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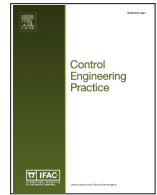
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Model-free control of direct air capture: Optimal rule-based policies tuned via Bayesian optimization

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ABSTRACT

Direct Air Capture is a promising carbon dioxide removal technology, offering safe, flexible, and scalable negative emissions. However, the capture costs and productivity of the Direct Air Capture process are highly influenced by fluctuations in weather conditions, such as ambient temperature and humidity, which hinder its large-scale adoption. Thus, in order to commercialize Direct Air Capture, it is vital to design a control method that reduces the capture costs and enhances productivity while considering the effects of local climate conditions. Due to the complexity of the coupled thermodynamic, heat, and mass transfer phenomena at the core of Direct Air Capture, the unavailability of a sufficiently efficient model, the intractability of parameter estimation, and the high computational costs of model-based approaches, we propose a model-free and data-driven rule-based control approach to improve the performance of Direct Air Capture and reduce its costs. The proposed rule-based control strategy functions as an online policy, continuously receiving feedback from ambient temperature and humidity, enabling dynamic adjustments. Therefore, to maximize the overall system performance, we need to obtain the optimal control law within the considered class of rule-based control schemes. To this end, we utilize a Bayesian optimization framework to optimally tune the parameters of the rule-based control strategy, leveraging data from the online operation influenced by climate conditions. We demonstrate that the proposed method increases the annual Direct Air Capture productivity by up to 16.7 % and lowers the annual capture costs by up to 10.3 % relative to the baseline method. We also observe that the proposed method achieves an annual productivity improvement of 13.59 % and an annual cost reduction of 9.30 % relative to the baseline in Amsterdam, outperforming a data-enabled predictive control method, which achieves 7.07 % productivity improvement and 8.25 % cost reduction, and a reinforcement learning-based controller, which achieves 3.63 % productivity improvement and 3.12 % cost reduction.

1. Introduction

Achieving the maximum 1.5 °C global warming goal of the Paris Agreement (Nations, 2015) may not be feasible without the active removal of CO₂. The development of renewable energy resources alone is not sufficient to meet this target, especially while distributed emissions from the transport and agriculture sectors remain (Group, 2021; Souvik & Ganguly, 2017). Consequently, it is essential for the Carbon Dioxide Removal (CDR) industry to grow towards gigatonne scale by 2050 (Höglund, 2025; National Academies of Sciences & Medicine, 2019).

Among CDR technologies, *Direct Air Capture* (DAC) is one of the most promising, as it offers a safe, flexible, and scalable solution for carbon dioxide removal (Fuss & Lamb, 2018; Kampman & Kruit, June 2023).

The most common DAC approaches capture CO₂ either in an aqueous alkaline solution, or on an amine-functionalized sorbent (Wiegner et al., 2022). In this work, we consider the latter, solid-sorbent process. The significant energy demand of DAC renders its current implementation economically impractical (Climeworks, 2022; National Academies of Sciences & Medicine, 2019). Hence, to further grow DAC technology towards commercial scale, it is essential to design a control method that reduces capture costs and enhances productivity.

A static optimization problem is proposed in Wiegner et al. (2022) to enhance DAC performance, highlighting the importance of considering environmental and operational conditions. In Lehtveer and Emanuelsson (2021), fluctuations in ambient temperature and humidity are introduced as key contributors to the high costs and low productivity of

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DAC systems. In Schellevis et al. (2021), the effects of environmental factors like temperature and humidity variations on DAC performance are assessed. The operational experience with a DAC pilot system, using a circulating amine-based solid sorbent concept, from different outdoor test campaigns, is discussed in Srinivas and Brilman (2024). In Sagel et al. (2024), an algorithm is designed to optimize the design and performance of a solid-sorbent fixed-bed DAC system. Although recent studies focus on improving sorbents and processes and emphasize climatic effects on DAC performance, they typically assume steady-state laboratory conditions and do not develop dynamic control and optimization methods.

One potential approach to improving the performance of the control systems, including DAC is to use models for control design, known as model-based methods. However, utilizing model-based approaches involves several challenges (Nobar et al., 2024; Novara & Formentin, 2017). Black-box models are often non-interpretable, poorly generalizable, highly nonlinear, and thus unsuitable for control design (Hassija et al., 2024; Rudin, 2019). On the other hand, constructing physics-based models requires expert knowledge in complex fields. Specifically, for the DAC system, the laws of thermodynamics, heat and mass transfer, and (co-)adsorption kinetics are required (Alesi & Kitchin, 2012; Bos et al., 2019; Sayari et al., 2012; Yu et al., 2017). Additionally, a parameter estimation method is usually needed for systems with parametric models, such as DAC systems. Thus, extensive data or full measurements, which are typically unavailable, are required for developing such models. Consequently, developing precise models for many systems, including DAC, is highly challenging, which affects their performance and cost-efficiency (Sagel et al., 2024; Schellevis et al., 2024, 2021; Schellevis, 2023). The physical behavior of some systems, such as DAC, is described by Partial Differential Equations (PDEs) (Schellevis et al., 2021; Schellevis, 2023). Thus, for these systems, tuning the model to match real-world conditions requires solving highly complex, nonconvex PDE constrained optimization problems. However, solving these problems is usually intractable and not applicable for real-time system operations. Hence, model-based methods are not suitable for the DAC system.

Data-driven control methods are introduced as alternative schemes for complex systems. These methods were first developed for linear systems (De Persis & Tesi, 2019; Van Waarde et al., 2020) and were later extended to special classes of nonlinear systems (Bisoffi et al., 2020; Guo et al., 2021; Luppi et al., 2022). However, a direct data-driven control approach for general nonlinear systems has yet to be established in the literature. The Bayesian Optimization (BO) based control is a well-known data-driven method that can be used for any nonlinear system. In this method, BO utilizes data to tune the parameters of the closed-loop controller through a series of limited experiments. BO can efficiently explore the solution space with minimal computational cost. It relies on a surrogate model to approximate the performance function (Garnett, 2023). To achieve this, Gaussian process regression (Seeger, 2004) is employed. BO is widely used in various applications (Guidetti et al., 2022; Khosravi et al., 2020, 2021, 2019a; König et al., 2024, 2023; Nobar et al., 2024; Khosravi, Mohammad and König, Christopher and Maier, Markus and Smith, Roy S. and Lygeros, John and Rupenyan, Alisa, 2022; Tang et al., 2024). In (Nobar et al., 2024), a guided BO algorithm is proposed, which iteratively tunes the parameters of the closed-loop controller using an event-triggered digital twin. Based on the BO method, a real-time data-driven approach for adaptive control of high-precision motion systems is presented in König et al. (2024). In König et al. (2023), a novel data-driven approach, which combines safe learning with risk-averse BO, is proposed for safe controller tuning. The BO optimization approach is applied to atmospheric plasma spraying and fused deposition modeling in Guidetti et al. (2022), minimizing the process cost. In Khosravi et al. (2019a,b), safe BO approaches are utilized for controller parameter tuning of a heat pump system. An automated data-driven method is presented in Khosravi, Mohammad and König, Christopher and Maier, Markus and Smith, Roy S. and Lygeros, John

and Rupenyan, Alisa (2022) for the safe tuning of proportional-integral-derivative cascade controller gains based on BO. A BO-based control strategy is proposed in Tang et al. (2024) for process quality control, using quality information with adaptive local convergence. Thus, BO can be used to optimally tune the parameters of the controller for the DAC system, enhancing its performance.

Following the above discussion, in this paper, we propose a model-free and data-driven rule-based control strategy for online regulation of the superficial gas velocity in DAC, according to the measured ambient temperature and humidity. The proposed rule-based control strategy significantly enhances the performance of the DAC process and overcomes the limitations of model-based techniques. To optimally tune the parameters of the controller, we utilize BO (Garnett, 2023), using data from the online operation of DAC. The contributions of the paper can be listed as follows:

- we propose a rule-based control strategy that operates as an online policy, continuously receiving feedback from ambient temperature and humidity to enable dynamic adjustments;
- the proposed rule-based control strategy improves DAC performance by enhancing its productivity and reducing capture costs, addressing the aforementioned challenges of model-based approaches;
- we utilize a BO framework to optimally tune the parameters of a rule-based control strategy for the DAC system, without relying on predefined models;
- the use of BO ensures adaptability to system variations while maintaining computational efficiency via a series of limited experiments, thereby avoiding additional expensive DAC process experiments.

The rest of the paper is organized as follows. Section 2 describes the DAC control design problem. Section 3 formulates the optimal rule-based control problem. Section 4 presents the BO-based tuning method. Section 5 introduces the high-fidelity model, Section 6 provides numerical results, and Section 7 concludes the paper.

Notation: The set of real numbers is denoted by $\mathbb{R}$. The set of non-negative real numbers is indicated by $\mathbb{R}_+$. Let $\mathbb{I}$ denote the identity matrix. A Gaussian process prior for the function $J : \mathcal{Z} \rightarrow \mathbb{R}$, is denoted by $\mathcal{GP}(\mu, \kappa)$, where $\mu : \mathcal{Z} \rightarrow \mathbb{R}$ and $\kappa : \mathcal{Z} \times \mathcal{Z} \rightarrow \mathbb{R}$ are the mean and kernel functions in the Gaussian process prior for J , respectively. Furthermore, $\mathbb{E}_{v \sim p_v}[\cdot]$ denotes the mathematical expectation with respect to the random variable v , where p_v is the probability distribution of v .

2. Control design for direct air capture

In this section, we present the control design problem for the solid-sorbent Direct Air Capture (DAC) unit utilizing a steam-assisted temperature vacuum swing adsorption process.

In the DAC process, increased temperature, reduced pressure, and a steam purge are applied to release the captured CO_2 in a concentrated stream, while the sorbent is regenerated for the next cycle of the process. This process consists of the following cyclic steps: (1) adsorption, (2) evacuation, (3) heating, (4) desorption, (5) cooling, and (6) repressurization. These cyclic steps are demonstrated in Fig. 1. More precisely, in the adsorption step, CO_2 molecules are adsorbed through chemical reactions with amine functional groups on the interior surfaces of the adsorbent. The subsequent steps (2) to (6) are referred to as *regeneration*, aimed at separating the concentrated CO_2 from the sorbent and preparing the adsorbent for the following cycles. The cyclic steps of the DAC process are governed by the laws of thermodynamics, resulting in coupled and nonlinear physics (Schellevis et al., 2021; Schellevis, 2023). The main challenge is that the high energy demand of DAC makes current implementations economically impractical (Climeworks, 2022; National Academies of Sciences & Medicine, 2019), while complex dynamics limit the applicability of existing performance optimization methods.

Recent research on DAC technology mainly focuses on improving sorbents, processes, and new designs for DAC under static and laboratory conditions (Lehtveer & Emanuelsson, 2021; Sagel et al., 2024;

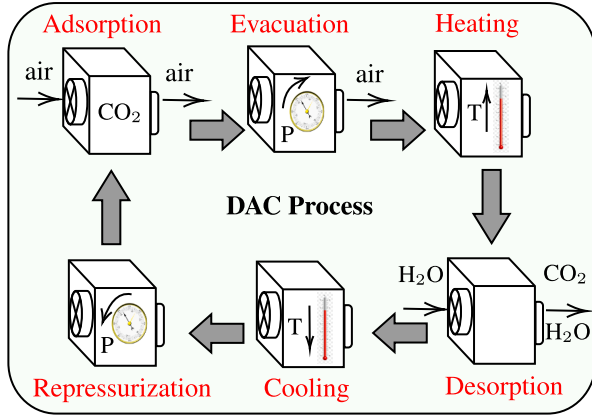


Fig. 1. The process steps of solid-sorbent DAC technology with pressure and temperature indicators. The cyclic steps are (1) adsorption, (2) evacuation, (3) heating, (4) desorption, (5) cooling, and (6) repressurization.

