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Environmental benefit-detriment thresholds for flow battery energy storage systems: A case study in California

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HIGHLIGHTS

- Combines life cycle analysis and electric grid dispatch modeling methods.
- Compares emissions reduced from battery use with emissions from battery production.
- Calculates net emissions reductions of flow batteries at increasing grid capacities.
- Capacity thresholds exist where emissions reduction benefits are maximized.
- Deploying too much flow battery capacity can reduce or negate emissions reductions.

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ABSTRACT

Energy storage systems are critical for enabling the environmental benefits associated with capturing renewable energy to displace fossil fuel-based generation, yet producing these systems also contributes to environmental impacts through their materials use and manufacturing. As energy storage capacity is scaled up to support increasingly renewable grids, the environmental benefits from their use may scale at different rates than the environmental impacts from their production. This implies the existence of capacity thresholds beyond which installing additional storage capacity may be environmentally detrimental. Identifying such thresholds are important for ensuring that energy storage capacity selection in future grids are consistent with net emissions reduction goals, but such thresholds have not been studied in the present literature. To identify such thresholds, here we combine electric grid dispatch modeling with life cycle analysis to compare how the emissions reductions from deploying three different flow battery energy storage types on a future California grid (>80% wind and solar) compare with emissions contributions from producing such batteries as total battery capacity installed on the grid increases. Depending on the type of battery and environmental impact indicator (greenhouse gas or particulate matter emissions), we find that the marginal environmental benefits of storage begin to diminish at deployed capacities of 38–76% of the mean daily renewable generation (256–512 GWh in our California scenarios) and reach zero at 105–284% of mean daily renewable generation (700–1810 GWh). Such storage capacities are conceivable, but upstream impacts of storage must be assessed in evaluating the environmental benefits of large-scale storage deployment, or they could negate the environmental benefits of regional electricity system decarbonization.

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1. Introduction and background

1.1. Context and literature review

Nomenclature

E3	Energy & Environmental Economics
GHG	Greenhouse Gas
GW	Gigawatt
GWh	Gigawatt-hour
HiGRID	Holistic Grid Resource Integration and Deployment model
IFB	Iron Flow Battery
kWh	Kilowatt-hour
LCA	Life Cycle Analysis
LCI	Life Cycle Inventory
MWh	Megawatt-hour
NREL	National Renewable Energy Laboratory
PM	Particulate Matter
SI	Supplemental Information
U.K.	United Kingdom
U.S.	United States
V2O5	Vanadium Pentoxide
VRFB	Vanadium Redox Flow Battery
ZBFB	Zinc Bromide Flow Battery

Energy storage systems have been identified as a key resource in enabling the increased use of variable renewable energy resources such as wind and solar power, which are cornerstones of many strategies for developing future energy infrastructure to reduce greenhouse gas (GHG) emissions, fossil fuel dependency, and environmental impact related to fossil fuel usage. Energy storage technologies provide an important degree of controllability for balancing the temporal profiles of electricity supply and demand in energy systems incorporating large capacities of wind and solar power. This role has been recognized by formal policies such as energy storage mandates in different states of the U.S [1]. While energy storage functions to further enable realizing the environmental benefits of wind and solar energy, these systems also contribute towards environmental impacts from their materials use, manufacturing, and end-of-life stages as well as their use-phase if interacting with fossil fuel-based electricity resources. Here, we refer to the "use-phase" as the part of the energy storage system's life cycle encompassing its operation on the electric grid to provide electricity system services, whereas "non-use phase" will refer to all other stages of the system's life cycle (materials extraction, manufacturing, and end-of-life).

1.1.1. Previous literature on energy storage operation in electric grids

Previous studies of the environmental effects of energy storage technologies may be grouped into two categories. The first are those that characterize how energy storage affects grid reliability and emissions under various technology pathways, but typically without accounting for emissions contributions from their production and supply chains. These evaluate how much energy storage is required to reach environmental goals given the reliability and operational needs of balancing electricity supply and demand. For example, Mahone et al. [2] investigated different energy technology portfolios for meeting California's goal of reducing emissions of GHGs across all economic sectors to 80% below year 1990 levels by 2050. Their study included pathways encompassing different energy carriers and primary energy sources. Across the wide array of pathways that comply with the goal, energy storage systems consisting of 17 to 32 GW of a mixture of 2-hour, 5-hour, and 8-hour batteries were required. Tarroja et al. [3] investigated the energy storage capacity needed to reach a 100% renewable energy penetration in California, finding that even with other complementary technologies such as dispatchable renewables and dispatchable loads, aggregated energy storage capacity of up to 0.6% of annual renewable energy production (2736 GWh) was required. Mileva et al. [4] investigated energy technology portfolios needed to reach an 80% GHG emissions reduction from the electricity sector across the entire Western U.S., which required energy storage capacities between 40 and 260 GW of 6-hr energy storage systems (240–1608 GWh). The National Renewable Energy Laboratory (NREL) [5] determined that between 100 and 152 GW of power capacity in energy storage systems consisting of a variety of storage types were required for the entire U.S. to reach 80% renewable energy penetration in the electricity sector. Similar studies highlighting energy storage have been conducted for other regions of the world such as Finland [6], Australia [7], and France [8]. Previous studies by Hittinger and Azevedo [9,10] characterized the emissions impacts of energy storage operation on near-term electric grids with energy storage operating to provide arbitrage.

These studies, however, focus only on environmental benefits or impacts provided during the operation of energy storage systems. Many of these studies do not account for the potential contribution of energy storage systems towards environmental impact or energy use from stages outside of energy storage operation, such as manufacturing and materials extraction to create the products for energy storage systems.

1.1.2. Previous literature on energy storage life cycle environmental impacts

The second are those that evaluate the life cycle environmental impact of storage, but typically without capturing the dynamics of their use on the electric grid. These use life cycle analyses (LCAs) to evaluate environmental impact related to materials use, manufacturing/production, use, and end-of-life stages of energy storage technologies. Weber et al. [11] performed an LCA of the vanadium redox flow battery, highlighting the contribution of the technology's electrolyte to environmental impact and the recyclability of this technology compared to lithium-ion batteries. Longo et al. [12] performed an LCA of the sodium/nickel chloride battery for stationary energy storage applications, finding that the use phase contributed the largest energy impact, but the manufacturing phase contributed to the largest environmental impact. Oliveira et al. [13] performed comparative LCAs of a suite of different energy storage technologies such as compressed air, lead-acid, lithium-ion, sodium-sulfur, hydrogen, pumped hydropower, and sodium-nickel-chloride energy storage systems for use in electric grid applications. This study compared these technologies under different electricity mixes and on different environmental impact indicators, highlighting how environmental performance depends strongly on application. Many studies have also focused on lithium-ion batteries due to interest in electric vehicle deployment, the most used LCI of lithium-ion batteries are sourced from Notter [14], Egede [15], and Majeau-Bettez [16] and are focused on batteries for electric vehicles. Additionally, studies by Dunn et al. [17,18] focus on recycling, and studies by the U.S. Environmental Protection Agency focusing on nanoscale technology application on lithium-ion batteries [19].

