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

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Cover Letter

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Dear Editor,

We would like to ask you to consider for publication the article entitled:

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

This paper aims at presenting a novel approach for capturing methane produced in livestock environment. The impact of methane on the environment is presented together with the state of the art on methane capture techniques. The paper then focuses on the description of the CH4rLiE project, highlighting in particular the expected impact of such an activity.

We affirm that the submission represent original work that is not currently being considered by another journal. Also, we confirm that each author has seen and approved the contents of the submitted manuscript.

Thanks for your consideration

Best Regards,

Ilaria Vai
University of Pavia and INFN

Declaration of Interest Statement

☒ 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.

☐ The author is an Editorial Board Member/Editor-in-Chief/Associate Editor/Guest Editor for this journal and was not involved in the editorial review or the decision to publish this article.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Capturing methane in a barn environment: the CH_4 Livestock Emission (CH4rLiE) project

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Abstract

The CH_4 Livestock Emission (CH4rLiE) project aims at developing a prototype for methane emissions capture in a barn environment. Methane has a higher global warming potential (GWP) with respect to CO_2, and methane emissions of human origin contribute about 23% to global warming. Emissions from livestock farms play a non-negligible role, as a single cow is capable of emitting about 110 kg of methane in a year. Several projects have tried to

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mitigate the problem by intervening on animal feed: CH4rLiE, in contrast, proposes to act on the methane already produced and diffused in the air, using a specially developed recovery system. The idea arose from the expertise acquired in the Large Hadron Collider experiments at CERN, where special gas recuperation systems are being developed to extract CF_4 from gaseous detectors' exhausted gas mixture. The project focuses on the study of gas adsorption by porous materials and on the development of a prototype system for methane capture, which will be installed in a real barn. This study is being supported by an initial phase of gas diffusion simulations and by a campaign of measurements of gas concentrations in different barn areas. CH4rLiE will also provide an opportunity to explore, for the first time, the feasibility of methane recovery from the farm environment without affecting the animals' feeding or living conditions. The social benefits are extremely interesting both in terms of developing and implementing low-impact farming production processes, but also in terms of recycling expensive or environmentally unfriendly gasses.

Keywords: methane, capture, livestock emissions, global warming

1. Introduction

"The Global Methane Budget 2000-2017" [1] published in 2020 quantifies the relevant role played by methane (CH_4) in global warming. Nowadays, CH_4 is the second most important human-influenced greenhouse gas in terms of climate forcing, just after carbon dioxide (CO_2). In addition, its atmospheric emissions and concentration continue to increase due to contributions from both natural and anthropogenic sources. An important role is played by the agricultural sector (around 56% of anthropogenic CH_4 emissions), and in particular the emissions from enteric fermentation [2] and manure contribute to more than half of the emissions included in the agricultural sector [3], as shown in Fig. 1.

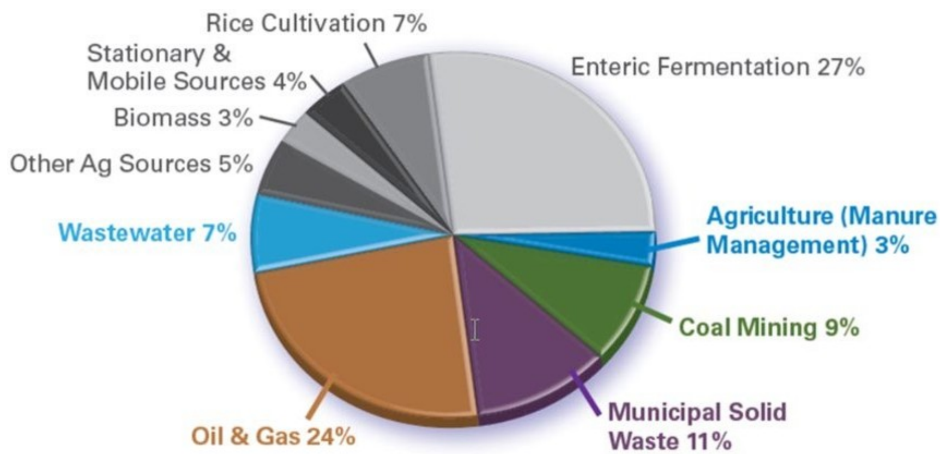

Figure 1: Estimated global anthropogenic methane emissions by source, 2020 [3].

