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Comment

Comment on Cabrera et al. A User-Friendly Tool to Characterize the Moisture Transfer in Porous Building Materials: FLoW1D. *Appl. Sci.* 2020, *10*, 5090

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Abstract: In July 2020, this journal published "A User-Friendly Tool to Characterize the Moisture Transfer in Porous Building Materials: FLoW1D". That article presents and applies a forward-Euler-based tool, implemented in Visual Basic for Applications in Excel, for simulating moisture transfer in building materials. In that tool, the building materials' hygric properties are described via the unimodal van Genuchten and Mualem equations. All parameters needed therein are inversely identified from only a capillary absorption experiment. These are three feats that, if valid, would strongly progress the state-of-the-art on moisture transfer simulation, hygric property description, and hygric property characterisation. This critique voices severe doubts, however, about the applicability of FLoW1D, the adequacy of the van Genuchten-Mualem equations, and (most fundamentally) the uniqueness of the inverse characterisation.

Keywords: hygric properties; moisture transfer; building materials; capillary absorption; inverse identification; moisture simulation; van Genuchten-Mualem

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Citation: Janssen, H. Comment on Cabrera et al. A User-Friendly Tool to Characterize the Moisture Transfer in Porous Building Materials: FLoW1D. *Appl. Sci.* 2020, *10*, 5090. *Appl. Sci.* **2022**, *12*, 1123. https://doi.org/10.3390/app12031123

Received: 7 September 2020
Accepted: 16 December 2021
Published: 21 January 2022

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1. Introduction

In July 2020, this journal published the article "A User-Friendly Tool to Characterize the Moisture Transfer in Porous Building Materials: FLoW1D" [1]. In this article, the authors present an Excel-based tool for the numerical simulation of moisture transfer in porous building materials, employing finite differences for the spatial discretisation and forward Euler for the temporal discretisation. The hygric properties of the material involved are parameterized via the unimodal van Genuchten and Mualem equations [2], which require five parameters, including porosity. Porosity is independently determined, and the four remaining parameters are obtained via inverse identification exclusively based on a capillary absorption test. In their 'featured application', the authors declare that "FLoW1D is a numerical tool that is designed as support for the analysis of the conventional experimental tests for the hygric characterization of porous building materials", hence alleging its general applicability for building materials. Their 'conclusions' say that "FLoW1D allows both the simulation of two WAC tests and the estimation of hygric parameters with robustness and ease", therefore averring the robust nature of their approach in general and their tool in particular. These feats—the forward-Euler-based moisture transfer simulation of building materials wherein the hygric properties were universally described with unimodal van Genuchten-Mualem equations, the parameters of which could be inversely identified exclusively based on a capillary absorption test—are impressive successes for research on moisture transfer simulation, hygric property description, and hygric property characterisation for porous building materials.

If valid, these three feats would indeed radically progress the current state-of-the-art on moisture transfer simulation, hygric property description, and hygric property characterisation for porous building materials. Regarding moisture transfer simulation, backward Euler with adaptive time stepping [3] is considered the least complex approach to overcome

the usual inefficiencies of forward Euler for the small time steps and/or element sizes [4] usually required in the simulation of the highly nonlinear moisture transfer equations. In relation to hygric property description, the unimodal van Genuchten equations are often assumed to be insufficient to accurately capture hygroscopic and capillary moisture storage [5], while the Mualem equations are usually seen as noncompliant with the complex variation of the moisture permeability [6]. Concerning hygric property characterisation, an extensive and time-consuming experimental campaign, comprising a myriad of measurements, is often deemed necessary [6–8]. The three feats reported in [1] would therefore form major steps forward in these three areas if valid. This critique does, however, formulate severe doubts on the reliability of [1]'s findings. Below, it is revealed that the applicability of the FLoW1D tool and the suitability of the unimodal van Genuchten-Mualem equations are limited. More crucially, however, this critique establishes that such an inverse identification of the hygric properties based on capillary absorption only cannot be unique and therefore cannot be considered robust or reliable.

Below, a concise digest of the disputed article is first given. Subsequently, the characterisation of the Cuenca limestone's storage and transport properties is critically examined by comparing it with other data on this material. The next section then discusses the non-uniqueness of parameterisations inversely identified from capillary absorption only and demonstrates that this non-uniqueness leads to non-unique hygrothermal performance evaluations. Next, the shortcomings of the unimodal van Genuchten-Mualem equations for the moisture storage and transport properties of porous building materials are substantiated by applying them to several sandstones. The final segment then demonstrates the instability of the FLoW1D tool when fed (slightly) different parameterisations. The critique is finalized with the discussion and the conclusions.

