

1. Methods

1.1. Energy System Model

Table S1 includes the technologies that are considered in each sector of the energy system. For consistency, we replace the life cycle inventories of passenger vehicle technologies used in the original model by Shu et al.¹ by using inventory data for the corresponding technologies from the *premise*² database, assuming medium-sized, European passenger vehicles, and the highest emission standards available for combustion engines. We further update the life cycle inventories of grid-scale batteries using inventory data for stationary batteries from the *premise* database that assume current market trends to continue. Further, we replace inventories for industrial heat pumps based on Khan et al.³ We assume that the energy system's demand for natural gas is met through imports from Norway.

Table S1: Technologies included in the considered sector-coupled energy system. The implementation of the technologies is described in detail by Baumgärtner et al.⁴ and Shu et al.¹

Sector	Technology
Power	Biogas-to-Power Geothermal Hard Coal Hydrogen Fuel Cell Lignite Natural Gas Combined Cycle Natural Gas Turbine Oil Photovoltaics Run-of-river Hydropower Waste Incineration Wind Offshore Wind Onshore Other Non-Renewables
Heat - Residential	District Natural Gas Boiler Electrode Boiler Thermal Insulation (Buildings) Thermal Insulation (District Heating) Heat Pump (residential) Natural Gas Boiler Oil Boiler
Heat – Low-temperature Industrial	District Natural Gas Boiler Electrode Boiler Heat Pump (Industrial) Natural Gas Boiler
Heat – Mid-temperature Industrial	District Natural Gas Boiler Electrode Boiler Natural Gas Boiler
Heat – High-temperature Industrial	District Natural Gas Boiler Natural Gas Boiler
Mobility	Battery Electric Vehicle Compressed Natural Gas Vehicle Diesel Vehicle Gasoline Vehicle Hydrogen Fuel Cell Vehicle Plug-in Hybrid Vehicle
Transmission & Storage of Electricity	220 kV Transmission Line 380 kV Transmission Line Stationary Battery Pumped Hydro Storage
CO ₂ Capture and Storage	Direct Air Capture Post-combustion Capture Pipeline Transport Geological storage of CO ₂
Power-to-X	Power-to-Diesel Power-to-Hydrogen Power-to-Methane

1.2. Allocation factors for private passenger mobility

Hartmann et al.⁵ do not provide a specific allocation factor for private passenger mobility, since private passenger mobility is not an economic sector, but a final consumption activity that relies on goods and services from multiple economic sectors. Therefore, we determine the allocation factor for private passenger mobility based on the allocation factors of the following sectors $j \in \mathcal{J}$: (i) sale, maintenance and repair of motor vehicles, motor vehicles parts, motorcycles, motor cycles parts and accessories; (ii) manufacture of motor vehicles, trailers, and semi-trailers; and (iii) retail sale of automotive fuel, as these sectors provide the goods and services required for private passenger mobility. For each sector j in each geographical scope $s \in \mathcal{S}$, we calculate the share of total output that is used to satisfy final demand in Germany $\varphi_{DE,j,s}$ (Eq. 1).

$$\varphi_{DE,j,s} = \frac{FCE_{DE,j,s}}{x_{j,s}}, j \in \mathcal{J}, s \in \mathcal{S} \quad (\text{Eq. 1})$$

In Eq. 1, $FCE_{DE,j,s}$ is the direct final consumption expenditure satisfied within Germany by sector j that operates within geographical scope s , and $x_{j,s}$ is the corresponding total output of the sector-scope pair j, s . Direct final consumption expenditure includes final consumption expenditure by households, non-profit organizations serving households and governments, as well as, gross fixed capital formation. We obtain the allocation factor for private passenger mobility in Germany ($SoSOS_{DE}^p$) by multiplying the resulting shares $\varphi_{DE,j,s}$ with the allocation factors for each sector-scope pair j, s , using the allocation factors based on total final consumption expenditure ($SoSOS_{j,s}^{tFCE}$) and total gross value added ($SoSOS_{j,s}^{tGVA}$), and aggregating across sectors and geographical scopes (Eq. 2).

$$SoSOS_{DE}^p = \sum_j \sum_s \varphi_{DE,j,s} \cdot SoSOS_{j,s}^p \text{ for } p \in \{tFCE, tGVA\} \quad (\text{Eq. 2})$$

1.3. Implementation of the parameter for material-related environmental impacts

We consider reductions in material-related environmental impacts associated with the primary and secondary production of steel, aluminum, and copper, considering reduction levels of 5 %, 10 %, 15 %, 20% and 25 %. To consider reductions, we first construct the technosphere matrix⁶ for each process in the ecoinvent 3.10.1 database⁷ using Brightway 2.5⁸ and assuming a functional unit of one. Within the technosphere matrix, we identify the row and column corresponding to each material-process, as listed in Table S2. To model changes in material-related impacts, we scale the respective technosphere matrix entries by multiplicative factors of 1.05, 1.10, 1.15, 1.20, and 1.25, representing increases in the material-processes' material output of 5 %, 10 %, 15 %, 20 %, and 25 %, respectively. Due to the linear nature of life cycle assessment models, an increase in the material-processes' material output equals a proportional decrease in material-related environmental impacts. Although, we model an increase in the material-processes' material output to consider a decrease in material-related environmental impacts, the same decrease can be also achieved through a reduction in the material-processes' environmental impacts, through a shift from primary to secondary materials or through a combination of effects. Using the modified technosphere matrix, we calculate the environmental impacts on each planetary boundary category for every process in the ecoinvent 3.10.1 database under each material-related impact reduction scenario. The resulting impact databases are subsequently implemented in the energy system model.

Table S2: Material processes in ecoinvent 3.10.1 that are influenced by the material-related environmental impacts parameter

Process name in ecoinvent 3.10.1	Reference product	Location
market for copper, anode	copper, anode	GLO
market for copper, cathode	copper, cathode	GLO
market for scrap copper	scrap copper	RoW
market for scrap copper	scrap copper	Europe without Switzerland
market for scrap copper	scrap copper	CH
market for waste reinforcement steel	waste reinforcement steel	RoW
market for waste reinforcement steel	waste reinforcement steel	CH
market for reinforcing steel	reinforcing steel	GLO
market for scrap steel	scrap steel	RoW
market for scrap steel	scrap steel	Europe without Switzerland
market for scrap steel	scrap steel	CH
market for steel in car shredder residue	steel in car shredder residue	RoW
market for steel in car shredder residue	steel in car shredder residue	CH
market for steel, chromium steel 18/8, hot rolled	steel, chromium steel 18/8, hot rolled	GLO
market for steel, chromium steel 18/8	steel, chromium steel 18/8	GLO
market for steel, low-alloyed, hot rolled	steel, low-alloyed, hot rolled	GLO
market for steel, low-alloyed	steel, low-alloyed	GLO
market for steel, unalloyed	steel, unalloyed	GLO
market for aluminium oxide, metallurgical	aluminium oxide, metallurgical	IAI Area, EU27 & EFTA
market for aluminium oxide, metallurgical	aluminium oxide, metallurgical	RoW
market for aluminium oxide, non-metallurgical	aluminium oxide, non-metallurgical	IAI Area, EU27 & EFTA
market for aluminium oxide, non-metallurgical	aluminium oxide, non-metallurgical	RoW
market for aluminium, cast alloy	aluminium, cast alloy	GLO
market for aluminium, primary, ingot	aluminium, primary, ingot	RoW
market for aluminium, primary, ingot	aluminium, primary, ingot	IAI Area, EU27 & EFTA
market for aluminium, primary, ingot	aluminium, primary, ingot	IAI Area, North America
market for aluminium, wrought alloy	aluminium, wrought alloy	GLO
market for scrap aluminium	scrap aluminium	RoW
market for scrap aluminium	scrap aluminium	Europe without Switzerland
market for scrap aluminium	scrap aluminium	CH

1.4. Modelling limitations

Goal & scope: The energy system model used in this study does not cover all sectors of final energy demand, as it omits the public mobility sector and corporate mobility sector. Additionally, the model solely covers private passenger mobility by private passenger vehicles and excludes other forms of transport.

Allocation: We determine system-specific sustainability thresholds by allocating a share of the safe operating space derived from the planetary boundary framework, as defined by Steffen et al.⁹ A more recent framework has since been provided by Richardson et al.¹⁰. However, there is no life cycle impact assessment method available to operationalize the framework. We consider the regional boundary for South Asia as a proxy for the global boundary, as no global boundary is available and as suggested by Ryberg et al.¹¹ We do not consider regional planetary boundaries, as environmental impacts along life cycles are not regionally disaggregated. Our study identifies the impact on atmospheric aerosol loading as critical. Hence, future research should focus on including regional planetary boundaries, e.g. for atmospheric aerosol loading. Furthermore, we do not consider the planetary boundaries for novel entities, as no impact assessment method is available¹², and we do not consider interactions between boundaries.

The functional unit, geographical scope, temporal scope and included processes of our energy system model and the economic sectors used for allocation do not exactly match, as required by Hartmann et al.⁵ For instance, our energy system model solely includes cars, while the sectors related to passenger mobility also include motorcycle and trailers. Additionally, aggregating allocation factors for economic sector could lead to possible double-counting.⁵ To this end, we provide a sensitivity analysis for a varying share of safe operating space.

Energy system model: We apply a greenfield optimization approach to determine cost-optimal energy system designs whose life cycle environmental impacts remain within an allocated share of safe operating space. However, future energy systems are not constructed from zero but rather build on legacy infrastructure that can be maintained, repurposed or that must be disposed of. Consequently, legacy infrastructure influences costs and environmental impacts of future energy systems.⁴

Legacy infrastructure can be considered in brownfield optimization approaches, such as transition pathway optimization. While transition pathway optimization is common to determine cost-optimal energy system and has been applied while considering emission constraint⁴, current planetary-boundary-based life cycle impact assessment methods do not cover temporally-explicit environmental impacts. Thus, absolute environmental sustainability and cost optimization of energy system transition pathways remains unfeasible.

Our model is based on projected final energy demand and life cycle inventories, drawing on technology learning curves and prospective databases for 2050. However, these inputs are subject to inherent data uncertainties. In addition, the prospective scenarios used to determine prospective databases² assume an evolution of the energy system that may differ from the energy system designs derived in this study, leading to potential inconsistencies.

Sensitivity analysis: We vary the material-related environmental impacts, mobility demand and CO₂ storage capacity. Our study does not consider how variations in these parameters can be achieved, but bases the intervals for variation on literature data (Section 4.4 of the main paper). We vary the material-related impacts for steel, aluminum and copper, as these materials are the main contributors to the impact on atmospheric aerosol loading and the phosphorus cycle. However, other materials also contribute to impacts in these categories. We focus on demand reduction for the mobility sector, as demand reduction can be achieved through the expansion of public and shared transport.¹³ Yet, demand reductions are possible beyond the mobility sector.

We constrain the part of CO₂ storage capacity that is explicitly modelled in our energy system model. However, as we consider economy-wide evolution of global life cycles based on prospective scenarios, supply chains of the energy system include CO₂ storage capacities² that are not explicitly constraint. Thus, the real energy-system-related demand for CO₂ storage capacity might be higher than the demand identified in this study.

2. Results

2.1. Energy system designs for the base case

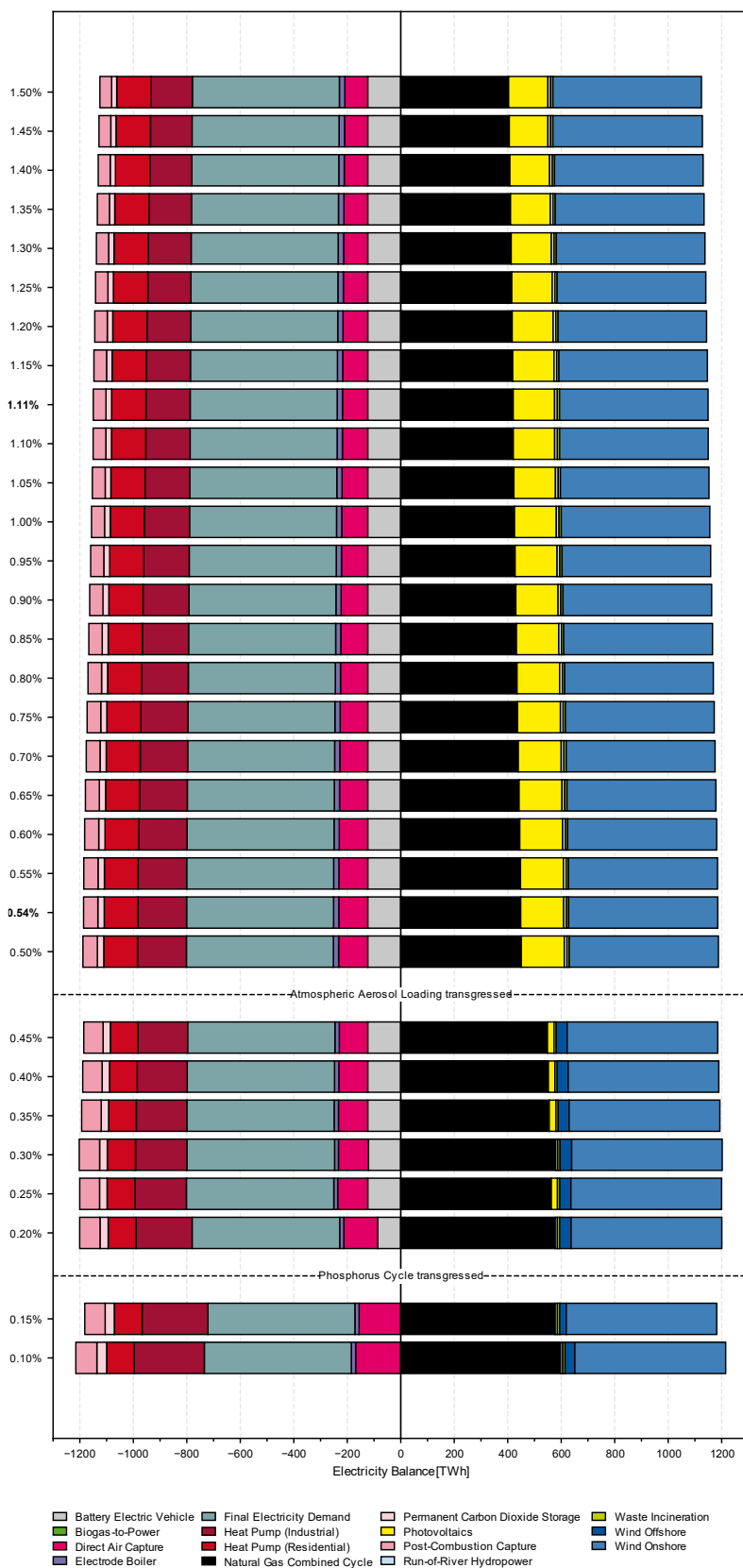


Figure S1: Base Case: Electricity balance for a varying share of safe operating space with technology contribution to electricity generation and electricity consumption. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