Schellevis et al., 2021; Srinivas & Brilman, 2024; Wiegner et al., 2022). Nevertheless, control design provides a practical, low-cost, and readily deployable strategy to optimize DAC performance, given its strong influence on system operation and its ability to enhance efficiency under varying operating conditions. For control design, the primary approach is model-based, relying on an accurate mathematical representation of DAC. Models obtained through complex black-box methods are non-interpretable, poorly generalizable, highly nonlinear, and therefore unsuitable for control purposes. (Hassija et al., 2024; Rudin, 2019). Extensive physics-based modeling methods require expert knowledge of DAC thermodynamics and involve parameter estimation that is computationally intractable and demands large, informative, and accurate set of measurement data, making them impractical for real-world application. Schellevis et al. (2024, 2021), Schellevis (2023). Also, simplified black-box and physics-based models, on the other hand, fail to capture the exact dynamics of DAC due to the high complexity of the process. Data-driven control methods, which are model-free and based directly on operational data, are typically designed for linear, noise-free, low-order systems (De Persis & Tesi, 2019; Van Waarde et al., 2020) and are therefore not suitable for DAC, given its complex physical dynamics.

Note that, in the DAC process, the adsorption time, heating time, desorption time, and cooling time are primarily treated as design parameters. On the other hand, the superficial gas velocity, denoted by u_g , is a variable that can be directly controlled in practical DAC systems. In particular, the airflow through the contactor can be regulated online by adjusting the blower fan actuation signal, e.g., through motor current control or the associated drive electronics. Accordingly, we consider u_g as the main control variable in this work, on which the overall performance of the DAC process depends. Following the above discussion, given the complexity of DAC, a suitable control design should be maximally model-free and adaptable to different settings. Thus, we employ a rule-based control strategy for online adjustment of u_g to achieve optimal DAC performance.

The CO₂ adsorption is highly sensitive to ambient temperature T_{amb} and relative humidity RH_{amb} (Bajamundi & Koponen, 2019; Silani et al., 2025; Wiegner et al., 2022), which are considered system disturbances, i.e., uncontrolled environmental input variables. More precisely, the ambient temperature appears in the initial condition of temperature during adsorption, while both temperature and the relative humidity affects the adsorption equilibria and rates of both CO₂ and water. Thus, we define the disturbance vector $v \in \mathcal{V}$ as

$$v := [T_{amb}, RH_{amb}]^T, \quad (1)$$

where $\mathcal{V} \subseteq \mathbb{R}^2$ denotes the set of admissible disturbances v . To improve the performance of the DAC system and mitigate the effect of climate conditions, we develop a rule-based control strategy for u_g for online

regulation of the superficial gas velocity according to the measured disturbance vector $v \in \mathcal{V}$. To this end, we consider a suitable dictionary of laws, namely $\{\psi^{(i)} : \mathbb{R}_+^2 \rightarrow \mathbb{R}_+ \mid i = 1, \dots, l\}$, as our set of base control rules, and subsequently, define a parametric class of control laws as

$$\mathcal{L}_\Psi := \left\{ \Gamma_\zeta := \Psi\zeta \mid \zeta \in \mathcal{Z} \right\}, \quad (2)$$

where Ψ is defined as $\Psi := [\psi^{(1)}, \dots, \psi^{(l)}]$, $\zeta = [\zeta^{(1)}, \dots, \zeta^{(l)}]^T$ is the vector of parameters characterizing the control laws, and $\mathcal{Z} \subseteq \mathbb{R}^l$ denotes the set of admissible controller parameters ζ . More precisely, any rule-based control law Γ_ζ in $\mathcal{L}_\Psi$ can be explicitly written as

$$\Gamma_\zeta = \zeta^{(1)}\psi^{(1)} + \dots + \zeta^{(l)}\psi^{(l)}. \quad (3)$$

For the basis functions $\psi^{(1)}, \dots, \psi^{(l)}$, any appropriate family of functions may be employed, including Radial Basis Functions (RBFs), polynomial bases, or other functional parameterizations customized to the context, that can be beyond the case discussed in this paper. Given $\psi^{(1)}, \dots, \psi^{(l)}$, the objective is to derive the *optimal control law* within the class $\mathcal{L}_\Psi$, and thereby, define the corresponding rule-based control strategy for u_g as

$$u_g := \Gamma_{\zeta^*}(v), \quad (4)$$

where ζ^* is the optimally tuned vector of controller parameters.

In this work, we primarily opt for polynomial functions of the ambient conditions as the basis for the control laws. Specifically, the basis functions are chosen as a third-order polynomial function of the ambient conditions, resulting in $l = 10$, that include constant, linear, quadratic, and cubic terms, and hence a parameter vector $\zeta \in \mathbb{R}^{10}$. The third-order polynomial basis function is selected to balance modeling flexibility and computational efficiency. Although basis functions such as RBFs could also be employed within the proposed framework, the polynomial basis functions were found suitable for capturing the dominant nonlinear effects of ambient conditions on DAC operation. Moreover, we will show in Section 6 that the third-order polynomial basis function outperforms lower-order polynomial basis functions and RBFs.

The operation of DAC units inherently follows a cyclic pattern, which enables the design of experiments that iteratively evaluate different control settings over successive cycles. To determine the optimal controller parameters ζ^* , candidate parameter vectors can be sampled from the feasible set associated with the class $\mathcal{L}_\Psi$ and evaluated directly on the system. To guide the resulting iterative sampling procedure, we define a suitable objective function that captures the desired operational criteria, and to efficiently explore the parameter space, we employ Bayesian Optimization (BO). Thus, we can leverage experimental data to update a surrogate model of the objective function and select informative parameter samples, requiring no explicit system model while maintaining low computational overhead. Additional methodological details are provided in the following sections.

3. Optimal rule-based control problem for direct air capture

In order to improve the performance of the DAC process, we should determine the optimal controller parameters ζ^* such that the total energy cost is minimized and the productivity of the DAC process is maximized. The total energy cost of DAC process, measured in J/kg_{CO₂}/d and denoted by C_{DAC} , is categorized into seven distinct components, including the energy required for the CO₂ endothermic desorption, energy required for the H₂O desorption, sorbent sensible heat, reactor sensible heat, latent and sensible heat of the purge gas, feed compression energy, and vacuum compression energy, which are all measured in J/kg_{CO₂}/d and denoted by E_{r,CO_2} , E_{r,H_2O} , $E_{sens,s}$, $E_{sens,r}$, E_{purge} , E_{feed} , and E_{vac} , respectively. Then, the total energy cost of the DAC process is given by

$$C_{DAC} := E_{r,CO_2} + E_{r,H_2O} + E_{sens,s} + E_{sens,r} + E_{purge} + E_{feed} + E_{vac}, \quad (5)$$

which is primarily a function of u_g and v when other parameters are held constant. Moreover, the productivity of DAC process measured in

$\text{kg}_{\text{CO}_2}/\text{kg}_s/\text{d}$ is denoted by P_{DAC} , which is similarly a function of u_g and v , under the mentioned conditions. Further details on costs and productivity are available in Appendix B, explicitly defined through (B.1)–(B.8). Let the rule-based control law $u_g = \Gamma_\zeta(v)$ be applied. Subsequently, let function $J_{\text{DAC}} : \mathbb{R}_+ \times \mathbb{R}_+^2 \rightarrow \mathbb{R}$ be defined as

$$J_{\text{DAC}}(\Gamma_\zeta(v), v) := W_1 C_{\text{DAC}}(\Gamma_\zeta(v), v) - W_2 P_{\text{DAC}}(\Gamma_\zeta(v), v), \quad (6)$$

where W_1, W_2 are positive weights. Accordingly, we define the objective function $J_{\text{DAC}}^{\text{av}} : \mathcal{Z} \rightarrow \mathbb{R}$ as

$$J_{\text{DAC}}^{\text{av}}(\zeta) := \mathbb{E}_{v \sim p_v} [J_{\text{DAC}}(\Gamma_\zeta(v), v)], \quad (7)$$

where p_v is the probability distribution modeling the uncertainty in v . Then, we obtain the optimal ζ^* to minimize the objective function. To do this, we solve the optimization problem formulated as

$$\begin{aligned} \min_{\zeta \in \mathcal{Z}} J_{\text{DAC}}^{\text{av}}(\zeta) \\ \text{s.t. } \Gamma_\zeta(v) \in [\underline{u}_g, \bar{u}_g], \end{aligned} \quad (8)$$

where $\underline{u}_g$ and $\bar{u}_g$ are the lower and upper bounds of the superficial gas velocity. Note that $J_{\text{DAC}}(\Gamma_\zeta(v), v)$ is an unbiased estimator for $J_{\text{DAC}}^{\text{av}}(\zeta)$.

To address the challenges associated with model-based methods, we propose a model-free and data-driven rule-based control strategy to enhance DAC performance and lower costs. The objective function $J_{\text{DAC}}^{\text{av}}(\zeta)$ is a black-box function. The value of the objective function $J_{\text{DAC}}^{\text{av}}(\zeta)$ can be approximately determined through the *oracle* $J_{\text{DAC}}(\Gamma_\zeta(v), v)$, i.e., by conducting an experiment in which the controller parameters are set to ζ . During the experiment, the system undergoes a full operational cycle, and the oracle is computed from the resulting complete cycle of the adsorption, evacuation, heating, desorption, cooling, and repressurization steps. In order to lower the number of invocations of this experimental oracle, BO is utilized (Gardner et al., 2014; Rupenyan et al., 2021; Shahriari et al., 2015), which is a powerful approach recognized for its utility in control process applications (Tang et al., 2024). Indeed, to optimize the parameters of the rule-based control strategy, we utilize the BO framework (Garnett, 2023), which efficiently explores the solution space while maintaining minimal computational cost.

4. Data-driven controller tuning via Bayesian optimization

For a given $\zeta \in \mathcal{Z}$ and $v \in \mathcal{V}$, the value of the black-box objective function $J_{\text{DAC}}^{\text{av}}(\zeta)$ can be approximately determined through the *oracle* $J_{\text{DAC}}(\Gamma_\zeta(v), v)$, i.e., by conducting an experiment, a full operational cycle of DAC, in which the controller parameters are set to ζ . During the experiment, the system undergoes a full operational cycle, and the oracle is computed from the resulting complete cycle of the adsorption, evacuation, heating, desorption, cooling, and repressurization steps. In order to lower the number of invocations of this experimental oracle, BO is utilized (Gardner et al., 2014; Krause & Ong, 2011; Rupenyan et al., 2021; Shahriari et al., 2015), which is a powerful approach recognized for its utility in control process applications (Tang et al., 2024). More precisely, BO is a method that leverages data to obtain optimal variables through a series of limited experiments. It relies on a surrogate model to approximate the objective function, guiding the optimization process efficiently (Garnett, 2023). To achieve this, Gaussian process regression (Seeger, 2004) is employed to create a surrogate model for the objective function while also assessing the process uncertainties and disturbances.

