

## Author responses to comments of Referee #1

We would like to thank the referee for the effort and time they put in to comment on our manuscript. We are grateful for their careful and considered comments and will make every attempt to fully address these comments in the revised manuscript.

In the following list, the points raised by the referee are written in bold characters, whereas our responses are shown in regular characters.

This manuscript presents a thorough imaging and modeling study on pore networks in peat samples collected at different depths from a drained peatland, and uses this analysis to discuss implications of peat porosity and pore network characteristics on gas transport and methane production in peatlands. The manuscript is well written and easy to follow. The methods are explained well, although more details of error analyses and impacts of errors on the results, as well as some reconsideration of the statistical analyses would be needed (see specific comments below). My main concern about the presentation is that the results seem mostly intuitively obvious. It makes sense that porosity increases in peat towards the surface just because of how increasing pressure would influence and deform peat structures the deeper one goes in the soil layer. Maybe this has not been shown with the methods used in this study before, but the manuscript fails to communicate the state of the art in this kind of studies. This, combined with that one of the main results, the hysteresis behavior, seems to be just a model result with no verification based on the experiments, and the superficiality of the discussion on the connections between the porosity and network parameters and their impact on methane emissions and gas transport makes it hard to understand the importance and impact of the study. Was the goal to demonstrate what the used techniques can do? Or was it more to showcase a way to understand CH<sub>4</sub> emissions?

In mineral soil, porosity decreases with depth because of compaction caused by overlying matter. However, in addition to physical factors such as pressure, air-filled porosity at a specific suction in peat is governed by the degree of peat decomposition. There was no decreasing trend in total porosity with depth (Table 1), which is consistent with other studies on drained peatlands (Minkinen and Laine 1998). With increasing peat decomposition, larger pores fracture and collapse and the volume fraction of smaller pores increases. Therefore, macroporosity (air-filled porosity under low suction conditions) decreases with depth. The degree of decomposition generally increases with depth, which decreases the macroporosity in deeper peat (Rezanezhad et al. 2016). We have discussed this fact in Discussion (Sect. 4.2), but we will include it briefly also in the Introduction.

We use more advanced methodology for the investigation of peat porosity and structure than most of the previous studies on the issue. The description of the pore space obtained through pore network analysis is more detailed in our study than in studies that have focused on the bulk properties of peat. The fact that our results are in line with an intuitive impression and the results of previous studies confirms the applicability of our method. A more detailed knowledge on pore space connectivity obtained by our method is needed for a better description of transport dynamics in peat.

Hysteresis, the dependence of the state of soil water on the process leading up to it, is a well-known phenomenon in soil science (Hillel 1998). It is observed as the difference between the volumetric water content of soil at a specific suction measured in sorption (wetting of soil when the water table is rising) and in desorption (drying of soil when the water table is falling). The experimental determination of the sorption curve is much more difficult than the determination of the desorption curve, and therefore it is seldom done. We show that pore network modelling can be successfully used to reproduce the hysteretic behavior of water retention and characterize the hysteresis effect in peat. Air-filled porosity is directly related to volumetric water content, and thus the evolution of connected pore space is a direct consequence of the evolution of water content. Therefore, hysteresis of air-filled porosity can be induced from hysteresis of water content. We will include a brief description of hysteresis in Introduction.

The aim of our work is to study the capability of complex network theory methods and metrics to characterize the physical structure of peat and its pore space and to qualitatively assess the obtained metrics and correlations between them in relation to gas transfer processes in peat. The study also shows the physical existence of anaerobic pockets and illustrates their capability to control the production and emission of gases from peat. A more thorough comparison between the pore network modelling method and experimental methods related to gas diffusion efficiency in peat will be the subject of further studies. We will reformulate and clarify the aims of the study in the last paragraph of the Introduction. We will also restructure the Discussion and Conclusions so that the conceptual nature of the assessment of methane-related processes will be indicated more clearly.