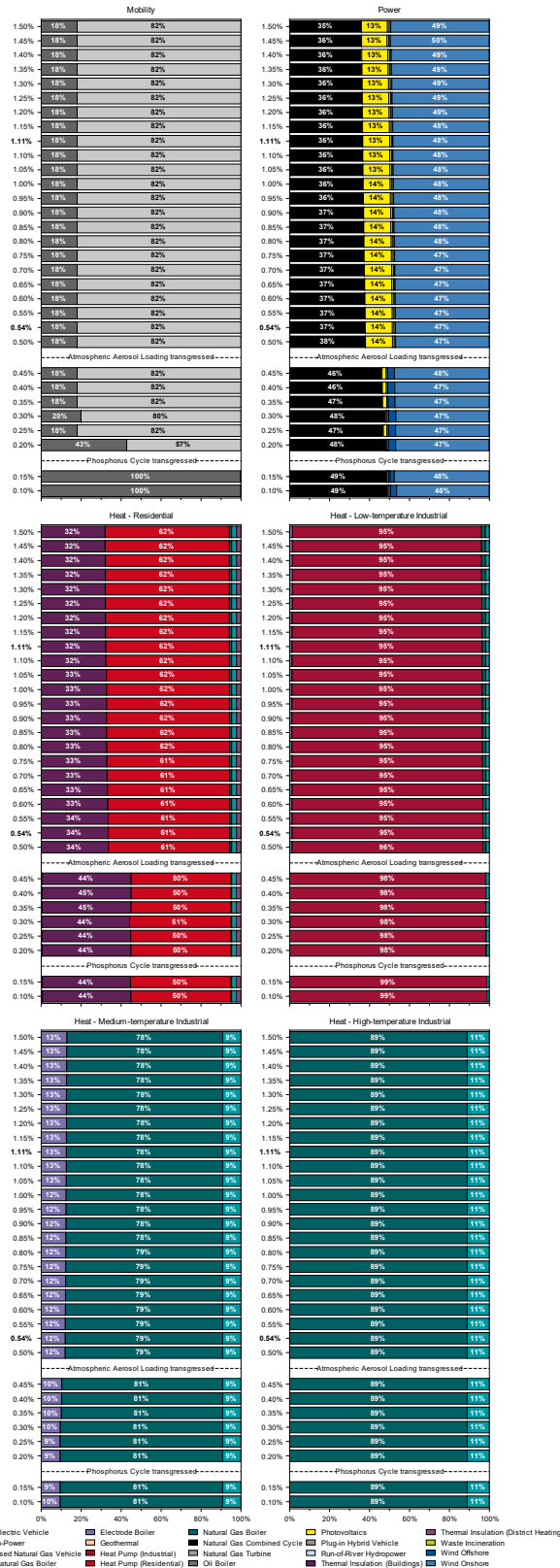


Figure S2: Base Case: Contributions of technologies serving final energy demand of the mobility, power, residential and industrial heat (low-temperature, medium-temperature, high-temperature) sectors for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54%) and total gross value added (1.11%) are marked in bold.

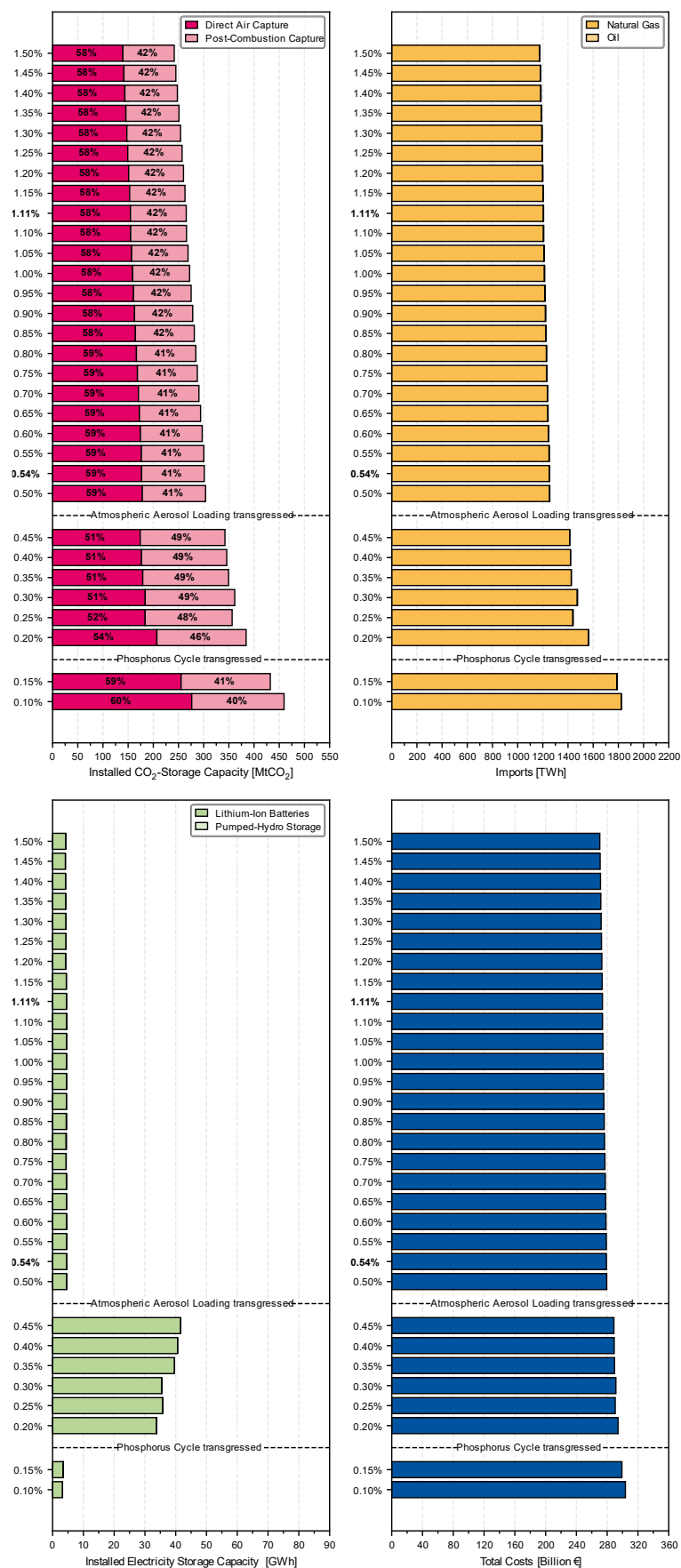


Figure S3: Base Case: Capacities for CO₂ storage, energy imports and electricity storage with technology contributions, and total system costs for a varying shares of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

2.2. Multi-tier contribution analysis for environmental impact categories

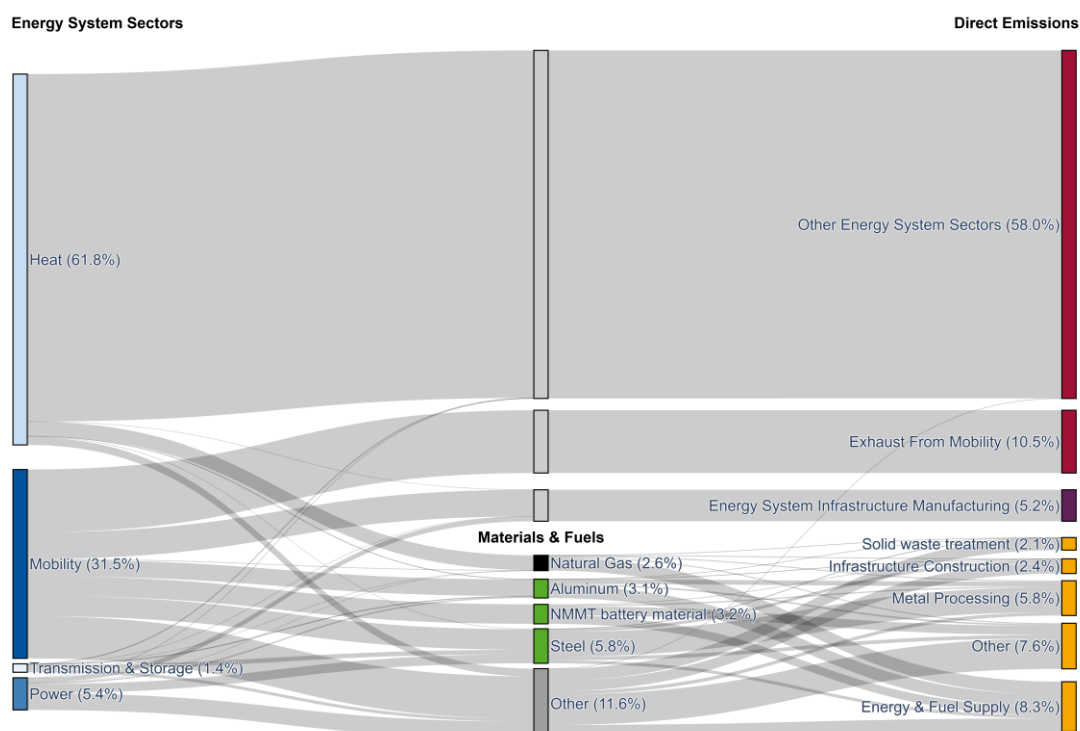


Figure S4: Multi-tier contributions to the climate change impact of the design based on total final consumption expenditure for Germany in 2050. Colors indicate the source of contributions: blue – energy system sectors; green – materials; black – fuels; grey – other materials and fuels; red – direct emissions from the energy system; purple – energy system infrastructure manufacturing; orange – material and fuels production. Contributions below 2 % to the impacts of materials, fuels, and their production are aggregated under “Other”.

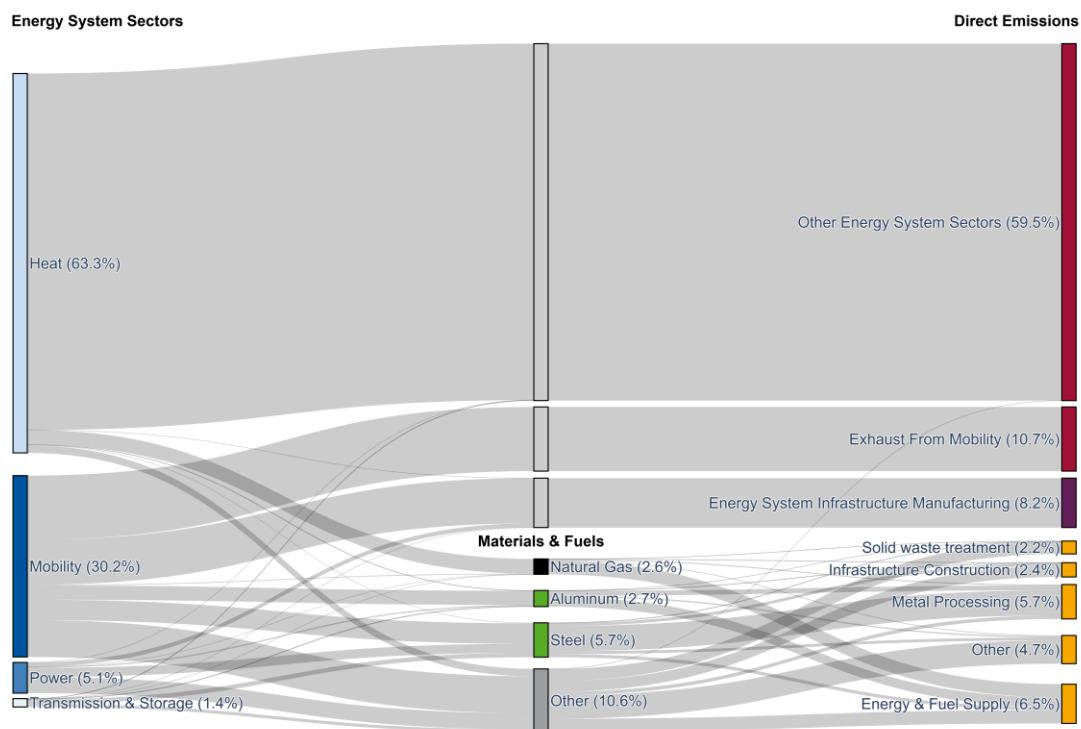


Figure S5: Multi-tier contributions to the ocean acidification impact of the design based on total final consumption expenditure for Germany in 2050. Colors indicate the source of contributions: blue – energy system sectors; green – materials; black – fuels; grey – other materials and fuels; red – direct emissions from the energy system; purple – energy system infrastructure manufacturing; orange – material and fuels production. Contributions below 2 % to the impacts of materials, fuels, and their production are aggregated under “Other”.

