We thank the reviewers for the constructive comments and remarks. We included the remarks in the revised version. Below are our responses to the reviewers’ comments and the protocol of changes.

Reviewer 1:

The article presents environmental scanning electron microscopy results as a complementary technique to scattering. In particular they study several non-dairy plant-based products with increasing fat content (milk, yogurt, cream cheese and cheese) as a function of the relative humidity and compare the ESEM to SAXS and discuss the complementarity to neutron scattering. Small angle scattering and in particular SANS is a very powerful technique for the structural characterization in soft materials, including industrially relevant materials such as the ones presented in this article. However, it has a mayor drawback which is that the results interpretation in subject to modelling and thus requires in most cases the use of complementary techniques. In this sense ESEM is a powerful technique and these results can be published as an example to other users who might see the potential use in their investigations.
There are several areas where the authors can improve the article, which are described in the following points:
• The phase behavior of the samples as a function of humidity should be specified. Many milk products phase separate upon drying and when studying the surface structuring upon drying it would be best to know if that is part of the sample itself or rather an upper phase resulting from creaming.

This question mainly plays a role for the ESEM images of plant drinks. The “high dry matter” products mainly lose water at the surface, leaving the structuring objects visible. We think that it is similar for the liquid drinks. A long term phase separation did not take place in our point of view, since the emergent structures disappeared again after re-humidification. In the ESEM part of 3.1 we included accordingly sentences describing this in more detail.

If creaming and destabilization would take place in milk and milk drinks, any process linked to increased dry matter content like downdraft evaporator before spray drying of emulsions would lead to destabilized emulsions, thus spray drying would be not sufficient and linked to extreme oxidation processes. However, that is not the case. Since we deal with commercial products, we believe that the drying conditions/ humidity changes are quite intermediate, thus we do not expose the liquid products to extreme environmental stresses causing creaming.

• The ESEM images are not analyzed at all. It would be ideal to present a size distribution of the observed features (in form of a histogram) or at least draw on them the feature size which is referred to in the text.

We thank the reviewer for this comment. We tried to analyze the pictures in more detail now. The globular features, where applicable, have been analyzed now in terms of size distribution, the “fractal surfaces” have been described in the text in a more detailed way. The results (mean, std dev.) are incorporated in the text, since histograms over a rather limited number of particles did not show much more details in our point of view. The corresponding particles discussed have been marked with arrows on the pictures.

• The comparison of the surface structure to the bulk SAXS structure should be discussed a little better. At least it should be indicated if the structure at the surface is representative of the bulk structure or they are disconnected. For instance, what I vs q slope would be expected from the type of structures observed in the ESEM micrographs and how do these compare to the slopes obtained from SAXS.

A paragraph discussing the differences in length scale and temperature/pressure/humidity conditions has been inserted at the end of section 3.1.

• In the outlook concerning neutron perspectives, it would be useful to know what kind of features are expected to be measurable that could not be accessed with SAXS in order to make a stronger case of the complementarity.

This section has been extended providing more comments on the complementarity to SANS

Minor questions:
Is the ESEM available to users?

Yes, the ESEM is accessible to users upon arrangement with the corresponding lab responsible.

The last two paragraphs of section 3.1 seem misplaced or out of context after the description of the SAXS results.

The sentences were indeed partly duplicates or misplaced, they have been removed or shifted (the liquid emulsion discussion sentence).

The caption of Figure 6 should include the bottom image.

Has been included now.

Figures of ESEM results should state the temperature of the experiment. Why is Figure 5 presented in terms of pressure instead of r.h.?

It should also be presented in terms of r.h., this has been corrected. The temperature of the peliter stage was 2-5°C for all ESEM images (section 2.1).

Revise the sentence: “The series of pictures are presented in Figures 1 and 2 for the liquid emulsions and Figures 3,4 and 5.”

Corrected.

Reviewer 2:

Since the same authors refer to a paper they recently published (Ref. [15]), duplication should be avoided. The samples, instruments and methods are already described in detail in Ref [15]. So this is nothing new. For better comparability, the same sample names as in Ref [15] should be retained and no new ones should be introduced. Table 1 on page 2 can therefore be omitted.

In terms of readability, we prefer to shortly explain the state of the samples presented here, we think this increases readability. Also, a description of the performed experiments is in our opinion required to make this paper readable on its own.
The results are not duplicates from ref 15, variation of temperature (SAXS) or r.h. (ESEM) are not shown in Ref. 15. Here our aim is to showcase a bit the possibilities and difficulties of such experiments.

