Answers to referee's comments to the manuscript: amt-2022-280 'Characterizing the automatic radon flux Transfer Standard system Autoflux: laboratory calibration and field experiments'

Comment on amt-2022-280', Anonymous Referee #1

The manuscript "Characterizing the automatic radon flux Transfer Standard system Autoflux: laboratory calibration and field experiments" porposes a new standard method for radon flux calibration which is an important contribution for those who works in this field mainly due to the lack of standardization. Writing is of good quality and provide sufficente data for those who wants to implement this metodoloy in their on laboratories.

We thank the referee for his/her time and we will introduce the comments and/suggestions as suggested.

I consider that just minor revisions of the manuscript are necessary for the publication

L396 - A typical measurement result is shown in Figure S10 – in fact is Figure S8

It has been changed as suggested, now is Figure S5.

L438 - Figure S8 of the supplementary - in fact is Figure S9

It has been changed as suggested, now is Figure S6.

L484 - Figure S13 shows the three main volumes - in fact is Figure S11

It has been changed as suggested, now is Figure S8.

Although discussed in the item 3.4 my main concern is due to the fact that all experiments were done with high Rn activity concentrations, which will not be found in environmental studies and I agree that more studies must be made in order to verify the validity of the proposed model.

The reviewer is right. As it has been reported into the conclusion section too the results obtained within this first study need now to be confirmed using an exhalation bed more similar to standard soils.

Answers to referee's comments to the manuscript: amt-2022-280 'Characterizing the automatic radon flux Transfer Standard system Autoflux: laboratory calibration and field experiments'

'Comment on amt-2022-280', Anonymous Referee #2

Overall, the manuscript is of good quality considering writing and technical contents. I believe that the presented methodology, approaches and results are of interest for the scientific community, aiming for and significantly contributing to a standardization of radon flux measurements.

Despite this, I believe that minor revisions of the manuscript and addressing of minor technical comments below can improve this work further. While there are some detailed technical comments/questions on the methodology below, these are presumably not critical and can be easily addressed by the authors.

We want to thank the referee #2 for his/her work to help with the improvement of this manuscript. In the following lines we have tried to answer one by one the referee's comments and we have improved the manuscript in agreement with them.

Comments on the contents:

L74: It is stated that calibration of Radon flux monitors requires a calibration exhalation bed type facility. I understand that this is the ideal situation. It seems from later given equations/methodology though, that the derivation of the flux boils down to geometric factors and the calibration factor of the Rn-monitors used inside of some sort of accumulation system. Can it not therefore be done simply by calibration of the Rn-monitors with conventional reference atmospheres and calibration of the geometrical components of the flux system – and if not, why? Have you checked if such a route would lead to similar results compared to the exhalation bed facility? (I presume that the geometry of the flux system can be easily determined to relatively good accuracy).

We thank the author to point out this useful reflection.

First of all, we have to take into account the difference between continuous radon monitors and continuous radon flux systems. Radon flux systems are composed by: i) a commercial continuous radon concentration monitor able to measure the volumetric activity (here named concentration) of the radon (Bq m^{-3}) within a detection volume; ii) an accumulation chamber which will be located on the top of the exhaling surface and which will be opened/closed between each measurement. In addition, several tubes and/or additional volumes are needed to dry the sampled air and/or to avoid the entry of thoron within the monitor detection volume as in the case of the presented Autoflux system.

The response of the radon monitor itself can be previously calibrated within a STAR (System for Test Atmospheres with Radon) by comparison with a known reference radon concentration. In addition, all external volumes making the radon flux system could be measured separately with their own uncertainties but exactly quantifying the internal volumes and tubes could lead to higher uncertainties. Comparing the radon flux systems response with a reference flux from the exhalation bed will allow to estimate the effective height of the system with the minimum uncertainty as it has been shown within the document. In addition, when the accumulation chamber is installed on the soil and the system starts working, the measured radon flux may be affected by several factors such the depth of the chamber installation into the soil (Gutierrez-Alvarez et. al., 2019), the environmental conditions of the soil (Yang et al., 2019), etc. All these previous effects cannot be estimated only with 'geometric' approach.

Thus, a reference exhalation bed, used as primary standard, with a constant and known radon flux, is needed to calibrate radon flux systems and their response under different environmental conditions and to ensure a robust metrology chain.