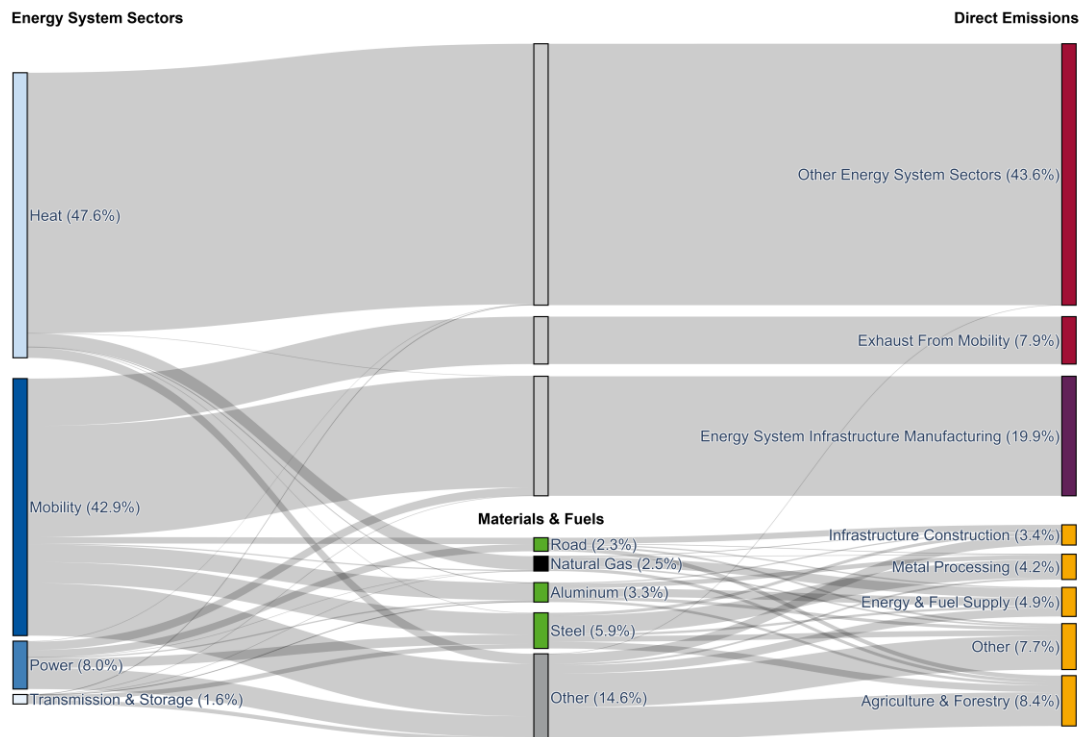


Figure S6: Multi-tier contributions to the change in biosphere integrity impact of the design based on total final consumption expenditure for Germany in 2050. Colors indicate the source of contributions: blue – energy system sectors; green – materials; black – fuels; grey – other materials and fuels; red – direct emissions from the energy system; purple – energy system infrastructure manufacturing; orange – material and fuels production. Contributions below 2 % to the impacts of materials, fuels, and their production are aggregated under “Other”.

2.3. Energy system designs for a decrease in material-related environmental impacts of 25 %

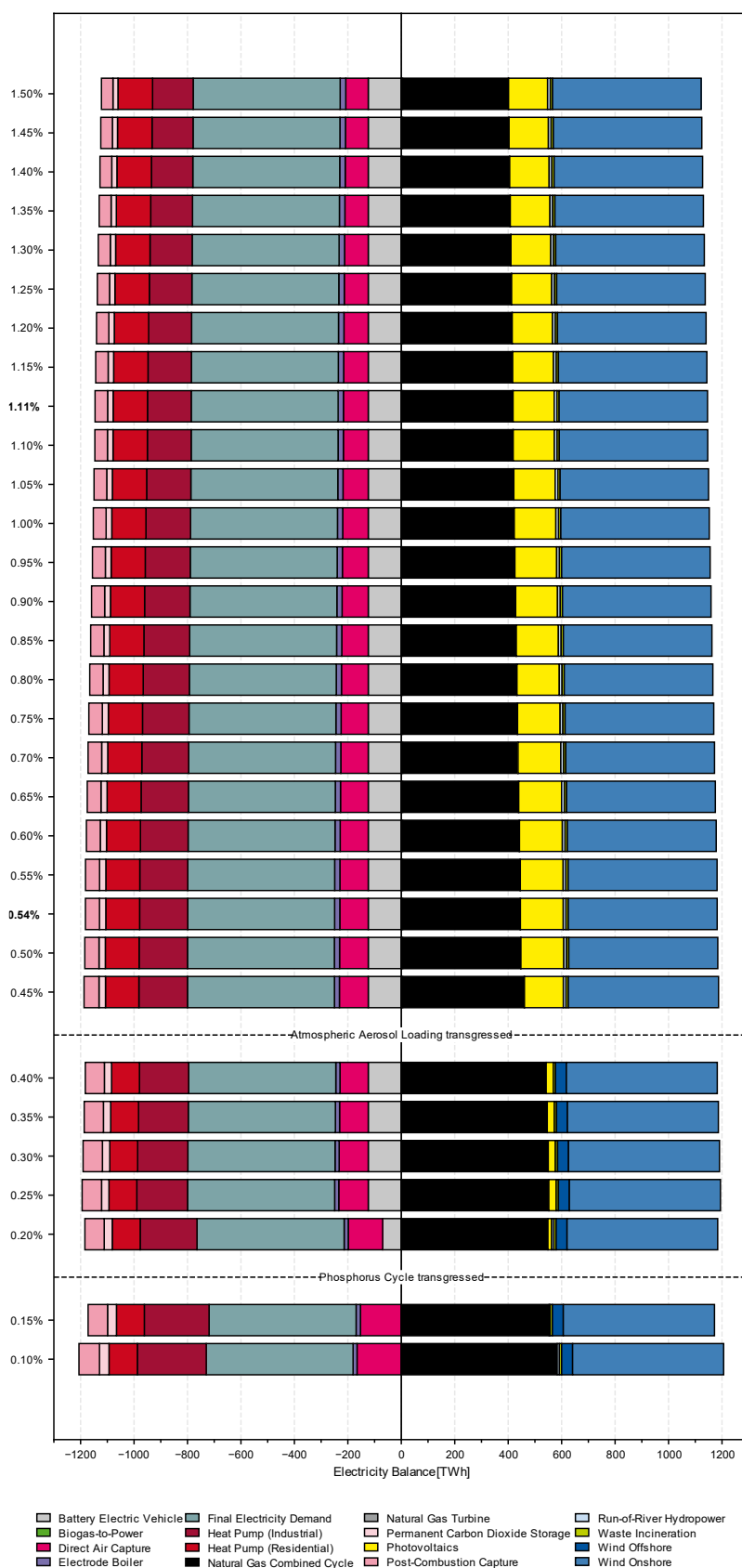
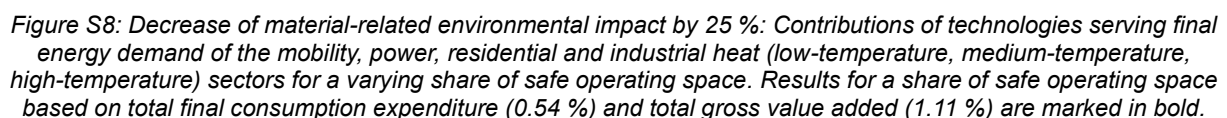


Figure S7: Decrease of material-related environmental impact by 25 %: Electricity balance for a varying share of safe operating space with technology contribution to electricity generation and electricity consumption. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.



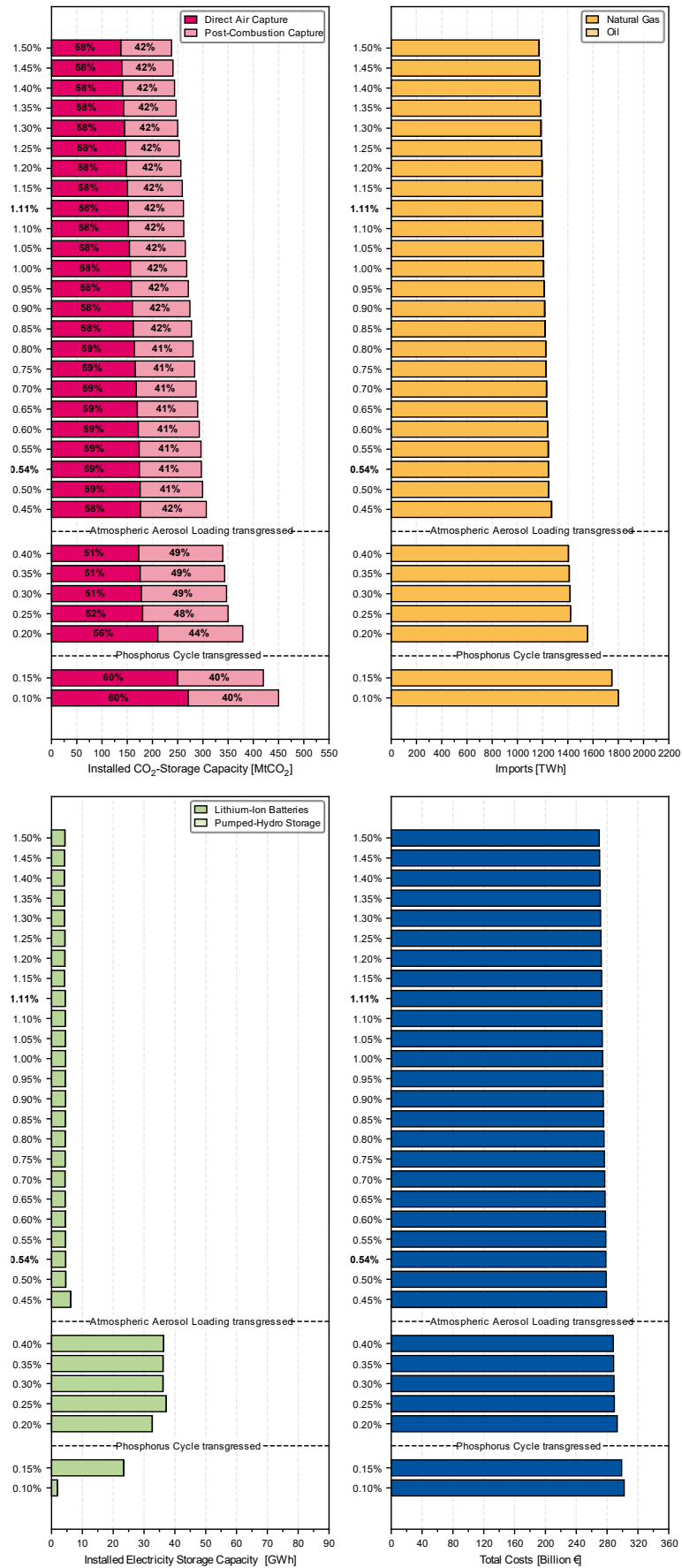


Figure S9: Decrease of material-related environmental impact by 25 %: Capacities for CO₂ storage, energy imports and electricity storage with technology contributions, and total system costs for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