2. Overview of Important Elements of Critiqued Article

The general isothermal moisture transfer equation adopted in [1] is:

$$\frac{dw}{dt} + \nabla g = \frac{d(w_w + w_v)}{dt} + \nabla(g_w + g_v) = \frac{d(\phi S_r \rho_w + \phi(1 - S_r)\rho_v)}{dt} + \nabla\left(-k\nabla p_c - \delta_p \nabla p_v\right) = 0 \quad (1)$$

where w (kg/m^3) is moisture content, g (kg/m^{2}s) is moisture flow, t (s) is time, ϕ (m^3/m^3) is porosity, S_r (–) is degree of saturation, ρ (kg/m^3) is density, k (kg/msPa) is liquid permeability, δ_p (kg/msPa) is vapour permeability, p (Pa) is pressure, and subscripts w, v, and c refer to water, vapour, and capillary, respectively. It has to be noted that the (negative) capillary pressure is used here instead of the (positive) suction in [1], as the former is more physically accurate. Flows are typically presumed to go from high to low flow potential values, along the negative of the potential gradient, whereas the inverse is true for suction. In that respect, it should be remarked that [1]'s Equation (9) is incorrect, so a negative sign was e added at the right hand side.

In complement to Equation (1), the liquid storage and transport properties of the material are described with the unimodal van Genuchten and Mualem [2] equations in [1]:

$$S_r(p_c) = \left(1 + (-\alpha \cdot p_c)^n\right)^{-m} \quad (2)$$

$$k(S_r) = K\frac{\rho_w}{\mu_w}\kappa(S_r) = K\frac{\rho_w}{\mu_w}S_r^{0.5}\left(1 - \left(1 - S_r^{\frac{1}{m}}\right)^m\right)^2 \quad (3)$$

completed with this equation for the vapour permeability:

$$\delta_p(S_r) = \delta_0 \tau \phi (1 - S_r) \quad (4)$$

where μ_w (Pas) is liquid water viscosity, K (m^2, is intrinsic permeability, δ_0 (kg/msPa) is the vapour permeability of still air, τ (-) is tortuosity correction (assumed equal to 1), and α (1/Pa), n (-), and m (-) are fitting parameters. The parameterisation of the moisture

storage and transport properties hence comprises five parameters: porosity ϕ, intrinsic permeability K, and fitting parameters α, n, and m.

In FLoW1D, the Excel-based simulation tool, Equation (1) is solved with finite differences for the spatial discretisation and forward Euler for the temporal discretisation, and the final implementation is verified by means of three qualification exercises, for all of which decent but not perfect agreements with the reference solutions are obtained; see [1]'s Figures 3 and 4.

As an exemplary application of FLoW1D, the model is used to characterize the moisture storage and transport properties of two limestones employed in the Cathedral of Sancta Maria and San Juan in Cuenca, Spain. For this, the parameters K, n, m, and α are inversely identified by fitting simulations to measurements of capillary absorption, while the parameter ϕ is independently determined from porosity measurements. For the 'OPS' limestone, the obtained parameter values are 0.38 for ϕ, 1.27×10^{-13} for K, 4.68×10^{-5} for α, 0.84 for n, and 0.70 for m. These values are also present in the Excel file 'FLoW1D.xlsm', provided via [1]'s supplementary material.

3. Conflicting OPS Limestone Hygric Property Characterisation

Figure 1 presents the moisture retention curve (left) and liquid and vapour permeability curves (right) for the OPS limestone based on the parameter values stated above. This vapour permeability is not δ_p, related to gradients in vapour pressure, but its translated version, linking it to gradients in capillary pressure. For this aim, the Kelvin equation was applied:

$$G_v = -\delta_p \nabla p_v = -\delta_p \frac{dp_v}{dp_c} \nabla p_c = -\delta_p \frac{p_v}{R_v T \rho_w} \nabla p_c = -k_v \nabla p_c \quad (5)$$

where R_v (J/kgK) is water vapour gas constant and T (K) is temperature. The last terms define k_v (kg/msPa), the vapour permeability for capillary pressure gradients, which is depicted in Figure 1 (right). This transformation was done to allow for a direct comparison of liquid and vapour permeabilities.

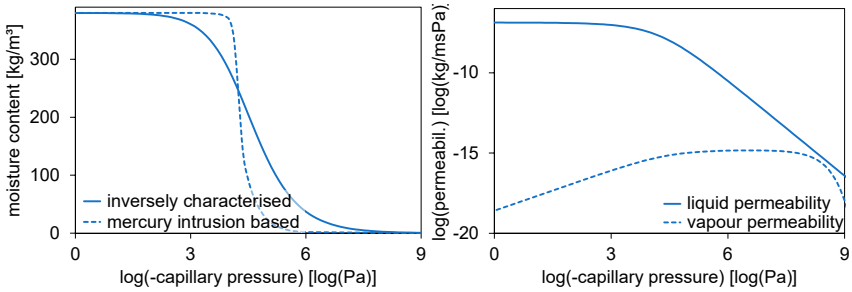

Figure 1. (**left**) Moisture retention curve for the OPS limestone, as inversely characterized or based on mercury intrusion; (**right**) liquid and vapour permeability (both related to gradients in capillary pressure) curves for the OPS limestone.

