3% were achieved, respectively. In any case, prediction models that showed improvements were always based on a combination of flour spectra and rheological subsets. Only in some cases, also analytical subsets were included in combinations of flour spectra and two subsets.

Spectra of different spectroscopic techniques contain information about flour constituents that show signals based on direct absorption or scattering of incident light, depending on the spectroscopic technique used. Consequently, spectra contain information about presence and concentration of specific molecular species and also about protein molecular arrangement to some extent [33, 34]. For example, Raman spectra contain various regions (e.g. disulfide bond regions, Amid I, II and III) with information about disulfide bond configuration and protein secondary structure [34]. Spectroscopy of flour samples can therefore be viewed more as a technique giving information about parameters related to analytical characteristics of the native flour sample. Many studies have shown that the predictive ability of flour spectroscopy regarding rheological parameters, such as farinograph or extensograph characteristics, is often quite poor [5, 10, 11]. It seems that regression models lack the ability to predict the rheological behavior of a flour sample in a certain process based on spectral data that is more related to analytical characteristics of the native flour sample. Results of this study have shown the importance of

Table 8 Cross-validation results for prediction of specific loaf volume from a combination of fluorescence, NIR or Raman flour spectra and one or two subsets of rheological and/or analytical data using PCR or PLSR

Combination			R^2_{CV}	RMSECV [mL/g bread]	Improvement of RMSECV I [%] (compared to)
Fluorescence +	1 subset	EX + Flour spectrum	0.86	0.20	13.0 (EX)
		FA + Flour spectrum	0.80	0.22	15.4 (FA)
		ML + Flour spectrum	0.76	0.25	7.4 (Flour spectrum)
	2 subsets	EX + ML + Flour spectrum	0.88	0.18	10.0 (EX + Flour spectrum)
		EX + AL + Flour spectrum	0.87	0.18	10.0 (EX + AL or EX + Flour spectrum)
NIR +	1 subset	FA + Flour spectrum	0.91	0.15	25.0 (Flour spectrum)
		EX + Flour spectrum	0.91	0.16	20.0 (Flour spectrum)
	2 subsets	FA + SRC + Flour spectrum	0.93	0.14	6.7 (FA + SRC or FA + Flour Spectrum)
		FA + AL + Flour spectrum	0.92	0.14	6.7 (FA + AL or FA + Flour Spectrum)
		EX + AL + Flour spectrum	0.91	0.15	6.3 (EX + Flour spectrum)
		AL + ML + Flour spectrum	0.89	0.17	10.5 (AL + ML)
Raman +	1 subset	EX + Flour spectrum	0.87	0.19	13.6 (Flour spectrum)
		FA + Flour spectrum	0.84	0.20	9.1 (Flour spectrum)
	2 subsets	EX + P/F + Flour spectrum	0.87	0.18	5.3 (EX + P/F or EX + Flour spectrum)
		ML + SRC + Flour spectrum	0.85	0.20	9.1 (ML + SRC)

Presented is also the improvement of the RMSECV I compared to the corresponding best result using either flour spectra on their own, individual subsets or a combination of two subsets (see Tables 3, 4 and 6). Only results of combinations with an RMSECV improvement $I > 5.0\%$ are shown. Abbreviations are explained in Table 1

rheological characteristics for the prediction of baking quality, as very good predictions were achieved using models based on various rheological subsets (see Tables 4 and 5). The novel approach of combining spectroscopic data with rheological and/or analytical sample characteristics was also able to achieve improved predictions here compared to just using flour spectra or rheological and/or analytical data alone. Mostly rheological subsets, especially the FA and EX subsets, were of great importance here. These results emphasize again the importance of rheological characteristics of flour samples for the prediction of specific loaf volume and show that complementary information is contained in spectroscopic data and rheological and/or analytical data.

Summary of overall best prediction models based on approaches using spectroscopic, rheological and analytical data of wheat

A summary of the overall best prediction models using all three novel approaches presented and discussed in the previous sections is provided in Table 9. For these models, additional information regarding e.g. preprocessing methods and number of used principal components are listed. A comprehensive list for all models presented in this study is provided in Supplementary Material S2. Furthermore, for some of the best models, fits of measured vs. predicted values of cross-validation are visualized in Fig. 3.