2.4. Energy system designs for a decrease in mobility demand by 40 %

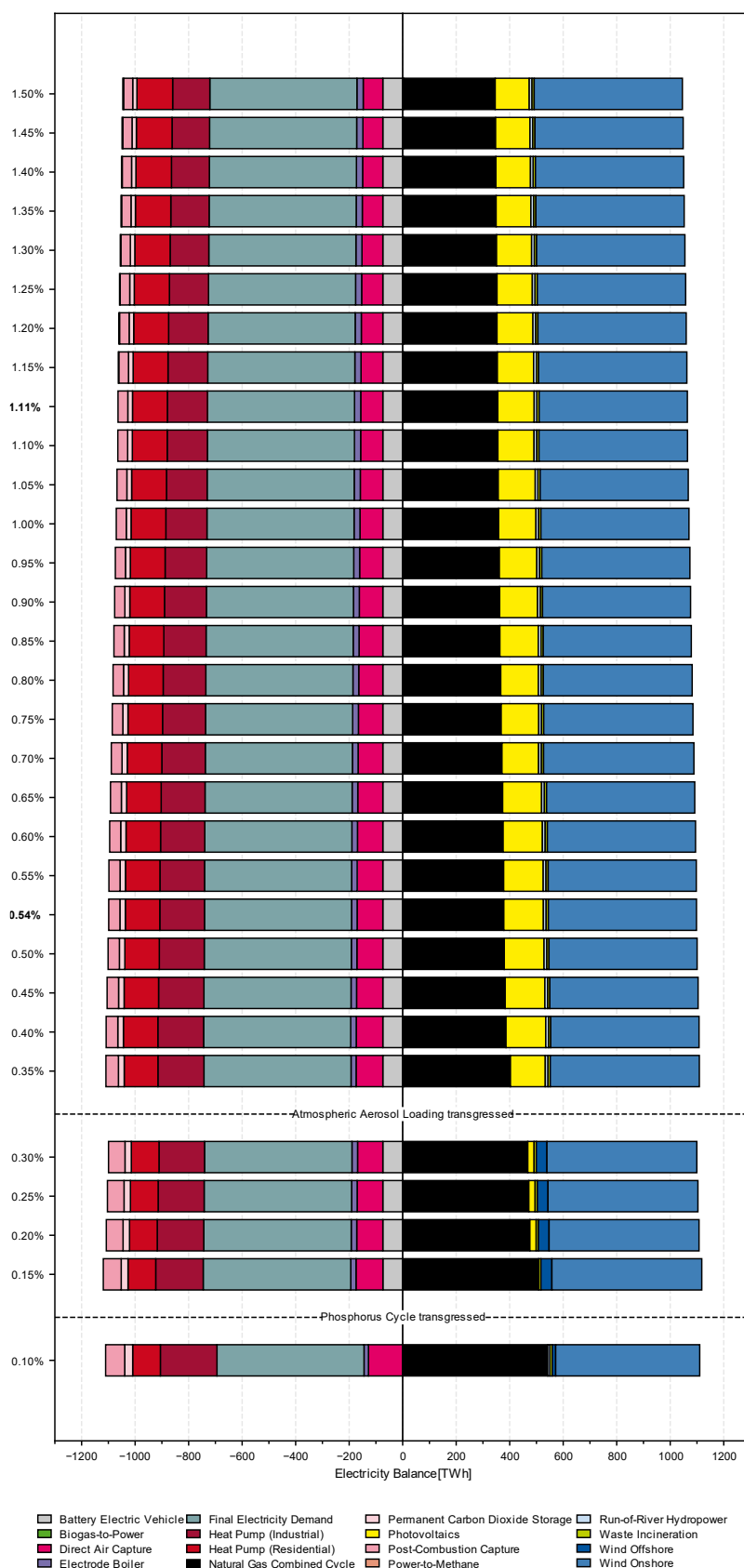


Figure S10: Decrease of mobility demand by 40 %: Electricity balance for a varying share of safe operating space with technology contribution to electricity generation and electricity consumption. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

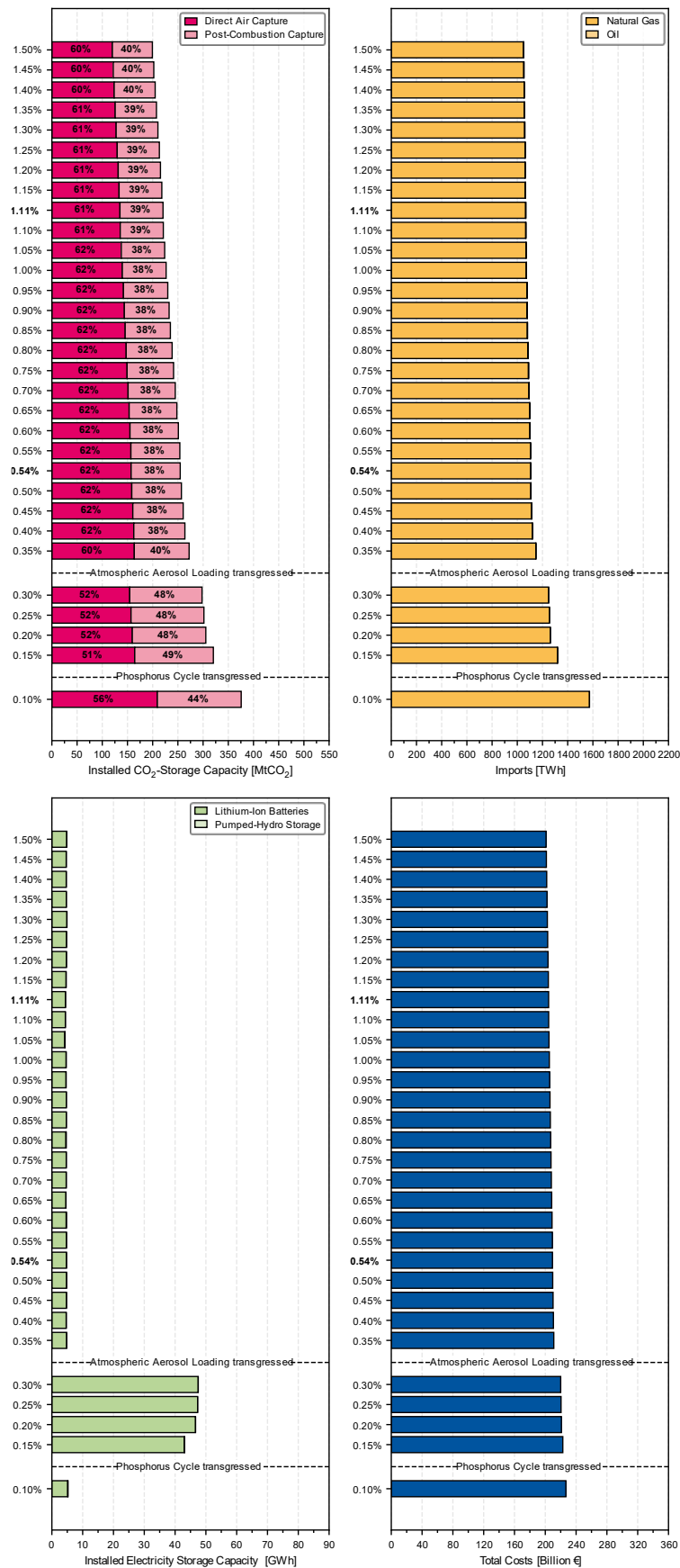


Figure S12: Decrease of mobility demand by 40 %: Capacities for CO₂ storage, energy imports and electricity storage with technology contributions, and total system costs for a varying shares of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

2.5. Energy system designs for a CO₂ storage capacity constraint to 179 Mt CO₂ annually

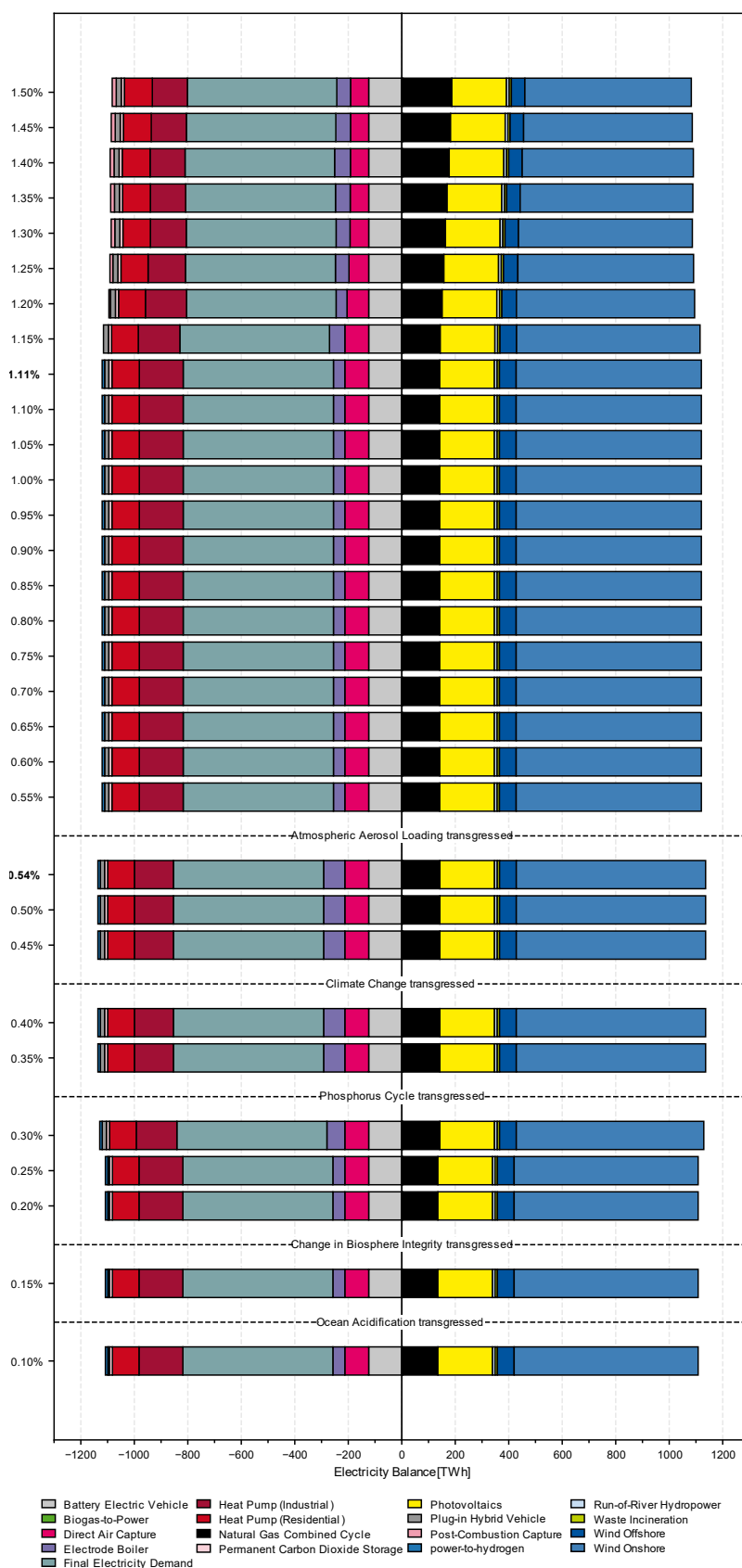


Figure S13: CO₂ storage capacity constraint to 179 Mt CO₂ annually: Electricity balance for a varying share of safe operating space with technology contribution to electricity generation and electricity consumption. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

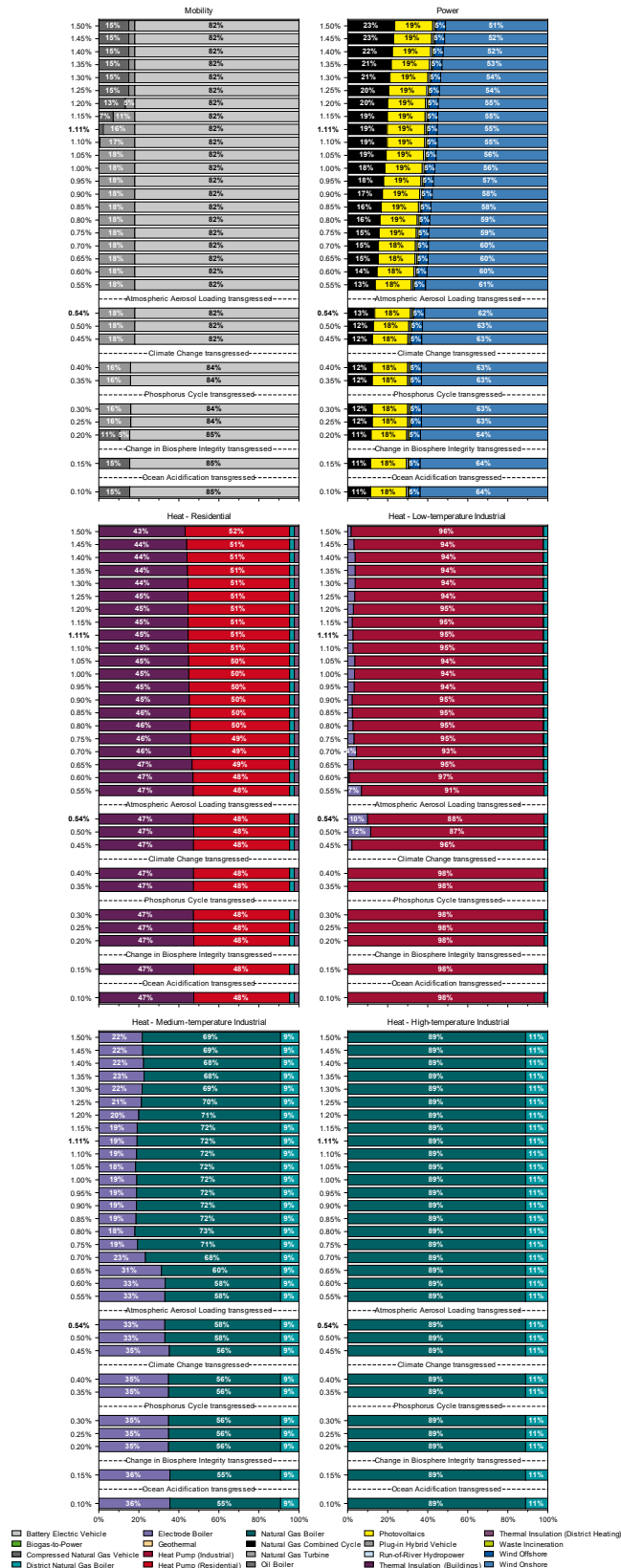


Figure S14: CO₂ storage capacity constraint to 179 Mt CO₂ annually: Contributions of technologies serving final energy demand of the mobility, power, residential and industrial heat (low-temperature, medium-temperature, high-temperature) sectors for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