The relevance of CH_4 production in livestock farming is explained by the increase in livestock numbers in Africa, the Middle East, China and South Asia [4]. In this framework, the CH_4 production happens mainly during the anaerobic digestion of ruminants (85% expelled from the mouth) and during the management of manure, which can release CH_4 by anaerobic fermentation, if rich in water. Otherwise, the aerobic fermentation of dry manure releases nitrous oxide (N_2O), which has a global warming potential (GWP) much higher than CH_4 (265 vs 28) [5]. The production of this last gas can anyway be avoided by disposing of manure in anaerobic digesters for the production of biogas. Addressing the problem of CH_4 emissions from enteric fermentation is therefore of primary importance, with the caveat, however, that any eventually proposed technology must also take into account the well-being of the animals. Recently the startup Zelp, part of the London Royal College of Arts, has been awarded for the development of a mask which traps CH_4 emitted by cows' mouths during digestion and its transformation in CO_2 by an oxidation process [6]. However, the use of a mask that must be worn directly by the animal raises questions of ethics and practical feasibility. An alternative solution must therefore be sought in recovering the CH_4 component from the gas once already emitted into the atmosphere of enclosed environments, such as stables, in which animals stay for long periods. The so-called Direct Air Capture (DAC) [7] techniques refers to a set of technological solutions designed to remove greenhouse gases already emitted into the atmosphere. They are being extensively tested for CO_2 removal, but could represent an interesting approach also for CH_4. One of the methodologies used in DAC foresees the physical capture, through adsorption on porous materials, of the relevant gas component. Air is passed through porous materials, such as zeolites, i.e. microporous, aluminosilicate minerals commonly used as commercial adsorbents and catalysts, which adsorbs the CH_4. The choice of using zeolites for CH_4 capture is due to the non-polarity of CH_4 and weak interaction with most materials. Liquid solvents have proven to be ineffective for CH_4 capture [8]. On the other hand, copper-doped zeolites have been tested for adsorption and oxidation of CH_4 to CO_2, in conditions of atmospheric and low concentration CH_4 levels, by heating the zeolite to relatively low temperatures (200-300 °C). This solution demonstrated a high conversion efficiency of CH_4 in CO_2 (Fig. 2). The material was tested in a simulated realistic atmosphere with 2 ppmv CH_4

concentration and 20% oxygen.

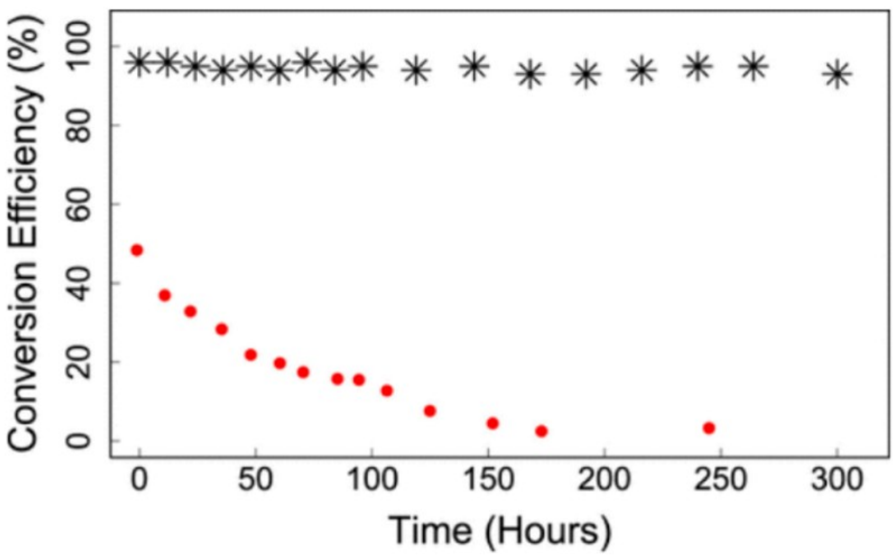

Figure 2: Conversion efficiency of CH_4 in CO_2. Low-level CH_4 (2 ppmv methane in 20% oxygen) was catalytically reacted over 300 h under continuous, isothermal operation at 310 °C (asterisks, following 8 h activation) versus a traditional two-step process (red circles; 450 °C, 30 min activation followed by 200 °C continuous reaction) [9].