1.1.3. Previous literature considering both use-phase and non-use-phase aspects

The studies in the previous subsections, however, lack two characteristics. First, they do not account for how the environmental benefits from the use of energy storage systems are affected by the temporal dynamics of electric grid operation and renewable electricity generation. Many of the studies parameterize the assumed electricity mix that interacts with the energy storage system, however, these do not capture how the deployment of energy storage changes the delivered electricity mix based on how it interacts with the electric grid. Second, LCA methods frequently assess technologies on a normalized basis (e.g., impact per one kWh of capacity or impact per 1 MWh electricity output from the battery) and do not provide a sense of the appropriate amount of energy storage capacity to realize potential environmental benefits.

More recent literature has started to focus on simultaneous accounting of both use-phase and non-use-phase environmental impacts. Schmidt et al. [20] assessed both use-phase and non-use-phase contributions towards emissions and costs associated with energy storage use in different regions. However, this study did not explicitly resolve the interaction between energy storage systems and the electric grid from a dynamic load balancing perspective. This aspect is critical to consider for capturing how the emissions reductions of deploying energy storage systems scale as more capacity is installed, as we show in the present study. To this end, Elzein et al. [21] proposed an optimized LCA method for more accurately capturing how the use phase affects energy storage life cycle footprints, and Ryan et al. [22] assessed how the use phase of lithium-ion batteries drives life cycle environmental impacts in frequency regulation applications. Chowdhury et al. [23] combined both grid operation and a simplified LCA to assess the environmental benefit of using lithium-ion batteries to displace combined-cycle natural gas turbines in the U.K. electric grid up to 2035. The present study further expands upon these concepts to assess how comparison of use-phase and non-use-phase contributions towards increasing or decreasing system-wide environmental impacts give rise to capacity thresholds for deploying energy storage 1) where the maximum net benefits occur as a policy target to aim towards and 2) where the net benefits reach zero as a capacity threshold to avoid in determining how much energy storage capacity to deploy.

1.2. Research gap and contribution of the current study

Thus, the existing literature reveals the following:

- Many studies characterize the environmental benefits of using energy storage at scale on the grid but do not simultaneously account for the non-use-phase environmental impacts of deploying that scale of energy storage.
- Conversely, many studies characterize the non-use-phase environmental impacts of producing energy storage systems but do not simultaneously account for the environmental benefits associated with their interaction with the electric grid at scale.
- A limited number of studies have focused on better-representing energy storage use in life cycle frameworks, but these have not been leveraged to gain a broader understanding of how energy storage deployment should be structured to maximize net environmental benefits.

Therefore, a study that accurately accounts for both the environmental benefits of energy storage provided during their use and the environmental impact of the life cycle of these systems when deployed to scale is currently lacking in the literature. This gap prevents the understanding of how the benefits and impacts scale relative to each other as energy storage deployment is increased and whether there exists a capacity threshold at which environmental impacts may outweigh benefits. We have filled this gap with the research described here by presenting a study that investigates the relative scaling of environmental benefits from energy storage operation versus impacts from energy storage production, using a case study of flow batteries deployed in a decarbonizing California grid as a representative example.

To effectively support decarbonization goals, it is important that substantial environmental benefits coincide with large energy storage capacities to ensure that 1) energy storage can be installed to the scale needed to meet decarbonization targets and 2) that regional decarbonization efforts do not inadvertently cause increases in greenhouse gas (GHG) emissions elsewhere in the world due to the production of energy storage devices. Differences in the rate at which the grid emissions reductions and life cycle pollution impacts scale as more energy storage is deployed imply that the environmental impact of additional storage production may outweigh the benefits during use when implemented beyond some level of installed capacity.

This study investigates how the emissions reduction benefits from the expanded use of energy storage in regional electric grids compare with the emissions contributions of producing these energy storage systems to scale, focusing on flow batteries. We develop a method for evaluating the marginal benefits of energy storage, identify thresholds of diminishing and negative environmental benefits for three different types of flow batteries: Vanadium-Redox (VRFB), Zinc-Bromide (ZBFB), and Iron (IFB), using the example of California's electric grid in 2045 and highlight drivers that apply to other storage technologies, grids, and locations. Details of our data sources and approach are given in *Materials and Methods*. We bring together two strands of analysis, combining dynamic electric grid modeling with LCA to examine the environmental trade-offs of deploying energy storage in future renewables-based electricity systems and the sensitivity of these tradeoffs to materials selection and supply chain choices.

2. Materials and methods

The schematic diagram of methods used in this study is presented in Fig. 1. Note that a summary of the approach is provided in this section. A detailed description of each step in the overall approach including assumptions, modeling choices, and parameter values are provided in the Supplemental Information (SI) that accompanies this manuscript.

The first step is to select a suitable scenario for representing a future electric grid configuration and a representative energy storage technology. We simulate a highly renewable electric grid in California in the year 2045 according to the Energy & Environmental Economics (E3) PATHWAYS study [2] using the "High Electrification" scenario. The High Electrification scenario represents a likely trajectory for the evolution of both electricity and non-electricity sector emissions. The E3 PATHWAYS study determined the evolution of energy system resources and demands each year from 2020 until reaching an 80% reduction in economy-wide (including both electricity non-electricity sectors) greenhouse gas reductions from year 1990 levels in California by the year 2050. Here, we select parameters for the year 2045 from this study since it coincides with newer California's electricity decarbonization goal codified by Senate Bill 100. The E3 PATHWAYS study accounted for changes in electric loads based on population growth, technology improvements, replacement rates of old technologies with new technologies, and the deployment of electric and hydrogen fuel cell vehicles. Additionally, changes in the energy resource mix for meeting these loads every year were determined in that study based on resource availability and economic cost. Parameters used from that study include the installed capacities of electricity generation technologies, the penetration level of complementary technologies such as electric vehicles and demand response, and the profiles of electric loads from industrial, commercial, residential, and transportation sectors and are detailed in the Supplemental Information: Tables S1 and S2. Note that the energy storage capacities from the E3 PATHWAYS scenario were not used, as this study varies that parameter. Visually, a representation of the major components of the E3 PATHWAYS scenario with respect to the electricity sector is presented in Fig. 2:

For the representative energy storage technologies, we model the deployment of three different flow battery types: vanadium redox flow batteries (VRFB), zinc bromide flow batteries (ZBFB), and iron flow batteries (IFB), based on the performance characteristics and LCA from He et al. [24]. Among flow batteries, the VRFB is currently the most mature [25] but both the ZBFB and IFB are commercially-available products. All three flow battery types, however, are still in their relatively early stage of development compared to conventional batteries and are still evolving in technical design, materials selection, and manufacturing strategies. While lithium-ion batteries are the leading candidates for grid-scale energy storage, we selected flow batteries due to the following reasons. First, from previous work by He et al. [24], we have access to consistent and harmonized life cycle inventory data for