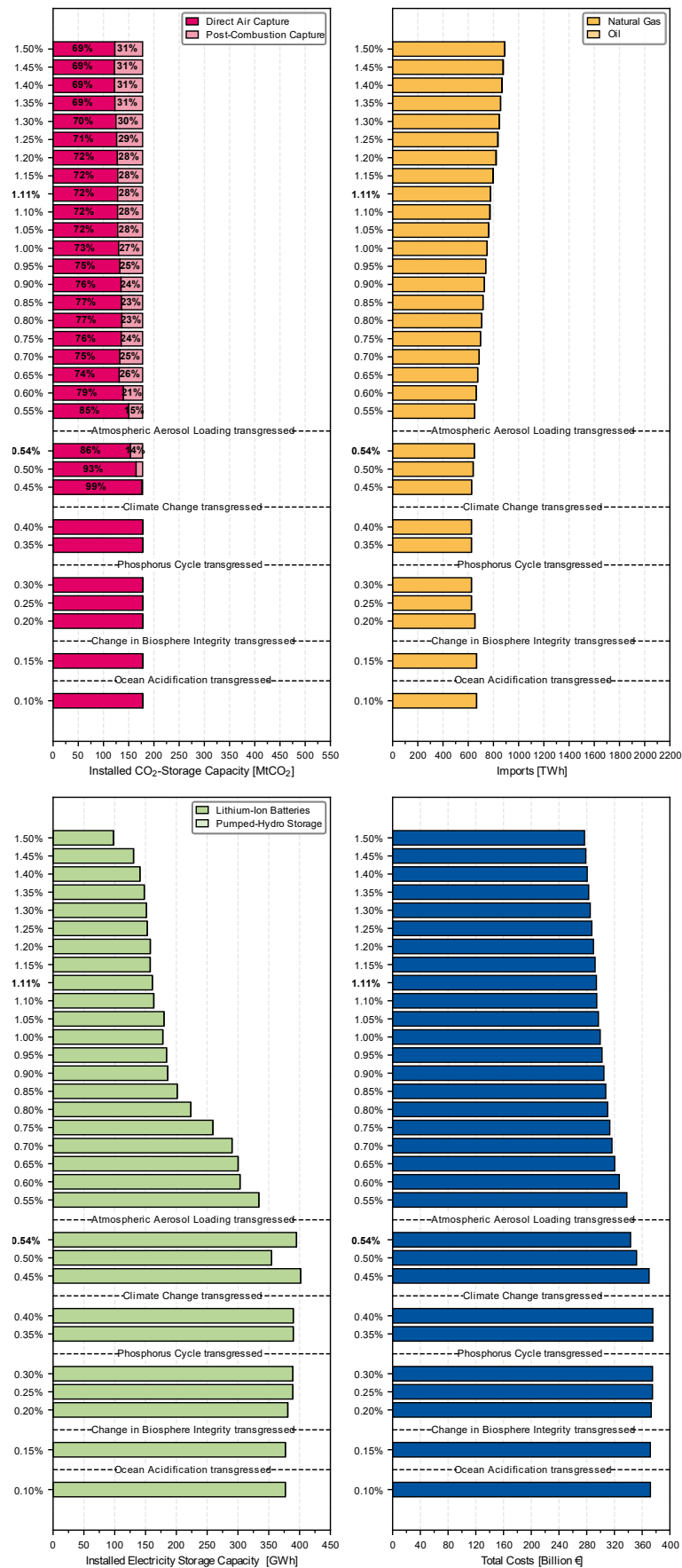


Figure S15: CO₂ storage capacity constraint to 179 Mt CO₂ annually: Capacities for CO₂ storage, energy imports and electricity storage with technology contributions, and total system costs for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

2.6. Energy system designs for a CO₂ storage capacity constraint to 146 Mt CO₂ annually

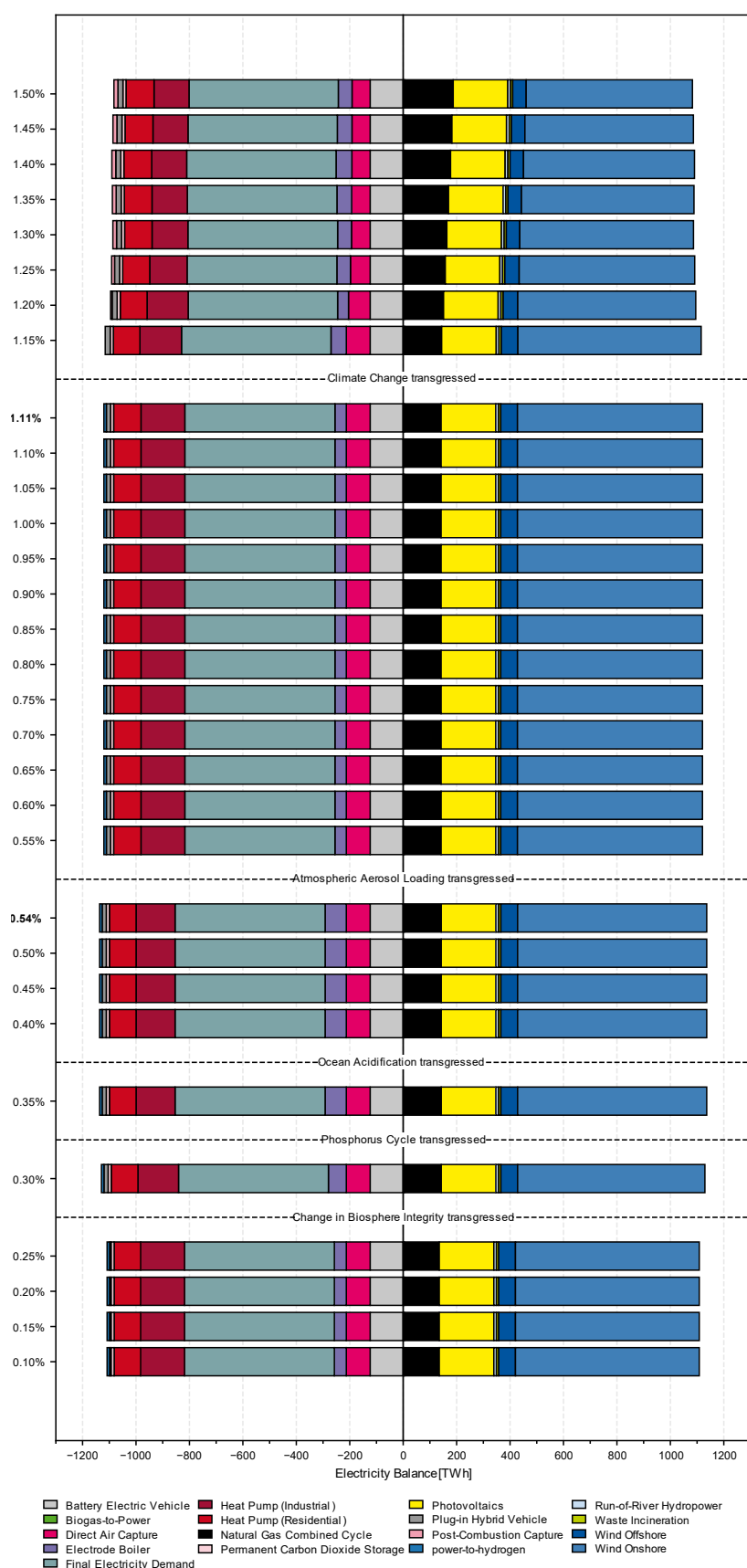


Figure S16: CO₂ storage capacity constraint to 146 Mt CO₂ annually: Electricity balance for a varying share of safe operating space with technology contribution to electricity generation and electricity consumption. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

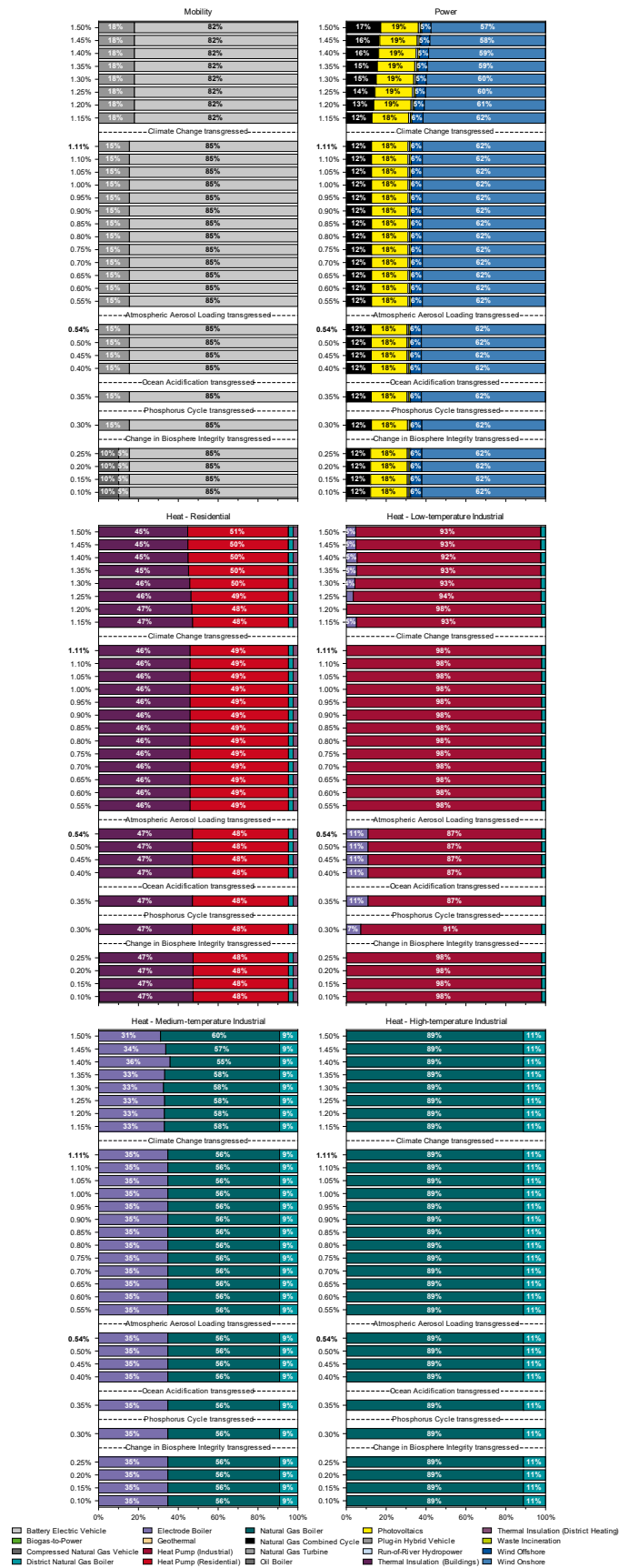


Figure S17: CO₂ storage capacity constraint to 146 Mt CO₂ annually: Contributions of technologies serving final energy demand of the mobility, power, residential and industrial heat (low-temperature, medium-temperature, high-temperature) sectors for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