This same unclarity in focus is apparent from the figures. Figure 3 is clearly a model vs. measurement comparison, but it is discussed in the results as a figure presenting characteristics of the peat samples. Figure 4, on the other hand, is presented in a format that emphasizes differences in the peat sample properties, but is discussed in the text as a model-measurement comparison. To improve the presentation, I suggest that the authors clearly define the purpose of the study, and then build the text and presentation of data in the figures to support that goal. For example, to support the statements about Fig 1, the changes in porosity between depths could be more effectively presented as a box or bar plot with appropriate error bars and statistical analyses. And for fig 4, the model-measurement comparison would be presented more clearly by an x-y -plot.

The discussion on Fig. 3 in Sect. 3.1 is intended to be focused on the comparison of the two methods of determination of the air-filled porosity of the samples, that is, on the assessment of the performance of image analysis. The procedures of the determination of air-filled porosity through water retention measurements and  $\mu$ CT imaging are different. The water retention measurements yield the volumetric water content of soil, and the air-filled porosity is then deduced using the estimated value of total porosity. By contrast, air-filled porosity is the quantity that can directly be determined from the binarized  $\mu$ CT images. Because the volumetric water content is the actual measured quantity and the air-filled porosity is a derived quantity, it is more reasonable to analyze the volumetric water content statistically. The changes in volumetric water content between depths are presented in Table 1 with statistical analysis.

The latter half of the discussion in Sect. 3.1 is not related to Fig. 3 but considers the effect of the pore network generation process on the volume of the pore network in comparison to the void volume of

the corresponding image section. The network volumes were smaller than the image section void volumes, which affected the results of the water retention simulation.

The graphs in Fig. 4 show the evolution of air-filled volume as a function of matric potential and the approximate shapes of the porosity–soil water potential curves. The graphs illustrate the variation between samples as well as the differences between simulation and measurements visually and explicitly. Fig. 4 is intended for qualitative analysis of the performance of the water retention simulations and of the pore network generation process behind them for each individual sample. Because the network domain used in the water retention simulations was smaller than the volume of the samples, it is not reasonable to accurately compare the measured and simulated values presented in Fig. 4 in more detail. Also, the measurement-based air-filled porosity values in Fig. 4 are not precise but estimates.

We will restructure, extend, and clarify the discussion in Sects. 3.1 and 3.2.

For the discussion, I suggest, again, focusing on the goal of the study. If the goal is CH<sub>4</sub> production and transport, then some simple calculations about the impact of the parameters measured here on the transport and emissions would be helpful. As it is, the discussion reads like a collection of separate paragraphs where the importance of more complex network parameters such as tortuosity or top-bottom betweenness centrality for CH<sub>4</sub> production and gas transport are left unexplained, while some relatively obvious results, like that higher porosity lead to higher connectivity (lines 436-444) are discussed in length.

The aim of the study is to qualitatively characterize the structure of peat and the changes in the structure with depth using methods adopted from complex network theory. Issues related to dynamic properties and processes and to comparison with experimental methods related to gas diffusion efficiency in peat will be the subject of further studies. We analyze how different network metrics are capable of describing network connectivity and how (1) porosity and connectivity measures and (2) local connectivity measures and global connectivity measures are related in a complex peat pore network. We state that the connection between porosity and average local connectivity may not always be straightforward in peat pore networks. Local connectivity may be high even if porosity or global connectivity is low. This might be the case, e.g., if the connected pore space does not extend over the whole network domain. We found that the applicability of local connectivity metrics to describe global network properties is dependent on the shape and topology of the connected pore space.

Geometrical tortuosity is, by definition, a direct proxy for network transport efficiency as it illustrates the tortuosity and length of flow paths through a network. Centrality measures (closeness centrality, betweenness centrality) are more advanced metrics adapted from complex network theory. Using geometrical tortuosity as a proxy, we could estimate how well the centrality measures and other network theory metrics are able to describe the transport properties of a network. Top-bottom betweenness centrality is, as it is defined in our study, an adaptation of the network measure betweenness centrality. Because of its direction-based definition, top-bottom betweenness centrality is more directly related to transport processes through a network than the other network theory metrics in the study. We will discuss further on this role of geometrical tortuosity in the performance

analysis and on the nature and relevance of top-bottom betweenness centrality in the Discussion section.