These moisture retention and permeability curves expose a number of abnormalities concerning the hygric characterisation of the OPS limestone. Firstly, the inversely characterized moisture retention deviates strongly from the moisture retention curve determined from mercury intrusion porosimetry. The latter has been determined with the Young–Laplace equation from the porosimetry results included in [1]'s supplementary material. This is a well-established technique [9], and its accuracy has recently been reiterated [8]. Whereas the mercury-intrusion-based curve presents a (very) steep slope, in agreement with this limestone's narrow 1–20 µm range of pore radii, the inversely characterized curve is far smoother, which does not concur with that narrow pore radii range. Secondly, the liquid and vapour permeability curves show that the liquid permeability always exceeded the vapour permeability, even at the very low capillary pressures and hence very low

moisture contents. In the capillary pressure range from -10^8 to -10^9 Pa, representing relative humidity values between 0.1 and 50% and moisture contents of about 1 to 2 kg/m^3 only, the liquid permeability is on average five times higher than the vapour permeability. This is unheard of, as it is widely accepted that vapour diffusion is the exclusive and dominant moisture flow mechanism at such low humidity [10]. When using these permeability curves for a simulation of a dry cup vapour diffusion test, the discusser acquires a moisture flow that translates to a vapour permeability δ_p of roughly 3×10^{-10} kg/msPa, 50% above the vapour permeability of air and about 40 times higher than typical limestone vapour permeabilities presented in the literature [11]. For that simulation, a 5 cm thick sample was placed in between environments at 54 and 12% RH, and the resulting steady-state moisture flow was translated to the reported vapour permeability.

The employed parameter values were obtained by fitting FLoW1D simulation results to the measured moisture mass evolution during a capillary absorption test. The resulting agreement of the simulated and measured moisture absorption is depicted in Figure 2 (left), and the resulting moisture content profiles as calculated with FLoW1D are shown in Figure 2 (right). Though the fit in Figure 2 (left) is acceptable, the moisture content profiles in Figure 2 (right) deviate from the sharp infiltration moisture fronts that are generally observed when building materials (certainly when having a narrow pore size distribution like the OPS limestone analysed here) undergo capillary absorption [12]. In [13], moisture content profiles during capillary absorption in a limestone—with similar porosity but a smaller dominant pore radius (near 2 µm) and thus a smaller capillary absorption coefficient (about 0.15 kg/m^{2}s$^{0.5}$)—were visualized with neutronography, exposing very distinct moisture fronts (see [13]'s Figures 6 and 7). Finally, Figure 2 (right) shows that the moisture contents during capillary absorption reached the saturated moisture content, though these were typically limited to the capillary moisture content due to air entrapment [12]. For the limestone studied in [13], the capillary moisture content was close to 220 kg/m^3; in contrast, its saturated moisture content was about 400 kg/m^3 (see [13]'s Figure 3).

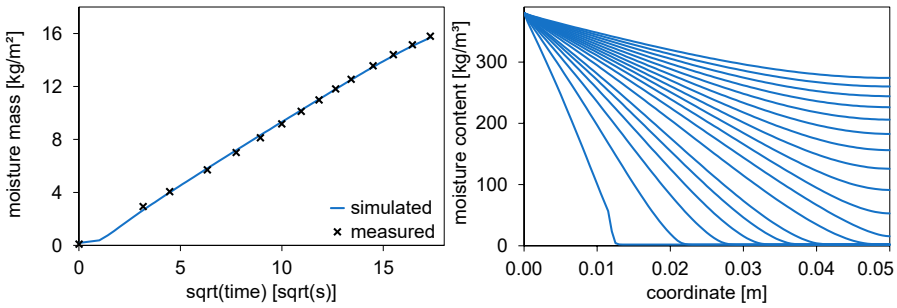

Figure 2. (**left**) Measurement and FLoW1D simulations of capillary absorption for the OPS limestone; the data are depicted as specific moisture mass versus square root of time, the common approach for this experiment, and are equivalent to [1]'s original data; (**right**) FLoW1D-simulated moisture content profiles during capillary absorption, shown every 10 s for the initial minute and then every 20 s for the four remaining minutes.

It must hence be concluded that [1]'s hygric characterisation of the OPS limestone indeed correctly reproduced its capillary absorption, but that it strongly conflicted with plausible expectations on its hygric behaviour for the rest. It must finally be noted that the first three conflicts between [1]'s hygric properties and the plausibly expected behaviour are all connected: a steeper moisture retention curve, e.g., comparable to the mercury-intrusion-based curve shown in Figure 1 (left), would yield a steeper moisture permeability curve, solving the currently excessive liquid permeability at low moisture contents. Additionally, the steeper moisture retention and liquid permeability curves would also result in sharper moisture content profiles during capillary absorption. The smooth moisture retention curve is a central issue in the following sections of this critique.

4. Non-Unique Characterisation and Hygrothermal Behaviour

It appears that the agreement between simulated and measured capillary absorption alone may not suffice to universally and dependably characterize the moisture storage and transport properties of building materials. This argument is further developed in this section, where the non-uniqueness of the parameter identification and its impact on hygrothermal behaviour are targeted. To do so, it is shown below that the measured capillary absorption can also be reproduced with different parameter values. Subsequently, it is demonstrated that these different parameter values yield deviating hygrothermal responses. This implies that the inverse identification based on capillary absorption exclusively suffers from non-identifiability [14,15], implying that the resulting characterisation of the material's moisture storage and transport properties may not be reliable.