Table 9 Overview of cross-validation metrics of overall best models for prediction of specific loaf volume using spectroscopy of flour and flour fractions, rheological and analytical data and a combination of both

		Number of samples	Combination	Preprocessing - model	Number of components	R^2_{CV}	RMSECV [mL/g bread]	$NRMSECV_{range}$ [%]
Rheological & analytical data	Linear regression	50	Protein content [%dm]	none - Linear regression	-	0.78	0.24	11.54
	Linear regression	49	EX extensibility 90 min [mm]	none - Linear regression	-	0.80	0.23	11.06
	1 subset	50	AL	autoscale - PCR	6	0.82	0.22	10.58
	2 subsets	49	FA + SRC	autoscale - PCR	6	0.91	0.15	7.22
	3 subsets	49	FA + OS + SRC	autoscale - PCR	8	0.94	0.13	6.25
Fluorescence Spectroscopy	Flour	50	-	detrend (1. order) + min-max normalization - PLSR	6	0.71	0.27	12.99
	Best fraction(s)	50	Gluten	detrend (1. order) + autoscale - PLSR	9	0.81	0.22	10.58
NIR Spectroscopy	Flour	50	-	smooth + highpass + normalization (1. norm) - PLSR	4	0.85	0.20	9.62
	Best fraction(s)	50	Gluten + Flour + Starch	smooth + highpass + normalization (1. norm) - PLSR	10	0.91	0.15	7.22
Raman Spectroscopy	Flour	50	-	smooth + baseline removal + highpass + autoscale - PCR	4	0.81	0.22	10.58
	Best fraction(s)	50	<32 µm + 75–100 µm	smooth + baseline removal + SNV + EMSC + autoscale - PLSR	9	0.91	0.15	7.22
Fluorescence Spectroscopy + Rheological & analytical data	Spectrum + 1 subset	49	EX + Flour spectrum	SNV (spectra); autoscale (all) - PCR	8	0.86	0.20	9.62
	Spectrum + 2 subsets	49	EX + ML + Flour spectrum	autoscale (all) - PLSR	8	0.88	0.18	8.66
NIR Spectroscopy + Rheological & analytical data	Spectrum + 1 subset	49	FA + Flour spectrum	smooth (spectra) + normalization (2. norm) (spectra); autoscale (all) - PLSR	10	0.91	0.15	7.22
	Spectrum + 2 subsets	49	FA + SRC + Flour spectrum	smooth (spectra) + normalization (2. norm) (spectra); autoscale (all) - PLSR	10	0.93	0.14	6.74
Raman Spectroscopy + Rheological & analytical data	Spectrum + 1 subset	49	EX + Flour spectrum	smooth (spectra) + SNV (spectra); autoscale (all) - PCR	10	0.87	0.19	9.14
	Spectrum + 2 subsets	49	EX + P/F + Flour spectrum	smooth (spectra); autoscale (all) - PCR	10	0.87	0.18	8.66