In order to clarify this point, the following paragraph will be added within the manuscript introduction:

‘The need of an EB facility is justified because, despite the fact that the response of the radon monitors itself can be previously studied within a STAR (System for Test Atmospheres with Radon) by comparison with a known reference radon concentration, and that geometries of external volumes making the radon flux systems could be measured separately with their own uncertainties, the total tubes and internal volumes estimation could lead to high uncertainties. Comparing the radon flux system response with reference exhalation bed will allow to characterize the effective height of the system, need for the flux calculation, with the minimum uncertainty.

You state also, that for the INTE_Flux, the employed Rn-monitor was already calibrated previously in L358. Also, this seems to be the way that the exhalation bed values have been characterized in the first place. I think it would therefore be beneficial to give some more explicit info on the significance of the exhalation bed (is it more for proficiency testing? What is the advantage of the EB? Also see comments about the Conclusion section)

As the referee said we declared that the continuous radon monitor, model DoseMan operating only in diffusion mode, was previously calibrated within the INTE-UPC radon chamber. This monitor was then installed within a metallic accumulation chamber composing the INTE_Flux system (the system is shown in Figure 2). As explained in the previous point, having a calibrated radon monitor does not mean to have the radon flux system calibrated as well. Actually, thanks to the presence of the radon exhalation bed and of the radon flux transfer standard, we were able to calibrate also the client system (INTE_Flux) by characterizing its effective height.

In this case we will not add any additional sentence into the document because it was already clarifying in the previous point.

L123-L124: You stated before that the emanation factor is given as the ratio between Rn activity that escapes from grains into the pore-space to the total Ra activity. Here, you state that it is given by the fraction of Rn activity escaping (i.e. that leaves from the pore space to ambient volume) to the Ra activity (Eq. 2). It appears to me that this is only valid for negligible bulk volume/masses, i.e. “small” enough or “well enough” distributed samples (as you state in L128). I suggest adding this information to the introductory text of Eq 2, i.e. in L122.

We thank the reviewer for this clarification. We have now added the sentence on the homogeneity and thick of the sample, previously presented in lines 150-151, before introducing the Equation 2.

L155/Eq 9: I believe the lambda without any subscript (in the denominator before the brackets) should be lambda_eff. (The formula in the brackets formally comes from a convolution integral over the non-time dependent flux term in the solution of associated ODE of the volumetric Rn activity in the “chamber of known volume”. This convolution integral should contain the kernel exp(-lambda_eff * (t – tau)) and be wrt. dtau, so it appears to me that this should be lambda_eff in the denominator).

The reviewer is right; it was a typographic error. We have now corrected it into Eq. 9.

In L187 and L188 you state that there is no “statistical difference” (between measured and theoretically/semi-empirically derived exhalation rates) between theoretical and experimental approaches given in other work, which I agree is indicated by their data. However, please include a statement at what uncertainties this is. It appears to me, that the theoretical approach in this cited work has uncertainties around 20 %. The way this is written here esp. considering L186 (lack of value/uncertainty for theoretical approach) suggests that there is some sub 2 % agreement between theoretical models and experimental values, which cannot be concluded from this other work.

We guess the reviewer comment refers to the work done by Gutiérrez-Álvarez et al. (2020a) presented in the section 'State of the art Exhalation Bed facilities'. We have now changed the paragraph adding the results they calculated with the corresponding uncertainties to avoid misunderstandings as reported in the following text:

'Gutiérrez-Álvarez et al. (2020a; 2020b) performed an experimental characterization of a soil exhalation rate using the accumulation method (Eq. 9). Two reference exhalation soils were prepared using phosphogypsum in rectangular polypropylene boxes with 6.0 cm and 13.0 cm soil thicknesses, respectively. Experimental means of the two beds exhalation rates were of 13.3 ± 0.42 mBq m-2 s-1 and 23.4 ± 0.53 mBq m-2 s-1 with an uncertainty for σ=1 of 2%-3%. These previous results were compared to exhalation rates determined by applying the theoretical approach (Eq. 3) which gave values of 12 mBq m-2 s-1 and 23 mBq m-2 s-1, respectively for the two exhalation beds, with a total uncertainty of about 20%'.