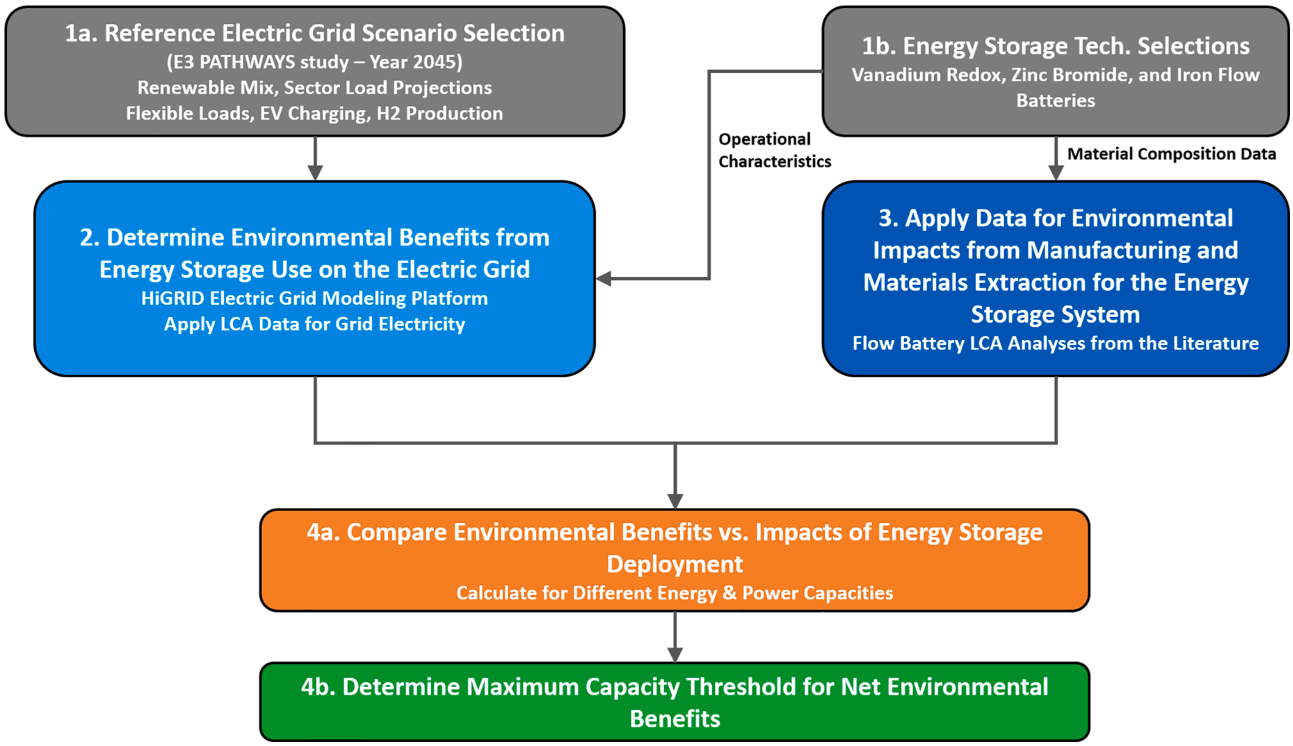

Fig. 1. Schematic overview of the research approach. Details on each step are provided in the Supplemental Information (SI).

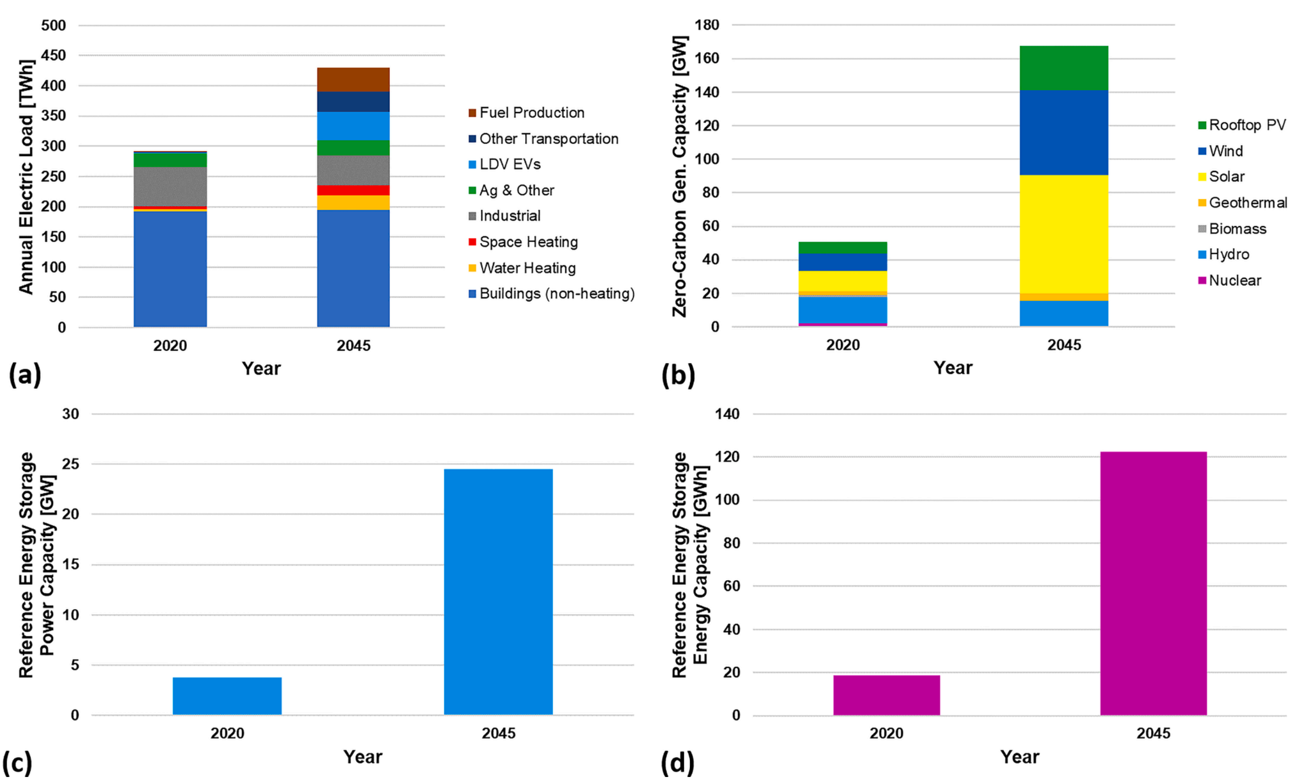

Fig. 2. Summary of the E3 PATHWAYS "High Electrification" scenario (19) contrasting differences in (a) annual electric load, (b) zero-carbon generation capacity, (c) energy storage capacity (power capacity basis), and (d) energy storage capacity (energy capacity basis) between the year 2020 (present-day) and the year 2045. Note that for the energy storage energy capacity, the PATHWAYS scenario uses a mixture of 2-hour, 5-hour, and 8-hour batteries, but does not explicitly specify the mix of these battery types, therefore the numbers in (d) assume a 5-hour average duration. The year 2045 parameters are used as inputs to the electric grid modeling conducted in this analysis except for the energy storage capacities, which are varied in this study. More detail is presented in the Supplemental Information.