The material appears to maintain its conversion efficiency for a long time (> 300 hours) if reacting isothermally at 310 °C, with a pre-activation period of 8 hours at the same temperature [9]. The study also highlighted how the production of these catalysts is based on synthesis from earth-abundant copper and clay aluminosilicates, with predicted costs of 0.33-1.82 \$/kg. Compared with CO_2, CH_4 recovery with DAC techniques from a barn environment presents a major challenge related to the fact that the barn is usually an open space, to ensure the ventilation needed for the animals' welfare, and the CH_4 concentration in this air is quite low (preliminary measures indicate an order of 20 mg/m^3). In this context, ongoing work on the recovery of pollutant gases used in mixtures for particle detectors exploited in high-energy physics experiments becomes relevant. In recent years, CERN [10] has been actively working to reduce pollutant emissions from its experimental activities [11]: 92% of these emissions were related to experimental activity, particularly due to the use of high-GWP gases in particle detectors. The most commonly used components, in this case, are $C_2H_2F_4$ (GWP 1430), SF_6 (GWP 22800) and CF_4 (GWP 7390). Along with extensive R&D aimed at finding alternative components, a mitigation strategy followed with extreme success was the implementation of "gas recuperation systems". Through these systems, the component of interest in the mixture is extracted, stored, and, when necessary, reused. The typical high level of complexity of such a plant is repaid by the excellent results that can be obtained: recent outcomes show that it is possible to recover up to 80% of $C_2H_2F_4$ present in the mixture of Resistive Plate Chambers [12], gaseous detectors widely used in high energy physics experiments.

2. The CH_4 Livestock Emission project

The CH_4 Livestock Emission (CH4rLiE) aims at adapting a gas recuperation system [13] developed in the framework of the Large Hadron Collider (LHC) [14] experiments at CERN to capture CH_4 emitted in a barn environment. To pursue this goal, the project has two main core activities: the study of gas adsorption by porous materials and the development of a prototype system for CH_4 capture, which will be then installed in a real barn. In addition, the study is being supported by an initial phase of gas diffusion simulations and by a campaign of measurements of gas concentrations in different barn areas.

2.1. Simulation of gas diffusion

The simulation activity focuses on the analysis of diffusion and concentration of CH_4 produced by ruminants in the test barn environment, shown in Fig. 3. This work aims at determining the most favorable areas for CH_4

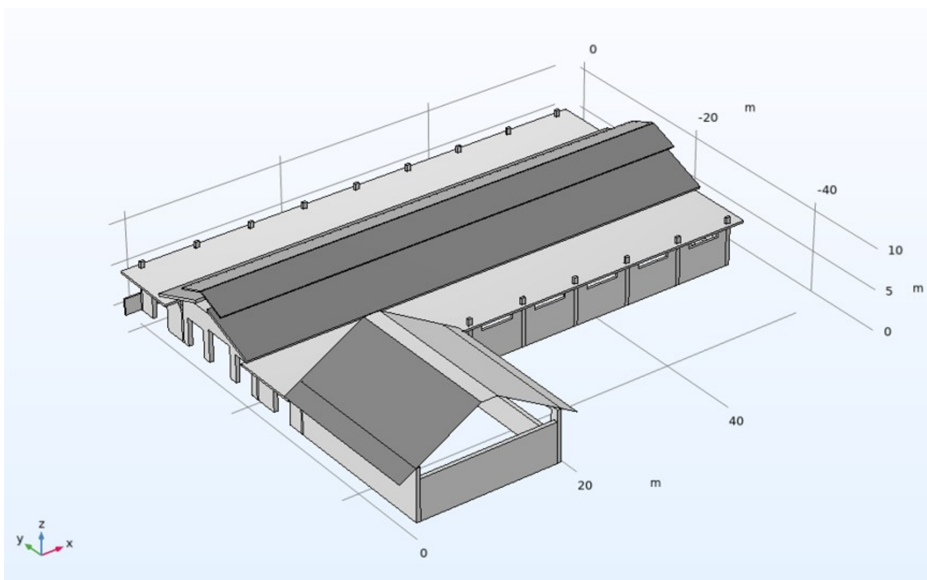

Figure 3: Layout of the barn simulated with COMSOL Multiphysics [15].