Let us define an initial set of the controller parameters as

$$\zeta_{\text{init}} := \{\zeta_i \in \mathcal{Z} \mid i = 1, \dots, n_{\text{init}}\}, \quad (9)$$

where ζ_{init} can be generated either by applying random feasible perturbations to its nominal values, or by using a Latin hypercube experimental design to create a maximally informative set (McKay et al., 2000). We consider the dataset

$$B_n := \{(\zeta_i, \rho_i) \mid i = 1, \dots, n\}, \quad (10)$$

where n is the index of iteration and ζ_i is the vector of controller parameters, and ρ_i is the obtained value for $J_{\text{DAC}}^{\text{av}}(\zeta_i)$ in the experiment i , i.e., we have

$$\rho_i := J_{\text{DAC}}(\Gamma_{\zeta_i}(v_i), v_i) = J_{\text{DAC}}^{\text{av}}(\zeta_i) + v_i, \quad (11)$$

where v_i denotes the uncertainty in estimating $J_{\text{DAC}}^{\text{av}}(\zeta_i)$ using ρ_i , modeled as a zero-mean noise term. We construct a surrogate model for $J_{\text{DAC}}^{\text{av}}$, at iteration $n \geq n_{\text{init}}$, using the dataset B_n and Gaussian Process Regression (GPR), denoted by $\hat{J}_{\text{DAC},n}^{\text{av}}$. More precisely, we consider a Gaussian process prior for the function $J_{\text{DAC}}^{\text{av}}$, represented as $\mathcal{GP}(\mu, \kappa)$, where $\mu : \mathcal{Z} \rightarrow \mathbb{R}$ and $\kappa : \mathcal{Z} \times \mathcal{Z} \rightarrow \mathbb{R}$ are the mean and kernel functions in the Gaussian process prior for $J_{\text{DAC}}^{\text{av}}$, respectively. To this end, we define the information matrices ζ_n and ρ_n respectively as

$$\zeta_n := [\zeta_1, \zeta_2, \dots, \zeta_n]^T, \quad (12)$$

and

$$\rho_n := [\rho_1, \rho_2, \dots, \rho_n]^T. \quad (13)$$

Furthermore, at iteration n , let the *mean function* $\mu_n : \mathcal{Z} \rightarrow \mathbb{R}$ and the *variance function* $\omega_n : \mathcal{Z} \rightarrow \mathbb{R}$ be respectively defined as

$$\mu_n(\zeta) := \mu(\zeta) + \mathbf{k}_n(\zeta)^T (K_n + \sigma^2 \mathbb{I})^{-1} (\rho_n - \mu(\zeta_n)), \quad (14)$$

and,

$$\omega_n^2(\zeta) := \kappa(\zeta, \zeta) - \mathbf{k}_n(\zeta)^T (K_n + \sigma^2 \mathbb{I})^{-1} \mathbf{k}_n(\zeta), \quad (15)$$

for any $\zeta \in \mathcal{Z}$, where $\mathbb{I}$ is the identity matrix and σ^2 is the variance of uncertainty in the computations of $J_{\text{DAC}}^{\text{av}}$. Thus, for any $\zeta \in \mathcal{Z}$, we have

$$J_{\text{DAC}}^{\text{av}}(\zeta) \sim \mathcal{N}(\mu_n(\zeta), \omega_n^2(\zeta)). \quad (16)$$

Moreover, let the vector $\mu(\zeta_n)$ and matrix K_n be respectively defined as

$$\mu(\zeta_n) := [\mu(\zeta_1), \dots, \mu(\zeta_n)]^T \in \mathbb{R}^n, \quad (17)$$

and

$$K_n := [\kappa(\zeta_p, \zeta_q)]_{p,q=1}^n \in \mathbb{R}^{n \times n}. \quad (18)$$

Also, let function $\mathbf{k}_n : \mathcal{Z} \rightarrow \mathbb{R}^n$ be defined as

$$\mathbf{k}_n(\zeta) := [\kappa(\zeta, \zeta_1), \dots, \kappa(\zeta, \zeta_n)]^T, \quad (19)$$

for any $\zeta \in \mathcal{Z}$. By utilizing these probabilistic surrogate models for the objective function $J_{\text{DAC}}^{\text{av}}$, we can obtain the parameters $\zeta_{n+1} \in \mathcal{Z}$ to be used in the next experiment and for future calculations of ρ_i . To balance exploration and exploitation, we define the acquisition function $E_n : \mathcal{Z} \rightarrow \mathbb{R}$, corresponding to the Lower Confidence Bound (LCB), as

$$E_n(\zeta) := \mu_n(\zeta) - \beta_n \omega_n(\zeta), \quad (20)$$

where β_n is the desired confidence level.

Using *global optimization methods*, namely genetic algorithm or particle swarm, the following optimization is solved to obtain the next vector of controller parameters

$$\begin{aligned} \zeta_{n+1} := \underset{\zeta \in \mathcal{Z}}{\text{argmin}} E_n(\zeta) \\ \text{s.t. } \Gamma_\zeta(v) \in [\underline{u}_g, \bar{u}_g]. \end{aligned} \quad (21)$$

When ζ_{n+1} is computed, a new experiment with the controller parameters ζ_{n+1} is executed. Subsequently, we update the dataset as

$$B_{n+1} = B_n \cup \{(\zeta_{n+1}, \rho_{n+1})\}. \quad (22)$$

The introduced procedure is repeated until convergence to the solution of (8) is achieved. Fig. 2 illustrates the data-driven tuning scheme of the proposed rule-based control strategy based on the BO method. The implementation of the proposed method is demonstrated in Algorithm 1. This method utilizes data from a high-fidelity model and is capable of operating with real-time data from an experimental DAC system, i.e., actual plant data.

There are different options for the selection of kernels, such as Matern, RBF, and linear kernels. In this work, we utilize the Matern kernel with automatic relevance determination, which uses different length

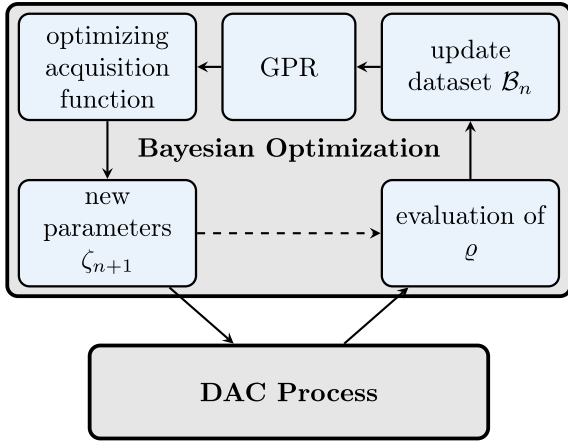


Fig. 2. The data-driven tuning scheme of the proposed rule-based control strategy based on the Bayesian optimization method.

Algorithm 1 The tuning method of the rule-based control strategy.

- 1: Choose the feasible set $\mathcal{Z}$, the dataset $\mathcal{B}_{n_{\text{init}}}$ given in (10), and the Gaussian process prior $\mathcal{GP}(0, \kappa)$, with the kernel function κ .
- 2: Using $\mathcal{B}_{n_{\text{init}}}$, the negative log marginal likelihood function is minimized to estimate the vector of hyperparameters for $\mathcal{GP}(0, \kappa)$.
- 3: $\mathcal{B}_n \leftarrow \mathcal{B}_{n_{\text{init}}}$ and $n \leftarrow n_{\text{init}}$.
- 4: **while** stopping criterion is not satisfied **do**
- 5: Obtain ζ_{n+1} via solving (21).
- 6: Set the parameters of the rule-based controller to ζ_{n+1} for the next experiment.
- 7: Compute ϱ_{n+1} given in (11).
- 8: Update the model of the Gaussian process by calculating μ_{n+1} and ω_{n+1}^2 given in (14) and (15), respectively.
- 9: Update the dataset $\mathcal{B}_{n+1}$ according to (22).
- 10: $n \leftarrow n + 1$.
- 11: **end while**

scale parameters for each input dimension. The hyperparameters of the Matern kernel are the signal variance σ_f^2 , smoothness parameter $\tilde{\chi}$, and diagonal matrix of length scale parameters $\tilde{\mathbf{P}} = \text{diag}(\tilde{\sigma}_1^2, \tilde{\sigma}_2^2, \dots, \tilde{\sigma}_D^2)$, where D is the input dimension and $\tilde{\sigma}_1^2, \tilde{\sigma}_2^2, \dots, \tilde{\sigma}_D^2$ are the length scale parameters. The distance between two inputs $\tilde{\mathbf{x}}$ and $\tilde{\mathbf{x}}'$ is defined as

$$\tilde{r} = \left((\tilde{\mathbf{x}} - \tilde{\mathbf{x}}')^T \tilde{\mathbf{P}}^{-1} (\tilde{\mathbf{x}} - \tilde{\mathbf{x}}') \right)^{\frac{1}{2}}. \quad (23)$$

The Matern kernel can then be expressed as

$$\kappa(\tilde{r}) = \tilde{\sigma}_f^2 \frac{2^{1-\tilde{\chi}}}{\tilde{\gamma}(\tilde{\chi})} \left((2\tilde{\chi})^{\frac{1}{2}} \tilde{r} \right)^{\tilde{\chi}} \tilde{K}_{\tilde{\chi}} \left((2\tilde{\chi})^{\frac{1}{2}} \tilde{r} \right), \quad (24)$$

where $\tilde{\gamma}(\cdot)$ is the gamma function and $\tilde{K}_{\tilde{\chi}}(\cdot)$ denotes the modified Bessel function. The smoothness parameter $\tilde{\chi}$ determines the smoothness of the kernel, smaller $\tilde{\chi}$ values produce less smooth functions, while larger $\tilde{\chi}$ values yield smoother ones (Maier et al., 2020). Then, the hyperparameters of kernel κ , collected in a vector $\theta \in \Theta$, are estimated by minimizing the negative log marginal likelihood (Khosravi et al., 2021; Rasmussen & Williams, 2006), i.e., we have

$$\hat{\theta} := \underset{\theta \in \Theta}{\text{argmin}} -\log p(\varrho_n | \zeta_n, \theta), \quad (25)$$

where $p(\varrho_n | \zeta_n, \theta)$ denotes the probability density function of the measurements. To ensure the convergence of the BO tuning procedure, we kept these values fixed after the initial training (Berkenkamp et al., 2023; Kavas, Barış and Balta, Efe C. and Tucker, Michael R. and Krishnadas, Raamadaas and Rupenyan, Alisa and Lygeros, John and Bam-bach, Markus, 2025; Khosravi, Mohammad and König, Christopher and Maier, Markus and Smith, Roy S. and Lygeros, John and Rupenyan, Alisa, 2022). This choice is consistent with standard Gaussian Process

(GP)-based BO theory, where convergence guarantees rely on a fixed GP prior with non-adaptive kernel hyperparameters. Furthermore, the design of the BO procedure reflects the intended real-world deployment of the proposed framework. In particular, it is designed to tune rule-based controller parameters through repeated real-world DAC operation cycles. In such operational settings, the stopping criterion in Algorithm 1 is defined as a fixed number of BO iterations, rather than a convergence threshold, to capture practical operational considerations. We set the mean function to zero, i.e., $\mu = 0$. If prior knowledge on the structure of $J_{\text{DAC}}^{\text{av}}$ exists, one can choose a different value for the mean function (Khosravi, Mohammad and König, Christopher and Maier, Markus and Smith, Roy S. and Lygeros, John and Rupenyan, Alisa, 2022).