In general, it should be made clear that the sample conditions in the ESEM analyses are completely different from those in the SAXS analyses. The ESEM analyses are carried out at a pressure of 1 - 8 mbar, while the SAXS analyses are carried out at 1,000 mbar. The SAXS measurements were carried out at 5 ⁰C and 25 ⁰C, but the ESEM measurements only at about 5 ⁰C.
The relative humidity in the ESEM measurements varies between r.h. 15% - 100%, whereas in the case of the samples enclosed in quartz capillaries, an r.h of 100% can be assumed for the SAXS measurements.
In addition, the SAXS measurements show information from the bulk of the sample, whereas the ESEM measurements only reflect the surface structure. That means, the drastic changes in morphology shown in the ESEM images when reducing the r.h from 100% to 50% could simply be due to "drying out" effects. Therefore, this publication should also show how the surface of the samples in the ESEM changes at constant r.h. of 100% changed over time and an intermediate step at r.h. 75% should be inserted. Since the measurements take place at 1 - 8 mbar, despite r.h. 100% incrustation or shrinkage of the surface cannot be ruled out. That is, what is responsible for the changed surface morphology? The low pressure in the ESEM, or the variation in r.h. ?

Both are somehow related to each other. In the current experimental setup, this cannot be separated. However, all samples were exposed to the same changes in pressure and humidity, a comparison between the samples analyzed ESEM is possible. Especially for the liquid systems, capillary effects should be taken into account. We talk about more than 80% of water, we won’t have a crust at the surface. There will be always an equilibrium of water between sample, its surface and the surrounding phase in the ESEM instrument. Also capillary effects and equilibrium between sample and surrounding play a role in the case of all samples. We hope our point of view is clearer now in section 3.1.

A real comparability of the SAXS and ESEM measurements can only be carried out to a very limited extent. (Would it be possible to carry out SAXS and ESEM measurements under comparable sample conditions?) I.e. SAXS and ESEM experiments show completely different sample conditions and thus different effects, which should be mentioned in the introduction or discussed in "3 Results and Discussion".

ESEM and SAXS are used as complementary methods. The results can not be compared directly, but both methods explain the structure in a broader way than one method alone. The revised version addresses these points in a more detailed way, pointing out the differences in ESEM (surface) and SAXS (bulk) measurements and the different conditions of the samples.

The ESEM images in Figures 4 and 5 show that the drying process is not reversible. I.e. after r.h. 15% back to r.h. 100 % the structures are preserved. This should be discussed. (Reasons). Does this effect also exist with samples PD2 and PY?

Yes, also PD2 and PY recover fully to a smooth surface when going back to 100% r.h., this has been described in the paper as well. The details of what is visible is difficult to say, since the full composition of the commercial products is not known in detail. Only the characteristic length scales can be obtained with these techniques presented here. The conclusion also addresses the need to investigate well defined model systems along the consumer product samples.

In Figure 2, 4, and 5., all figures should be of the same scale and, if possible, show the same position.

This has been tried as much as possible, sometimes due to charging it was not possible to acquire all pictures under the same conditions.

It should be worked out to what extent the results from the ESEM analyses are complementary to those of the SAXS measurements and possibly confirm or contradict each other.
For this it would be useful to show ESEM images with higher resolution. For example, Ref [15] shows ESEM images from other samples that reveal structures < 200 nm and thus overlap with the SAXS size range. This should also be shown with the samples discussed here. The SAXS measurements from Ref [15] also go further into the smaller Q range, which should also be shown here in order to compare them with features down to 200 nm in size from the ESEM images.

Low Q SAXS data are unfortunately not available for all samples (that is the USAXS setup of KWS-X, which was not used for all samples). Since the samples are not storable, this can not be repeated on the very same samples.
The length scale range in ESEM is complementary between SAXS and ESEM, and lower than in Ref 15, but it shows best the changes with relative humidity.

In general, the ESEM images could also be subjected to an analysis (grain size, 2D-FFT...) with suitable software in order to obtain feature sizes and size distribution. Then they would have more meaning. At the very least, the different morphologies should be discussed. Thus, sample PD1 and PCC show at r.h. 15% globular structures, whereas sample PC and PD2 have sharp-edged structures. Sample PY shows the weakest surface change. What are these effects due to? (fat content, type of fat, emulsifier, etc.?)