L185 why are these experimentally determined values considered to be an "estimate", despite the very low cited uncertainties? Also appears at several different points in the text, i.e. L213, L438, 448. Isn't the experimental method a "determination" rather than an "estimate"? (I agree on the theoretical one being an estimate though). Since the "experimental method" was what apparently determined the "reference value", I don't think this should be called an estimate, especially not in L448. Please also give indication more explicitly there, that this is the "go to" value for the rest of the work (i.e. the one that was used for the actual calibration in L529).

We agree with the reviewer comment and we will change the term within the text when referred to experimental measurements.

L241 following, Eq 10: It appears to me that this "linearization" approach is not a main contribution of this paper, but no references are given (also used/derived in cited work by Gutierrez-Alvarez et. al.). I understand that in practice, lambda_eff is not known and therefore, Eq. 9 is linearized for it to "cancel out" in Eq 10. Please include this information explicitly, because otherwise, it is not clear why the equation needs to be linearized in the first place. (i.e. if lambda_eff was known, Eq 9 would already be linear wrt. to 1-exp(…)). Is there an estimate on the error limits introduced by the truncated terms (I presume this scales with lambda_eff*t), maybe also elsewhere, which can be cited? (Error estimate can also be in L304 where specific integration time is mentioned)

As correctly said by the reviewer the linear approximation shown by Eq. 10 is absolutely not our paper contribution. Actually this method is quite old (i.e. Morawska, 1989) and it is not used because the lambda_eff is unknown but to benefit of the fact that it can be negligible during short time measurements. Short time measurements are needed also to validate high resolution radon flux models. The new reference has been added into the manuscript together with a sentence to explain its importance.

Morawska, L., 1989. Two ways of 222Rn determining the emanation coefficient. Health Phys. 57, 481-483,

L266: You state that the Radon monitors influences the "system response" time by diffusion. This is very true, but another factor is for the (not rare) systems which measure the Rn progeny (as a proxy) rather than the volumetric Rn. For those, the response characteristics are even way different because of the progeny ingrowth, but since their detection limits are typically way smaller, it may be tempting for future work to use such a monitor. I think it would be beneficial information to include here explicitly that the approach can only reasonably be applied to Rn monitors which measure actual volumetric Rn.

If we correctly understood the referee comment, we agree that there are monitors which calculate the radon concentration using the measured counts from its progeny (such as ^{214}Po) but using these type of monitors will make much more complex the use of the linear fit method. We clarify that direct radon monitors may be used in the requirements list.

L278: Shouldn't the required detection limit depend on the volume of the accumulation chamber and be therefore expressed in terms of a proportionality factor?

It is correct that the radon concentration increase observed within the accumulation chambers during radon flux measurements is related to the radon exhaled from the studied soil and to the ratio between the total system volume, where the exhaled radon is moving, and the accumulation chamber surface, which is practically the effective height (V_{eff}/A) of the total system presented in the Eq. 10.

As an example here for the referee, we may consider that the literature shows typical soil fluxes around 50-100 Bq m^{-2} h^{-1} (Grossi et al., 2011; Karstens et al., 2015). If these fluxes are measured using a radon monitor, with a time resolution of 10min, within a system with a typical h_{eff} = 0.15 m, the radon monitor should be able to catch a minimum radon concentration increase of 56 -111 Bq m^{-3}, respectively.

Thus, in order to generalize the manuscript sentence, we have now added in the requirement that the MDC (minimum detectable concentration) of the monitor should be low enough to observe single time step radon increase with an uncertainty not bigger than 20%.

L382, 383: Celaya et. al. is not in the reference list and therefore, I could not look it up there: Please include which gamma-emissions have been used for determining the Ra-226 activity). Since it is stated that the sample was equilibrated beforehand, I presume that the progeny emissions (e.g. 352 keV) have been used rather than the 186 keV Ra-226 line. I assume that is due to interference with Uranium, which could have been corrected for using its other emissions. Please include info on this explicitly, otherwise it is unclear why the sample had to be sealed beforehand.

