Dear Professor Guadagnini,

We would like to take the opportunity to thank you and the reviewers for the insightful comments that led to improvement of our manuscript. Though it is challenging to satisfy an overly critical reviewer, we did our very best to revise the manuscript according to the reviewer's and your suggestions. Below please find a detailed response to all inquiries and a description of all changes made during the revision process.

We strongly believe that besides an excellent dataset that is now provided to other research groups in supplementary Table S1, the parameterization of air and water flow models with parameters that are directly derived from X-Ray CT observations is quite novel and as demonstrated also leads to significantly improved predictive capabilities.

We hope that our revisions are satisfactory and the manuscript acceptable for publication in HESS.

Sincerely,

Markus Tuller

#### Editor

**Comment 1:** The manuscript has been reviewed by two of the reviewers who were involved in the previous round of reviews. One of them is still very negative, his/her main point being linked with the apparent lack of predictive capability of the type of correlations they come up with. While this might be true, I urge the authors to clearly state this aspect in the body of the manuscript so that there is no ambiguity to it.

**Reply:** We tested the Revil Cathles (1999) model and various forms of the Kozeny-Carman equation; the latter was parameterized with pore parameters directly derived from X-ray CT observations for prediction of water and gas permeabilities and diffusivity. We found that the simplest form of the Kozeny-Carman equation proposed by Ahuja et al. (1984) with CT derived parameters predicted measured permeability and diffusivity data quite well. However, we are also aware that our study is only a first step, and further investigations are needed to confirm validity for a wide range of soil textural classes.

This has now been clearly stated in in the Materials and Methods section (Page 12 L10-21). Results are depicted in Fig. 9 and discussed in the Results and Discussions section. (P20 L22-25, P21 L1-12). We have now also clearly stated that the proposed model needs to be evaluated independently for different soil textures to confirm robustness (P21 L9-12).

**Comment 2:** The second reviewer still lists some major concerns about the PTF-based nature of the work as well as about the appropriate identification of the scale, which is of relevance to the study. The reviewer also lists a series of other points, which are relevant, even as they can be considered as minor.

**Reply:** In the first part of the presented study we evaluate the predictive performance of existing pedotransfer functions/models for saturated permeability, air permeability, and gas diffusivity and demonstrate their rather poor predictive capabilities. We acknowledge that it

has been previously demonstrated that water flow in macropores cannot be accurately predicted with empirical models from basic soil properties. However, at the same time we find that there is only very little published work related to gas diffusivity. Furthermore, existing pedotransfer functions/empirical models only consider basic soil properties and usually ignore important pore network characteristics.

In the second part of this study we derived novel macropore network characteristics for saturated permeability, air permeability, and gas diffusivity from X-ray CT observations and demonstrate their utility for improving accuracy of gas and water flow predictions based on the Kozeny-Carman equation.

We agree to the concerns about identification of the appropriate scale and have removed all references to the "field scale" in the revised paper. Furthermore, a disclaimer was added (P15 L22-25).

Finally, all minor comments were addressed as suggested (see response to reviewer 2 below).

**Comment 3:** After my own reading, I do concur that the data set can be of potential value for HESS, even as the model does not appear to have a convincing predictive capability. This said, I also noted that the details of the variogram analysis underlying the kriging application (for example, the type of variogram considered as well as the way its parameters are estimated, including a quantification of the associated uncertainty) are missing from the manuscript.

**Reply:** The entire dataset is provided to other research groups in supplementary Table S1. The details about semivariogram analysis are now provided in Table 2.

**Comment 4:** Incidentally, is the exponent "9" correct in Equation (1)? Just to make sure

**Reply:** Thanks for catching this error. Equation 1 was corrected in the revised manuscript.

**Comment 5:** All in all, I do concur with the less negative reviewer and I am willing to give the authors a very last opportunity to strengthen their work. Note that I am still recommending major revisions to be implemented, according to the spirit of my assessment above. In doing these revisions, the authors are also encouraged to avoid leaving out amendments which they claim have been undertaken. To me, it has to be clearly stated that the contribution of the work to the advancement of our knowledge is not too large and it is compensated by the presentation of the valuable data-set that the authors analyze. If this is acceptable to the authors, then I suggest they prepare a revised manuscript along these lines. The latter will then most likely be re-reviewed.

**Reply:** As mentioned above, it is always challenging to satisfy overly critical reviewers. We strongly believe that besides an excellent dataset that is provided to other research groups in supplementary Table S1, the parameterization of air and water flow models with parameters that are directly derived from X-Ray CT observations is quite novel and as demonstrated also leads to significantly improved predictive capabilities, mainly because directly derived pore network characteristics are considered. We think that the scope of our study with all its limitations is clearly stated in the last paragraph of the introduction as well as throughout the manuscript.