This non-uniqueness should of course not come as a surprise. The capillary absorption by a building material is typically described with a single value: the capillary absorption coefficient, which quantifies the slope of the linear moisture mass evolution in function of the square root of time. Fitting three independent parameters (K, m, and n; α is determined via the initial moisture content) to one single target value is a textbook example of non-identifiability [14,15]. It is furthermore suggested below that these four parameters featuring in the unimodal van Genuchten-Mualem equations often do not suffice to capture the full complexity of the moisture storage and transport properties of building materials. In [16], the hygric properties of brick and mortar were described via independent bimodal and trimodal curves, comprising some 15–20 parameters, which would obviously only worsen the non-identifiability.

Though demonstrating the uniqueness of inverse characterisation procedures can be somewhat complex [14,15], establishing their non-uniqueness is far more straightforward. If it can be confirmed that multiple sets of parameter values all minimise the objective function, then it must be concluded that the characterisation is non-unique and hence unreliable. Table 1 shows three parameter value that all satisfactorily reproduce the capillary absorption. The first is [1]'s OPS limestone original, and the two others are variations. The changes in their parameters values primarily reflect somewhat steeper moisture retention curves, still centred though at the same capillary pressure as [1]'s original. The intrinsic permeabilities were subsequently calibrated such that the variants reproduce the capillary absorption. Figure 3 illustrates the resulting moisture retention and permeability curves. It should be noted that the two variants were deliberately kept quite similar to the original to demonstrate the large impacts of small deviations on the hygric response. Compared to the hygric properties for other building materials in the literature [3–6,8,14,16], these are certainly not exceptional hygric property curves, implying that all are physically viable.

Table 1. Parameter values for the original OPS limestone from [1] and two variations on this material.

Material	ϕ	K	α	n	m
OPS limestone original	0.38	1.27×10^{-13}	4.67×10^{-5}	0.842	0.709
OPS limestone variant 1	0.38	1.77×10^{-13}	5.61×10^{-5}	1.000	0.585
OPS limestone variant 2	0.38	4.29×10^{-13}	8.42×10^{-5}	2.000	0.280

For reasons discussed in Section 6, K fitting was not performed with FLoW1D; instead, the well-known hygrothermal simulation tool Delphin was used [17]. These simulations were conducted for the same configuration as in [1]: a 5 cm thick slab, initially at 32% RH, with bottom water contact. Complementarily, three more hygrothermal cases were simulated: moisture redistribution, isothermal drying, and hygroscopic adsorption. In hygroscopic adsorption, the 5 cm thick slab started at 32% RH, and was exposed to a 100% RH environment with a 2×10^{-7} kg/m^{2}sPa transfer coefficient at one surface. For isothermal drying, the 5 cm thick slab started at 100% RH and was exposed to a 32% RH environment with a 2×10^{-7} kg/m^{2}sPa transfer coefficient at one surface. For moisture

redistribution, the lower 1 cm and upper 4 cm of the 5 cm slab started at 100% RH and 32% RH, respectively, and both surfaces were impermeable.

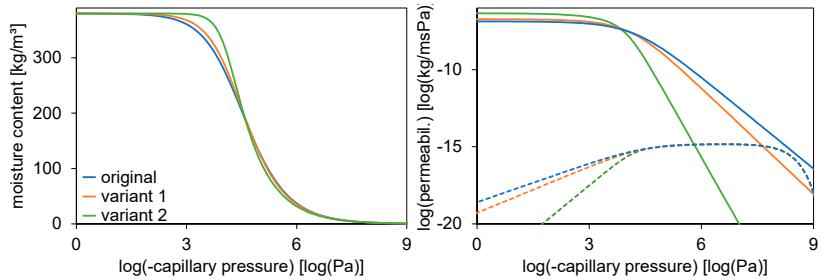

Figure 3. (**left**) Moisture retention curves of the original OPS limestone and two variants; (**right**) liquid and vapour permeability (both related to gradients in capillary pressure) curves of the original OPS limestone and two variants.

The outcomes of the Delphin simulations are collected in Figure 4. For the capillary absorption, hygroscopic adsorption, and isothermal drying cases, the moisture mass evolution in the 5 cm slab is shown; for moisture redistribution, the moisture mass evolution in the top 4 cm of the slab is depicted. Figure 4 (top left) verifies that the three parameter sets all accurately reproduced the measured capillary absorption—all three can hence be assumed to comprise a suitable characterisation of the moisture storage and transport properties of the OPS limestone, at least if adhering to [1]'s inverse identification approach. Unfortunately, the other simulations in Figure 4 reveal that the moisture behaviours of the different hygric property characterisations were very much different. This implies that the inverse identification based on capillary absorption alone is non-unique and thus that the hygric property characterisation cannot be considered robust or reliable. Figure 4 also exposes that minor changes in that characterisation may yield major changes in the hygric response, highlighting the need for an accurate description of the hygric properties, which is the topic of the following section.