different flow battery types that enables comparisons across different battery chemistries. This avoids the uncertainty associated with needing to harmonize different data from the literature on different energy storage types, which typically comprise different vintages and system boundaries between studies. Second, flow batteries can physically separate their power and energy capacity subsystems, enabling us to track how the net benefit of energy storage scales with increasing power capacity, energy capacity, or both. The main insights of this study, however, are not only specific to flow batteries and are conceptually generalizable to other energy storage types, with the main difference being the numerical values for the total capacities where net benefit thresholds occur. Parameters for the efficiencies and other characteristics of the three different flow battery types are provided in the Supplemental Information.

The second step is to determine the environmental benefits provided by the deployment and operation of energy storage at different power and energy capacity combinations through the changes that these systems induce on the mix of utilized electricity. Energy storage provides environmental benefits by enabling the electric grid to uptake additional renewable energy that would otherwise have been curtailed which displaces fossil fuel-based electricity generation. The extent of these benefits depends on the total power and energy capacity of the aggregated energy storage fleet installed on the grid. The inputs from the E3 PATHWAYS model are used in an electric grid dispatch model called the Holistic Grid Resource Integration and Deployment (HiGRID) model [26]. The HiGRID model determines the hourly dispatch of electricity generation and complementary technologies on the electric grid subject to the constraints of balancing supply with demand, providing sufficient reliability services, and transmission and distribution losses. Hourly electric load that is unable to be met with specified resources (renewables, hydro, nuclear, energy storage discharge, demand response) is met with natural gas combined-cycle generation to ensure that the load is balanced. Descriptive detail on HiGRID as it is implemented in this study is provided in the Supplemental Information: Figures S2 and S3, Table S4, and their accompanying text. As outputs from these processes, HiGRID produces metrics for environmental impacts such as fuel usage and the annual delivered energy by resource type. These are then translated into annual GHG emissions and criteria pollutant emissions, accounting for both emissions from their use in power plants as relevant and cradle-to-gate emissions associated with the relevant electricity generation technologies obtained from the EcoInvent database. For this study, the reference electric grid configuration is simulated with energy storage systems ranging in energy capacity from 0 to 2880 GWh and power capacity from 0 to 360 GW. Different energy storage sizes interact differently with the electric grid and impart different changes in the delivered electricity mix.

The third step is to obtain and apply data for the environmental impact associated with the materials extraction and manufacturing of electric grid resources and the three different flow batteries. For the VRFB, ZBFB, and IFB, we draw upon the inventory and environmental impact analysis conducted by He et al. [24] that performed an LCA to characterize the environmental impact associated with the materials extraction and manufacturing processes of three flow battery chemistries to provide an up-to-date understanding of the life cycle impacts of these emerging technologies. He et al. [24] utilized and harmonized material data inputs from actual flow battery manufacturers for the three different chemistries and drew on studies in the literature for cell stack, electrolyte storage, and balance-of-plant components. They coupled these data with reference life cycle inventory datasets from EcoInvent [27], to assess eight different environmental impact categories using the ReCiPe 2016 impact assessment method. Detail on the EcoInvent database for the resources considered here as well as the ReCiPe 2016 application are provided in the Supplemental Information.

The environmental impacts from the materials extraction and manufacturing of flow batteries depend on the configuration of their supply chains and production methods. To demonstrate how such choices affect the primary results, we apply the emissions factors for alternative production pathways of the VRFB electrolyte from He et al. [24]. However, we do not attempt to project changes in other aspects of the flow battery supply chains or production processes, and the inventories used here represent the current state of each technology. Moving into the future, these aspects may change for reduced environmental footprint. However, these changes are difficult to predict consistently over a 25-year timeframe since these will depend on market conditions and manufacturer choices.

For the present study, we focus on a subset of environmental impact indicators from the ReCiPe 2016 impact assessment method [28] as implemented in SimaPro, consistent with the data from He et al. [24]: 1) greenhouse gas emissions (GHG) and 2) particulate matter formation (PM). These indicators are reflective of metrics used to assess energy system environmental performance in electric grid planning studies in the context of GHG emission reduction goals and criteria pollutant emissions (PM). Additionally, while there is a wide range of available environmental impact indicators from ReCiPe 2016, GHG and PM were selected since the mechanisms for how energy storage reduces these emissions in their use but also contributes towards these emissions in their production are well understood. While there are other environmental impact indicators that can be calculated by default methods such as ecotoxicity, there are many default assumptions in how these are calculated which can obscure a mechanistic understanding of how each behaves when deploying increasing capacities of energy storage. Mechanistically understanding why an energy storage system contributes towards or reduces the emissions causing environmental impacts is important for having confidence in the results. Finally, the scope of our LCA does not include the end-of-life stage of the flow battery technologies. While literature exists regarding the effect of VRFB recycling [11] on life cycle greenhouse gas emissions, comparable analyses are not available for the ZBFB and IFB technologies. The implications of flow battery end-of-life on the main results will be discussed in Section 4.

The fourth step is to compare the benefits from electric grid integration against the environmental impact associated with flow battery deployment as the capacities of these systems are scaled up. The results for the different environmental impact indicators from the LCA described in Step 3 are scaled up linearly based on the energy and power capacity levels examined in Step 2 and compared against the corresponding calculated grid emissions reduction. The net environmental effect for each environmental impact indicator – defined as the difference between the benefit and impact – is calculated for each combination of power and energy capacity for the energy storage system. This exercise produces a two-dimensional contour map of the net environmental effect for each indicator: energy and power capacity comprise the two independent variables, while the net environmental effect comprises the contour levels. Results for a given power capacity but varying energy capacity are extracted from these maps to present results as shown in Fig. 3, whereas the entire map is shown in Fig. 5. The results reveal for each environmental impact indicator: 1) whether a contour corresponding to a value of zero for the net environmental benefit exists and if so, 2) at what power and energy capacity values this occurs. Such a contour represents the maximum threshold values for the power and energy capacities of the aggregated energy storage system beyond which installing more capacity would cause a net environmental detriment, and therefore should be set as the limiting capacity for energy storage deployment in sustainable energy system planning, given the assumptions of this study.