## **Reviewer 1**

**Comment 1:** The authors revised the paper and provided an improved version. Nevertheless, I am sorry, but I think that after these changes the manuscript still does not fully satisfy the quality standard for publication on HESS. The data processing yields a set of one-to-one functional relationships between CT-based indices and physical properties of the samples (saturated permeability, air permeability and gas diffusion at two different values of suction). My personal opinion is that this is still far from a "predicting model", which can be used to derive physical properties from structural data, including a description of the macropore region.

**Reply:** We tested the Revil Cathles (1999) model and various forms of the Kozeny-Carman equation; the latter was parameterized with pore parameters directly derived from X-ray CT observations for prediction of water and gas permeabilities and diffusivity. We found that the simplest form of the Kozeny-Carman equation proposed by Ahuja et al. (1984) with CT derived parameters predicted measured permeability and diffusivity data quite well. However, we are also aware that our study is only a first step, and further investigations are needed to confirm validity for a wide range of soil textural classes.

This has now been clearly stated in in the Materials and Methods section (Page 12 L10-21). Results are depicted in Fig. 9 and discussed in the Results and Discussions section. (P20 L22-25, P21 L1-12). We have now also clearly stated that the proposed model needs to be evaluated independently for different soil textures to confirm robustness (P21 L9-12).

**Comment 2:** Please, check the style for expressing values of quantities, in particular when a range of values is indicated (I suggest to conform to the recommendations of section 7.7 of the NIST guide to the SI; see <u>http://www.nist.gov/pml/pubs/sp811/</u>).

**Reply:** We have revised the manuscript accordingly. All quantities are now expressed in SI units.

**Comment 3:** I agree with comment #4 by Referee #3 of the first version: saturated permeability is independt of the fluid, so that "water" should be erased from the expression "saturated water permeability" throughout the whole paper. Along the same line, I suggest to denote this quantity with "ksat" instead of "kw", not only in the text, but also in the tables and figures. Notice that "saturated permeability" could be substituted with the symbol "ksat" at several places in the text.

**Reply:** The notation has been changed throughout the manuscript and in Figures.

**Comment 4:** Page 7, lines 21 to 24. Isn't it possible to provide a quantitative index to separate and label columns with biopore- or matrix-dominated flow?

**Reply:** It is challenging to suggest a quantitative index/cut-off point to discern between biopore-dominated and matrix dominated-flow based on X-ray CT observations. This is mainly because biopores are not isolated and usually connected with other smaller pores.

**Comment 5:** Page 9, line 10. Correct the dimensions of the pressure difference.

# **Reply:** Corrected (P11 L19).

**Comment 6:** Page 11. (a) Lines 7 to 9. The sentences "Although... distributions." can be erased, without loss of information. (b) Lines 20 to 22. The sentences "Large... distribution." can be shortened.

**Reply:** Corrected following the reviewer's suggestions in the revised manuscript.

**Comment 7:** Page 13, line 20. Substitute "pores" with "pore diameters". Similar expressions are used in other sentences and should be changed.

**Reply:** Corrected

**Comment 8:** Table 1. (a) Correct the measurement units.

(b) Are the significant digits used for each measured quantity coherent with the measurement resolution and accuracy? For instance, I am afraid that hydraulic conductivity cannot be measured with 5 significant digits. The same comment applies to the supplementary material.

(c) Please, check whether the number of significant digits for means values is coherent with the standard deviation.

**Reply:** Measurement units are now corrected in table 1. Now 2 significant digits are reported for saturated hydraulic conductivity. Table 1 and supplementary data are coherent now

#### **Reviewer 2**

**Comment 1:** The authors received comments on the matter of 'field scale'. They agreed to remove this term from the title, but the question points beyond the title. The study still suggests that this is field scale, while e.g. earlier studies (PTF studies) were concerned of a much smaller scale. The fact is that both types of studies are concerned of the same scale. Indeed, many PTF studies stop at testing findings at the same sample scale, but many others attempt to conclude about information or phenomena at a larger scale, even above field scale. Both those PTF studies and this study are supported by samples of the same size and scale. In fact, pF rings are typically even taller than those used in this study, i.e. lots of PTF studies rely on larger samples. This needs to be resolved, preferably by removing the field scale notion from the paper entirely – unless that aspect is really well supported.

**Reply:** We agree to the concerns about identification of the appropriate scale and have removed all references to the "field scale" in the revised paper. Furthermore, a disclaimer was added (P15 L22-25).