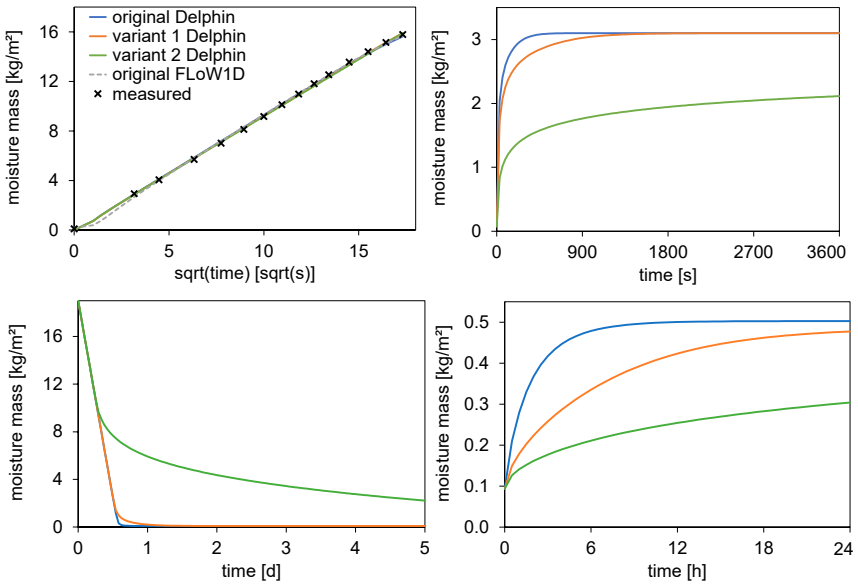

Figure 4. (**top left**) Delphin simulations of capillary absorption for the original OPS limestone and two variants, complemented with original measurements and FLoW1D calculation (blue and orange lines are hidden underneath the green line); (**top right**) Delphin simulations of moisture redistribution; (**bottom left**) Delphin simulations of isothermal drying; (**bottom right**) Delphin simulations of hygroscopic adsorption.

5. Inadequate Unimodal van Genuchten-Mualem Equations

Various sensitivity analyses [18–20] have stressed the necessity of using accurate hygric properties for the reliable simulation of the hygrothermal response of building materials. Defraeye et al. [19] showed that minute changes in the capillary range—5% on moisture retention and 1% on liquid permeability—already substantially affect the drying behaviour. Van Belleghem et al. [20] revealed that minor variations in the hygroscopic range—20% on sorption isotherm and 20% on vapour permeability—significantly impact the buffering behaviour. Any parameterized description of the moisture storage and transport properties of building materials thus needs to be precise. Additionally, it is shown below that the unimodal van Genuchten-Mualem equations mostly do not meet this criterion. Accordingly, we fitted independent hygric properties with these equations, revealing sizeable deviations between the originals and the unimodal van Genuchten-Mualem fits.

Figure 5 shows the moisture retention and liquid permeability curves of three sandstones (Cotta, Posta, and Hildesheim), the properties of which were taken from the Delphin material database. The figure additionally depicts the unimodal van Genuchten-Mualem fits. For these, a least-squares-optimisation was executed, wherein deviations of saturation degree and liquid permeability were brough together in linear and logarithmic fashions, respectively. This confrontation of original and fitted data clearly reveals that van Genuchten-Mualem description was lacking, as it could not capture all complexities of the hygric properties of building materials. It should be noted that the original moisture retention curves, which were desorption curves, did not start from the saturated moisture content but instead from a lower effective maximum moisture content [6]. To not handicap the fitting from the start, these effective maximum moisture contents—which were lower than the saturated moisture contents—were adopted for the parameter fitting. It can also be shown that the use of bimodal van Genuchten-Mualem equations was not satisfactory. In the discusser's experience, the van Genuchten-Mualem equations led to far better results when working with two or three modes while simultaneously employing independent parameters for the moisture retention and liquid permeability curves; however, this necessitates a far larger set of parameters [16].

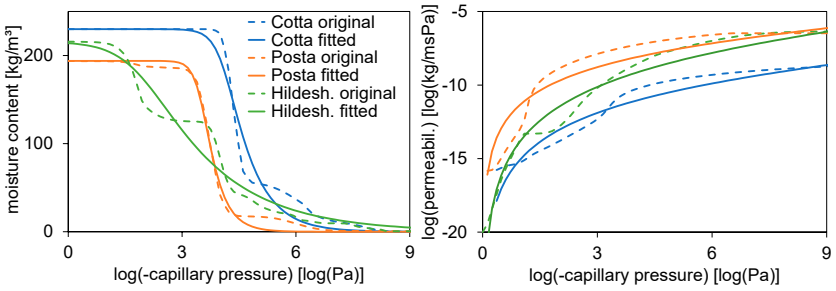

Figure 5. (**left**) Original (from Delphin material database) versus van Genuchten-Mualem-fitted moisture retention curves for three sandstones; (**right**) original (from Delphin material database) and van Genuchten-Mualem-fitted liquid permeability curves for three sandstones.

A second fallacy of [1]'s hygric property description regards vapour permeability; see Equation (3). Predictions of the vapour permeability at 25% RH based on this equation yielded values of 5.4×10^{-11}, 4.2×10^{-11}, and 4.7×10^{-11} kg/msPa for the Cotta, Posta, Hildesheim sandstones, respectively. Measurements in the Delphin material database provided values of 1.0×10^{-11}, 1.2×10^{-11}, and 1.4×10^{-11} kg/msPa, respectively—roughly 3–5 factors lower.