A coarse particle size analysis has now been included in the text. We tried a FFT analysis, but it was not straightforward since the images represent a 3D view of a surface and not, as in TEM, a 2D projection of the electron density. Also artefacts from charging prevented a FFT analysis. We therefore think it is not easily feasible in an artefact-free way and abandoned it.

The individual features of the SAXS measurements have already been analysed and discussed in detail in Ref. [15] (however, the temperature specification is missing here. 5 ⁰C or 25 ⁰C ?). In the draft presented here, the focus is on the smaller Q range (further away from the ESEM range) and on changes from 5 ⁰C to 25 ⁰C. However, these effects are only described phenomenologically here. A discussion is missing why, for example, the Bragg peak (A, size 4 at Q approx. 2\*10-1 Å-1 ) is more pronounced at lower temperatures, while the increase in crystallinity (atomistic distances that arise from transient glassy states of the fat molecules ) can only be found in the sample PCC for very large Q. A second x-axis in the SAXS measurements, which indicates the feature size or s-space according to 2π/Q, would be helpful.

A length scale axis is not shown, since it might be misleading especially with Bragg peaks from crystalline structures. We therefore prefer to stay with the reciprocal axis.
The discussion of what causes the different structural observations is difficult at the moment for commercial food products, this is only at the beginning of further investigations. This discussion has been improved.

In summary, this work shows different features and effects for plant-based emulsions, namely:
\* Large surface structures > 1 µm with ESEM at < 8 mbar, r.h. 15% - 100% and 5°C.
\* Small bulk features < 100 nm with SAXS at 1,000 mbar, r.h. 100% and 5°C or 25°C.
To what extent are these measurement conditions and observed effects relevant for the food industry?
The observed effects should therefore be discussed more scientifically and not just described.

Studies of plant based food products is a rather new topic and only scarce information are available so far. The use of commercial food products where not all details about the samples are known limits the results to a descriptive discussion.

The main aim of this paper is to show some experimental aspects of what can ESEM and SAXS be used for in such systems. For the future, we aim to use the methods shown in Ref 15 and the current proceedings article to explore the meaning for the food industry. We analyze the structure via ESEM and SAXS, the structure affects the mouth feel and sensory acceptance of the consumers. Sensory studies do not include a structural resolution of the samples like ESEM and SAXS. This motivation is very well explained in Ref 15. Later on, we aim to compare sensory aspects (mouthfeel,…) with SAXS and ESEM studies. We could explore, what structures are linked to what ingredients and to what kind of sensorial impressions, thus consumer acceptance. These correlations need to start with model systems, and complexity must increase with time and number of studies. Ref 15 and this study, are kick off studies.

An overlap of the ESEM and SAXS measurement ranges under identical measurement conditions could increase the significance.
The title of the work should include both measurement methods ESEM and SAXS.

SAXS has been included into the title.

Further errors:

Page 1:
ESEM images were obtained at JCNS MLZ using a Thermo Fischer Quattro S ESEM (Thermo Fischer Scientific,
Eindhoven, Netherlands) equipped with a 500m (???) aperture gaseous secondary electron detector (GSED).

(µm, corrected)

During observation, the temperature of the samples was controlled with a Peltier cooling system. \*)

\*) not the sample temperature, but the temperature of the ESEM stage is probably controlled. The sample temperature is not measured. Since the sample is far away from the temperature control of the stage, its temperature might be higher than 5C.

The sample stage was controlled, this has been corrected. Prior to each measurement, the samples were equilibrated for 15 min to reach the temperature.

page 2:
All ESEM experiments were carried out under the following operating conditions: beam accelerating voltage
10 kV for samples PD1”Drink1”, PD2 ”Drink1”,PCC and PC, 5 kV was used for sample PY to diminish beam damage, spot size 3.0, \*\*) sample working distance 6.7 +/- 1.2 mm, and and 5–10 us dwell time.

\*\*) it is better to indicate the actual sample current (measured in the Farraday cup) here. Spot size says nothing.

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The WetSTEM stage used for the presented results doesn´t have a Faraday cup as it is meant to receive a STEM detector below.

The sample currents measured in the Faraday cup of the standard sample at high vacuum were

80 pA and 110 pA for 5 and 10 kV respectively, which we think is also valid for the ESEM stage given the relatively short working distance. This has been included into the ESEM section.