Specific comments:

Line 14: What was the hysteresis in relation to? What were the parameters that showed the hysteresis behavior?

The hysteretic behavior was seen as the difference in the volumetric water content of a sample at a specific soil water potential between drying and wetting processes. The effect is also reflected in the volume fractions of air-filled pore networks in Fig. 9. Under the same pressure conditions, the volume fraction of water in the pore space was considerably smaller during wetting than during drying. We will clarify the context in the Abstract.

Line 23: Please be more specific about how peatlands modulate or are modulators of hydrological and biogeochemical cycles. Why are they globally important?

Vast quantities of water are stored in peatlands, and they may either accelerate or attenuate flooding in watersheds (Holden 2005). Peatlands store one third of global soil carbon (Kang et al. 2018), and boreal and subarctic peatlands store 300-400 Pg carbon as peat (Turunen et al., 2002), which corresponds to nearly half of the carbon currently held in the atmosphere (Limpens 2008). A large proportion of dissolved organic carbon in freshwater ecosystems originates from watersheds with peatlands (Kang et al. 2018, Asmala et al. 2019). We will clarify the reasoning at the beginning of the Introduction.

Line 28: The line starts with repetition.

We will reformulate the related sentences and remove the repetition.

Line 39: Capability instead of characteristic?

In soil science, the functional relationship between soil water potential and soil moisture is called, e.g., the soil-moisture retention curve, the soil-moisture release curve or the soil moisture characteristic (Hillel 1998); the water release characteristic (Townend et al. 2001); the soil-water retention curve (van Genuchten 1980); or the water retention characteristic (Raats 2001).

Line 96: Why was the diameter of the sample only measured to determine shrinkage? Where the samples all the time in the plastic tubes? Why was the vertical shrinkage not considered?

As the manuscript text states, also the height of the sample was measured and the vertical shrinkage was considered. In most samples, vertical shrinkage was greater than horizontal shrinkage. The samples were not removed in the acrylic tubes during the water retention measurement process.

Line 104: How good is the value of 1500 kg m<sup>-3</sup> for particle density for these samples? Does it accurately represent all the sample depths?

The value of 1500 kg m<sup>-3</sup> is a widely used reference value for particle density in boreal peat soil (Paavilainen and Päivänen 1995). The variation in particle density with depth (classified by bulk density) is very small in peat (Päivänen 1973).

Line 106: The assumption that air-filled porosity was zero in the saturated state is probably not quite true. Please give an estimate for how much that might be off in your samples and what kind of an error it would lead to in your results.

The mass of the saturated sample was not used in the determination of the total porosity, as the total porosity was calculated using the measured dry mass and estimated particle density (Eq. 2). Therefore, the assumption that air-filled porosity was zero in the saturated state did not affect the air-filled porosity calculations at other matric potentials. The assumption was only reflected in the fitting of the van Genuchten model, where we separately state the saturated water content was assumed to equal the total porosity that we had estimated. This assumption was also used in Weiss et al. (1998). In addition, the assumption was used in Fig. 4 as the air-filled porosity value at -0.1 kPa matric potential, but this value is not essential in the analysis of the water retention properties presented in the figure. Because the assumption is used only for illustrative purposes in Fig. 4, we will remove the statement from this section and state the choice and give reasons for it in the caption of Fig. 4.

The deviation from total saturation is naturally dependent on the method of sample saturation. An estimated range of the air-filled porosity in the saturated state in laboratory conditions is about 2...3% (Päivänen 1973). The larger the deviation from zero air-filled porosity in the saturated state, the smaller are the values of the parameters  $\alpha$  and  $n$  in the optimal fit. However, because the parameter values are not of primary importance in the study, we consider that a more thorough error analysis would be beyond the scope of the study.

Eq 3. Why is this equation needed? Why is not the calculation using eq 1 not enough for the comparisons? How was this equation fitted to data? What were the fitted variables that were measured, and how were they measured?