The proposed framework is conceptually related to recent developments in safe BO (Khosravi et al., 2019a; König et al., 2023; Khosravi, Mohammad and König, Christopher and Maier, Markus and Smith, Roy S. and Lygeros, John and Rupenyan, Alisa, 2022), where safety constraints are incorporated. However, for the considered DAC application, defining a meaningful safety constraint is not straightforward. For instance, imposing a minimum productivity requirement would conflict with the intended flexible operation of the DAC unit, which is designed to adapt to the availability of renewable or low-carbon electricity and to prioritize cost efficiency. Consequently, enforcing hard constraints on productivity could restrict economically and operationally viable solutions.

5. High-fidelity model for numerical experiment

We introduce the detailed, physics-based model of a solid-sorbent DAC system based on Lewatit®VP OC 1065 sorbent (Schellevis et al., 2021; Schellevis, 2023; Schellevis et al., 2020; Yu & Brilman, 2020). This model acts as an accurate ground truth where we will validate our proposed data-driven approach. The process is shown schematically in Fig. 1. The model was previously introduced by Schellevis et al. (2021), Schellevis (2023), Schellevis et al. (2020), Yu and Brilman (2020), and represents each stage in the DAC cycle as a set of differential equations. Here we repeat the foremost governing equations, while the detailed description of all parameters is given in Appendices A and B for brevity. The sorbent properties, such as CO₂ adsorption capacity, reaction kinetics, and chemical stability, are described in Alesi and Kitchin (2012), Bos et al. (2019), Sayari et al. (2012), Schellevis et al. (2021), Yu et al. (2017).

5.1. Adsorption

The adsorption model is a transient, 1-dimensional column model, characterized by a system of time-dependent PDEs (Schellevis et al., 2021; Schellevis, 2023; Schellevis et al., 2020; Yu & Brilman, 2020). A key assumption in this model is that the gas velocity remains constant during adsorption, given the relatively low concentrations of the adsorbed species. This leads to the following mass balance equations for CO₂ and H₂O in the gas phase

$$\partial_t c_{m,g} = \frac{1}{\varepsilon_r} \partial_z (D_{a,m} \partial_z c_{m,g} - u_g c_{m,g}) + \frac{1 - \varepsilon_r}{\varepsilon_r} \rho_s \partial_t q_m, \quad (26)$$

where $m \in \{\text{CO}_2, \text{H}_2\text{O}\}$. Furthermore, the mass balance equations for CO₂ and H₂O in the solid phase are expressed as

$$\partial_t q_m = R_m \eta_m, \quad (27)$$

where R_{CO_2} , η_{CO_2} , $R_{\text{H}_2\text{O}}$, and $\eta_{\text{H}_2\text{O}}$ are given in (A.1), (A.7), (A.9), and (A.16) in Appendix A, respectively. Additionally, the model includes the thermal energy balance equation, which is given by

$$\begin{aligned} \partial_t T = & \frac{1}{(1 - \varepsilon_r) \rho_s C_{p,s} + \varepsilon_r \rho_g C_{p,g}} \left(Q_{\text{source}} \right. \\ & + \partial_z (\lambda_{s,\text{eff}} \partial_z T - u_g \rho_g C_{p,g} T) \\ & \left. + (1 - \varepsilon_r) \rho_s \sum_m \Delta H_{r,m} \partial_t q_m \right), \end{aligned} \quad (28)$$

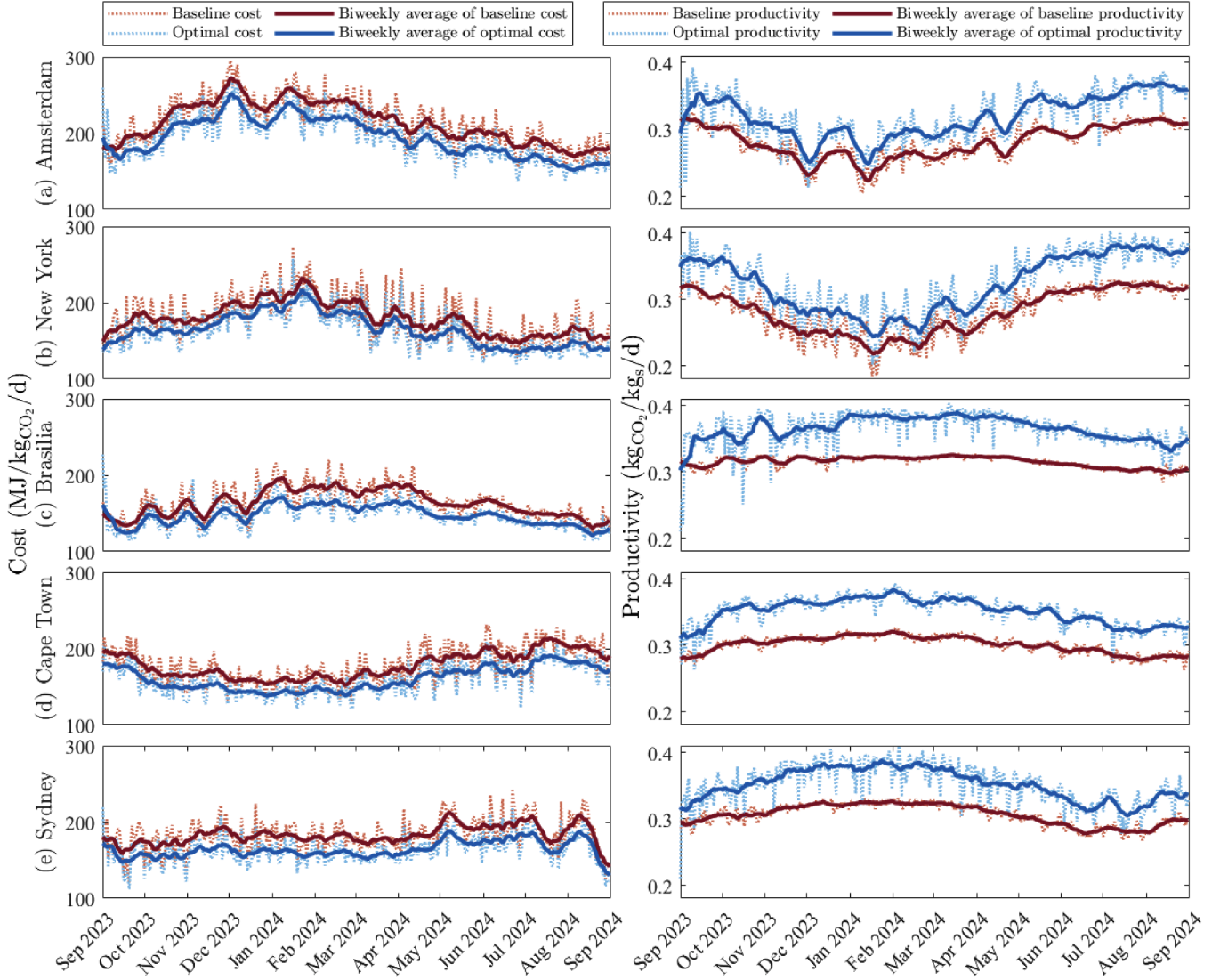


Fig. 3. Cost and productivity in different cities: comparing the proposed and baseline method. Figures (a)–(e) correspond to Amsterdam, New York, Brasilia, Cape Town, and Sydney, respectively. For each city, the left figures show the DAC capture cost, and the right figures show the DAC productivity from September 1, 2023, to September 1, 2024. The baseline method is shown with dotted red lines, and its biweekly averages with solid red. The proposed method is shown with dotted blue lines, and its biweekly averages with solid blue. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

where $Q_{\text{source}} = 0$ for adsorption. Moreover, Danckwerts boundary conditions are applied to the PDEs, which are given in (A.23)–(A.28). The initial conditions are taken from the final conditions of the previous stage, given in (A.29)–(A.33), respectively.

5.2. Heating and cooling

During the heating and cooling phases, only the thermal energy balance is solved

$$\partial_t T = \frac{1}{(1 - \varepsilon_r)\rho_s C_{p,s} + \varepsilon_r \rho_g C_{p,g}} (\lambda_{s,\text{eff}} \partial_z^2 T + Q_{\text{source}}), \quad (29)$$

where Q_{source} , the boundary and initial conditions are given in (A.34)–(A.36) in Appendix A, respectively.

5.3. Evacuation and repressurization

Additionally, to model the evacuation and repressurization stages, we use the ideal gas law

$$c = \frac{p}{RT}, \quad (30)$$

where p is the reduced pressure applied by the vacuum pump in the evacuation and desorption stages or ambient pressure in the repressurization stage, and c is the overall concentration in these stages.

5.4. Desorption

The desorption model operates on the same foundational principles as the adsorption model concerning mass and thermal energy transport. Hence, the dynamic model for desorption is the same as (26)–(28) with Danckwerts boundary conditions (A.23)–(A.28) and initial conditions (A.29)–(A.33), where Q_{source} is given in (A.34) and $\eta_{\text{CO}_2} = 1$. However, the assumption of a constant gas velocity does not hold, particularly during desorption at lower absolute pressures. Thus, to obtain the changing gas velocity, we consider

$$\partial_t T = \frac{RT^2}{\varepsilon_r p} \left(\frac{p}{R} \partial_z \left(\frac{u_g}{T} \right) + (1 - \varepsilon_r) \rho_s \sum_m \partial_t q_m \right), \quad (31)$$

which is derived from the ideal gas law and total mass balance.

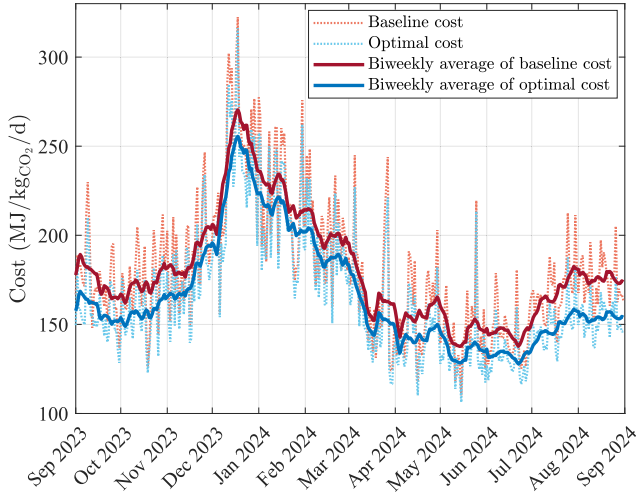


Fig. 4. Cost in Beijing: comparing the proposed and baseline method. The baseline method is shown with dotted red lines, and its biweekly averages with solid red. The proposed method is shown with dotted blue lines, and its biweekly averages with solid blue. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

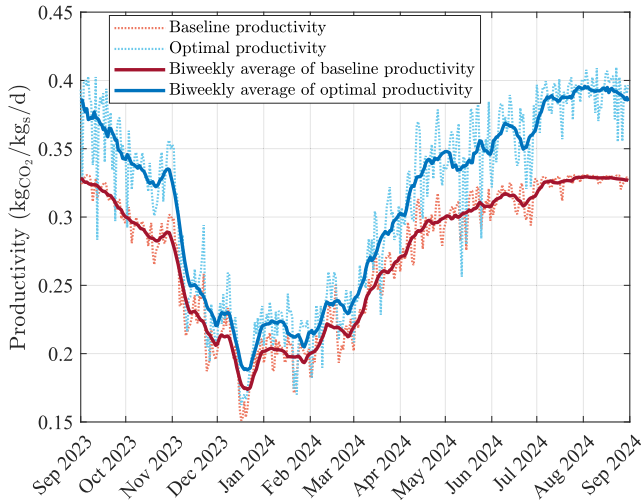


Fig. 5. Productivity in Beijing: comparing the proposed and baseline method. The baseline method is shown with dotted red lines, and its biweekly averages with solid red. The proposed method is shown with dotted blue lines, and its biweekly averages with solid blue. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

5.5. Energy penalty and productivity

The energy consumption of the steam-assisted temperature–vacuum swing adsorption process, measured in $\text{J/kgCO}_2/\text{d}$, is categorized into seven distinct components: the energy required for the CO_2 endothermic desorption, denoted by E_{r,CO_2} ; the energy required for the H_2O desorption, denoted by $E_{r,\text{H}_2\text{O}}$; the sorbent sensible heat, denoted by $E_{\text{sens},s}$; the reactor sensible heat, denoted by $E_{\text{sens},r}$; the latent and sensible heat of the purge gas, denoted by E_{purge} ; the feed compression energy, denoted by E_{feed} ; and the vacuum compression energy, denoted by E_{vac} . These cost components are defined in (B.1)–(B.7) in Appendix B. Furthermore, the productivity of the process, expressed as $\text{kgCO}_2/\text{kg}_s/\text{d}$ and denoted by P_{DAC} , characterizes the efficiency of CO_2 extraction from the air and is given in (B.8) in Appendix B.