collection. The simulation is being performed with the software COMSOL Multiphysics [15], implementing a simplified version of the geometry of the barn and simulating the animal emission as point sources at ground level. The concentration of the different components can be then evaluated in several positions of the barn, such as at ground level, at different heights in the center of the barn and in regions under the roof, especially close to the openings for air circulation. The simulation is being initially performed in a static environment on a single gas component, with the goal of understanding the time needed for a complete dissipation of the gas itself from the barn volume. In a second phase, we will include the effects of environmental parameters (eg. temperature) and different air circulation conditions (forced ventilation, wind or static atmosphere) for a more realistic overview of the system. The simulation will be then finalized evaluating the contribution from all the gases expected in the barn, for a complete overview of the foreseen operational conditions. The goal of this analysis is to identify the most suitable areas for the collection of CH_4, quantifying at the same time the presence of other gases (CO_2, N_2O, NH_3) in the same areas and the variation in their concentration, depending on the wind conditions. The innovative aspect of these simulations is that they allow a complete overview of the barn area, from the ground to the roof. The role of openings on the roof will be investigated too, in order to understand if the chimney draught can offer favorable conditions for CH_4 collection.

2.2. Direct characterization of the barn atmosphere

A proper characterization of the barn atmosphere is therefore crucial for the success of the project. Complementary to the simulations described in Sec. 1, 24h-long periodic measurements in the different seasons of the year are being performed through a sensitive photoacoustic analyser based on quartz-enhanced photoacoustic spectroscopy (QEPAS) [16]. The instrument is coupled with multipoint sampling devices which will allow the monitoring of CO_2, N_2O, CH_4 and NH_3 in different positions of the barn. The activity is also aimed at evaluating the impact of the project, from the ecological point of view (reduction of greenhouse gasses and possible generation of waste materials), but also from the economical (strategies for reusing collected CH_4) and barn management sides. The evaluation of the emissions will be based on the CO_2 mass balance method. Specifically, the ventilation flow rate will be estimated according to CO_2 concentration indoors and outdoors, and the relative animal activity approximated on the base of the sinusoidal equation as reported in [17]. The ventilation flow will then be multiplied by the measured CH_4, N_2O and NH_3 concentration difference to estimate the emissions. In addition to this, monitoring stations are being installed in different fixed areas of the barn available for the project to continuously analyze (24h/7) the concentration of gases during several months of data taking. This will allow a complete understanding of concentrations expected in different seasons of the year and in different weather conditions, with the goal of identifying the optimal position for the installation of a prototype for the capture of CH_4. The two stations prepared are equipped with pressure, temperature, humidity and gas sensors (especially CO_2 and CH_4, which are the most abundant components). In parallel, a data acquisition-system dedicated to the readout of the monitoring stations has been developed. The hardware system is based on commercial cards, such as Arduino [18] for the control of the sensors and Raspberry Pi [19] for the delivery of data to a remote computer via network. A custom software has been developed both for the remote control and data readout of the stations. The installation of the monitoring stations in the barn will be carried out with the goal of minimizing the impact of the presence of the stations themselves on the normal operation of the barn, as well as preserving the lifetime of sensors, minimizing the maintenance needs.

The data taking will cover several months to inspect a variety of environmental conditions which could affect the diffusion of gases, such as different temperatures, pressures or the presence of wind. The data collected will be analyzed daily to keep track of the evolution of data and to correlate them with the activities performed in the barn. We expect to be able to identify the most interesting regions in the barn for the CH_4 prototype installation after a data taking period of about 6 months with the monitoring stations. Anyway, if the operational conditions in the barn allow it, we could continue the monitoring also in the following period to complete the barn atmospheric mapping in a whole year time period.