3. Results

3.1. Net emissions benefits and thresholds: Fixed power capacity

Fig. 1 shows the relationship between net reductions in GHG (Fig. 3a, c, and e; using 100-year global warming potentials [29]) and PM (Fig. 3b, d, and f, represented by $PM_{2.5}$-equivalent) emissions with

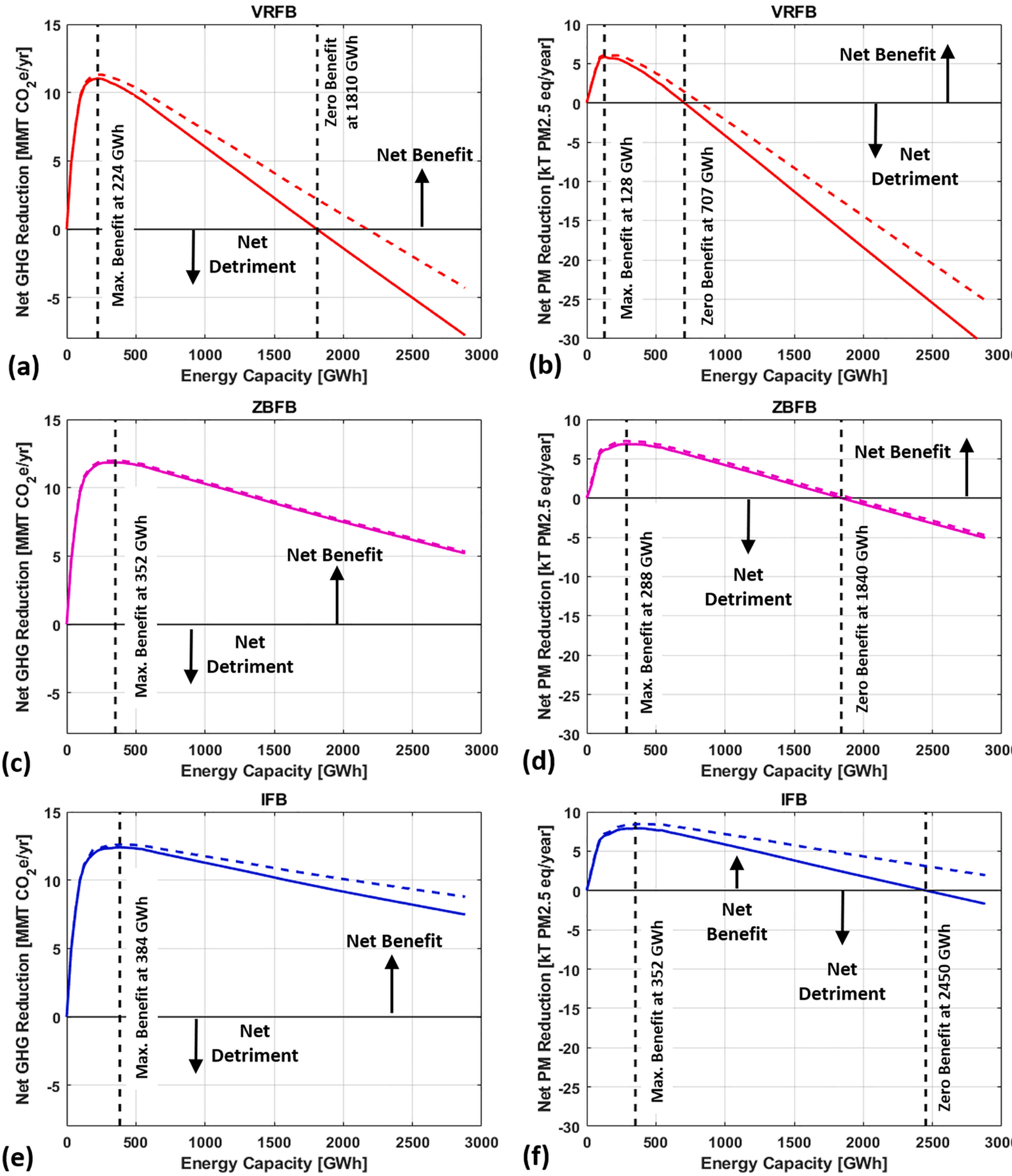

Fig. 3. Net environmental benefits as a function of varying energy capacity and a fixed power capacity of 24 GW when deploying each of the three different flow battery types: Vanadium-Redox (VRFB), Zinc-Bromide (ZBFB), and Iron (IFB) on two different environmental impact indicators: Greenhouse Gas Emissions [million metric tons of CO2-eq per year] and Particulate Matter (PM) [kilotons of PM2.5-eq per year]. (a) VRFB for GHGs, (b) VRFB for PM, (c) ZBFB for GHGs, (d) ZBFB for PM, (e) IFB for GHGs, (f) IFB for PM. Solid lines represent results using the reference life cycle inventories for battery manufacturing. Colored dashed lines represent the sensitivity of the results to decarbonized electricity inputs in battery manufacturing. Black dashed lines emphasize that, in each case, marginal environmental benefits begin to diminish after less than 350 GWh of batteries are deployed, and in several cases additional deployment has a detrimental effect on emissions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

deployed energy capacity for the three types of flow batteries. Such net reductions reflect the difference between the environmental benefits of deploying energy storage capacity on the electric grid and the emissions associated with the production of the storage system (including the acquisition of raw materials and production of the battery, from “cradle-to-gate”). Because energy and power capacity of flow battery energy storage systems may be independently sized, these results reflect a constant power capacity of 24 GW, since this is the energy storage power capacity specified for the year 2045 in the E3 PATHWAYS study [2] for California that we use as our representative modeled scenario. In each case shown, the net benefits of storage are substantial at energy capacities of 100–350 GWh. However, such benefits reach a maximum (i.e., the marginal benefits diminish) as installed energy storage capacity increases above 350 GWh. In the modeled scenario, 350 GWh corresponds