The referee is right. The reference has been added now. The radium activity was actually determined using the 214Pb photopeak (351.9 KeV). The information has been now added into the document as follow:

'The average radium activity concentration of the soil in the EB was obtained by gamma spectrometry analysis of 5 separate samples. The samples were extracted from the center and each of the four corners of the EB at a depth of 10-15 cm. Samples were hermetically sealed in a cylindrical container for one month to allow the 226Ra of reaching the secular equilibrium with its short-lived progeny (214Pb and 214Bi). After this time, the radium activity was determined using the 214Pb photopeak (351.93 keV) with a high-resolution gamma HPGe coaxial detector (model GL-2015-7500, Canberra, USA) following Celaya et al., (2018). The mean 226Ra activity concentration was 19130 ± 350 Bq kg-1'.

L391: So, was the stated uncertainty of epsilon just determined as the standard deviation of the three identical experiments, or have other uncertainty contributions been included/considered too? Which of these two is the major contributor, i.e. is the "across samples" variation comparable to the uncertainties theoretically derived for each of the samples? If it isn't, what might be the cause for this (also aiming into the direction of why the three identical replicas were needed in the first place)? I consider this important also because the determined epsilon together with the relatively high uncertainty is a big reason to why you can state in several other places (namely e.g. L505, 506) that all values are consistent with each other wrt. to given uncertainty, especially considering the semi-empirical/theoretical approach (which seems to me only compatible because of this high uncertainty).

As correctly understood by the referee, the emanation factor of the soil sample was obtained as ratio of the radon activity and the radium activity of the sample. The radon activity concentration was measured by repeating three times the accumulation experiment into a known volume and fitting the curve to obtain the unknown variables. Thus the emanation factor of 0.18 was obtained as mean value of the mean values of the three experiments and its uncertainty 0.03 is actually the uncertainty of the mean, obtained for sigma = 1 from the standard deviation of the mean of 3 experiments divided by the square root of 3. The parameters shown in Equation 11 were calculated as the average of every parameter of every single experiment. This information has been now added in the manuscript.

Eq 11, L395, L396: This refers to figure S5 of the supplement rather than figure S6. Was the correlation of the lambda_eff and phi (both appear in the associated equation) resulting from the regression considered in the uncertainty propagation for epsilon (or is this rather just stated as the standard deviation, see previous comment)?

The experiment in Figure S5 was presented as example of the three experiments carried out and the reported values are the averages of the parameters obtained from all experiments.

L429 please explicitly state the “water saturation values” for which this applies

We thank the reviewer to indicate it. Actually the initial emanation factor, now indicated as ε_0 , was estimated under standard temperature conditions and for dried soil. It has been indicated within the manuscript too.

Table 2: It appears to me that the “experimental approach” to determine the exhalation rate was carried out using Eq 10 (L448). However, the budget given here contains some symbols that are not included in Eq 10. What is their significance?

We thank the reviewer for this comment and he/she is right. The second part of the Table 2 indicate the environmental conditions during the experiments (T, wc and RH) and the parameters (h_{eff}) used within the Eq. 10 to calculate the radon exhalation rate from the Exhalation bed. We have deleted the radon emission rate variable because we actually used directly the measured radon concentration during the experiments.

Eq 15, 16, 17: Am I misunderstanding or are these equations not considering the ingrowth-/decay of Rn in each of the compartments. If so, why not? Is it because this is insignificant on the considered time-scales and since these equations are just to “understand which data should be analyzed” and to thus keep the modeling simpler? If so, I think this should definitely be mentioned in the text.

As correctly understood by the reviewer the radon decay term was not considered here because the equations were applied to simulate and to understand the behavior of the radon concentration within the different volumes of the Autoflux system only during the first hour (Figure S9 in the supplement material). During this first hour the loss of the radon concentration due to its decay within the Autoflux volume is negligible. However, we have now added a sentence to justify this within the text.

How is “F_Th_AF” in L504 determined? Is this just an estimate “by eye”?

As explained in the Lines 537-546, the F_{Th_AF} was obtained by fitting the experimental concnetrations measured within the AlphaGUARD monitor of the Autoflux system (black dotted line in Figure 5) with the ones theoretically obtained by applying the Equations 15/16 and 17 (blue dotted line in Figure 5). We have now tried to make this sentence clearer within the manuscript.