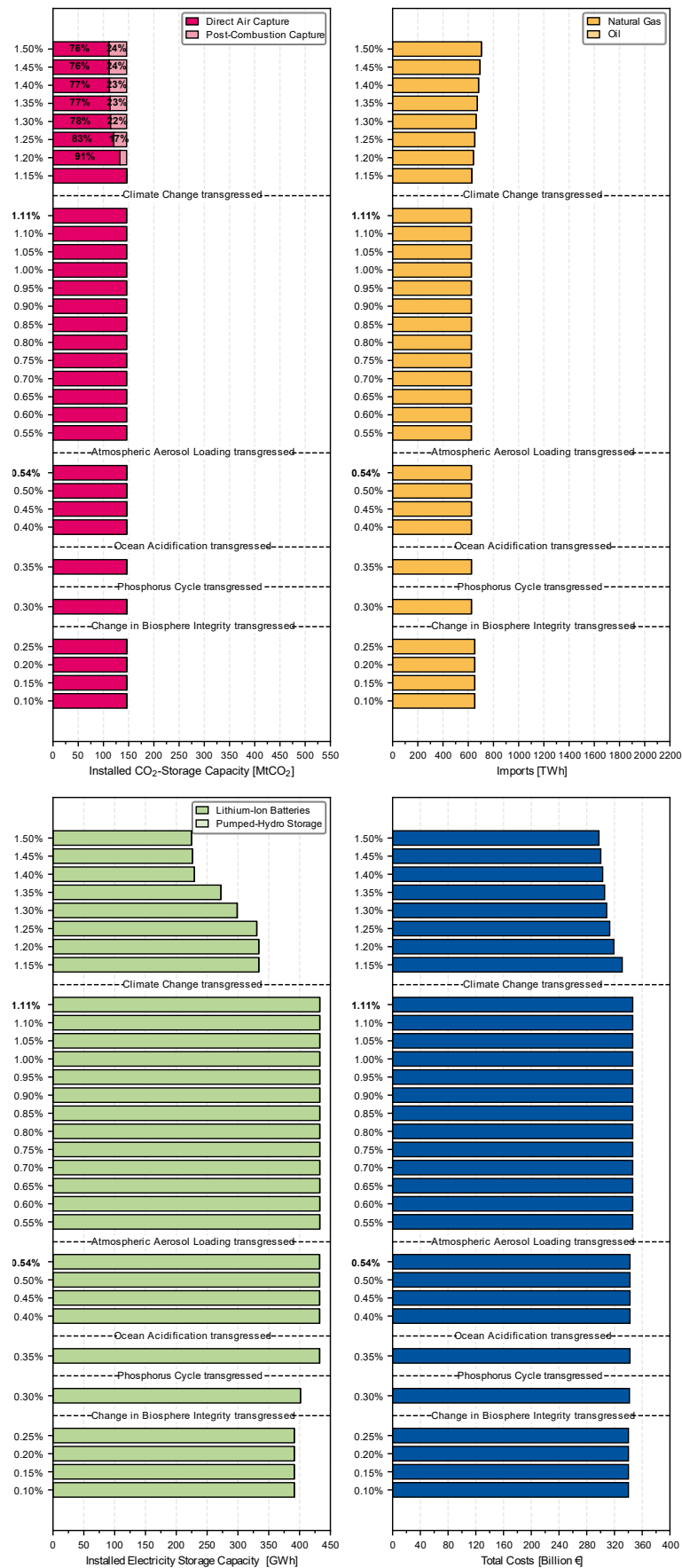


Figure S18: CO₂ storage capacity constraint to 146 Mt CO₂ annually: Capacities for CO₂ storage, energy imports and electricity storage with technology contributions, and total system costs for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

2.7. Sensitivity analysis for the natural gas price

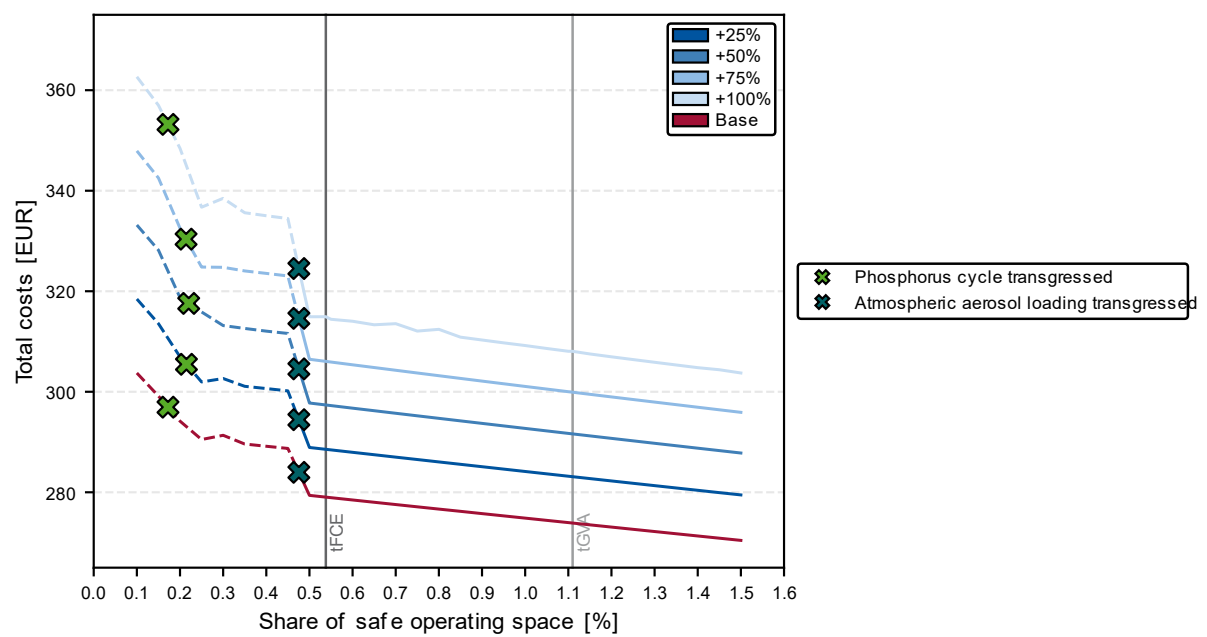


Figure S19: Sensitivity analysis for an increasing natural gas price.

2.8. Energy system design for an increase of the natural gas price by 100%

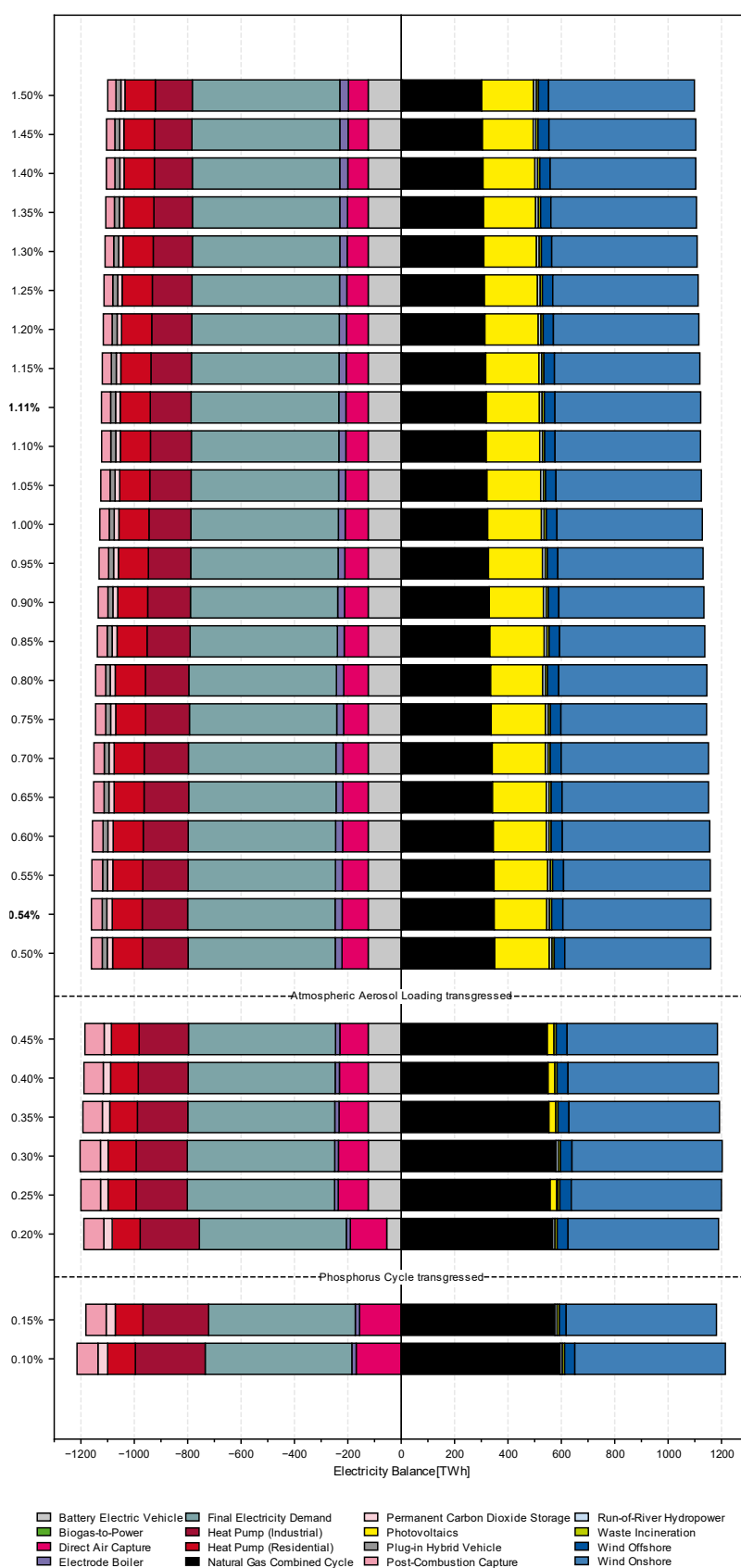


Figure S20: Natural gas price doubled: Electricity balance for a varying share of safe operating space with technology contribution to electricity generation and electricity consumption. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

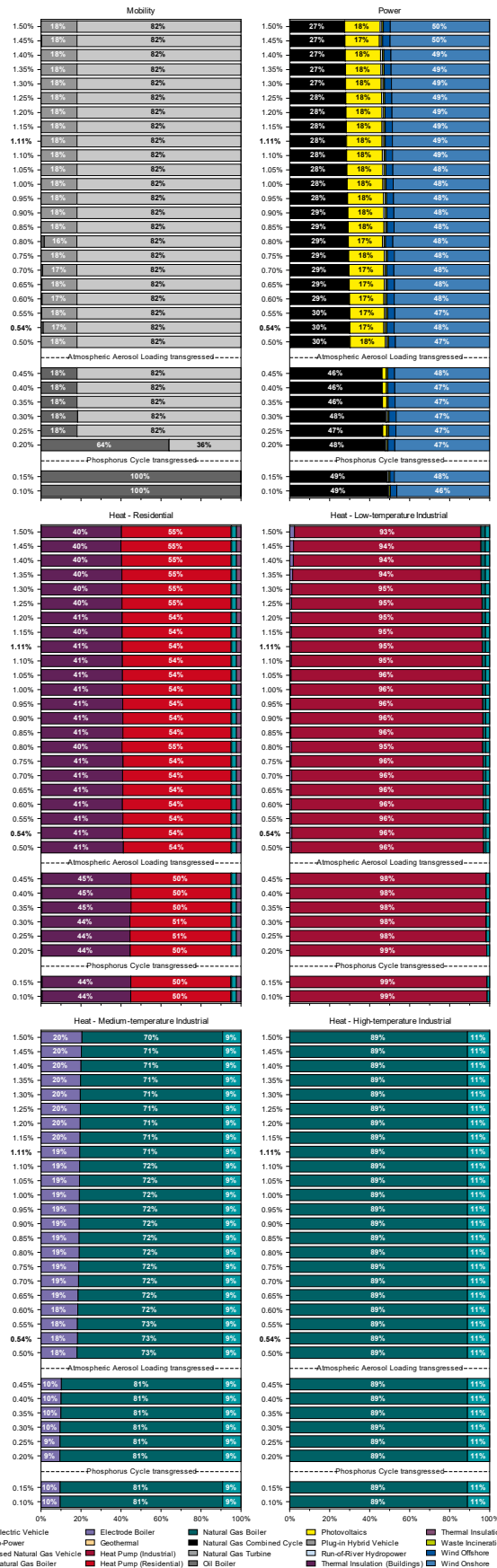


Figure S21: Natural gas price doubled: Contributions of technologies serving final energy demand of the mobility, power, residential and industrial heat (low-temperature, medium-temperature, high-temperature) sectors for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

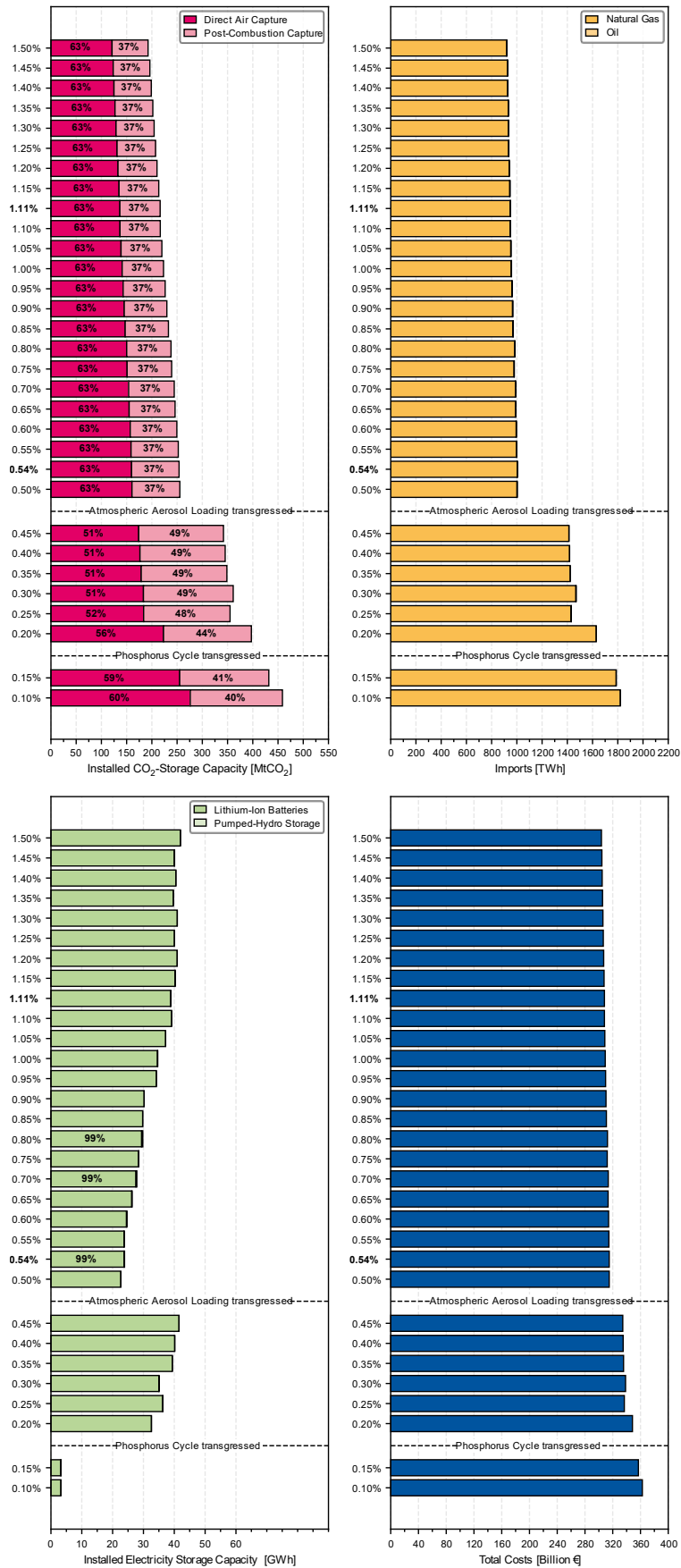


Figure S22: Natural gas price doubled: Capacities for CO₂ storage, energy imports and electricity storage with technology contributions, and total system costs for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