2.3. Research on adsorption material

A great effort is focused on the testing of different candidates as CH_4 adsorbers materials. We will evaluate the ability of these materials to select CH_4 among other gaseous pollutants present in the barn in significant concentrations. A first set of tests will be carried out using commercial molecular sieves, already adopted for the capture of other gas components, to understand their ability to capture CH_4. In parallel, new materials will be tested at the Laboratory of Radiation Chemistry and EPR Spectroscopy of the University of Pavia. In particular, the investigation will focus on porous materials, such as pyrolytic carbon materials and zeolites [8][20], which represent good candidates for the purpose. Carbon-derived materials have the advantage of being hydrophobic, therefore minimizing concurrent water adsorption. On the other hand, zeolites are more polar but their loading efficiency is higher with respect to carbon materials. The best candidate zeolites to be tested are summarized in Table 1.

Zeolite code	Formula	Crystallographic Info	Syntesys Info	Name
SBN	$[(MAH+)_4]_2[Ga_4Ge_6O_{20}]_2$-MAH+ =tetramethylammonium ion	[21]	[24]	UCSB-9
ZON	$[(TMA+)_4]_2[Zn_4Al_{12}P_{16}O_{64}]_2$-	[22]	[25]	ZAPO-M1
FER	$[Mg_2Na_2(H_2O)_{18}][Al_{16}Si_{30}O_{72}]$-	[23]	[26]	Ferrierite

Table 1: Most promising candidate zeolites to be tested within the project.

Zeolites are renowned for their high thermal and chemical stability. Their synthesis is straightforward, and it is based on the correct ratio about the mixture of oxides inserted into the appropriate vessel. Temperature and processing time are important factors for the synthesis. In our project, the preparation will be performed by inserting the needed components in a Teflon-lined autoclave and bringing them to the proper temperature for the needed time [27]. Usually, the synthesis of zeolites is carried out in hydrothermal conditions and starts from simple and low-cost materials. Other techniques might be also used for their preparation, such as alkali leaching methods or sol-gel [27]. Their basic structure is composed of hollow cavities of different dimensions, whose structure depends mainly on the ratio of components Si/Al. 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. Tailoring of the characteristics will be possible by measuring directly the adsorption capacity and their selectivity. In addition to zeolites, pyrolytic carbon compounds will be investigated, with a special attention, sustainability-wise, on the choice of the starting material. Vegetable-derived and waste biomasses will be the first choice. Among them, we will consider lignin, kraft lignin and other ligno-cellulosic material. For the preparation of these compounds, the material is placed under a high vacuum inside a quartz tube, and the temperature might be increased up to 1100 °C for the required time. The resulting compound's nature depends on the composition of the starting material, the presence of inorganic salts, the final temperature reached in the process and its increase rate. After preparation, these new materials will undergo an adsorption measurement, performed by inserting the adsorber and a gas mixture in a plastic bag (1 liter). The adoption of this system allows us to work close to ambient pressure. This will be useful for the comparison between synthetic and sampled gas. The gas is sampled and analyzed with a quadrupole gas analyzer at ambient temperature and pressure to quantify the material adsorption.

2.4. Preparation, testing and deployment of a prototype for CH_4 capture

The main target of the project is the design, assembly and deployment of the CH4 capture prototype. The prototype, sketched in Fig. 4, will be designed on the basis of an already existing CF4 recovery plant [13][28][29].

