



Research on environmental aspects of retention/release of pollutants in soils and sorbents. What should be next?

Avelino Núñez-Delgado

Dept. Soil Science and Agricultural Chemistry, Engineering Polytechnic School, University of Santiago de Compostela, Campus Univ. s/n, 27002, Lugo, Spain

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ABSTRACT

Although studies dealing with adsorption/desorption (and/or retention/release) of pollutants present in environmental compartments is a classical field of research, recent papers are focusing on some weak points of investigations and publications within the area. In addition, an increasing number of works are being published related to new possibilities and alternatives in this kind of research works, many of them in relation to the use of artificial intelligence (AI). Considering the existence of eventual controversies, eventual mistakes, and the convenience of suggesting alternatives to go ahead in the future, in this work, after taking into account some relevant publications in the previous literature, a simple workflow is proposed as a kind of protocol to revise successive steps that could guide the direction to follow when programming research dealing with the retention/release of pollutants in soils and sorbent materials.

1. Introduction

Research focused on adsorption/desorption (and, more widely, on retention/release) started many years ago, as indicated in Robens (1994), while works specifically dedicated to the adsorption and desorption of pollutants have also a long history. For instance, some publications dealing with the theme are those by Smith (1908), Lewis and Andervont (1927), Dhar (1930), Volker et al. (1940), Thornton and Moore (1951), Weber et al. (1963), Gupta and Chen (1978), Dzombak et al. (1987), Kan et al. (1994), Pan et al. (2009), Awad et al. (2019), and Tee et al. (2022).

In fact, research teams where I have collaborated have been involved in a variety of investigations and papers related to that, including López et al. (1998), Arias et al. (2006), Cutillas-Barreiro et al. (2014), Anastopoulos et al. (2020), Conde-Cid et al. (2020a), and Ahmed et al. (2021).

Recently, different authors have commented on some limitations and mistakes found in previous works involving adsorption/desorption (or retention/release) studies, such as Tran et al. (2017), Kopinke et al. (2018), Inglezakis et al. (2019), Hubbe (2022), Opotu et al. (2022), and Wang et al. (2024a,b), whereas certain scholars are proposing new ways to improve aspects of the research carried out in this field, including some related to the use of AI. Within these, there are papers such as those by Alam et al. (2022) and by Lakshmi et al. (2021), where the authors indicate that, comparing experimental results and those from AI models,

it is suggested that these tools can make good predictions regarding “the adsorption capacity or removal efficiency of the adsorbents”. Also interesting are many other works, such as those by Huang et al. (2023), Jaffari et al. (2024), and Zhao et al. (2024), and, with special focus on environmental pollution due to antibiotics and its removal by means of sorbent materials, some examples of publications dealing with the use of AI in this specific field are the works by Zhu et al. (2021), Okolie et al. (2022), and Wang et al. (2024a,b).

However, I think that an even more basic consideration could be needed to make clear how to proceed, in a step-by-step procedure, when programming research tasks in the field of retention/release of pollutants in environmental compartments.

In view of that, the objective of this manuscript is to propose a workflow as the basis for implementing a step-by-step procedure to go ahead when programming investigations related to retention and release of pollutants in soils and sorbent materials.

2. Proposed protocol to guide and/or assess the programming of research on retention/release of pollutants in soils and sorbents

I would like to suggest, as preliminary, to consider the “Armoricano Eco-ethical Principles” proposed in the conclusions chapter corresponding to a recent book (Núñez-Delgado, 2024) (see Supplementary Material).

After that, and specifically for the current work, I would propose the

E-mail address: avelino.nunez@usc.es.

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so-called *NuDe-2* protocol.

1. For that, the first step would be programming the characterization of the previously sampled soils and/or sorbent materials, following standard procedures. It is not needed to show an exhaustive list of parameters and determinations to be considered, as it is a rather routine task to carry out as many quantifications as possible to reach an in-depth knowledge of the soils or sorbents subjected to any investigation. Obviously, basic physicochemical characterizations, such as pH, electric conductivity, total contents of elements, as well as available, extractable, and fractionation of key elements, would be performed as extensively as possible. Also, as far as possible, it would be desirable to get data from specific devices, such as X-Ray diffractometer, FT-Infra-Red apparatus, Scanning Electron Microscopy, Nuclear Magnetic Resonance, etc. Some determinations could be appropriate for specific soils or sorbents, but not for others, which would be decided by the researchers.
2. The second step would be preparing samples for batch-type experiments. In all cases, rather standardized procedures could be followed, facilitating the comparison of results. It would be decided which range/s of sieving would be used, also facilitating comparisons. Using different meshes for sieving would be applicable to soils, and to a variety of sorbents, but maybe not to any kind of sorbent material.