2.9. Sensitivity analysis for electricity import capacities

Imports of natural gas can be displaced by the import of alternative energy carriers. Here, we show the impact of electricity imports on sustainable energy system design, under the simplified assumptions of firm import availability, fixed constant prices and environmental impacts, and an annual import capacity limit. We assume environmental impacts of the average European electricity grid in 2050, modelled by using the life cycle inventory for '*marked group for electricity, medium voltage*' in '*EUR*' in 2050. We further assume import prices based on average European electricity prices in 2050 in the EU reference scenario.¹⁴

In practice, the availability of electricity imports may vary, limiting their ability to fully substitute for electricity generation from natural gas. Additionally, both environmental impacts and costs of electricity imports are time-dependent, affecting the integration of electricity imports in energy system designs.

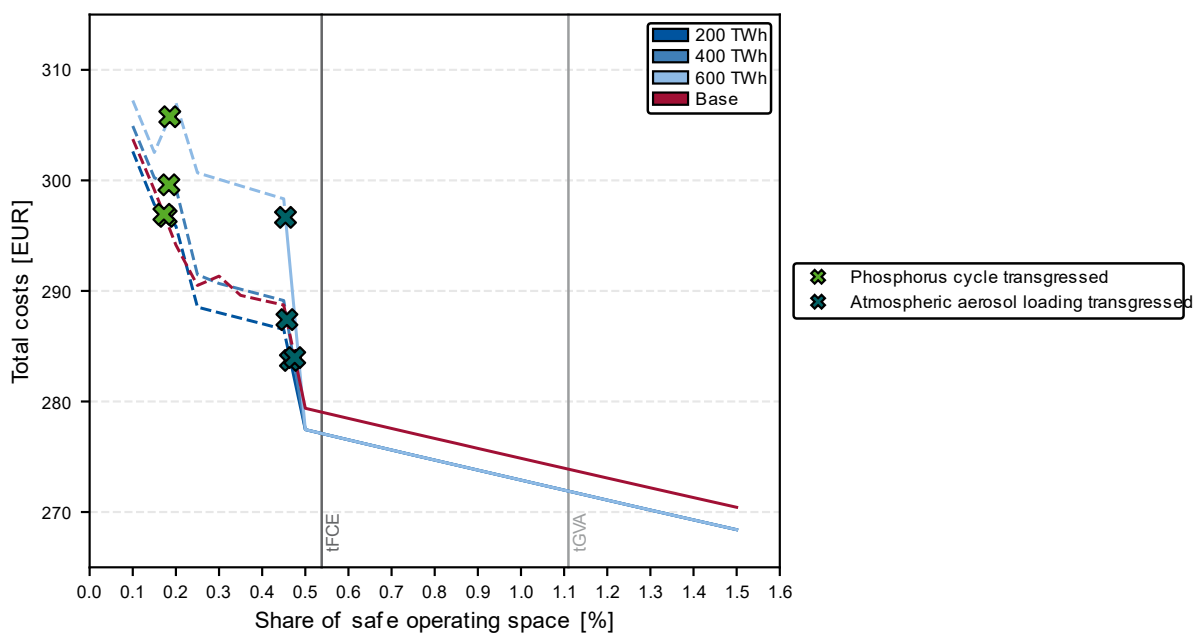


Figure S23: Sensitivity analysis for increasing annual electricity import capacity limit, assuming firm import availability, fixed constant prices and environmental impacts.

The resulting design for a scenario assuming electricity imports of 600 TWh are shown in the following Section 2.10.

2.10. Energy system designs for an electricity import capacity of 600 TWh per year

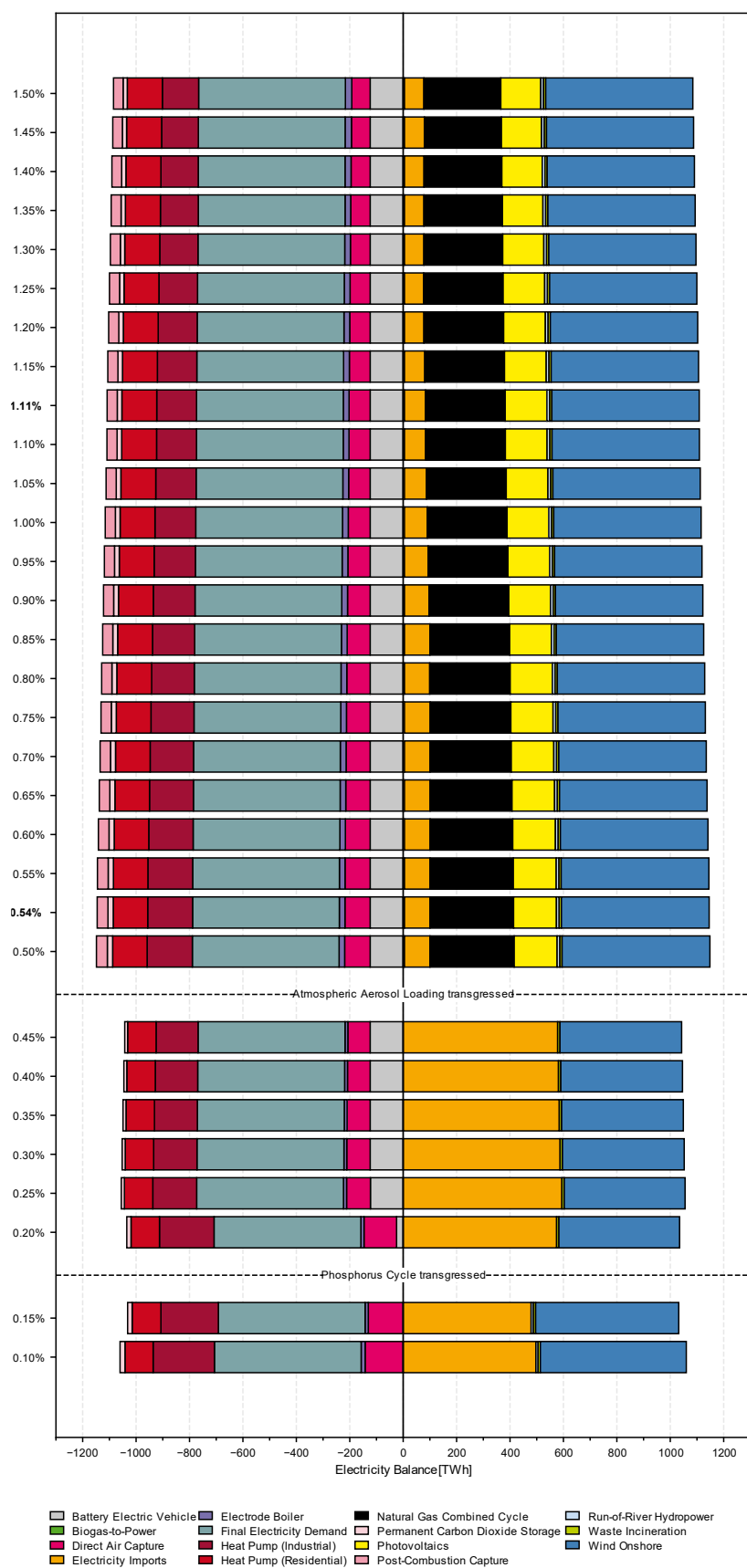


Figure S24: Electricity import capacity of 600 TWh per year: Electricity balance for a varying share of safe operating space with technology contribution to electricity generation and electricity consumption. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

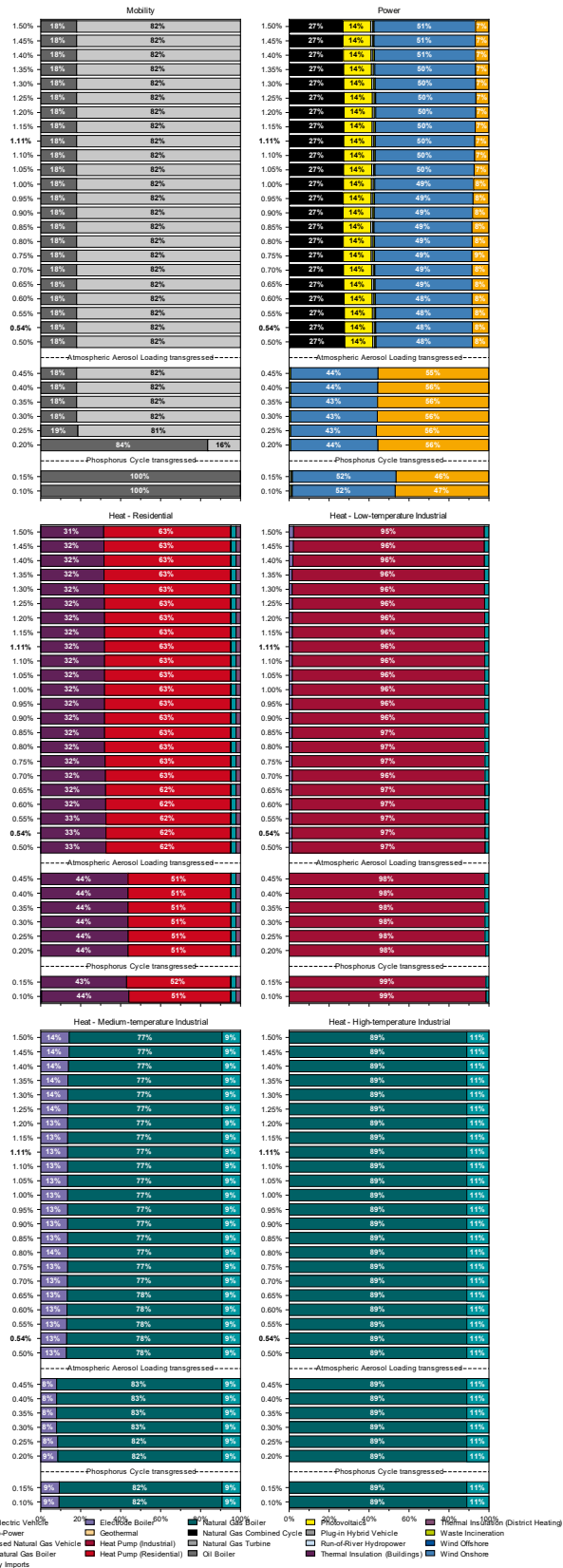


Figure S25: Electricity import capacity of 600 TWh per year: Contributions of technologies serving final energy demand of the mobility, power, residential and industrial heat (low-temperature, medium-temperature, high-temperature) sectors for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