Conclusions section: I believe this should be extended wrt. to several points, especially considering the exhalation bed that takes up a significant portion of the work. It is not mentioned/clear if this exhalation bed is now good/better than others. Further, it is not clear what the purpose of the “theoretical approach” is in the first place, since the results are not discussed, put into perspective nor used in any further calculations in the calibration, correct? Please discuss this aspect of the work, especially, in my opinion, the stated agreement between the theoretical and the actual values considering the relatively high associated uncertainties. Also, the conclusions seem to be somewhat more connected with the overarching goals of the project rather than the main parts (calibration, transfer standard, exhalation bed) of the presented work. I suggest putting more focus on the actual main contents of this work,e.g., why is the ANSTOFlux a reasonable transfer standard compared to others etc.?

We agree the reviewer for this comment. We improved the conclusion section now explaining that our exhalation bed was designed and built on the basis of the experience of our colleagues from the literature in order to take into account their lessons. We also explained that the theoretical approach was used to confirm the reliability of our experimental results and to better understand the radon exhalation rates variability associated with soil water content. Finally, we comment the utility the option of using the Autoflux system as transfer standard and possible improvement of the system.

L635 and Figure 8: I think it is surprising that the model driven by the ERA5 data produces considerably "better" results than the model driven by the actually determined (i.e. local) parameters. Wouldn't one expect, for a correct/working/accurate model, to work best on the best (i.e. the local?) data?

The reviewer is right, theoretically we would expect a better agreement between experimental radon flux data and radon flux model applied using local parameters/variables. However, the radon flux equation is strongly influenced by the soil water content and past studies (Karstens et al., 2015) indicate that when ERA or GLDAS Noah data are better catching the experimental soil water content at a specific site, the radon fluxes model based on one of them also fits better with experimental radon flux values. This is why it is really important validating the model input. The soil water content at the ELSE site was measured using a one-point sensor which will not offer a total column depth representatively.

We have explained better this results within the manuscript now.

Beyond scope comments:

Would it not be possible to derive an analytical equation for the regression analysis from Eq 15, 16, 17 in order to include the now excluded data? (i.e. similar to the "blue values" in Figure 5, but with the reference values as F and fitted other parameters?)

We thank the reviewer suggestion and we try here to clarify this point. The purpose of the formalization and solution of these three equations was actually understanding why the Autoflux system was initially underestimating the radon exhalation rates measured during the calibration experiments at the Cantabria University laboratory in comparison with well (theoretically and experimentally) characterized exhalation bed. Thanks to these equations we actually discovered that the thoron delay volume was not initially included in the ANSTO Autoflux manual thus not only the effective height of the system was different but the first two points were erroneously included into the linear fit calculation. These two points are not representative of what is being measured within the detection volume of the AlphaGUARD of the Autoflux and they cannot be included.

Formal comments:

Line 28 (L28): I think, given the definitions used in the paper, that "exhalation" would be want is meant here instead of "emanation"

Thank you. It has been changed.

L42: I suggest keeping the germs "noble" and "gas" together (i.e. "radioactive, noble gas")

Thank you. It has been changed.

L85: I think “equipped with [..]” instead of “provided with [..]” is better wording

Thank you. It has been changed.

L117: A reference for the Rn decay-constant should be given, in my opinion.

Thank you. It has been added.

L239: “ionization chamber” rather than “ion chamber”

Thank you. It has been changed.

L326: Please include “nominal volume V_D” or include an uncertainty (since many digits are given).

Thank you. It has been added.

L341: I think information on the specifics of IT (i.e. raspberry Pi ethernet, “Bitwise” client) can be omitted, since it serves no further purpose.

The Autoflux system was initially built by ANSTO without any remote control and data download system. It was implemented in this work and we think the information is useful for researchers and readers interested in using an Autoflux system in the future and doing the remote connection themselves.

L437: I believe it should be S6 rather than S5 here.

Thank you. It has been changed.

Figure 5: Please correct typo “withing” in the labels.

Thank you. It has been done.

L595: Typo in “properties”

Thank you. It has been changed.

L620: I believe this should be “purposes” rather than “proposes”

Thank you. It has been changed.

L622: “To” rather than “for”

Thank you. It has been changed.

L626: “properties”

Thank you but the word was correctly used in this case and revised my a native English.