**Comment 2:** I feel that the authors push to position themselves away from PTFs. This seems to be apparent by the sum of comments made about earlier PTF works, e.g. on how much data is needed, what 'scale' those are developed for, and most importantly those they cannot predict macropore flow. Latter is true, but the authors need to remain

objective and modest about what this paper really is. This is just as much a PTF studies as all those referred earlier studies are. It carries all those characters, all the strength and all the weaknesses that those earlier studies do. The only substantial difference is in what the inputs and outputs are, and the methodology how those were obtained. Otherwise, the sample size/scale is the same, the study is based on a collection of 65 local samples (vs. limited data – see minor comment below), and the authors present regression equations between soil properties (Table 2, Figure 8). Please also see additional comment below on P12, L23. Please revise the manuscript to remove the 'us' and 'them' attitude and present this study by what it is: a PTF work with novel data support. This means revising the text everywhere where there is commenting on PTF data support, scale, etc.

**Reply:** In the first part of the presented study we evaluate the predictive performance of existing pedotransfer functions/models for saturated permeability, air permeability, and gas diffusivity and demonstrate their rather poor predictive capabilities. We acknowledge that it has been previously demonstrated that water flow in macropores cannot be accurately predicted with empirical models from basic soil properties. However, at the same time we find that there is only very little published work related to gas diffusivity. Furthermore, existing pedotransfer functions/empirical models only consider basic soil properties and usually omit pore network characteristics.

In the second part of this study we derive novel macropore network characteristics for saturated permeability, air permeability, and gas diffusivity from X-ray CT observations and demonstrate their utility for improving accuracy of gas and water flow predictions based on the Kozeny-Carman equation.

There is absolutely no intention to belittle other contributions, but it is obvious that when directly derived pore network characteristics are employed for model/PTF parametrization the predictive capabilities improve.

We think that the scope of our study with all its limitations is clearly stated in the last paragraph of the introduction as well as throughout the revised manuscript.

**Comment 3:** Some of the earlier reviewer comments given were responded to as 'Done', while it is apparent from the manuscript's new version that they were not done. I assume they were forgotten, e.g. comments 5, 8, and 29 of reviewer 4. They all should be checked again.

**Reply:** Comment 5: In figure 9 (now figure 8), the choice of the fitting function depends upon its accuracy of predictions. The power function was preferred over simple linear regression if it resulted into greater R<sup>2</sup> value. It has been clarified now P13 L7-10. Comment 8: It has been correct now, P2 L4. Comment 29: We have replaced Kw with Ksat now wherever I was mentioned previously that referred studies predict Kw e.g. P5 L12, P15 L12.

Comment 4: P2, L4: due to its inherently

**Reply:** Corrected P2 L4.

**Comment 5:** P3, L2: for the partitioning

**Reply:** Corrected P4 L2.

**Comment 6:** P3, L23: 'higher' should preferably be replaced with 'greater' in this context. This is applicable to the whole manuscript, please check consistently.

**Reply:** This has now been corrected throughout the manuscript.

**Comment 7:** P4, L21.23: here I have a problem with the 'field scale'

**Reply:** This sentence has been revised now. P6 L1-2.

**Comment 8:** P4, L24: development of new imaging techniques or developments in imaging

**Reply:** Corrected P6 L3.

**Comment 9:** P5, L12-13: neither is your study conducted at the field scale!

**Reply:** This sentence has been removed from the manuscript.

**Comment 10:** P5, L12-13: how many samples mean 'limited', and how many don't? Better avoid such statements I think, because with the technology developing, 65 samples will soon be referred to as 'limited data'.

**Reply:** This sentence has been removed from the manuscript.

**Comment 11:** P5, L22-26: again, the 'limited number' statement would be good to avoid.

**Reply:** This sentence is removed now from the manuscript as suggested.

**Comment 12:** P5, L22.26: To me this sentence is misplaced, and should better be built in somewhere before the 'second and novel part' sentence (or even earlier, maybe in the previous paragraph) as something identified from the literature search, which you will then address in this study.

**Reply:** This sentence has been moved to an earlier paragraph P6 L6-12.

**Comment 13:** P6, L15: How does the 15 by 15 grid cited here compare to the grid presented in Figure 2? Or is 15m the spacing, between 2 points in Fig 2? Then state that you had a 12x5 rectangle with 15m spacing + 5 points in between, apparently.

**Reply:** This has been revised now P8 L14-16.

**Comment 14:** P8, L22-23: Soil texture was determined from disturbed soil samples using a combination of wet sieving and the hydrometer method, after passing the sample through a 2-mm sieve.

# **Reply:** Corrected P11 L5-6.

**Comment 15:** P10, L9-10: this does not answer the original comment – still no statement similar to what has been in the response file.