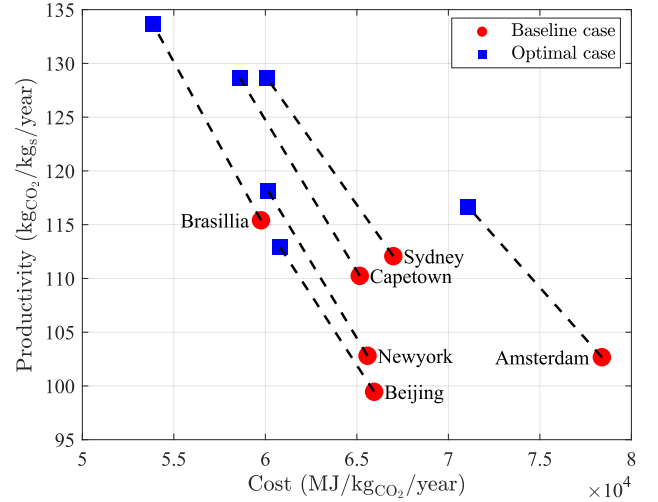


Fig. 6. The comparison of the proposed and baseline method in different cities for one year. The red circles show the baseline method, and the blue squares indicate the proposed method. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

5.6. Experimental validation for the high-fidelity model

The high-fidelity DAC model introduced in this section is a physics-based model that has been previously reported and experimentally validated in Schellevis (2023). More precisely, Schellevis (2023) presents an extensive experimental investigation of this DAC system, which is based on a Lewatit® VP OC 1065 sorbent unit. The experimental campaign includes representative measurements of CO_2 concentration, temperature, and pressure throughout the different stages of the DAC cycle. Both the adsorption and desorption phases are experimentally validated, with particular emphasis on the reproducibility of the results across multiple operating cycles and across individual sorbent beds. The time-dependent profiles of CO_2 concentration, temperature, and pressure are carefully analyzed and shown to be consistent from cycle to cycle. Moreover, a series of experimental studies are conducted to assess the sensitivity of the DAC process to key operational parameters. These studies further examine the influence of parameter choices and the robustness of the model with respect to uncertainty in operating conditions. Consequently, the experimental results reported in Schellevis (2023) demonstrate that the high-fidelity model provides an accurate and reliable representation of the underlying DAC process dynamics, thereby providing a solid experimental foundation for the control design and performance evaluation presented in this work.

Furthermore, due to the data-driven and model-free nature of the proposed control approach, it is inherently well-suited for operation with real-world DAC experimental setups. The controller does not rely on an explicit process model and can therefore be directly deployed on experimental DAC platforms once such setups become available.

6. Numerical results

In this section, the performance of the proposed model-free and data-driven rule-based control approach is verified in simulation. We used the DAC process model described in Section 5 (Schellevis et al., 2021; Schellevis, 2023; Schellevis et al., 2020; Yu & Brilman, 2020) as our ground truth model for performing the experiments. The parameters of the model are described in Table A.1 in Appendix A. To solve PDEs (26)–(29), (31), the domain is discretized spatially for 15 points using the forward differencing method. Then, the resulting system of discretized differential equations is solved in MATLAB using the ode15s solver. The basis function for the control law is chosen as a third-order polynomial function of the ambient conditions, i.e., temperature and

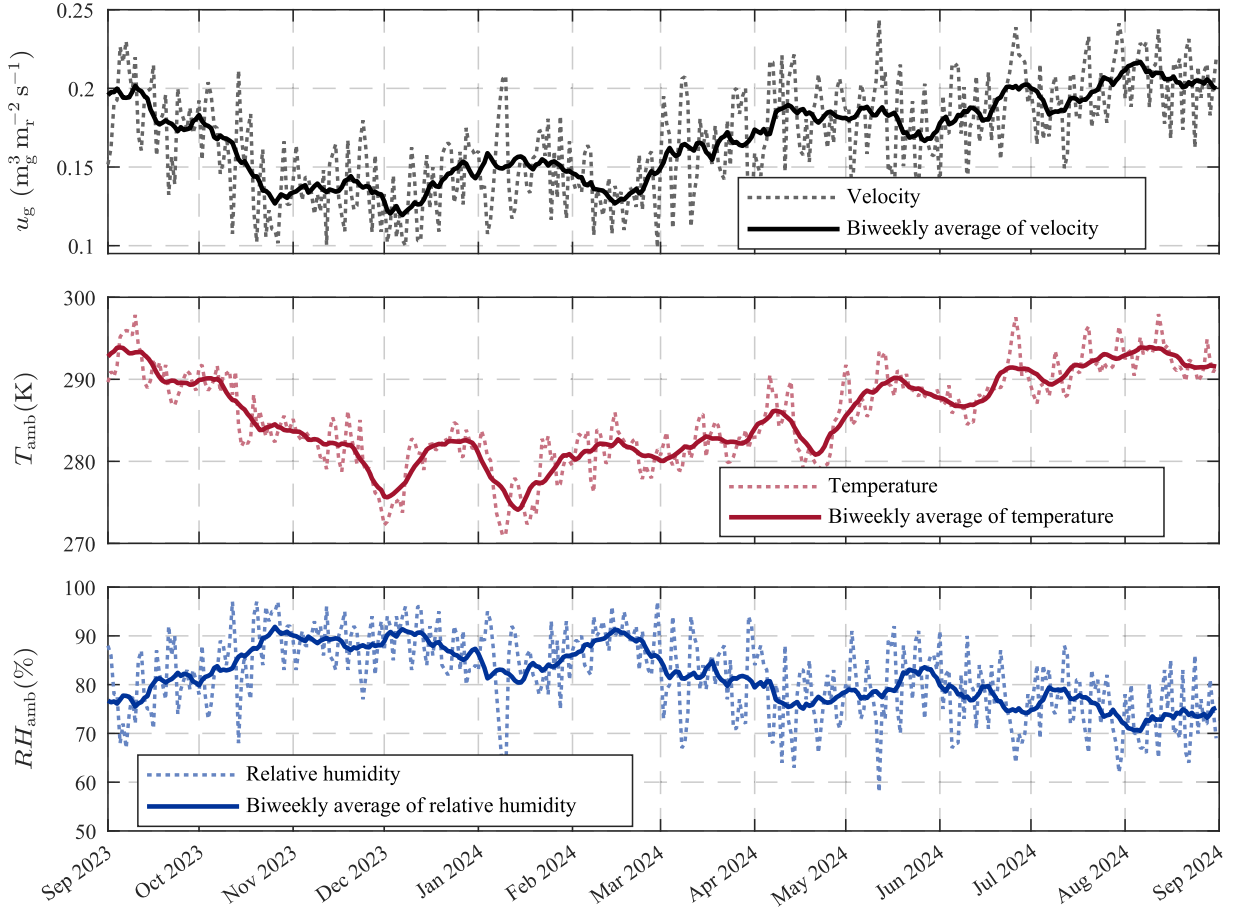


Fig. 7. The variation of superficial gas velocity, ambient temperature, and relative humidity in Amsterdam. The dotted lines show the values, while the solid lines indicate their biweekly averages. The three figures respectively illustrate (top) superficial gas velocity, (middle) ambient temperature, and (bottom) ambient relative humidity.

Table 1

The cost reduction and productivity improvement in different cities for one year: comparing the proposed and baseline method. The proposed method improves the annual DAC productivity by up to 16.69 % and reduces the annual capture costs by up to 10.34 %.

City	Cost reduction	Productivity improvement
Amsterdam	9.30 %	13.59 %
New York	8.27 %	14.88 %
Brasilia	9.86 %	15.78 %
Cape Town	10.05 %	16.69 %
Sydney	10.34 %	14.82 %
Beijing	7.80 %	13.52 %

Table 2

The cost reduction and productivity improvement for different basis functions for one year in Amsterdam: comparing the proposed and baseline method.

Basis function	Cost reduction	Productivity improvement
Linear polynomial basis	9.29 %	12.84 %
Second-order polynomial basis	9.33 %	12.93 %
Third-order polynomial basis	9.30 %	13.59 %
RBFs	9.20 %	9.98 %

relative humidity, resulting in a controller parameter vector $\zeta \in \mathbb{R}^{10}$. The weights W_1 and W_2 are selected as $1/(1.5 \times 10^8)$ and $1/0.3$, respectively, such that both the DAC capture cost and productivity are nor-

malized. This normalization ensures that the two objectives contribute comparably to the overall performance metric under typical operating conditions. The desired confidence level is set to $\beta_n = 2$. The initial dataset size is set to $n_{\text{init}} = 5$, and the initial samples are generated via random sampling by applying randomized perturbations, rather than using Latin hypercube sampling. For performing the BO tuning procedure, we employ the GPML package (Rasmussen & Nickisch, 2024) in MATLAB. We utilize the genetic algorithm method to optimize the acquisition function and the Matern kernel with automatic relevance determination. We use ambient temperature and relative humidity data from September 1, 2023, to September 1, 2024, from the weather stations in Amsterdam, Beijing, Brasilia, Cape Town, New York, and Sydney as the disturbance in the model (KNMI, 2024; Time and Date, 2024; Visual Crossing, 2024). Each BO iteration corresponds to a DAC operating cycle evaluated under different ambient conditions, ensuring that the reported results are not based on a single simulation run. Furthermore, the constraint $\Gamma_{\zeta}(\nu) \in [u_g, \bar{u}_g]$ in (8) is enforced in the optimization by approximating the disturbance set $\mathcal{V}$ with a sufficiently dense grid of temperature and humidity values. For each disturbance sample, upper and lower bounds on the controller output are imposed, resulting in a set of linear inequality constraints within the optimization step (21). This formulation ensures that the optimized controller parameters satisfy the physical limits. Moreover, we note that the results presented in this section are based on numerical simulations using the high-fidelity DAC model introduced in Section 5. However, this model has been experimentally validated in Schellevis (2023).