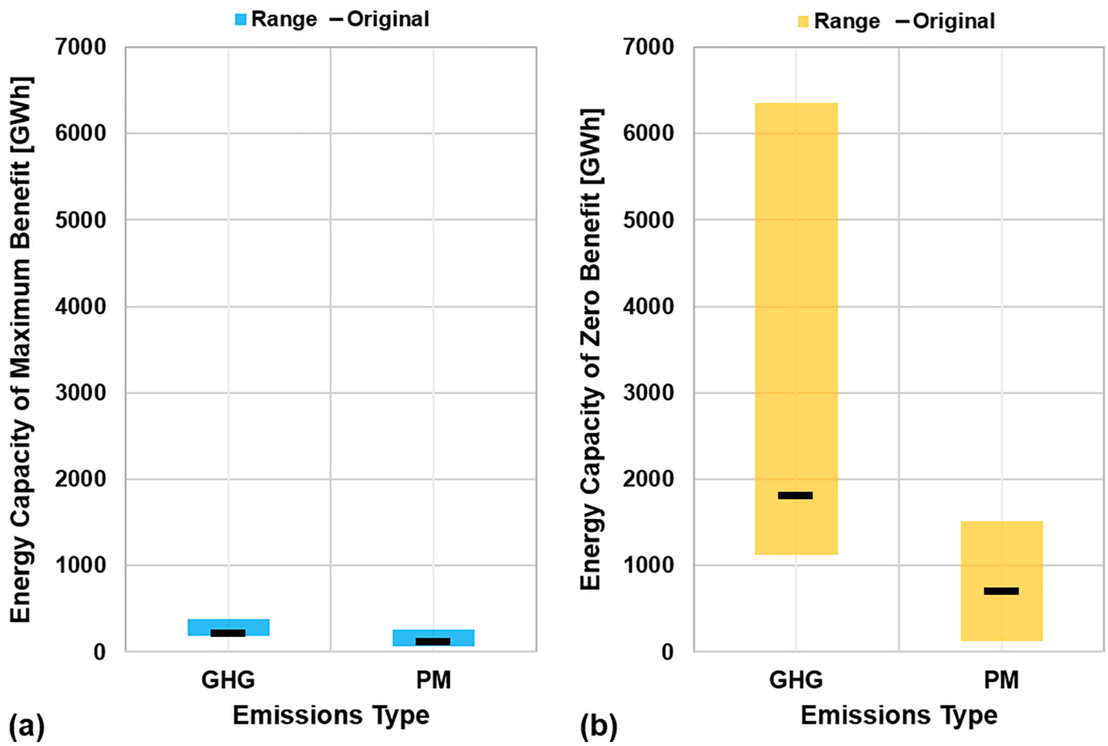

Fig. 4. Values of the energy storage capacities where (a) maximum emissions reduction occur and (b) zero-emissions reduction occur for the Vanadium Redox Flow Battery using different production pathways for the vanadium pentoxide electrolyte on different environmental impact indicators. Total power capacity is fixed at 24 GW for the electric grid of California. GHG = Greenhouse Gas; PM = Particulate Matter. The base case values consistent with Fig. 1 are presented as black lines.

to 53% of the mean daily generation from renewable sources. Indeed, in several of the modeled cases, we find that marginal benefits eventually reach zero—where additional storage capacity increases emissions on a net basis (Fig. 3a, b, d, and f).

Diminishing net benefits occur because the majority of wind and solar generation are misaligned with the electric load profile on a timescale of 4–12 h. Therefore, the initial units of energy storage capacity are sufficient to shift renewable generation across this timescale. When misalignment on the hourly timescales is alleviated, misalignment between wind/solar generation and the electric load on longer timescales must be addressed. Therefore, the energy storage system will need to become capable of shifting renewable generation across weeks, months, and seasons, and will need to become significantly larger to do so. This effect is explored in more detail in electric grid modeling studies [3,30,31]. Thus, as the negative life cycle impacts of energy storage increase linearly in proportion to energy capacity, the positive grid benefits associated with such increasing energy capacity increase asymptotically. Combined, these result in diminishing and eventually zero marginal net benefits of energy storage capacity deployed (black dashed lines in Fig. 3).

The specific thresholds of energy capacity where net benefits reach zero depend strongly on environmental impact indicator and battery type. For GHG emissions, the VRFB exhibits a threshold of 1810 GWh (271% of mean daily renewable generation), while the ZBFB and IFB do not exhibit such a threshold in the range of energy storage capacities examined here. This is because the IFB and ZBFB materials extraction and manufacturing processes have relatively low GHG emissions, while the VRFB contributions are much higher. For PM, however, all three flow battery types exhibit marginal thresholds of 707, 1840, and 2450 GWh (106%, 275%, and 357% of mean daily renewable generation) for the VRFB, ZBFB, and IFB, respectively. The thresholds of energy capacity where net benefits reach the maximum also depend on the environmental indicator and battery type, however, there is less variance in capacities of the maximum net reduction between these factors than the capacities when net reduction reaches zero. The capacities of the maximum net reduction for all three batteries fall in the range of 224–384 GWh (33–57% of mean daily renewable generation) for GHG emissions and 128–352 GWh (19–52% of mean daily renewable generation) for PM emissions.

3.2. *Effect of production process choices on maximum- and zero-benefit thresholds*

These threshold values will depend on the materials and manufacturing process selections represented in the life cycle inventories of the different flow batteries as well as the composition of the regional grid in which these storage systems operate. To highlight the effect of changing life cycle inventories on these thresholds, here we investigate two different sensitivities.

The first assesses the effect of fully decarbonizing the electricity inputs for the battery materials extraction and manufacturing processes, specifically with wind power, represented by the colored dashed lines in Fig. 1. Existing life cycle inventories for these processes characterize the emissions intensity of electricity inputs based on the electric grid mix of the region where the process takes place at the time the inventory was created. However, many regions around the world are moving towards decarbonized electricity systems, affecting the emissions intensity of these processes. Decarbonizing the electricity inputs for materials extraction and manufacturing has relatively minimal effects for the ZBFB but increases the net benefits for the VRFB and IFB most notably after the capacity of maximum benefit. This serves to extend the threshold where zero benefit occurs to a higher energy storage capacity, enabling more energy storage to be installed while still providing net benefits.

The second assesses different production pathways for the VRFB electrolyte. Vanadium pentoxide (V_2O_5) is the major electrolyte component and also the largest contributor to VRFB environmental impact [24], but this material does not have a consistent life cycle