Abbreviations are explained in Table 1

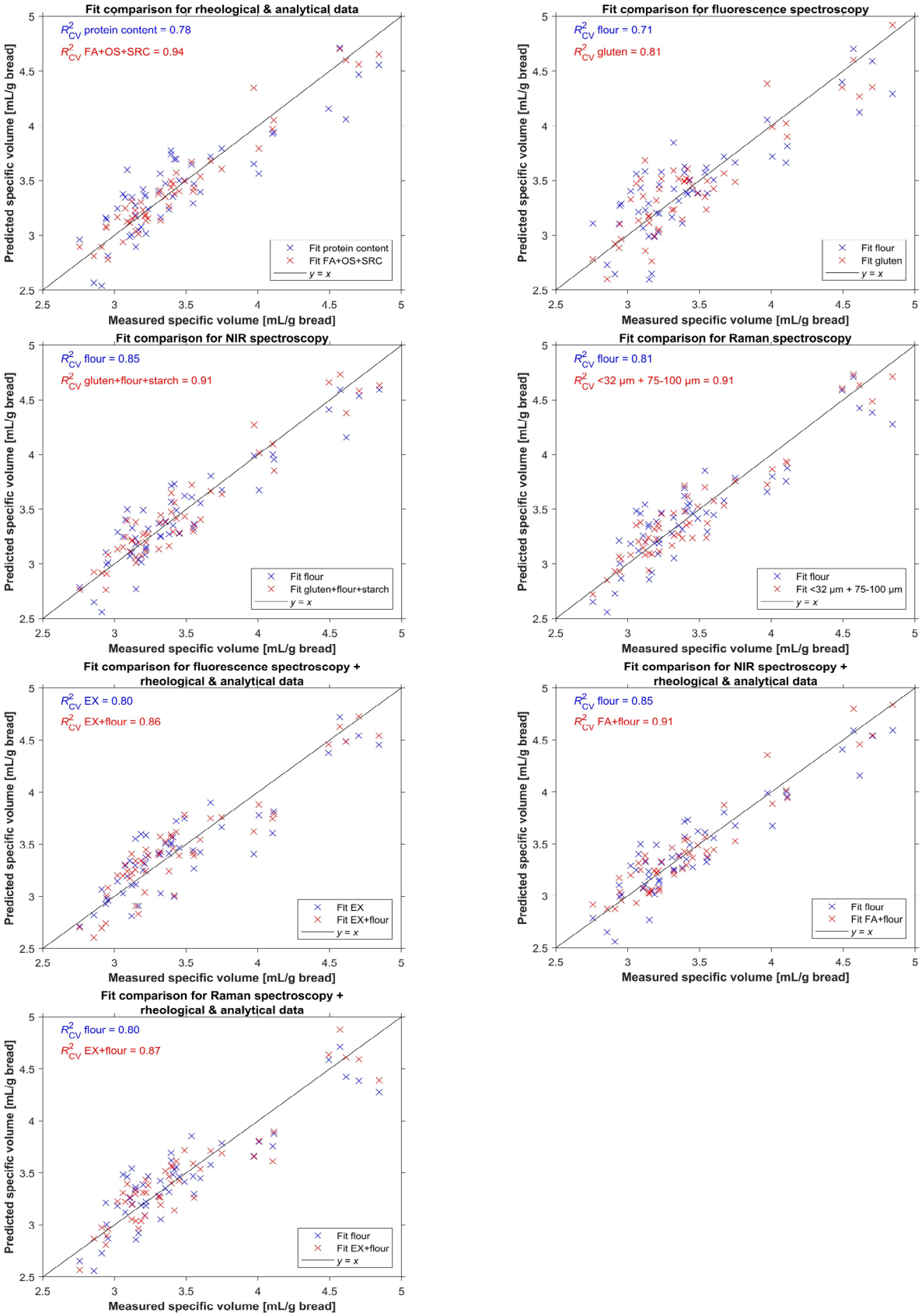

Fig. 3 Measured vs. predicted values of cross-validation for best models predicting specific loaf volume using spectroscopic, rheological and analytical data of wheat as well as combinations thereof

Conclusion

This study investigated the potential of three novel approaches to improve the prediction of wheat baking quality for commercially available wheat flour samples. Baking quality was characterized by the specific loaf volume obtained using an optimized baking test that focused on optimal hydration and dough development for every flour sample. Different data fusion methods and their influence on the predictive performance of models were tested. The three approaches were based on building regression models using a large variety of different rheological and analytical tests as well as fluorescence, NIR and Raman spectroscopic analyses of flour, flour fractions and dough.

All three novel approaches showed great potential to improve the prediction of baking quality compared to the currently used indicator protein content and also compared to regression models just based on flour spectroscopy. For all three approaches, the best models achieved R^2_{CV} of 0.91 and higher. Combining different data types (rheological and analytical subsets, spectra or both) yielded improved predictions. This shows the importance of data fusion for the prediction of baking quality, as various measurement methods can provide complementary information. The results also showed the high importance of rheological measurements for the prediction of baking quality, because many different rheological tests, with farinograph, extensograph and alveograph being the most important ones, always contributed to the best regression models. In addition, flour fractionation significantly altered spectral signals, enabling an improved prediction quality although the exact underlying mechanisms remain unknown.

For the optimized baking test used in this study, very good prediction models were generated based on a large variety of different rheological, analytical and spectroscopic methods that all claim to capture specific flour quality characteristics. Also, a good correlation of specific loaf volume and protein content was achieved with an R^2_{CV} of 0.78 in linear regression. The fact that a large number of these flour characteristics captured by the many different measurements were successfully related to the baking test result shows that this baking procedure was able to adequately capture the breadmaking potential of the flour samples used. This emphasizes the importance of an optimized standard baking test in the future and shows that the baking test method should always be considered when comparing results of different studies.

In future studies, the three novel approaches presented here can be tested using a larger number of flour samples and potentially also other types of regression models. Furthermore, studies focusing on the influence of specific parameters or wavelengths on the prediction result could give more insights into the relationship between flour quality, as captured by different measurements, and baking quality. In general, all three approaches show great potential for practical application, especially because many different predictor variables can be used for modeling and models can therefore be adapted depending on available instruments. A decision can also be made as to whether simpler models or more complex models should be used, depending on the amount of flour, manpower and time and the number of devices available and what prediction accuracy is to be achieved.

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Declarations

Conflict of interest The authors declare no conflict of interest.

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