Using the BO approach introduced in Section 4, we solved the problem (8), which involves minimizing DAC costs and maximizing its

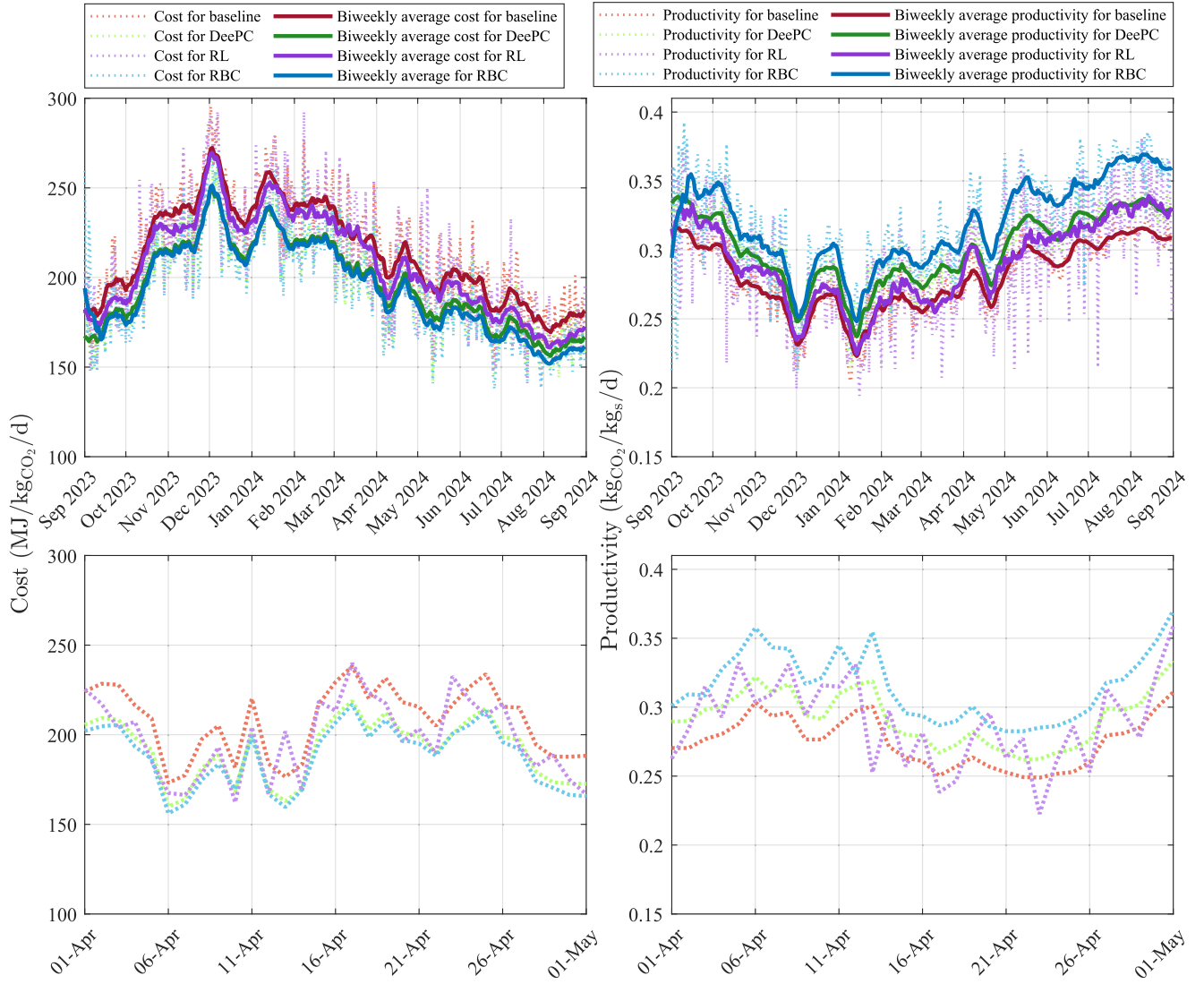


Fig. 8. Cost and productivity in Amsterdam: comparing the proposed Rule-Based Control (RBC), baseline method, data-enabled predictive control (DeePC), and Reinforcement Learning (RL)-based control. The top panel shows the DAC capture cost and productivity from September 1, 2023, to September 1, 2024, while the bottom panel depicts the results for the representative month of April.

productivity. The results are compared with the baseline method described in Schellevis (2023). Fig. 3 (a–e) demonstrates the capture cost and productivity in Amsterdam, New York, Brasilia, Cape Town, and Sydney, respectively. Similarly, Figs. 4 and 5 depict respectively the capture cost and productivity for Beijing. We can observe that during spring and summer months in the Northern Hemisphere, DAC productivity improves significantly, while in fall and winter, the improvement is mild. We can note that during the spring and summer months in the Southern Hemisphere, productivity improvement is slightly greater than during fall and winter. Low temperatures can lead to reduced productivity and increased heating costs. Therefore, in cities where the temperature difference between cold and warm seasons is high, the variation in both cost and productivity is also more pronounced. For instance, in Beijing, the minimum temperature is -13°C in winter with 57.1 % humidity, where the productivity is 0.162 and 0.151 $\text{kgCO}_2/\text{kg}_\text{s}/\text{d}$ and the cost is 316.21 and 322.47 $\text{MJ}/\text{kgCO}_2/\text{d}$ for the proposed and baseline methods, respectively. However, in Brasilia, the minimum temperature is 18.3°C in winter with 55.5 % humidity, where the productivity is 0.342 and 0.301 $\text{kgCO}_2/\text{kg}_\text{s}/\text{d}$ and the cost is 138.95 and 154.02 $\text{MJ}/\text{kgCO}_2/\text{d}$ for the proposed and baseline methods, respectively. In addition, at the beginning of the proposed data-driven method, DAC

productivity may drop slightly, and costs may increase marginally. However, after a few iterations, the parameters of the rule-based control scheme are updated, improving DAC productivity and reducing costs.

Table 1 illustrates the annual cost reduction and productivity improvement in different cities. We can note that in Amsterdam, the costs are reduced by 9.30 %, while DAC productivity increases by 13.59 %. For New York, the capture costs decline by 8.27 %, with a corresponding productivity improvement of 14.88 %. Moreover, DAC costs are lowered by 9.86 %, 10.05 %, 10.34 %, and 7.80 % while productivities are elevated by 15.78 %, 16.69 %, 14.82 %, and 13.52 % in Brasilia, Cape Town, Sydney, and Beijing, respectively.

Fig. 6 depicts the capture cost and productivity values in different cities over one year. The red circles indicate the baseline cost and productivity, while the blue squares represent the values obtained using the proposed method. We can observe that the proposed method enhances the DAC performance by shifting the red circle points to the blue square ones. The magnitude of this shift varies with climate, such that Cape Town, Brasilia, and Sydney demonstrate the largest improvements, while Beijing and Amsterdam exhibit smaller but still consistent gains.

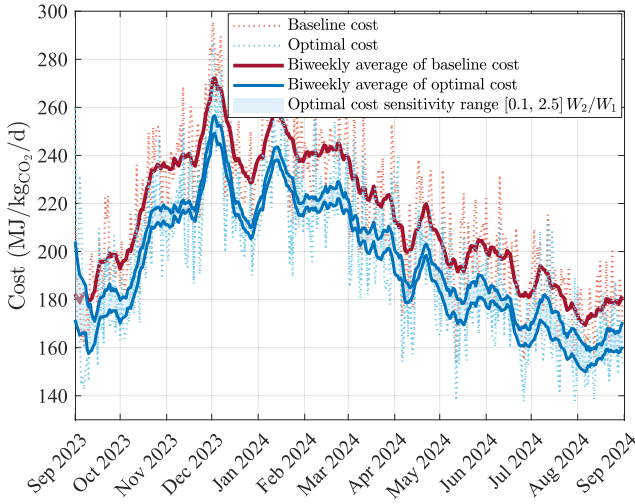


Fig. 9. Cost in Amsterdam: sensitivity analysis with the weights varied over a wide range, from $0.1 \times W_2/W_1$ to $2.5 \times W_2/W_1$. The baseline method is shown with dotted red lines, and its biweekly averages with solid red. The minimum and maximum values of the proposed method across all values within the sensitivity range are shown with dotted blue lines, and its biweekly averages with solid blue. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

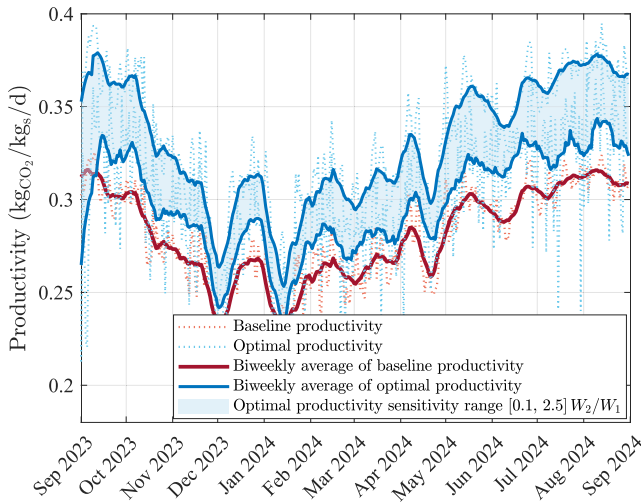


Fig. 10. Productivity in Amsterdam: sensitivity analysis with the weights varied over a wide range, from $0.1 \times W_2/W_1$ to $2.5 \times W_2/W_1$. The baseline method is shown with dotted red lines, and its biweekly averages with solid red. The minimum and maximum values of the proposed method across all values within the sensitivity range are shown with dotted blue lines, and its biweekly averages with solid blue. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 7 illustrates how variations in ambient temperature and relative humidity influence the selected superficial gas velocity in Amsterdam. We can observe that the periods of higher ambient temperature generally correspond to higher selected gas velocities. In contrast, we can note that both very low and very high humidity levels tend to reduce the selected velocity. The highest velocities are therefore observed when humidity remains within a moderate range while temperatures are high. Consequently, the trends in the figure highlight how the controller adjusts the superficial gas velocity in response to the effects of ambient temperature and relative humidity to improve DAC productivity and reduce its capture costs throughout the year.