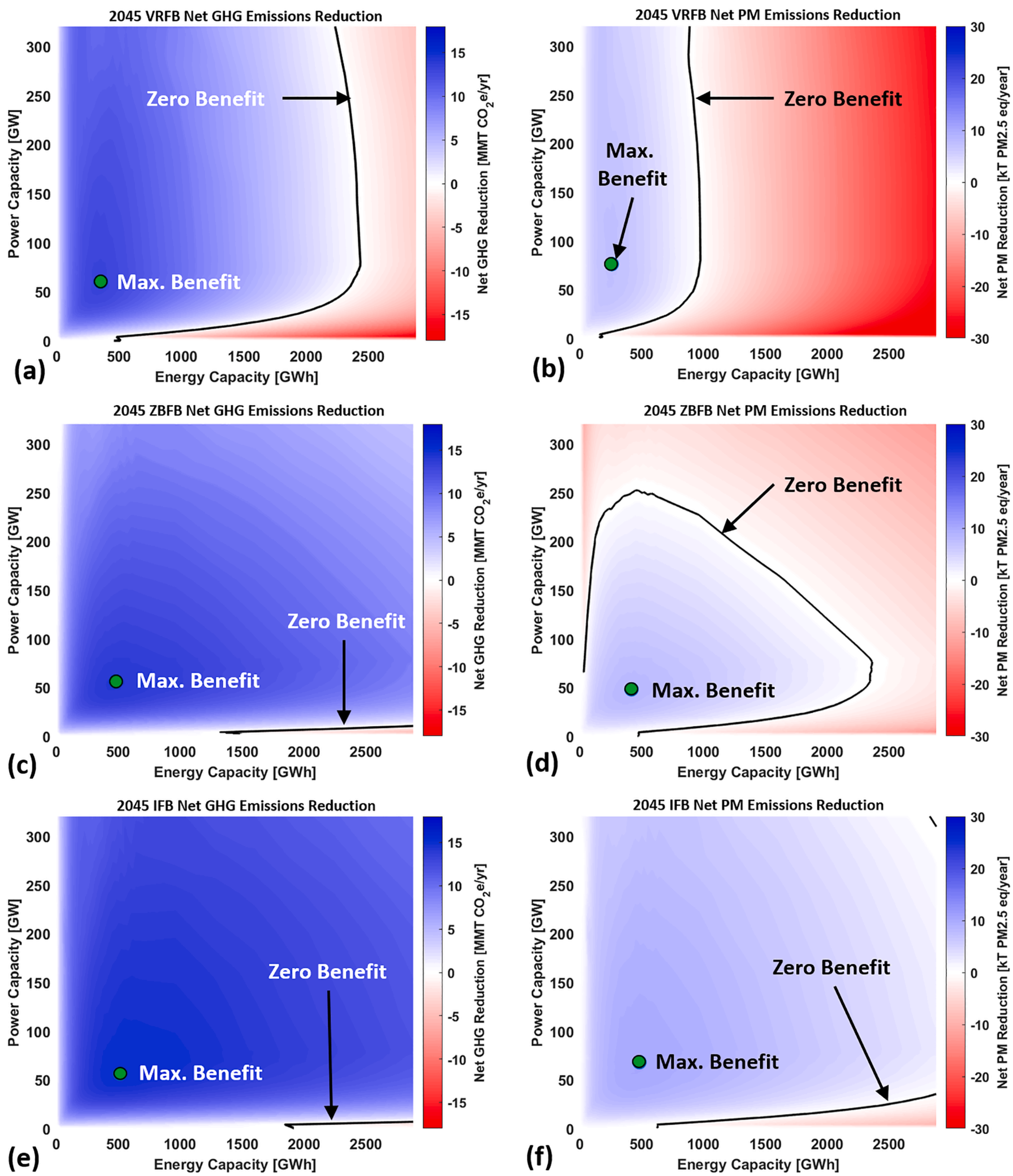

Fig. 5. Net environmental benefits as a function of varying both power and energy capacity when deploying each of the three different flow battery types: Vanadium-Redox (VRFB), Zinc-Bromide (ZBFB), and Iron (IFB)on two different environmental impact indicators: Greenhouse Gas Emissions (GHG) in million metric tons of CO_2-eq per year and Particulate Matter (PM) in kilotons of PM2.5-eq per year. (a) VRFB for GHGs, (b) VRFB for PM, (c) ZBFB for GHGs, (d) ZBFB for PM, (e) IFB for GHGs, (f) IFB for PM. Solid black lines represent the threshold beyond which additional capacity causes a net emissions increase. Solid green dots represent the energy and power capacity combinations where the net emissions reduction is maximized. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

inventory that is agreed upon in the literature. To understand how the uncertainty in the materials extraction and manufacturing process for the vanadium pentoxide electrolyte affects the threshold values, we conducted the analysis using different life cycle inventories for vanadium pentoxide production as presented by He et al. [24] and determined the corresponding range of these threshold values as presented in Fig. 4 for a fixed power capacity of 24 GW.

Fig. 4 shows that depending on the environmental impact intensity of the materials extraction and manufacturing processes of the evaluated flow batteries, the thresholds of energy capacity where net benefits reach a maximum and where net benefits are zero can vary between 18–226% and 33–953% of mean daily renewable generation, respectively. This variability highlights the sensitivity of net emissions reductions to the life cycle emissions associated with energy storage systems. In particular, net reductions in PM emissions may reach zero at relatively low levels of deployed capacity (e.g., 122 GWh or 18% of mean daily renewable generation) in the case of VRFB due to the intensity of PM emissions in the production of vanadium pentoxide [24]. However, net reductions of GHGs may persist to much larger storage capacities (e.g., 6360 GWh or 953% of mean daily renewable generation) if the materials extraction and manufacturing pathway for vanadium pentoxide is one with low GHG emissions intensity.

3.3. Net emissions benefits and thresholds: Variable power and energy capacities

A major feature of flow batteries is that their power capacity and energy capacity are decoupled. Different components in the battery are responsible for providing power and energy capacity so that they can be sized independently. Fig. 5 thus shows the sensitivity of net reductions in GHG and PM emissions in our California scenario to both energy and power capacity. For each type of flow battery and emission, black contours indicate the combination of energy and power capacities where net benefits reach zero, and green circles indicate the capacity combination that maximizes net emissions reductions. Net reductions in emissions are largest at power capacities between 56–60 GW for GHGs and 48–76 GW for PM (38–41% and 33–52% of available generating capacity, respectively), indicating that there are diminishing benefits in sizing the power capacity of grid-wide energy storage beyond this level (at least in this California scenario). Meanwhile, the greatest net reductions correspond to energy capacities of 352–512 GWh for GHGs and 256–480 GWh for PM (53–76% and 38–73% of mean daily renewable generation, respectively). These power and energy capacity ranges represent target specifications for the fleet of flow battery energy storage systems that maximize the environmental benefits.

Since the benefits of storage to the electric grid at a given power and energy capacity do not depend on the storage technology between the flow battery types, the net reductions in emissions as storage is deployed are due to differences in the emissions intensity of different batteries' materials extraction and manufacturing processes. For VRFB, in which the electrolyte is the major contributor to environmental impact, net reductions in emissions are considerably more sensitive to energy capacity than power capacity (evident in the nearly vertical orientation of contours in Fig. 5a and b), but the power capacity must be at a minimum level to ensure that benefits are not negated. In contrast, for ZBFB almost any combination of energy and power capacity provides net reductions in GHG emissions (Fig. 5c), but net reductions in PM occur only over a relatively narrow band of energy and power capacities. (Fig. 5d). This is because the production of both the electrode assembly and electrolyte for ZBFB are PM-emissions intensive. Finally, life cycle GHG and PM emissions of IFB are lower than the other types, enabling a net reduction in emissions across a wide range of power and energy capacities.

