Date: Apr 23, 2025
To: "Ilaria Vai" ilaria.vai@unipv.it
From: "Energy Nexus" NEXUS@elsevier.com
Subject: Decision on your submission to Energy Nexus

Manuscript Number: **NEXUS-D-25-00102**

Capturing methane in a barn environment: the CH4 Livestock Emission (CH4rLiE) project

Dear Dr Vai,

Thank you for submitting your manuscript to Energy Nexus.

I have completed my evaluation of your manuscript. The reviewers recommend reconsideration of your manuscript following revision. I invite you to resubmit your manuscript after addressing the comments below. Please resubmit your revised manuscript by **May 23, 2025**.

When revising your manuscript, please consider all issues mentioned in the reviewers' comments carefully: please outline in a cover letter every change made in response to their comments and provide suitable rebuttals for any comments not addressed. Please note that your revised submission may need to be re-reviewed.

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Energy Nexus values your contribution and I look forward to receiving your revised manuscript.

Kind regards,
Pietro Bartocci, Ph.D
Editor-in-Chief
Energy Nexus

Editor and Reviewer Comments:

Reviewer #1: The authors carried out systematic studies for capturing methane in a barn environment: the CH4 Livestock Emission (CH4rLiE) project. The manuscript is recommended for publishing after Major revision. More details are as follows:

1. Did the author do repetitive experiments? How many times? Is there an error line in Figs?
2. In the end of abstract, the significance of research can be described.
3. How to improve the accuracy of the model?
4. Why author said: " Second generation zeolites can replace Si and/or Al with other elements of the same group. Further modifications can then be made, by changing the acidity of the structure, the counter cations or inserting other chemicals. The project will start with commercially available zeolites, then moving on to research other synthetic materials."?

5. What is the long-term performance under the optimized condition?
6. Check out the subscript, superscript and italics throughout the manuscript.
7. Check out the typographical and grammatical errors throughout the manuscript.
8. Some of the latest studies about environmental protection can be summarized in "introduction", such as: Water Research 188 (2021) 116518; Journal of Membrane Science, 663 (2022) 120979.
9.Could author add some examples of practical applications?
10.Could author discuss the development prospects?
11.Please check references. There are some missing data.
12.Please check references. There are some irregularities.

Reviewer #3: Comments on NEXUS-D-25-00102: Capturing methane in a barn….Angiulli et al.

This is a very interesting foray into the important problem of capturing methane in cattle barns, but it is essentially written as a project proposal not as a scientific paper, and as such is not publishable in its present form. The science presented in the paper is thoughtful and interesting, but it reads as if it's clipped straight from a proposal. While 'ideas' papers are often triggers for major new work, this paper is not written as such. Moreover, the paper takes very little note of previously published work on these problems.
My recommendation is that the paper should be rejected, but with an invitation to resubmit either 1) rewritten as an 'ideas' paper, not as a project proposal, or else 2) with the inclusion of some real results to demonstrate the problem.
I have a number of specific comments.
Lines 1-32 need to be significantly updated and focussed more on agroicultural methane from livestock, rather than generalities. It's also paradoxical that the text focusses on livestock in Africa, as well as South Asia, where animals are typically not in barns. Maybe the authors could consider citing newer literature - for example Nisbet et al. 2025, Royal Society Proc A. 481, 2309: 20240390. Methane emissions from barns are tough to quantify - see Laubach, J., et al. (2024) Agri.& Forest Meteorol 350: 109971, and also: https://www.itv.com/news/channel/2021-11-18/jersey-pioneers-testing-new-methane-measuring-technology
Lines 32 onwards on Direct air capture etc: see also Nisbet-Jones, Peter BR, et al. (2022) Phil Trans Royal Society A 380.2215: 20210108.
Line 115 - low precision acoustic sensor? Why not a standard cavity-based optical system like Picarro/Los Gatos or Licor?
L122 - mass balance based on CO2 measurement and then proportionation to CH4 fluxes? Why not simply do this by measuring CH4 directly?
L 111-227 This is simply a proposal text - The methodology seems interesting but without direct practical evidence. As the authors are Italian, maybe Ricerca sul Sistema Energetico - RSE SpA, Milan, Italy would have advice (Zazzeri, G., et al. 2015. Plume mapping and isotopic characterisation of anthropogenic methane sources. Atmos Enviro. 110 (2015): 151-162.
General comments
This paper is interesting and addresses a major problem, but at the moment it is very much a proposal and not a scientific paper. The work needs to be rewritten either as a review of the problem or preferably backed up with some actual measurements, perhaps in collaboration with someone active in the field with measurement expertise. Perhaps the authors should talk to someone in the field - maybe at the upcoming EGU meeting or else Dr G Zazzeri in Ricerca sul Sistema Energetico, Italy.