To further assess the effectiveness of the proposed data-driven Rule-Based Control (RBC) method, we expanded the performance evaluation

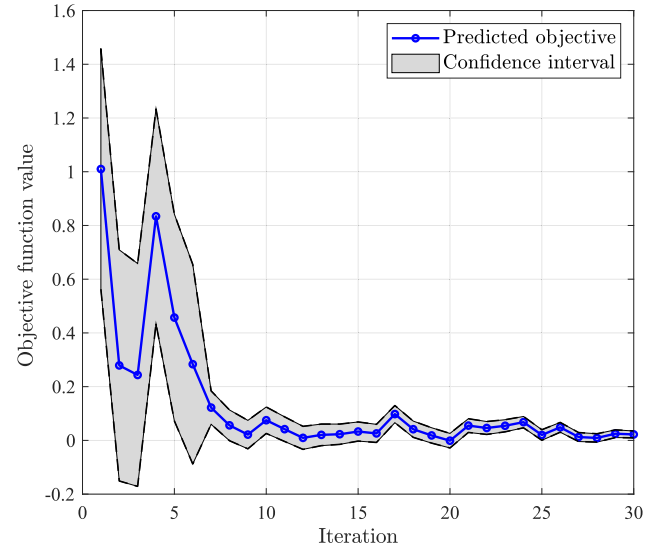


Fig. 11. Predicted objective function value and corresponding confidence interval over BO iterations. The predicted objective is shown in blue (solid line with circular markers), while the gray shaded region represents the confidence interval. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

by including two additional benchmark controllers: (i) a data-enabled predictive control (DeePC), and (ii) a Reinforcement Learning (RL)-based control scheme. More precisely, the DeePC benchmark follows a fully data-driven predictive control paradigm and does not rely on a system model (Coulson, Jeremy and Lygeros, John and Dörfler, Florian, 2019). Instead, it constructs Hankel matrices from persistently exciting input-output data collected from the high-fidelity DAC simulator. At each control step, a finite-horizon optimization problem is solved to compute the superficial gas velocity by minimizing both tracking error, defined as the deviation of the total CO_2 sorbent loading from its capture capacity, and control effort subject to input constraints and data consistency conditions. For the RL method, an actor-critic learning framework was employed to directly learn a control policy through interaction with the simulated environment (Han, Minghao and Zhang, Lixian and Wang, Jun and Pan, Wei, 2020).

Fig. 8 compares the performance of the baseline, DeePC, RL, and proposed RBC method in terms of DAC capture cost and productivity in Amsterdam. Specifically, the top panel of Fig. 8 represents the DAC capture cost and productivity over one year, while the bottom panel shows the corresponding results for the representative month of April. We can observe from Fig. 8 that the DeePC method yields a cost reduction of 8.25 % and a productivity improvement of 7.07 %, while the RL-based controller achieves a cost reduction of 3.12 % and a productivity improvement of 3.63 % compared to the baseline method. However, the proposed RBC method achieves a cost reduction of 9.30 % and a productivity improvement of 13.59 % relative to the baseline method, outperforming both DeePC and RL. Although DeePC provides a model-free predictive control framework, its performance in this application was limited by the high dimensionality and strong nonlinearities of the DAC process. Consequently, the resulting cost reduction and productivity improvement of the DeePC method are lower than those achieved by the proposed RBC method. Moreover, due to the high dimensionality, strong nonlinear dynamics, and stiffness of the DAC process dynamics, the RL method did not outperform the proposed RBC approach. Therefore, the proposed data-driven RBC method delivers superior performance compared to both DeePC and RL-based benchmark controllers for the considered DAC system.

To evaluate the robustness of the proposed method with respect to the weight selection, a sensitivity analysis was conducted by varying

the weights over a wide range, from $0.1 \times W_2/W_1$ to $2.5 \times W_2/W_1$, as shown in Figs. 9 and 10. The results demonstrate that the proposed method consistently outperforms the baseline method across the entire range of weight variations in terms of both DAC capture cost reduction and productivity improvement. More precisely, the minimum cost reduction achieved with respect to the baseline method is 6.73 %, while the maximum cost reduction is 10.29 % across all values within the sensitivity range. Similarly, the minimum productivity improvement relative to the baseline is 6.10 %, and the maximum productivity improvement is 17.72 % across all values within the sensitivity range. These results indicate that the performance gains of the proposed approach compared with the baseline method are not sensitive to the specific choice of weights, thereby confirming the robustness of the method under a wide range of weighting configurations.

Table 2 summarizes the impact of different basis function choices in the proposed method on DAC capture cost reduction and productivity improvement over one year of operation in Amsterdam, compared to the baseline method. For polynomial bases, the achieved cost reductions are similar across different polynomial degrees, whereas the third-order polynomial basis consistently delivers a higher productivity improvement compared to the linear and second-order cases. Moreover, Radial Basis Functions (RBFs) do not outperform the third-order polynomial basis. Consequently, the third-order polynomial basis achieved the best overall performance. More complex or higher-degree polynomial basis functions lead to convergence difficulties in the BO procedure and do not yield consistent performance improvements.

Figs. 11 illustrates the evolution of the predicted objective function value and its associated confidence interval over BO iterations. At the initial stages, the predicted objective exhibits relatively large variations accompanied by wide confidence bounds. As additional samples are acquired, the objective value decreases rapidly. Subsequently, we can note that the BO algorithm has effectively converged.

7. Conclusions

In this work, we proposed a model-free and data-driven rule-based control strategy for a solid-sorbent Direct Air Capture (DAC) system. The proposed rule-based control strategy operates as an online policy, continuously receiving feedback from ambient temperature and humidity to enable dynamic adjustments. We used the Bayesian Optimization (BO) method to optimally tune the parameters of the rule-based control strategy for the DAC system. We demonstrated that the proposed method shows promising performance improvement across different cities and various days of the year. It enhances the annual productivity of the DAC system by 13.52–16.69 % and reduces its annual energy costs by 7.80–10.34 % relative to the baseline across the different considered cities under varying weather conditions. It also achieves higher performance than other data-driven control approaches, achieving an annual productivity improvement of 13.59 % and an annual cost reduction of 9.30 % relative to the baseline in Amsterdam, while the data-enabled predictive control (DeePC) method reaches 7.07 % productivity improvement and 8.25 % cost reduction, and the Reinforcement Learning (RL)-based controller achieves 3.63 % productivity improvement and 3.12 % cost reduction. Additionally, the proposed method is model-free and data-driven, meaning that it does not rely on a detailed system model. Therefore, it addresses the challenges of model-based approaches, like the unavailability of a complex DAC model, the intractability of parameter estimation, and high computational costs.

CRedit authorship contribution statement

Amirreza Silani: Conceptualization, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing; **Tim M.J. Nijssen:** Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Supervision, Validation, Visualization, Writing – review & editing; **Mohammad Khos-**

ravi: Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing.

Declaration of competing interest

None.

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Appendix A. Description of high-fidelity model

The values of R_{CO_2} , k_T , q_{eq,CO_2} , b , t_h , q_s , η_{CO_2} , ϕ_{th} , R_{H_2O} , RH , $p_{H_2O}^{sat}$, q_{eq,H_2O} , C_{GAB} , k_{GAB} , k_{LDF} , and η_{H_2O} are given by

$$R_{CO_2} = k_T \left(RT c_{CO_2} \left(1 - \left(\frac{q_{CO_2}}{q_s} \right)^{t_h} \right)^{\frac{1}{t_h}} - \frac{q_{CO_2}}{b q_s} \right) \quad (A.1)$$

$$k_T = k_{0,CO_2} \exp \left(- \frac{E_{act,CO_2}}{RT} \right) \quad (A.2)$$

$$q_{eq,CO_2} = \frac{q_s b R T c_{CO_2}}{(1 + (b R T c_{CO_2})^{t_h})^{\frac{1}{t_h}}} \quad (A.3)$$

$$b = b_0 \exp \left(\frac{\Delta H_0}{R T_0} \left(\frac{T_0}{T} - 1 \right) \right) \quad (A.4)$$

$$t_h = t_{h,0} + \alpha \left(1 - \frac{T_0}{T} \right) \quad (A.5)$$

$$q_s = q_{s,0} \exp \left(\chi \left(1 - \frac{T}{T_0} \right) \right) \quad (A.6)$$

$$\eta_{CO_2} = \frac{\int \int \int R_{CO_2}(c_{CO_2}, q_{CO_2}) dV}{R_{CO_2}(c_{bulk}, q_{av})} \approx \frac{3}{\phi_{th}} \frac{\phi_{th} \coth(\phi_{th}) - 1}{\phi_{th}} \quad (A.7)$$

$$\phi_{th} = r_p \left(\frac{\rho_s k_T R T \left(1 - \left(\frac{q_{CO_2}}{q_s} \right)^{t_h} \right)^{\frac{1}{t_h}}}{D_p} \right)^{\frac{1}{2}} \quad (A.8)$$

$$R_{H_2O} = k_{LDF} R H (q_{eq,H_2O} - q_{H_2O}) \quad (A.9)$$

$$R H = \frac{R T c_{H_2O}}{p_{H_2O}^{sat}} \quad (A.10)$$

$$p_{H_2O}^{sat} = 611.21 \exp \left(\left(18.678 - \frac{T - 273.15}{234.5} \right) \left(\frac{T - 273.15}{257.14 + (T - 273.15)} \right) \right) \quad (A.11)$$

$$q_{eq,H_2O} = \left(C_m C_{GAB} k_{GAB} R H \right) / \left((1 - k_{GAB} R H) (1 + (C_{GAB} - 1) k_{GAB} R H) \right) \quad (A.12)$$

$$C_{GAB} = C_{0,GAB} \exp \left(\frac{\Delta H_C}{R T} \right) \quad (A.13)$$

$$k_{GAB} = k_{0,GAB} \exp \left(\frac{\Delta H_k}{R T} \right) \quad (A.14)$$

$$k_{LDF} = k_{0,H_2O} \exp \left(- \frac{E_{act,H_2O}}{R T} \right) \quad (A.15)$$

$$\eta_{H_2O} = 1. \quad (A.16)$$

Moreover, we have

$$D_{ax,m} = \frac{u_g d_p}{P e_{ax}} \quad (A.17)$$

Table A.1

Description of symbols and parameters (for parameters with two values or equations, the first is for carbon dioxide and the second for water).

ΔH_0	Tóth heat of adsorption (J mol ⁻¹)	95.3×10^3	k_{GAB}	multilayer correction factor in GAB isotherm (-)	Eq. (A.14)
$\Delta_c H$	difference in enthalpy between monolayer and multilayer (J mol ⁻¹)	-8.69×10^3	$k_{0,GAB}$	Arrhenius pre-exponential factor for K_{GAB} (-)	0.92
$\Delta_k H$	difference in enthalpy between bulk liquid and multilayer (J mol ⁻¹)	-8.2×10^3	k_0	Arrhenius pre-exponential factor for reaction rate (mol kg ⁻¹ bar ⁻¹ s ⁻¹ , s ⁻¹)	$3.5 \times 10^{-2}, 450$
$\Delta_r H_m$	reaction heat of component m (J mol ⁻¹)	$75 \times 10^3, 43 \times 10^3$	k_{LDF}	linear driving force reaction rate constant (s ⁻¹)	equation (A.15)
a_s	specific surface area (m ² m ⁻³)	150	k_T	Tóth reaction rate constant (mol kg ⁻¹ bar ⁻¹ s ⁻¹)	equation (A.2)
b	Tóth affinity constant (Pa ⁻¹)	Eq. (A.4)	p	pressure (Pa)	(-)
b_0	Tóth affinity constant at reference temperature (Pa ⁻¹)	93×10^{-5}	q	sorbent loading (mol kg ⁻¹)	equation (27)
C_0	Arrhenius pre-exponential factor for C_{GAB} (-)	100	q_s	Tóth maximum capacity (mol kg ⁻¹)	equation (A.6)
C_{GAB}	Guggenheim constant in GAB isotherm (-)	equation (A.13)	$q_{s,0}$	Tóth maximum capacity at reference temperature (mol kg ⁻¹)	3.40
C_m	monolayer capacity in GAB isotherm (mol kg ⁻¹)	5.55	R	ideal gas constant (J mol ⁻¹ K ⁻¹)	8.31446
$C_{p,g}$	gas specific heat capacity (J kg ⁻¹ K ⁻¹)	1005	R_m	reaction rate of component m (mol kg ⁻¹ s ⁻¹)	equations (A.1), (A.9)
$C_{p,s}$	solid specific heat capacity (J kg ⁻¹ K ⁻¹)	1580	T	temperature (K)	Eq. (28)
c	concentration (mol m ⁻³)	Eq. (26)	T_0	Tóth reference temperature (K)	353.15
D_{ax}	axial dispersion coefficient (m ² s ⁻¹)	Eq. (A.17)	t_h	Tóth heterogeneity constant (-)	Eq. (A.5)
E_{act}	activation energy (J mol ⁻¹)	15.2×10^3	$t_{h,0}$	Tóth heterogeneity constant at T_0 (-)	0.37
h_i	heat transfer coefficient (W m ⁻² K ⁻¹)	150	z	axial direction (m)	(-)
u	superficial gas velocity (m ³ m ⁻² s ⁻¹)	[0.05, 0.3]	δ	identification of energy point source in 1D model (-)	binary
α	temperature dependency of t_h (-)	0.33	η	effectiveness factor (-)	[0,1]
ϵ_p	void fraction of sorbent (m ³ m ⁻³)	0.23	ρ	density (kg m ⁻³)	1.204
ϵ_r	void fraction of reactor (m ³ m ⁻³)	0.4	Φ	gas flow rate (g _{purge} min ⁻¹)	1.04
$\lambda_{s,eff}$	axial thermal conductivity (W m ⁻¹ K ⁻¹)	0.121	g	gas phase	
χ	temperature dependency of q_s	0	m	components (CO ₂ and H ₂ O)	
			r	reactor	
			s	solid phase	