The water retention properties of soil are characterized by the water retention curve, that is, the functional dependence of volumetric water content on soil matric potential. The functional form of the dependence is used in process-based models to characterize the properties of soil in different wetness conditions. The van Genuchten parameterization of the water retention curve (van Genuchten 1980) is widely used in soil science, and the van Genuchten parameters can be used to classify the soil properties and illustrate the characteristics of a specific soil sample.

The fitted variables were soil matric potential  $\psi$  and volumetric water content  $\vartheta$ . The determination of  $\vartheta$  is described in Sect. 2.2 and the formula is presented in Eq. 1. The soil matric potentials were equivalent to the pressures applied to the peat samples pressure plate apparatus. The fitting was performed for the average values of  $\vartheta$  of the samples from each depth by applying the Levenberg–Marquardt algorithm using the Python library SciPy designed for scientific computing. As the result of the fitting, a function  $\vartheta(\psi)$  was obtained by determining values for the parameters  $\alpha$  and  $n$  in Eq. 3.

Line 113-114: Why is it important to tell that the pressure range was so narrow that the fitting procedure needed to be modified? Why was a larger pressure range or a more appropriate fitting algorithm not used?

The study was focused on macroporosity (pore diameter > 100  $\mu\text{m}$ ) because smaller pores are usually water-filled in peatlands where the water table depth is generally less than 1 m; thus, gas diffusion in

air occurs only in macropores. In addition, it is the pore size range that can be investigated through  $\mu$ CT imaging so that the sample size remains reasonably large. Macropores are supposed to be emptied of water at a matric potential of -10 kPa; therefore, the retention measurements were performed up to -10 kPa potential. The shape of the van Genuchten retention curve is such that this potential range does not cover the whole shape, therefore the end points must be fixed. This also fixes two of the model parameters, the saturated water content and the residual water content. The main focus of the work was not to study the retention properties of peat on a larger range, and the retention model fit was a supplementary result in the study. We will elaborate the description in the text.

Line 118: Please move “noninvasively” to the end of the sentence.

We will correct the word order.

Line 123-125: Please give an error estimate for what this darkening might do to your results.

We state in the Discussion (Sect. 4.1) that the darkening of the  $\mu$ CT images due to image reconstruction defects near the top and bottom of the sample increased the void fraction slightly in all binary images. It is very difficult to give a quantitative estimate of the effect of the darkening on the obtained void fractions of the images. This error only concerns the air-filled porosity estimates of the whole samples presented in Fig. 1 because the top and bottom regions were excluded from the pore network domains.

Line 136: How did you deal with that the contrast between air-filled and water containing regions or organic materials was low? How did this affect your results? What kind of and how large an error could it cause?

We used a standard binary segmentation algorithm, Otsu's method, to perform the solid-void classification. This way the segmentation result contains a minimal amount of operator bias. The fraction of void space in the image may have been over- or underestimated depending on if the threshold intensity was high or low, respectively. The magnitude of the error is very difficult to estimate. Further, each stage in the workflow from  $\mu$ CT imaging to pore network generation is prone to contain some inherent discrepancy. We find that a thorough analysis of image segmentation and the sources of discrepancy in it is beyond the scope of this study. We will, however, return to this issue qualitatively in Discussion.

Line 154: Why did you use the sum of distances? It is hard to visualize how this calculation was performed and what it means.

The centroids of adjacent pores and the centroid of the throat between them are usually not collinear, and the linear distance between the pore centroids is therefore smaller than the length of the route from the centroid of pore 1 through the connecting throat to the centroid of pore 2 (see Fig. AR1). We will clarify the definition.