In the testing phase, a gas mixer is used to prepare the desired test gas mixture with air, CH_4 and other pollutants which are present in the barn (CO_2, N_2O, NH_3 and humidity). The quality of the gas mixture can be checked using a dedicated extraction line for the analysis. A first column is used to remove humidity from the gas mixture, down to a concentration of a few ppm. A second column is then filled with the adsorbing material selected for CH_4 capture. Finally, an extraction line can be delivered to a gas

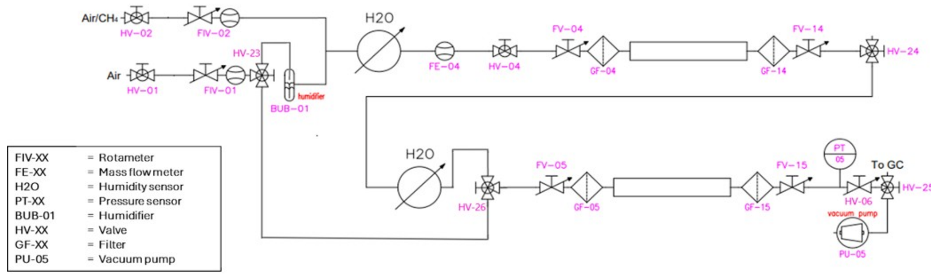

Figure 4: Setup used to test CH_4 capture and filtration of pollutant gases. Details in the text.

chromatograph for analysis. When the system is operated, the gas is flushed inside the columns: target molecules (e.g. water in the first column) remain trapped in the adsorber material by the effect of Van Der Waals interaction of gas molecules with the porous material. When the material has reached full adsorption capacity, it loses its adsorption power and has to be regenerated. This operation can be performed either by extracting the trapped molecules with a vacuum pump, or, if they are too strongly bound to the material, by heating up the cartridge. With this setup, we plan to investigate:

1. the selectivity of columns to different pollutants;
2. the time of exhaustion of a column and the procedure needed to recover it.

This investigation process will take place in the laboratory where the system will be assembled, calibrated and tested, while in a second moment a data taking campaign in the barn will then take place for a few months and data will be analyzed to understand the real efficiency of the prototype in realistic and controlled environmental and operational conditions.

3. Evaluation of the impact of the project

As mentioned in Sec. 1, livestock heavily contributes to the negative environmental impact of agriculture, especially on water/air quality. According to FAO, gas emissions from global livestock production amount to 7.1 Gt/yr of CO_2-equivalent [30], i.e. about 15% of all the anthropogenic GreenHouse Gases (GHG) emissions. Ruminants are the main producers of GHG by enteric fermentation, but also livestock housing and manure storage and management are responsible for NH_3, CH_4 and N_2O emissions. These emissions amount to 3.6 Mt/yr of NH_3, 185 N_2O Mt/yr and 245 CH_4 Mt/yr [31]. In this scenario, planning and validating new systems to capture NH_3 and GHG to recycle as fertilizers and renewable energy sources is a new eco friendly and circular approach that could help in reducing the environmental impact arising from livestock. According to this vision, in the short term, we aim at validating a novel technological strategy accessible to and practicable by farms of any size to reduce GHG emissions from barns. This would result in a high positive impact on global challenges such as climate change, renewable energy and animal welfare in a perspective of circular bioeconomy and one-health approach. To build up scientific knowledge about this hypothesis, a demonstration in a real operational environment is expected as a result from this project. It could open perspectives for further environmental and economic benefits arising from livestock management, falling within the framework of the Green Supply Chain Management (GSCM) and Sustainable Supply Chain Management (SSCM) concepts [32].

4. Conclusions

Fighting CH_4 emissions is crucial in the climate change mitigation effort. In this context, emissions from livestock farming pose a twofold difficulty. First of all, emissions should be reduced without affecting animal life. Furthermore, capturing CH_4 after it has been released into the atmosphere, where its concentration is very low, is highly challenging. Hence, the CH_4 Livestock Emission project proposes an interesting and low-cost technological alternative that has already been validated in the very different context of the Large Hadron Collider. Pursuing research in this area is crucial, from a climate perspective, but also from an energetic point of view, because captured CH_4 could later provide a clean resource from further harmful emissions.