3. The third step would be performing kinetic studies, with special focus on determining the time required to reach the equilibrium in adsorption (or sorption) processes, following standard procedures.
4. The fourth step would be carrying out batch experiments to get data to plot adsorption curves (essentially for changing concentrations of the pollutant/s under study). It would be interesting to test the effects of different variables, such as pH, ionic strength, and any other of fundamental relevance for the specific case.
5. The fifth step would be making all the experiments needed to study desorption (or release).
6. For cases where it could be considered appropriate, stirred flow chamber experiments could be carried out.
7. For the seventh step, some differences could be taken into account depending on certain characteristics of the soils or sorbents being studied. In fact, this would be the first step where the use of laboratory columns should be considered, and it would have a clear relevance. In this step, different kinds of lab columns could be used, going from some of small size (more appropriate for ground or small particle diameter materials) to others of increasing size (large and diameter) that could fit better experiments where the columns would be filled with structured and/or porous materials keeping a partial integrity (such as low or medium-size soil aggregates maintained even after sieving through rather high diameter meshes).

However, for this step I would like to propose an option, with some

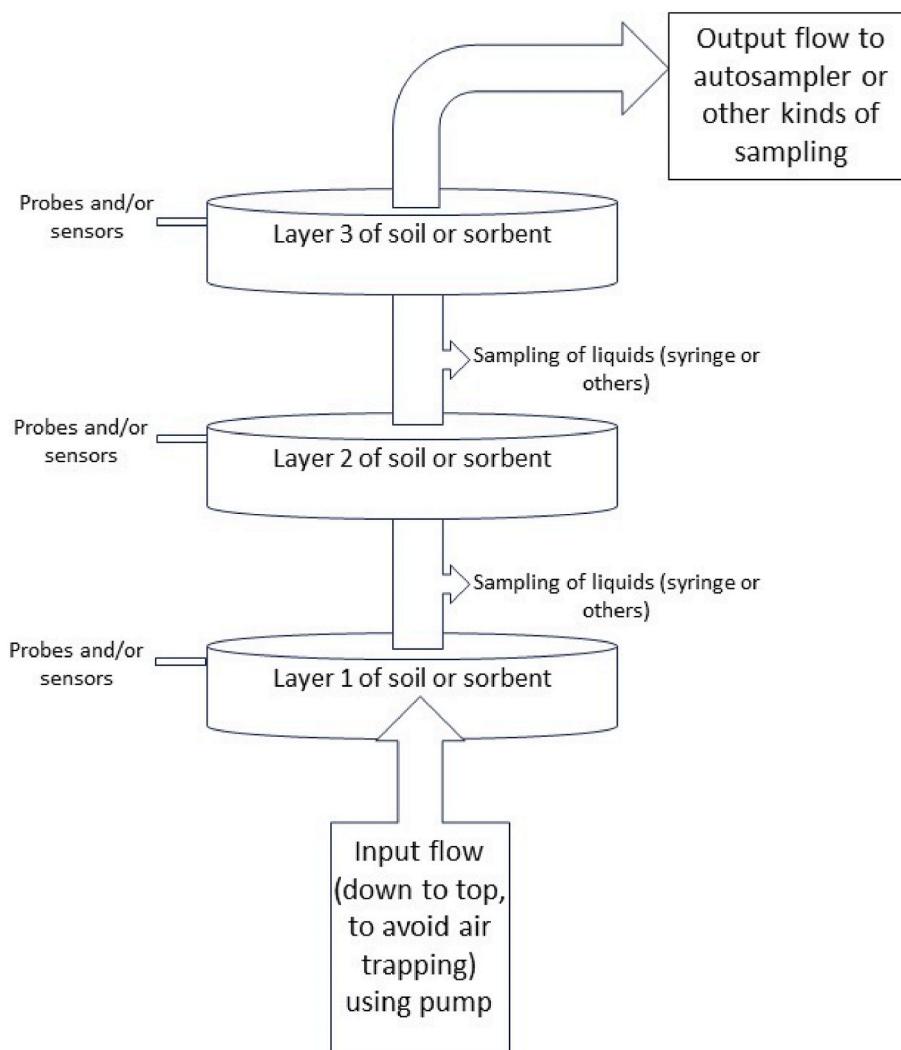


Fig. 1. Schematic overview of key alternatives for experimental design in laboratory studies investigating pollutant retention and release in soils and sorbents.

details very schematically shown in Fig. 1. It does not try to be exhaustive, but rather strictly schematic, as a starting point for what could be shown in much more developed forms and varieties. In fact, it should be seen as a simple scheme showing the basis of certain alternatives in experimental setups to be considered when programming

laboratory experiments related to the study of retention/release of pollutants in soils or sorbent materials. Any case, the main objective of this kind of experimental setup would be to allow the analysis and quantification of the parameters and variables needed to reach a detailed characterization of the processes taking place, eventually