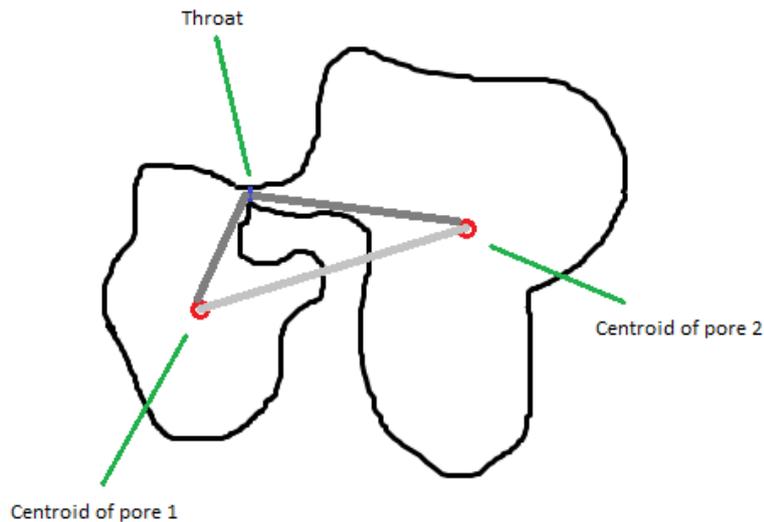


Figure AR1. Schematic 2D illustration of two pores and a throat. The linear distance (lighter gray) is smaller than the length of the actual route between the pore centroids through the throat (darker gray).

Line 158-160: Does this mean that the shrinkage of the samples was not taken into account here at all?

It was assumed that the peat sample filled the acrylic cylinder completely in the initial, saturated state. The void volume, that is, the volume inside the boundaries of the cylinder not occupied by the peat matrix, was completely filled with water (and a little amount of air). When the sample started to dry and compress, the pore spaces inside the sample started to collapse and become smaller. This means that the air-filled regions inside the sample were effectively transferred into the space between the sample and the cylinder boundaries. The images were taken at -10 kPa matric potential when the sample was shrunk. If it is assumed that the solid volume remained constant during the shrinkage process, the volume of the non-solid matter inside the boundaries of the cylinder also remained constant. If the volume fraction of air-filled space is now determined with respect to the total volume of the cylinder, the shrinkage of the samples will be taken into account. We will clarify in the text that this procedure ensures that the effect of shrinkage is included.

Line 164: What does the “largest of these clusters” mean? How was size defined? How does taking this part under analysis solely affect the interpretation of the porosity and connectivity results from the samples? Can this size be somehow standardized between samples to make them comparable? Can there be more than 1 large enough network in a sample that all of them should be considered?

The size of the pore clusters was defined by their volume. The largest connected cluster was the cluster that extended through the network domain in the vertical direction; other clusters were not connected to both open ends of the network domain. As is seen from the results in Sect. 3.1, the largest cluster contained on average about 90% of the total pore space volume. Therefore, the transport processes that occur in the pore system are restricted only to the largest cluster, that is, the connected network. The connected network is the relevant space when assessing the transport properties of the pore system. The impact of this limitation is discussed in Results in Sect. 3.1: the

network volumes were smaller than the corresponding image void space volumes. If there were two or more connected networks that extended through the whole domain, all of them should be included in the analysis, but this was not the case in this study. We will emphasize in the text that the largest pore clusters were the only pore clusters that extended through the network domain in each pore system.

Line 202: See previous comment. How does the defined region affect the connectivity analysis. How can you account for non-connectivity in a standardized sample size?

Due to computational limitations, we needed to assume that the selected subregion was representative of the whole sample and that the generated network was representative of the network that would have been generated using the whole sample as the domain. The largest cluster contained on average about 80% of the total pore space volume of the cubical-domain networks and was always the only cluster that extended through the network domain in the vertical direction. Therefore, the smaller pore clusters would not have taken part in transport processes through the network. We will clarify the reasoning for the selection of only the largest cluster for network analysis and the dominance of the volume fraction of the largest cluster in the text.

Line 234: Please explain more clearly how the one-way anova analysis was performed. Were all the depths analyzed simultaneously or did you perform pairwise tests between each. Would a multi-way anova with interactions included or a version of linear models give you more comprehensive results?