It should thus be concluded that the parameterisation suggested in [1], based on van Genuchten-Mualem for liquid storage and transport and on a non-correction for tortuosity for vapour transport, cannot accurately represent the hygric properties of building materials. Given that small deviations in the hygric properties may yield highly different moisture responses [18–20] (see also Section 4), such inaccuracies should be avoided. The authors of [1] could argue though that researchers are free to adopt more complex descriptions—

with (many) more parameters—if desired. This would, however, be inconsistent with their previous and future work on this topic [21,22]. The functional structure put forward in [21] (in theory more complex as here, as two van Genuchten-Mualem modes were used, though only one mode had a substantial contribution in practice) explicitly targets a reduction of the number of parameters: the authors of [21] openly stated that these "new functions reduce the parameters needed to simulate the moisture transfer". The hygric characterisation in [22] was irrefutably based on the conceptual model of Section 2, and the authors of [22] concluded by stating "that the hygric properties of both lithotypes have been consistently characterized and that the proposed mathematical model is appropriate". The firm confidence of [1]'s authors in the (unimodal) van Genuchten-Mualem equations as passepartout for hygric property description of building materials in [21,22] is quite clear. This being said, adopting a more complex description with (many) more parameters would not only exacerbate the non-identifiability discussed above but also make the task of the Excel solver far more difficult.

6. Inefficient Forward-Euler-Based Simulation with FLoW1D

The sections above have formulated serious concerns on the hygric property description and hygric property characterisation presented in [1], but the moisture transfer simulation with the Excel-based FLoW1D tool may still have value for research on moisture transfer in building materials. For this aim, the tool should efficiently yield reliable results for typical moisture transfer problems. Unfortunately, it is demonstrated here that this may not be the case.

The moisture retention and liquid permeability curves shown in Figure 3 (with parameter values in Table 1) were recycled here and used for FLoW1D simulations of capillary absorption. FLoW1D should be able to tackle this type of simulation, as it is the core of [1]'s inverse identification strategy. Figure 6 reveals, though, that this was not the case: when fed the variant 1 and 2 parameter sets, FLoW1D failed to provide reliable results. For variant 1, an increase in the absorbed moisture was still obtained, but it strongly deviated from the typical square-root-of-time behaviour, which was expected for such simple moisture diffusion problem. The reason for this ill-conditioned behaviour can (probably) be observed in the related moisture content profiles; see Figure 6 (bottom left). The (only marginally) steeper moisture retention and permeability curves led to the typical sharp moisture front, and FLoW1D evidently could not numerically cope with this issue. This was confirmed by simulation outcomes for variant 2, where FLoW1D failed to produce any absorption. This non-robust behaviour should not be surprising: it is well-known that explicit (forward-Euler-based) temporal schemes easily suffer from instability. These observations probably explain why [1]'s characterisation of the OPS limestone led to the smoothly declining moisture retention and liquid permeability curves presented in Figure 1: with the element size and time step stipulated in the supplied FLoW1D version (5×10^{-4} m and 1×10^{-2} s), the steeper moisture retention curves would not allow for successful simulations and hence not permit minimising the objective function. The analysis in Section 3 reveals the conflicts between the obtained hygric characterisation and other data, while the discussion in Section 4 exposes why this deviant characterisation may still adequately reproduce the capillary absorption process.

The authors of [1] could argue that such instability can be remedied by using a finer spatial and temporal discretisation. The author of this research achieved an acceptable solution for OPS limestone variant 1 by using an element size of 6.25×10^{-5} m and a time step of 1×10^{-4} s, which were 8 and 100 times lower, respectively, than those provided in the original version of FLoW1D. A single calculation of that moisture absorption process did, however, take more than half a day (on a Dell Latitude 7480 with an Intel Core i7 7600U quadcore processor). It should in this regard be remarked that the same Delphin simulation only took a number of seconds. However, the OPS limestone variant 1 is not the worst case scenario yet because the hygric properties defined for OPS limestone variant 2 and the plausible actual hygric properties of the OPS limestone are (far) still more

challenging, probably requiring a (much) finer spatial and temporal discretisation and thus needing far more time than that "half day". If a single simulation already takes this much time, the author of this research can only wonder how much time would be needed for the inverse characterisation of the K, m, and n parameters, wherein Excel's solver executes multiple repetitions of the capillary absorption simulation. These considerations conflict with [1]'s 'conclusions' that "FLoW1D allows both the simulation of two WAC tests and the estimation of hygric parameters with robustness and ease". It has been demonstrated that the forward-Euler-based simulation strongly hinders the efficiency and thus the applicability of FLoW1D. Additionally, it would appear that [1]'s authors arrived at that same conclusion since they reverted to the use of Comsol, instead of FLoW1D, in their next paper on this topic [22].