$$\frac{1}{P_{\text{ex}}} = \frac{0.72}{Re Sc} + \frac{0.52}{1 + \left(\frac{9.0}{Re Sc}\right)} \quad (\text{A.18})$$

$$Re = \frac{\rho_g u_g d_p}{\mu_g} \quad (\text{A.19})$$

$$Sc = \frac{\mu_g}{\rho_g D_{M,m}} \quad (\text{A.20})$$

$$\frac{1}{d_{\text{pore}}} = \frac{\tau}{\epsilon_p} = \frac{1}{D_{M,m}} + \frac{1}{D_{Kn}} \quad (\text{A.21})$$

$$D_{Kn} = \frac{d_{\text{pore}}}{3} \left(\frac{8RT}{\pi M_{W,m}} \right)^{\frac{1}{2}}. \quad (\text{A.22})$$

The Danckwerts boundary conditions and initial conditions are given by

$$u_{g,\text{in}} c_{\text{CO}_2,\text{in}} = -D_{\text{ax},\text{CO}_2} \partial_z c_{\text{CO}_2,g} \Big|_{z=0} + u_{g,\text{in}} c_{\text{CO}_2,g} \Big|_{z=0} \quad (\text{A.23})$$

$$u_{g,\text{in}} c_{\text{H}_2\text{O},\text{in}} = -D_{\text{ax},\text{H}_2\text{O}} \partial_z c_{\text{H}_2\text{O},g} \Big|_{z=0} + u_{g,\text{in}} c_{\text{H}_2\text{O},g} \Big|_{z=0} \quad (\text{A.24})$$

$$u_{g,\text{in}} \rho_g C_{p,g} T_{\text{in}} = -\lambda_{s,\text{eff}} \partial_z T \Big|_{z=0} + u_{g,\text{in}} \rho_g C_{p,g} T \Big|_{z=0} \quad (\text{A.25})$$

$$\partial_z c_{\text{CO}_2,g} \Big|_{z=L} = 0 \quad (\text{A.26})$$

$$\partial_z c_{\text{H}_2\text{O},g} \Big|_{z=L} = 0 \quad (\text{A.27})$$

$$\partial_z T \Big|_{z=L} = 0 \quad (\text{A.28})$$

$$c_{\text{CO}_2,g} \Big|_{t=0} = c_{0,\text{CO}_2}(z) \quad (\text{A.29})$$

$$c_{\text{H}_2\text{O},g} \Big|_{t=0} = c_{0,\text{H}_2\text{O}}(z) \quad (\text{A.30})$$

$$T \Big|_{t=0} = T_0(z) \quad (\text{A.31})$$

$$q_{\text{CO}_2} \Big|_{t=0} = q_{0,\text{CO}_2}(z) \quad (\text{A.32})$$

$$q_{\text{H}_2\text{O}} \Big|_{t=0} = q_{0,\text{H}_2\text{O}}(z). \quad (\text{A.33})$$

The value of Q_{source} , boundary and initial conditions for the heat transfer model (29) are expressed as

$$Q_{\text{source}} = \delta(z) h_i a_s (T_{\text{ht}} - T) \quad (\text{A.34})$$

$$\partial_z T \Big|_{z=0} = \partial_z T \Big|_{z=L} = 0 \quad (\text{A.35})$$

$$T \Big|_{t=0} = T_0(z), \quad (\text{A.36})$$

where T_{ht} is the temperature of heat or cool sources, and the function $\delta(z)$ is characterized as a Dirac-delta function, which takes a value of one precisely at the positions of heat sources and zero elsewhere.

B. Detailed description of energy costs and productivity

The energy required for the CO₂ endothermic desorption measured in J/kg_{CO₂}/d is expressed as

$$E_{r,\text{CO}_2} = \frac{\Delta_r H_{\text{CO}_2}}{M_{W,\text{CO}_2} t_{\text{cycle}}}, \quad (\text{B.1})$$

where $\Delta_r H_{\text{CO}_2}$ is the CO₂ reaction enthalpy, M_{W,CO_2} is the CO₂ molar mass, $t_{\text{cycle}} = (t_{\text{ads}} + t_{\text{vac}} + t_{\text{heat}} + t_{\text{des}} + t_{\text{cool}} + t_{\text{repr}})/(24 \times 60)$, and t_{ads} , t_{vac} , t_{heat} , t_{des} , t_{cool} , and t_{repr} are the adsorption, vacuum, heating, desorption, cooling, and repressurization times expressed in min, respectively. The energy required for the desorption of H₂O measured in J/kg_{CO₂}/d is given by

$$E_{r,\text{H}_2\text{O}} = \frac{\Delta_r H_{\text{H}_2\text{O}}}{S_{\text{CO}_2} M_{W,\text{CO}_2} t_{\text{cycle}}}, \quad (\text{B.2})$$

where $\Delta_r H_{\text{H}_2\text{O}}$ represents the H₂O reaction enthalpy, $S_{\text{CO}_2} = \Delta q_{\text{CO}_2}/\Delta q_{\text{H}_2\text{O}}$ is the selectivity of CO₂ towards H₂O, Δq_{CO_2} is the CO₂ working capacity, and $\Delta q_{\text{H}_2\text{O}}$ is the H₂O working capacity. The sorbent sensible heat measured in J/kg_{CO₂}/d is expressed as

$$E_{\text{sens},s} = \frac{C_{p,s} \Delta T}{\Delta q_{\text{CO}_2} M_{W,\text{CO}_2} t_{\text{cycle}}}, \quad (\text{B.3})$$

where $C_{p,s}$ denotes the sorbent specific heat capacity and ΔT is the temperature difference between adsorption and desorption. The reactor sensible heat measured in J/kg_{CO₂}/d is given by

$$E_{\text{sens},r} = \frac{R_{\text{sens},r} E_{\text{sens},s}}{t_{\text{cycle}}}, \quad (\text{B.4})$$

where $R_{\text{sens},r} = (m_r C_{p,r} + m_{ht} C_{p,ht}) / (m_s C_{p,s})$, m is the mass, and C_p is the specific heat capacity of the reactor segment, denoted by r , heat transfer medium, denoted by ht , and sorbent bed, denoted by s . The total energy required for the purge gas measured in $\text{J/kg}_{\text{CO}_2}/d$ includes both sensible and latent heat is expressed as

$$E_{\text{purge}} = \frac{R_{\text{purge}}}{M_{\text{W,CO}_2} t_{\text{cycle}}} (M_{\text{W,p}} C_{p,L} \Delta T_L + M_{\text{W,p}} C_{p,g} \Delta T_g + \Delta_{\text{vap}} H_{\text{purge}}), \quad (\text{B.5})$$

where $C_{p,L}$ and $C_{p,g}$ are the specific heat capacities of the liquid and gas phases, ΔT_L and ΔT_g are the temperature differences of these phases, respectively. Moreover, $\Delta_{\text{vap}} H_{\text{purge}}$ is the evaporation enthalpy and $R_{\text{purge}} = n_{\text{purge}} / (m_s \Delta q_{\text{CO}_2})$ is the purge gas ratio, where $n_{\text{purge}} = (\phi_{V,g,\text{des}} t_{\text{des,av}}) / (RT_{\text{des,av}})$ is the total amount of purge gas introduced into the reactor during the desorption, $\phi_{V,g,\text{des}}$ is the air volumetric flow rate during the desorption, $t_{\text{des,av}}$ is the average desorption pressure, R is the gas constant, and $T_{\text{des,av}}$ is the average desorption temperature. The feed compression energy measured in $\text{J/kg}_{\text{CO}_2}/d$ is calculated by

$$E_{\text{feed}} = \frac{\Delta p}{\eta_f c_{\text{CO}_2} \eta_{\text{gas}} M_{\text{W,CO}_2} t_{\text{cycle}}}, \quad (\text{B.6})$$

where $\Delta p = (150 \mu_g L (1 - \epsilon_r)^2 u_g) / (d_p^2 \epsilon_r^3) + (1.75 L \rho_g (1 - \epsilon_r) u_g^2) / (d_p \epsilon_r^3)$ is the pressure drop based on the Ergun equation, μ_g is the dynamic viscosity of air, L is the bed thickness, d_p is average particle diameter, η_f is the efficiency of the air blower, $\eta_{\text{gas}} = (m_s \Delta q_{\text{CO}_2}) / (\phi_{V,g,\text{ads}} t_{\text{ads}} c_{\text{CO}_2})$, $\phi_{V,g,\text{ads}}$ is the air volumetric flow rate during the adsorption, and c_{CO_2} is the CO_2 concentration of the air. The vacuum compression energy measured in $\text{J/kg}_{\text{CO}_2}/d$ is expressed as

$$E_{\text{vac}} = \frac{RT_{\text{des,av}}}{M_{\text{W,CO}_2} \eta_{\text{vac}} t_{\text{cycle}}} \left(\frac{k}{k-1} \right) \left(\frac{1}{x_{\text{CO}_2}^{\text{prod}}} + \frac{1}{S_{\text{CO}_2}} + R_{\text{purge}} \right) \left(\left(\frac{p}{p_{\text{des,av}}} \right)^{\frac{k-1}{k}} - 1 \right), \quad (\text{B.7})$$

where $x_{\text{CO}_2}^{\text{prod}} = \Delta q_{\text{CO}_2} / (\Delta q_{\text{CO}_2} + \Delta q_{\text{H}_2\text{O}})$ is the CO_2 fraction in the product, R_{purge} is the purge gas ratio, η_{vac} is the vacuum pump efficiency, k is the C_p/C_v ratio, and p is the product gas pressure. The productivity for the steam-assisted temperature vacuum swing adsorption process expressed in $\text{kg}_{\text{CO}_2}/\text{kg}_s/d$ is given by

$$P_{\text{DAC}} = \frac{\Delta q_{\text{CO}_2}}{t_{\text{cycle}}}. \quad (\text{B.8})$$

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