We determined if (1) water retention characteristics differed between depths, (2) if pore sizes differed between depths, and (3) if network metrics differed between depths. We thus analyzed the influence of one categorical variable (depth) of one continuous dependent variable at a time. There was only one categorical variable, and the use of a multi-way ANOVA was therefore not possible. The main interest was the variability of each of the listed properties with depth. There may have been interactions between variables, but the number of degrees of freedom was too low for a more thorough analysis. Also, obvious correlations between independent variables (especially water retention characteristics and pore sizes) may weaken the power of linear models. We will clarify the description of ANOVA in the text.

Line 242: Figure 3 actually shows a consistent underestimate of porosity by the model.

The void fractions obtained from the images were indeed slightly lower than their measurement-based counterparts. We will change the wording.

Line 254: Does water content here refer to the original samples, or saturated samples or dried samples, and at what depth?

As it is stated in the text, water content in all studied matric potentials was clearly lowest in the top layer and highest in the deepest studied peat layer. Water content refers to the samples in each state during the retention measurement. Water content was lowest in the top layer in the -1 kPa conditions, and it remained lowest in all the progressively drier (-3 kPa, -6 kPa and -10 kPa) conditions. Correspondingly, water content remained highest in the deepest layer in all matric

potential conditions. The water content of the original samples was not measured, and the water content of a dried sample is zero by definition.

Line 260: The hysteresis effect seems to be only a modeling result. Is that a feature of the model, or is there real experimental evidence for that this hysteresis happens?

The phenomenon that different water saturations are reached at the same capillary pressure, depending on whether the wetting fluid (wetting/imbibition/sorption) or the nonwetting fluid (drying/drainage/desorption) is the displacing phase, is a well-known issue both in porous media in general (see Dullien 2000) and in soil (e.g., Haines 1930, Poulouvassilis 1962). The experimental determination of the sorption curve is much more difficult than the determination of the desorption curve, and therefore it is seldom done (Hillel 1988). For most practical applications, only the desorption curve is used and the effect of hysteresis is ignored (Townend et al. 2001). Because air-filled porosity is obtained by subtracting the volumetric water content from total porosity, hysteresis of volumetric water content translates directly into hysteresis of air-filled porosity.

Figure 6: I think combining the cumulative volume figures to one figure where each depth is described by one average line and error bars (or error shading) would be a more effective way to present this data. It is not necessary to present data for individual samples here.

We believe that the figure is most illustrative in its current form. The panels c-e in Fig. 6 show the large variation of both the total network volumes and the sizes of the largest pores between samples.

Lines 371-380: How do the results of this study relate to this text?

We showed that the volume of air-conducting pathways in peat is dependent on its wetness and that the volume of the connected pore space decreases with increasing water content. An increasing fraction of the peat pore space therefore becomes isolated from the atmosphere, and these locations become favorable for anaerobic processes. We will restructure the Discussion and state more explicitly that we are assessing conceptual implications of the pore network structure of peat for methane-related processes.

Line 442: Can anything else happen in networks than shorter pathways leading to more alternate routes?

When local network connectivity was high (implying that a random pore was connected to several other pores), the average traveling distance from a random pore to another random pore was relatively short. When pore connectivity is higher, there are more alternative routes available between two random pores, some of which may be spatially rather straight. Therefore, a higher number of alternate routes is the cause and a shorter pathway is the effect in this case.

Lines 454-459: How is this related to gas transport of CH<sub>4</sub> production?

The ratio of the average clustering coefficient and the average coordination number may give insight on the efficiency of gas transport through a peat pore network. As it is stated in the text, the ratio of the two metrics may reflect the abundance of long and narrow pore conduits in the network, which may govern the number of alternative transport routes through the network. A low number of

alternative routes (a low ratio) may lead to a rapid decrease of gas transport if one of the routes gets blocked, and a high ratio may enhance the resilience of the network to local blockages. We will clarify the text.

Line 494-495: How do you get to the conclusion that orientation in diffusion paths does not change transfer towards atmosphere?

We meant that the diffusion paths are oriented in such a way that gas transfer towards the atmosphere is neither promoted nor restrained in deeper peat layers. We will correct the sentence.

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