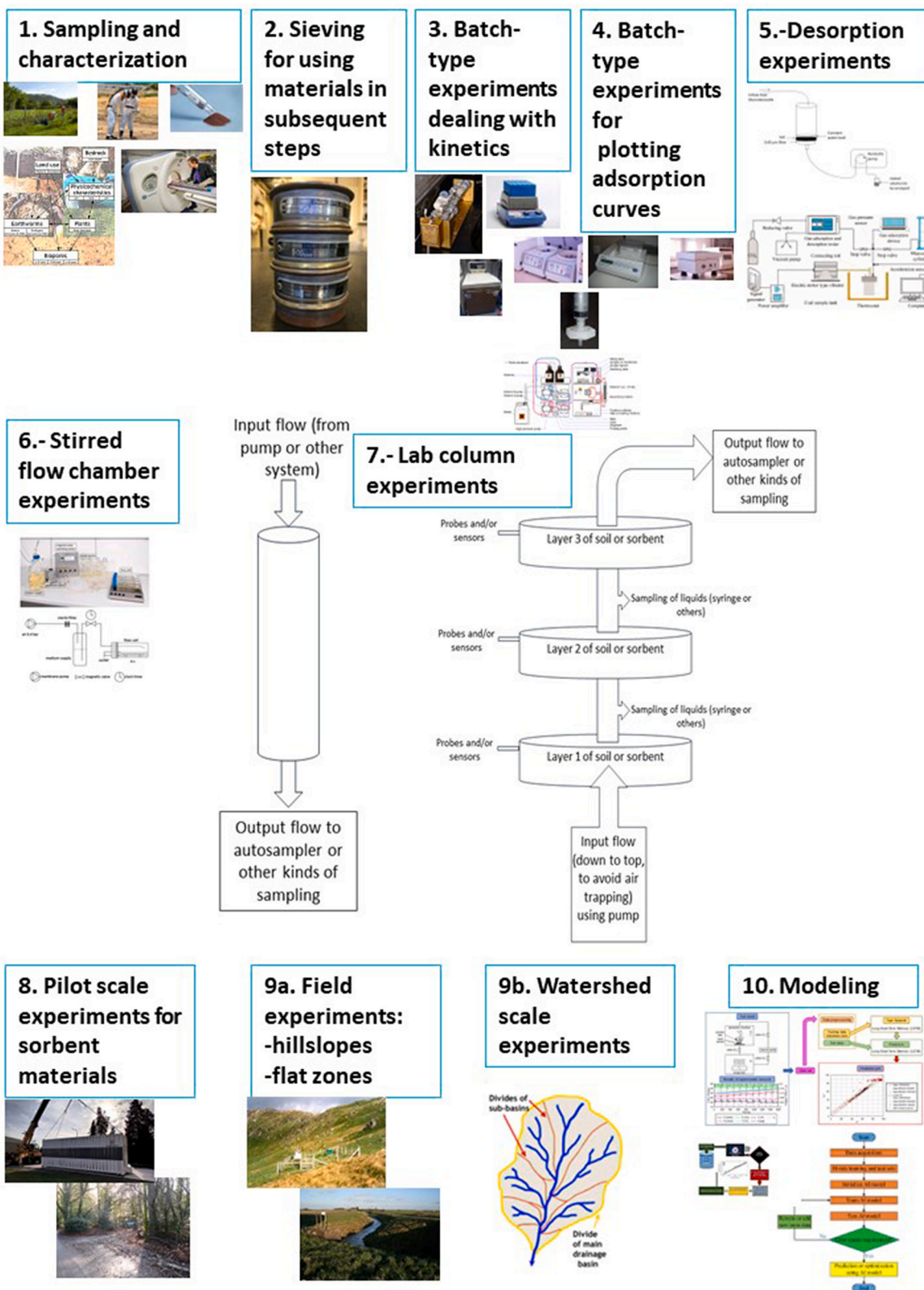


Fig. 2. An overall view of the steps corresponding to the proposed NuDe-2 protocol.

providing details about the mechanisms involved in the retention and release of the pollutants being investigated. Additional comments will be included below, when treating modeling.

8. When dealing with sorbent materials, in case of being considered as potential means to remove pollutants from waters (including wastewaters), pilot scale experiments should be implemented. In case of considering the potential of these materials to retain/remove pollutants after being spread on/into soils, not just laboratory columns should be necessary, but also field experiments.
9. When dealing with soils, even after performing laboratory column studies using a certain degree of undisturbed aggregates (which should be considered essential), a larger scale would be needed to go further. In this regard, field studies would be performed in hillslopes, in flat areas, and at the level of watersheds.