Reviewer #4: Review Comments
This article proposes the application of the gas recovery technology from CERN's high-energy physics experiments to methane capture in agricultural environments, showcasing remarkable interdisciplinary innovation. The study combines the development of gas adsorption materials, diffusion simulations, and on-site measurements to systematically explore the technical feasibility. After reviewing the entire manuscript, I propose the following suggestions:
1.Supplementary experimental data on the adsorption efficiency of adsorption materials such as copper-doped zeolite under low-concentration methane should be provided to clarify their performance in the actual environment of cattle barns. Additionally, conduct a selective analysis of coexisting gases to verify the methane capture ability of the materials in a gas mixture. Moreover, the experiment should clearly state the number of repetitions and supplement the error range or confidence interval to enhance the credibility of the data. Meanwhile, it is necessary to quantify the full life cycle costs and methane recovery benefits, and discuss the potential challenges of the large-scale application of the technology.
2.Regarding the problem of airflow disturbance in the open space of cattle barns, it is recommended to optimize the layout of the adsorption system in combination with the simulation results of COMSOL, and supplement the adsorption efficiency data in a dynamic environment. Evaluate the impact of the operation of the prototype on the temperature, humidity, and ventilation rate in the cattle barn, and monitor the behavior of the animals to ensure that the technology has no negative impact on animal welfare. It is advisable to cooperate with livestock experts to carry out short-term observational studies to verify the practical applicability of the system.
3.Conduct an in-depth comparison with existing methane emission reduction technologies to highlight the innovation of this solution from the dimensions of efficiency, cost, and animal adaptability. Cite recent research on the adsorption of low-concentration methane to fill the gaps in the literature, and supplement classical references to support the material design theory. Strengthen the connection with direct air capture technology and cite the latest reviews to clarify the technological positioning.
4.There are problems with the format of the charts in the article. Standardize the chart annotations and unify the use of terms. Correct the grammatical errors to ensure that

the language is concise and professional. The citation format of the references should also be carefully compared to make it more in line with academic norms.
In summary, while this paper offers some valuable contributions, there are aspects that can benefit from further refinement and optimization. It is hoped that the author will incorporate the aforementioned suggestions to enhance the overall quality of the paper. As such, my recommendation is to reject the manuscript at this stage. I trust that my feedback will be beneficial to you, and I eagerly anticipate your future scientific endeavors and discoveries.

Reviewer #5: This paper presents a comprehensive account of an innovative project aimed at capturing methane emissions within livestock barn environments. The manuscript is well-structured, with a logical flow of content and a robust scientific foundation, underscoring its significance in environmental and social contexts. While there is room for enhancement regarding the presentation of experimental outcomes and cost analysis, the concepts and methodologies proposed exhibit substantial potential and value. The following recommendations are suggested to refine the paper further:

1. The paper references several existing technologies for methane emission reduction, such as adjusting animal feed, yet it lacks an in-depth comparative analysis of these technologies. It is advised to incorporate a detailed evaluation of the advantages and disadvantages of current technologies to better highlight the superiority and uniqueness of the CH4rLiE project.

2. The methodology section outlines the general framework for gas diffusion simulations and field measurements but omits specific experimental details. For instance, the specific parameters employed in the simulations (e.g., temperature, humidity, wind speed) require detailed specification. Additionally, the models of equipment, sampling frequencies, and data processing techniques utilized in the field measurements need more comprehensive description.

3. The simplified geometric model and point source assumption employed in the simulations necessitate further validation of their rationality. A sensitivity analysis of the simulation parameters is recommended to evaluate the impact of varying parameters on the results.

4. The field measurement section mentions the installation of multiple monitoring stations but lacks detailed information on their exact locations and installation methods. Providing these specifics is essential to ensure the reproducibility of the experiment.

5. The paper notes the use of custom software for data acquisition and analysis but does not elaborate on the specific data processing methods. A detailed description of the data processing workflow, including the handling of noise and outliers, is advised.

6. While the paper mentions the analysis of gas concentration variations under different environmental conditions, it does not adequately discuss how these data correlate with methane capture efficiency. A detailed description of the data analysis methods and a discussion on optimizing the methane capture system using these data are recommended.

7. The paper refers to the use of several charts (e.g., COMSOL simulation figures and experimental setup diagrams) but does not provide the specific content of these charts, nor are the simulation effects fully demonstrated. It is essential to include and clarify these visual elements to better support the text.

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