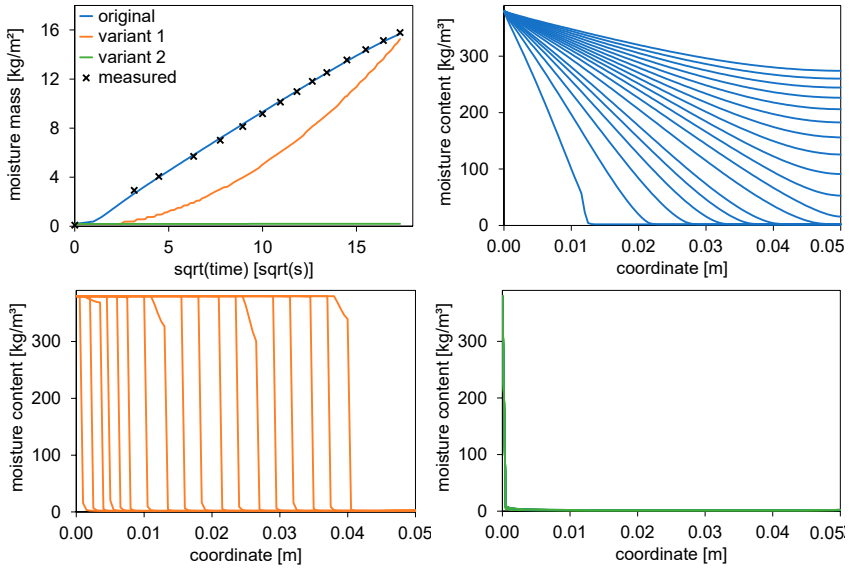

Figure 6. (**top left**) FLoW1D simulations of capillary absorption in the original OPS limestone and two variants, plus original measurements; (**top right**, **bottom left**, **bottom right**) FLoW1D-simulated moisture content profiles during capillary absorption, shown every 10 s for the initial minute and then every 20 s for the four remaining minutes.

7. Discussion

The article "A User-Friendly Tool to Characterize the Moisture Transfer in Porous Building Materials: FLoW1D" [1] proposes three major innovations: the forward-Euler-based moisture transfer simulation of building materials wherein the hygric properties are universally described via unimodal van Genuchten-Mualem equations, of which the parameters can be inversely identified based on a capillary absorption test only. This paper presents severe doubts regarding the applicability of FLoW1D, the adequacy of the van Genuchten-Mualem equations, and (most fundamentally) the uniqueness of the inverse characterisation. The authors of [1] are invited to soothe these doubts in their reply. In concrete terms, the following 10 questions should be addressed:

1. Section 4 shows that many parameter value sets allow for the reproduction of the capillary absorption, revealing that the inverse characterisation of multiple van Genuchten-Mualem parameters based on the capillary absorption coefficient only is non-unique and implying that the ensuing hygric property characterisation is also non-unique and therefore not reliable or robust. Could [1]'s authors counter by verifying that their method satisfies the requirements for structural and practical identifiability [14,15]? Additionally, could [1]'s authors establish that this remains valid even if researchers choose a more reliable hygric property description involving more parameters?
2. Section 3 has illustrates that the achieved hygric properties for the OPS limestone disagree with other data on various fronts. Could [1]'s authors verify their smooth

moisture retention curve with experimental data, possibly resulting from pressure plate tests [8]? Could [1]'s authors support their smooth liquid permeability curve with experimental data, possibly resulting from moisture diffusivity experiments based on visualised moisture content profiles [8,12]? Could [1]'s authors validate their smooth moisture content profiles during capillary absorption, possibly with visualised moisture content profiles again? Could [1]'s authors confirm that the moisture content during the capillary absorption indeed goes up to the saturated moisture content, possibly with visualised moisture content profiles again or otherwise by prolonging the capillary absorption experiment well into the second absorption phase? Could [1]'s authors establish that the liquid permeabilities in the hygroscopic range are well above the vapour permeabilities, possibly via dry and wet cup tests? Could [1]'s authors verify the tortuosity correction being equal to 1, possibly via dry cup tests again?

3. Section 6 demonstrates that FLoW1D suffers from inefficiency due to its instable forward-Euler-based solution. Could [1]'s authors demonstrate that FLoW1D can simulate capillary absorption in the OPS limestone variant 2, report which element size and time step would be needed for that, and report how much computation time is needed for that simulation?
4. Section 5 reveals that the unimodal van Genuchten-Mualem equations may be inadequate for the accurate hygric property description of building materials. Could [1]'s authors confirm that these equations do accurately describe the hygric properties of the OPS limestone, possibly with the outcomes of the experiments that are suggested in relation to point 2 above?

In short, the authors of [1] were the first to propose that the hygric properties of building materials may be fully characterized exclusively based on the moisture mass increase measured during capillary absorption. Such a ground-breaking thesis, which strongly contradicts the state-of-the-art, must be supported with more and stronger evidence than currently provided.