In the Supplementary Material, some examples show references of studies presenting some experimental designs corresponding to various of the previous steps, as well as additional considerations.

10. Modeling. It would be desirable going beyond the classical black-box empirical models, not based on physical aspects and parameters directly related to measurable variables with real physical meaning. It would facilitate avoiding the introduction of just “constants” or any other kind of ad-hoc values used to achieve fits between one and another side of the equations. Instead, as many as possible of the physical, chemical and physicochemical variables (letting those of biological nature for eventual ulterior phases) known to have clear influence on the retention/release processes would be included in the equations.

This kind of objective was something where I had some involvement decades ago, in relation to studying and modeling the drift of pollutants downslope in runoff, as well as in vertical flows taking place in experimental plots. At that time, although working with a research team, I personally received requests to feed and try wide-range models, dealing with erosion and pollution in hillslopes and watersheds, even though I had just limited data, and just for some of the cases requested. In view of that, I personally started to test some of the models, with some data I had available, but I did not dedicate enough time to improving or contributing to develop better models in collaboration with the international specialists and institutions (mainly from the USA) that proposed it. Any case, since then I am aware of the need of going further than just performing empirical adjustments, and of the convenience of searching for physically based (white box) models as much as possible.

An overall view of the ten steps corresponding to the proposed protocol is shown in Fig. 2.

Being more specific, and restricted to laboratory columns, in line with the details very schematically presented in Fig. 1, I would suggest to bear in mind, when preparing the experimental set-up, that it would facilitate all the sampling and determination tasks required to allow the quantification of the parameters involved in the retention and release of the pollutants studied, and to make easier the elucidation of the mechanisms involved. It would affect to surface (interface) interactions (mainly liquid-solid, solid-solid, liquid-liquid), as well to convective (and mainly to diffusive) flows directing dissolved pollutants (or very small particle-size suspended pollutants, in some cases) towards small diameter pores, where some liquids could remain strongly retained, which should be counted as a temporal retention of pollutants, such as certain adsorption (or even sorption) processes taking place onto solid sorbents, which sometimes result in occlusion, added to or complementing precipitation.

As shown in Fig. 1, the basic set-up would include successive columns, linked with tubing allowing sampling. Previous analyses of the materials introduced into the columns, as well as data from eventual probes and sensors, and the subsequent analyses of the materials

removed from the columns after the end of the flowing experiments, would facilitate getting explanations for the events taking place during the circulation of the polluted solutions through the soils and sorbents contained into the columns. Analyses of parameters affecting retention/release, as well as determinations and classical calculations relative to convective and diffusive flows, as well as retained mass of pollutants (from concentrations and volumes) would be key.

At this juncture, having gathered results for all pertinent parameters and variables, particularly when applied to a vast array of soils and sorbents, we can generate a reliable dataset from numerous trustworthy experiments to support the modeling process. This ultimately leads to the derivation of equations that can predict pollutant retention or release based on key influencing variables. Multiple regression techniques may be employed following established statistical protocols, as discussed by Conde-Cid et al. (2019a, b, c; 2020b). Additionally, the integration of AI tools could offer beneficial alternatives or complement current methods, enhancing the accuracy and efficiency of our predictive capabilities.

Obviously, in addition to physical, chemical, and physicochemical processes affecting retention and release of pollutants, biological processes are also relevant, and need specific focus on and procedures to quantify, elucidate and model them. In addition, in cases where the experiments are carried out in the field, or in conditions where environmental variables such as those related to weather are not fixed, specific details must be taken into account, such as eventual volatilization, photodegradation, etc. This is not an exhaustive protocol or revision of all the possible situations, variables, and eventualities potentially affecting the theme, but just a brief call on what could be used to guide successive steps in an overall procedure that would need further development, as it is really wide and with multiple options and pathways to follow.

3. Conclusions

Research dealing with adsorption and desorption (or with retention and release) of pollutants in the environment has been carried out for years, providing very interesting and relevant results. However, some publications have recently stressed the need for checking some mistakes found in certain cases, regarding rather classical procedures, while others have highlighted the potential of new tools, as well as the convenience of implementing them as much as possible, with AI-based tools being among the most promising. In view of that, the current work made a brief overview of some of these aspects, and suggested a kind of protocol that could be followed to guide a step-by-step advancement when programming research focused on retention/release of pollutants in environmental compartments, and specifically involving soils and/or sorbent materials. This simple procedure (called NuDe-2 protocol) could help to guide and/or assess research proposals in this field.

CRedit authorship contribution statement

Avelino Núñez-Delgado: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envres.2024.118593>.

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