4. Discussion and conclusions

Our findings demonstrate the potential for negative life cycle impacts to substantially diminish or even outweigh the environmental benefits of deploying energy storage technologies, suggesting the importance of analyzing trade-offs when transitioning to renewables-heavy electricity systems. Although the thresholds of energy and power capacity of storage technologies we identify are specific to the storage technologies, raw materials acquisition and production processes, and details of the energy system we model, there are general lessons represented by the following key findings:

- In deploying energy storage, the largest marginal emissions reductions are provided by the first units deployed. This marginal benefit diminishing sharply after a certain capacity of energy storage is installed since the emissions contributed to producing the next unit of energy storage begins to exceed the emissions reductions provided from using that unit of energy storage on the grid.
  – In these results, the point of negative marginal returns (i.e., less than the maximum reduction in emissions) occurring at relatively modest storage capacities—38–76% of mean daily renewable generation with the technologies and scenario we analyzed.
- Without accounting for the relative scaling of the emissions benefits from energy storage use versus the emissions contributed from energy storage production, it is possible to install energy storage capacity to the point where net emissions are increased.
  – In these results, deploying large energy storage capacity (e.g., >106% of mean daily renewable generation) caused overall emissions to increase. This is a key conclusion for policymakers in jurisdictions such as California, where ambitious legislation targets carbon neutrality of a renewables-dominated electric grid by 2045: energy storage systems in support of such goals must be sized such that net environmental benefits are maximized—or at least net positive.
- Reducing the emissions intensity of the non-use-phase of energy storage systems via manufacturing or materials improvements extends the range of energy storage capacity that can be installed while still providing net emissions reduction benefits.
  – In these results, we use the example of lower-emissions sourcing of the VRFB electrolyte to show how it extends the threshold where marginal emissions benefits begin to decrease.

Our analysis raises questions in three major aspects:

1) How would advances in technological maturity and consequential material use, etc. affect impact scaling with increases in installed capacity? This analysis is based on a snapshot of technological maturity for each flow battery and the life cycle inventories for their supply chain and composition. The potential for improvements in the manufacturing of the different battery types and their in-use performance as the technology matures and as the infrastructure supporting their supply chains improve would shift the thresholds. For example, improving the roundtrip efficiency of the battery system or adopting a more environmentally benign manufacturing and materials usage profile will increase the ratio of environmental benefits relative to impact and will widen the capacity range where net benefits are realized. Alternatively, extending the lifetime of the energy storage system will also increase the benefits provided per unit installed by reducing the per-kWh environmental impact of the unit. In addition, the improvements in reducing the uncertainty in the life cycle inventory data could alter the results. In this study, for instance, uncertainty is known to exist for the boundary and process chosen for vanadium pentoxide, as explained in more detail by He et al. [24]; the relevant data used for the present study are presented in the Supplemental Information. Further uncertainty is based on the exclusion of the end-of-life stage of the flow battery life cycle in this analysis. As battery technologies mature, implementing recycling processes will offset the environmental impacts of raw materials extraction, but consequently will impose environmental impacts of its own. If the reduction in the former is larger than the contribution of the latter, this reduces the non-use-phase impact of the batteries and will increase the capacity levels at which the zero-benefit and maximum benefits occur, and vice versa. Weber et al. [11] showed that implementing recycling for the VRFB can reduce its GHG emissions by up to 50% when using wind resources to supply process electricity needs. Similar results are obtained for lithium-ion batteries, as demonstrated by Dunn et al. [18].
2) How would thresholds where the net reduction reaches a maximum or zero change if this analysis was conducted for a different region? California already exhibits a relatively low-carbon grid that will continue to decrease in GHG emissions, for example, and the benefits provided by energy storage are based on displacing natural gas with renewables. In other regions of the U.S. or the world, the displacement of other resources currently in use such as coal or fuel oil will

provide higher benefits compared to those shown here, and consequently, the range of capacities where net benefits are provided will be widened. Additionally, the thresholds vary for different environmental indicators due to differences in the spatial scale over which the impacts occur. The impacts of GHG emissions occur on a global scale and are not sensitive to the location of contributing emissions. By contrast, PM and other benefits and impacts can be regionally separated. For example, the California electric grid already exhibits low PM emissions and in the year 2045 scenario, the PM emissions are only sourced from natural gas power plants that have relatively low PM emissions intensity relative to other fossil fuels. Therefore, the reduction in PM from electric grid operation due to the use of these energy storage systems is relatively small compared to that contributed by the materials extraction and manufacturing processes for the flow battery systems in this study. While the increases of PM can occur in different regions than the reductions in PM and therefore certain populations can still benefit from improved air quality, increasing PM emissions on a net basis may still be undesirable.

3) How would changes in energy policy in other regions affect the capacity where the maximum or zero benefit thresholds occur? We demonstrate the effect of decarbonizing electricity inputs in materials and manufacturing processes in this study, finding that decarbonizing the electricity inputs in regions where materials and manufacturing processes take place increase the net environmental benefits and extend the range of capacities where energy storage provides a net positive environmental benefit. However, there are many forms of energy other than electricity that are used as inputs to these processes to provide functions such as high-grade heat, such as direct combustion of natural gas or coal. Electrifying these inputs in combination with a decarbonization of the regional grid will decrease the emissions intensity associated with these processes. This will reduce the cradle-to-gate emissions associated with flow batteries, increasing their net benefit and subsequently the thresholds where maximum benefit is reached or where zero benefit occurs.

Future work will involve assessing a wider range of current and emerging energy storage technologies beyond flow batteries. Such work could support policy targets that consider in tandem the deployment of renewable generation and energy storage that achieve maximum decarbonization benefits, not just renewable energy penetration targets. Moreover, using LCAs that consider a suite of environmental impacts can provide a more holistic picture of environmental benefits or impacts of not only battery storage technologies, but also grid decarbonization strategies. Further, investigation of how the cost-effectiveness of using energy storage to reduce emissions when taking both in-use benefits and life cycle impacts into account is warranted, as the emissions reductions per dollar profile of these systems will differ between frameworks accounting for in-use emissions reduction benefits vs. net emissions reduction benefits. Finally, once a larger suite of studies that characterize the non-use-phase impacts vs. use-phase benefits for a diverse array of energy storage systems is available, further research can abstract robust characteristics from these studies to develop a more general mathematical model for optimally planning large-scale battery deployments for supporting grids with high solar and wind penetrations.

Despite these limitations, our results demonstrate the need to evaluate both environmental impact and benefit, which depend on both the life cycle of technologies and the complex dynamics of electricity systems. Without such convergence of energy systems and LCA, decision-makers risk following pathways and deploying technologies that fail to reduce emissions as much as they expect, if at all.

CRediT authorship contribution statement

Shan Tian: Methodology, Formal analysis, Investigation, Writing - original draft. **Haoyang He:** Formal analysis, Investigation. **Alissa Kendall:** Writing - review & editing, Conceptualization. **Steven J. Davis:** Writing - review & editing, Conceptualization. **Oladele A. Ogunseitan:** Writing - review & editing, Conceptualization. **Julie M. Schoenung:** Writing - review & editing, Conceptualization, Resources, Supervision. **Scott Samuelsen:** Supervision, Resources. **Brian Tarroja:** Conceptualization, Methodology, Resources, Writing - original draft, Writing - review & editing, Funding acquisition.

Declaration of Competing Interest

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

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apenergy.2021.117354.

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