Author Contributions

- Conceptualization: I. Vai, S. Calzaferri, P. Salvini
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- Methodology: M. C. Arena, D. Biagini, E. Dinuccio, D. Dondi, R. Guida, B. Mandelli

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- Writing – original draft: I. Vai
- Writing – review and editing: I. Vai, P. Montagna, L. Finco, C. Aimè, S. Calzaferri, M. Brunoldi, F. A. Angiulli

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Competing Interests

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

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Research highlights

- Methane released from anthropogenic activities contributes 23% to global warming
- Livestock emissions are difficult to handle due to low concentration and animals welfare
- Technologies developed for high energy physics can be applied to fight climate change

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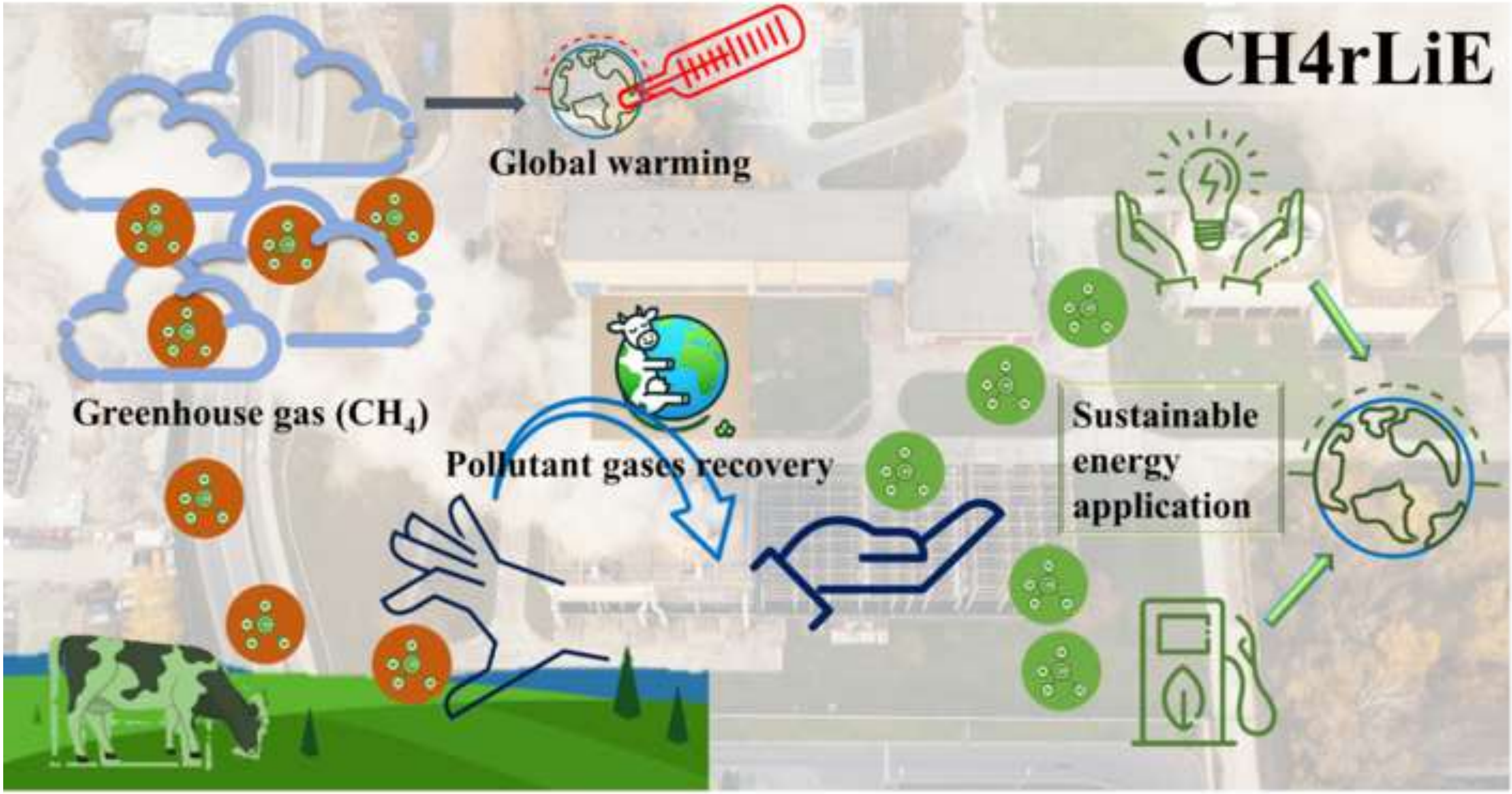

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