**Reply:** This has been better explained now on Page 13 Line 6-11.

**Comment 16:** P11, L4: from 14 to 19%

**Reply:** Corrected P14 L4.

Comment 17: P11, L26: out of the 4 marked

**Reply:** Corrected P14 L25.

**Comment 18:** P12, L11: still incorrect, those studies did not predict kw, you converted them from Ksat.

**Reply:** Corrected P15 L12.

**Comment 19:** P12, L15-16: narrow range of saturated water permeability was predicted.

Reply: Corrected P15 L15-16

Comment 20: P12, L16: ...a wide range

**Reply:** This has been revised now P15 L17.

**Comment 21:** P12, L17: The primary reason for the failure of the...

**Reply:** Corrected P15 L18.

**Comment 22:** P12, L23: PTFs are not developed for the scale of a horizon necessarily, just samples are taken from horizons for some practical considerations. In the same sense, all of these samples from 5-8.5 depth are representative of the A horizon, so there is just as much reason to classify your study under the same umbrella with those PTF studies. This is again a reason why I don't see this study to be so far from PTF studies that the authors try to distance it from.

**Reply:** Regarding PTF-based nature of the work. We agreed that this is also a PTF-based study, where in first part we have tested the predictive performance of the existing empirical models/PTFs and in the second part we have modified and tested the existing empirical model (Ahuja et al. 1984) by using the novel X-ray CT derived input parameters. This has been clarified now in the objectives of the manuscript. P6 L21-25-25, P7 L1-9.

Comment 23: P12, L24-25: ...developing empirical models for the prediction...

**Reply:** Corrected P16 L1-2.

Comment 24: P12, L26: From those, we have tested ...

**Reply:** Corrected P16 L3.

**Comment 25:** P13, L5: instead of 'past century' use 'previous century', or '20th century'

**Reply:** Corrected P16 L8-9.

**Comment 26:** P13, L5: models for the prediction

**Reply:** Corrected P16 L9.

Comment 27: P15, L2: in the soil column

**Reply:** Corrected P18 L11.

**Comment 28:** P15, L6: CT-derived macroporosity (Figure 8).

**Reply:** Corrected P18 L15, P19 L1.

**Comment 29:** P15, L6-7: I still think that there should be a better term found for "two-branch system data trend"

**Reply:** We would like to use this notion now as it has been referred at various places in the manuscript, and also that we could not find the one that better describes our data.

**Comment 30:** P15, L8: by 'upper branch', are you referring to the branch 'showing greater permeability' (or whatever applies)? Use more precise references to the data.

**Reply:** Revised now P18 L17

**Comment 31:** P15, L10 and L12: they are first marked in Figure 2, aren't they? That is where you introduce them.

**Reply:** Yes, they are first marked in Figure 2 but we have mentioned Figure 3 here because they are visually shown in Figure 3. This has been clarified now at P18 L19 and L22.

**Comment 32:** P15, L10-12 (two instances): replace 'fall under this branch' with e.g. 'are members of this branch' – or something similar in meaning.

**Reply:** Done P18 L20 and L22.

**Comment 33:** P15, L13: for the two categories

Reply: Done P18 L23.

Comment 34: P15, L14: replace 'reflected' with 'suggests'

**Reply:** Done P18 L24.

**Comment 35:** P16, L4-5: Significant strong power regressions... this sentence should be revised/replaced for various reasons (also for two-branch system)

Reply: Done, this sentence is revised for its clarity. P19 L16-18

Comment 36: P16, L5: Similarly

## Reply: Done, P19 L18

**Comment 37:** P16, L5-7: This significant increase is relative to what? Include in the sentence.

**Reply:** Done, this sentence is revised now. P19 L18-21.

**Comment 38:** P16, L19: What is an 'independent significant power regression'? Rephrase.

**Reply:** Done, this is revised now to make it clear, P20 L8-10.

**Comment 39:** P17, L21: was found to be the key predictor that yielded the highest adjusted R2 value... among all macropore flow parameters....

**Reply:** This paragraph has been removed now to include Ahuja et al. (1984) model predictions.

**Comment 40:** P18, L1: predictors of macropore flow in the future

**Reply:** This paragraph has been removed now to include Ahuja et al. (1984) model predictions.

**Comment 41:** P19, L6-7: Existing empirical models overpredicted saturated water permeability in case of .... And underpredicted it in case of.... (However, I need to add that your models are empirical too, so a different term may be better).

**Reply:** Done, Page 22, L8.

Comment 42: P19, L16: Total macroporosity

**Reply:** This has been revised now, P22 L18-23.

Comment 43: P19, L23: replace 'data' with 'image'

Reply: Done, P21 L13

**Comment 44:** Figure headings: are Figure 4 an 5 headings mixed up?

**Reply:** They are corrected now.