8. Conclusions

In July 2020, this journal published "A User-Friendly Tool to Characterize the Moisture Transfer in Porous Building Materials: FLoW1D" [1]. This critique has formulated severe doubts on multiple elements of that publication, and the author of this paper invites [1]'s authors to placate these doubts in their reply. Refuting these doubts will underline the validity of [1]'s claims in relation to moisture transfer simulation, hygric property description, and hygric property characterisation for porous building materials

Funding: This research was in part supported by the FWO Odysseus grant (G.0C55.13 N), which is gratefully acknowledged.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Cabrera, V.; López-Vizcaíno, R.; Yustres, Á.; Ruiz, M.Á.; Torrero, E.; Navarro, V. A user-friendly tool to characterize the moisture transfer in porous building materials: FLoW1D. *Appl. Sci.* **2020**, *10*, 5090. [CrossRef]
2. Van Genuchten, M.T. Closed-form equation for predicting the hydraulic conductivity of unsaturated soils. *Soil Sci. Soc. Am. J.* **1980**, *44*, 892–898. [CrossRef]
3. Janssen, H.; Blocken, B.; Carmeliet, J. Conservative modelling of the moisture and heat transfer in building components under atmospheric excitation. *Int. J. Heat Mass Tranf.* **2007**, *50*, 1128–1140. [CrossRef]
4. Janssen, H. Simulation efficiency and accuracy of different moisture transfer potentials. *J. Build. Perform. Simul.* **2014**, *7*, 379–389. [CrossRef]
5. Derluyn, H.; Janssen, H.; Carmeliet, J. Influence of the nature of interfaces on the capillary transport in layered materials. *Constr. Build. Mater.* **2011**, *25*, 3685–3693. [CrossRef]
6. Scheffler, G.A.; Plagge, R. A whole range hygric material model: Modelling liquid and vapour transport properties in porous media. *Int. J. Heat Mass Transf.* **2010**, *53*, 286–296. [CrossRef]
7. Feng, C.; Janssen, H.; Feng, Y.; Meng, Q. Hygric properties of porous building materials: Analysis of measurement repeatability and reproducibility. *Build. Environ.* **2015**, *85*, 160–172. [CrossRef]

8. Feng, C.; Janssen, H. Hygric properties of porous building materials (VII): Full-range benchmark characterizations of three materials. *Build. Environ.* **2021**, *195*, 107727. [CrossRef]
9. Krus, M.; Kießl, K. *An Experimental Comparison between Pore Size Distributions Obtained with Mercury Porosimetry and Pressure Extractors*; Report FtB-11, Fraunhofer-Institut für Bauphysik: Holzkirchen, Germany, 1991.
10. Islahuddin, I.; Janssen, H. Pore structure based determination of unsaturated hygric properties of porous materials. *Transp. Porous Med.* **2019**, *130*, 675–698. [CrossRef]
11. Pia, G. Fluid flow in complex porous media: Experimental data and IFU model predictions for water vapour permeability. *J. Nat. Gas Sci. Eng.* **2016**, *35*, 283–290. [CrossRef]
12. Ren, P.; Feng, C.; Janssen, H. Hygric properties of porous building materials (V): Comparison of different methods to determine moisture diffusivity. *Build. Environ.* **2019**, *164*, 106344. [CrossRef]
13. Randazzo, L.; Venuti, V.; Paladini, G.; Crupi, V.; Majolino, D.; Ott, F.; Ricca, M.; Rovella, N.; La Russa, M.F. Evaluating the protecting effects of two consolidants applied on Pietra di Lecce limestone: A neutronographic study. *J. Cult. Herit.* **2020**, *46*, 31–41. [CrossRef]
14. Vereecken, E.; Roels, S.; Janssen, H. Inverse hygric property determination based on dynamic measurements and swarm-intelligence optimisers. *Build. Environ.* **2018**, *131*, 184–196. [CrossRef]
15. Deconinck, A.-H.; Roels, S. Is stochastic grey-box modelling suited for physical properties estimation of building components from on-site measurements? *J. Build. Phys.* **2017**, *40*, 444–471. [CrossRef]
16. Janssen, H. Adaptive Kronrod-Patterson integration of non-linear finite element matrices. *Int. J. Numer. Methods Eng.* **2010**, *81*, 1455–1474. [CrossRef]
17. Delphin Website. Available online: http://bauklimatik-dresden.de/delphin/index.php?aLa=en (accessed on 5 September 2020).
18. Van Belleghem, M.; Steeman, H.-J.; Steeman, M.; Janssens, A.; De Paepe, M. Sensitivity analysis of CFD coupled non-isothermal heat and moisture modelling. *Build. Environ.* **2010**, *45*, 2485–2496. [CrossRef]
19. Defraeye, T.; Blocken, B.; Carmeliet, J. Influence of uncertainty in heat–moisture transport properties on convective drying of porous materials by numerical modelling. *Chem. Eng. Res. Des.* **2013**, *91*, 36–42. [CrossRef]
20. Van Belleghem, M.; Steeman, M.; Janssen, H.; Janssens, A.; De Paepe, M. Validation of a coupled heat, vapour and liquid moisture transport model for porous materials implemented in CFD. *Build. Environ.* **2014**, *81*, 340–353. [CrossRef]
21. Cabrera, V.; López-Vizcaíno, R.; Yustres, Á.; Ángel Ruiz, M.; Torrero, E.; Navarro, V. A functional structure for state functions of moisture transfer in heritage building elements. *J. Build. Eng.* **2020**, *29*, 101201. [CrossRef]
22. Cabrera, V.; Yustres, Á.; López-Vizcaíno, R.; Merlo, Ó.; Ángel Ruiz, M.; Torrero, E.; Navarro, V. Determination of the hygric properties of the heritage stone of the Cathedral of Cuenca through the water absorption by capillarity test. *J. Cult. Herit.* **2021**, *48*, 186–195. [CrossRef]