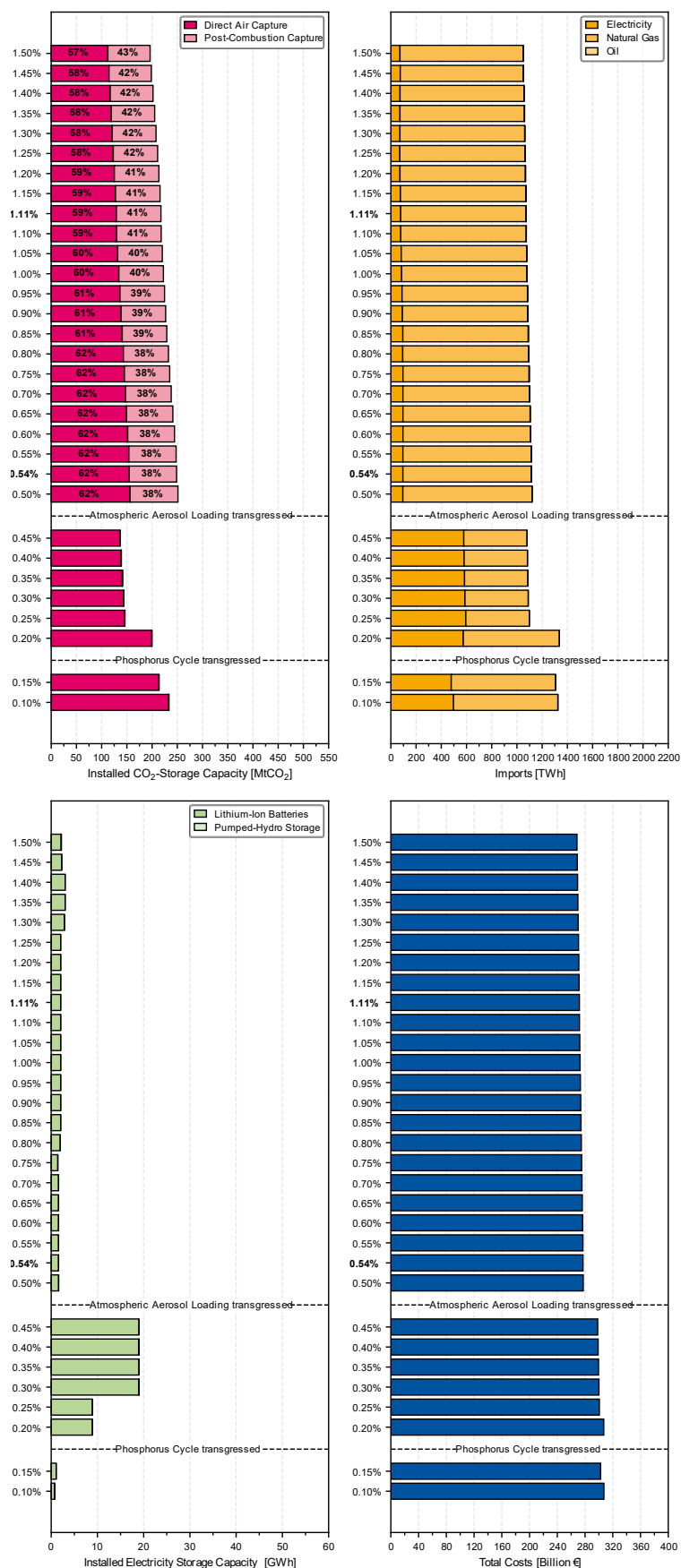


Figure S26: Electricity import capacity of 600 TWh per year: Capacities for CO₂ storage, energy imports and electricity storage with technology contributions, and total system costs for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

2.11. Sensitivity analysis for scenario used to consider the evolution of global supply chains

In this study, we assume the evolution of global supply chains to follow the SSP2-NDC scenario, where SSP2 refers to a “Middle-of-the-Road” shared socio-economic pathway and NDC to a policy implementation corresponding to a mean average surface temperature increase of 2.0-2.5°C by 2100 under this socio-economic pathway. Here, we demonstrate the sensitivity of our results to the SSP2-PkBudg650 scenario, representing a highly stringent mitigation pathway compliant with 1.5°C target, and the SSP2-NPi scenario, representing a low-ambition baseline that assumes only the continuation of currently implemented national policies, leading to a temperature increase of 3.0°C or more by 2100. All scenarios are based on the integrated assessment model REMIND 3.5.¹⁵

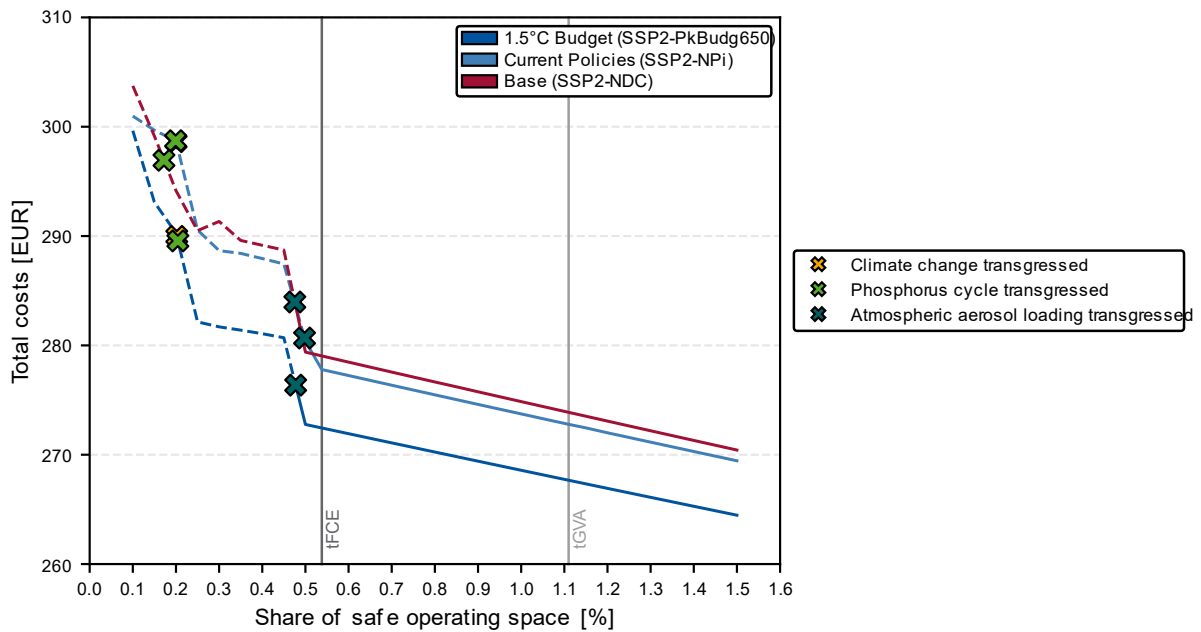


Figure S27: Sensitivity analysis for scenario used to consider the evolution of global supply chains.

The energy system designs that correspond to the SSP2-PkBudg650 and SSP2-NPi scenarios are shown in Section 2.12 and Section 2.13, respectively.

2.12. Energy system designs for the SSP2-PkBudg650 scenario

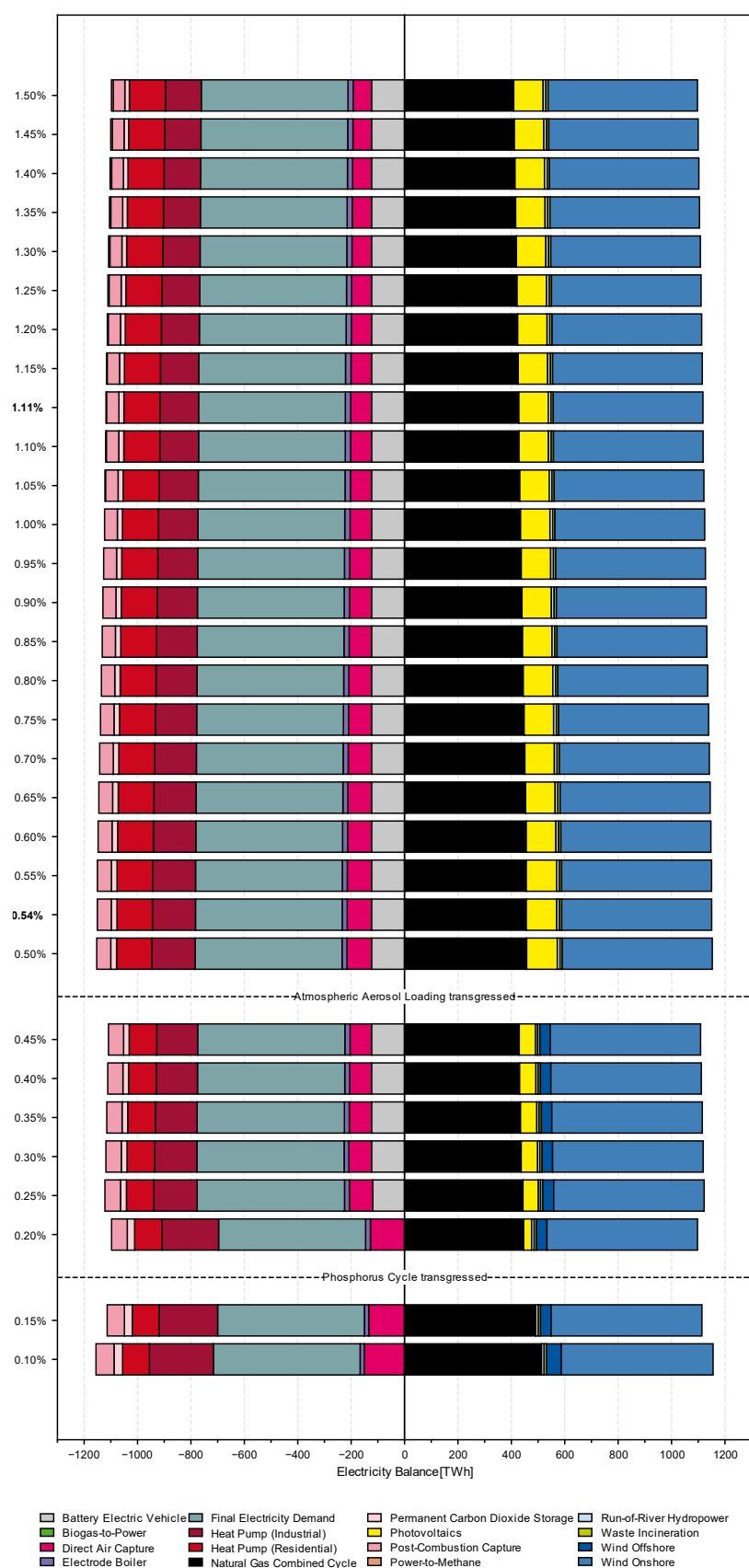


Figure S28: SSP2-PkBudg650 scenario: Electricity balance for a varying share of safe operating space with technology contribution to electricity generation and electricity consumption. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

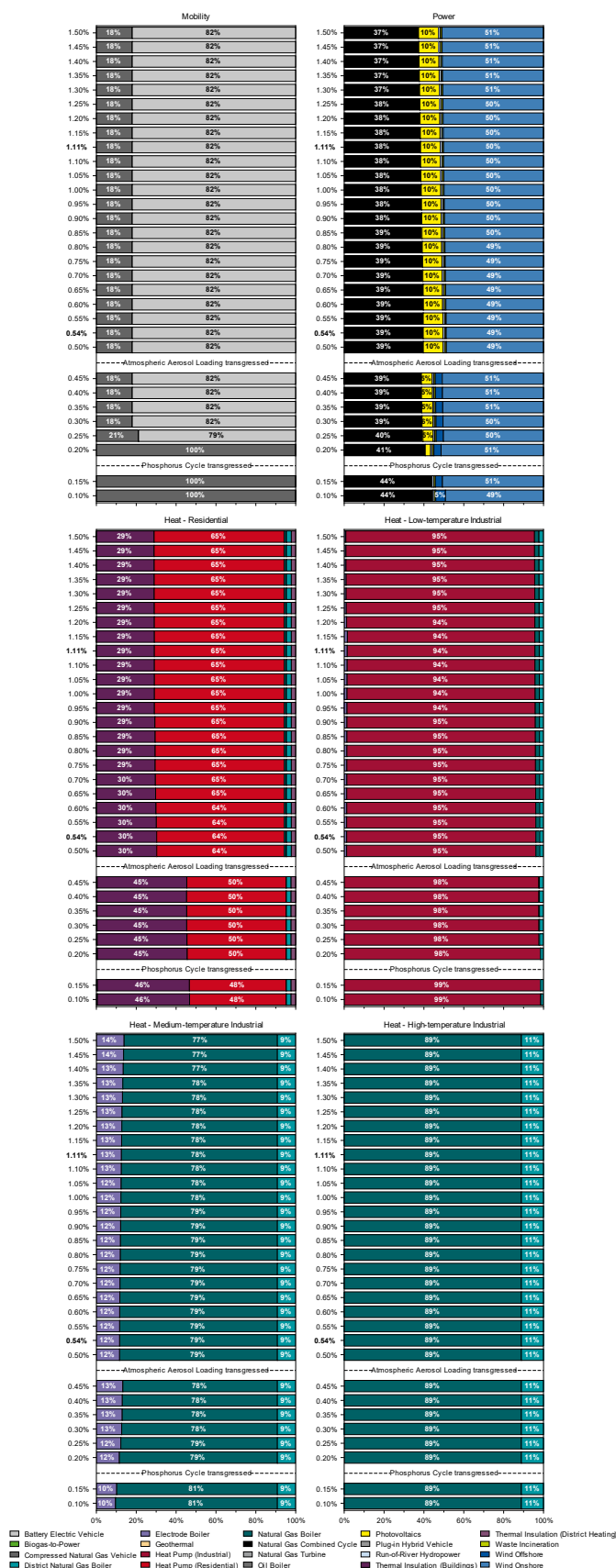


Figure S29: SSP2-PkBudg650 scenario: Contributions of technologies serving final energy demand of the mobility, power, residential and industrial heat (low-temperature, medium-temperature, high-temperature) sectors for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

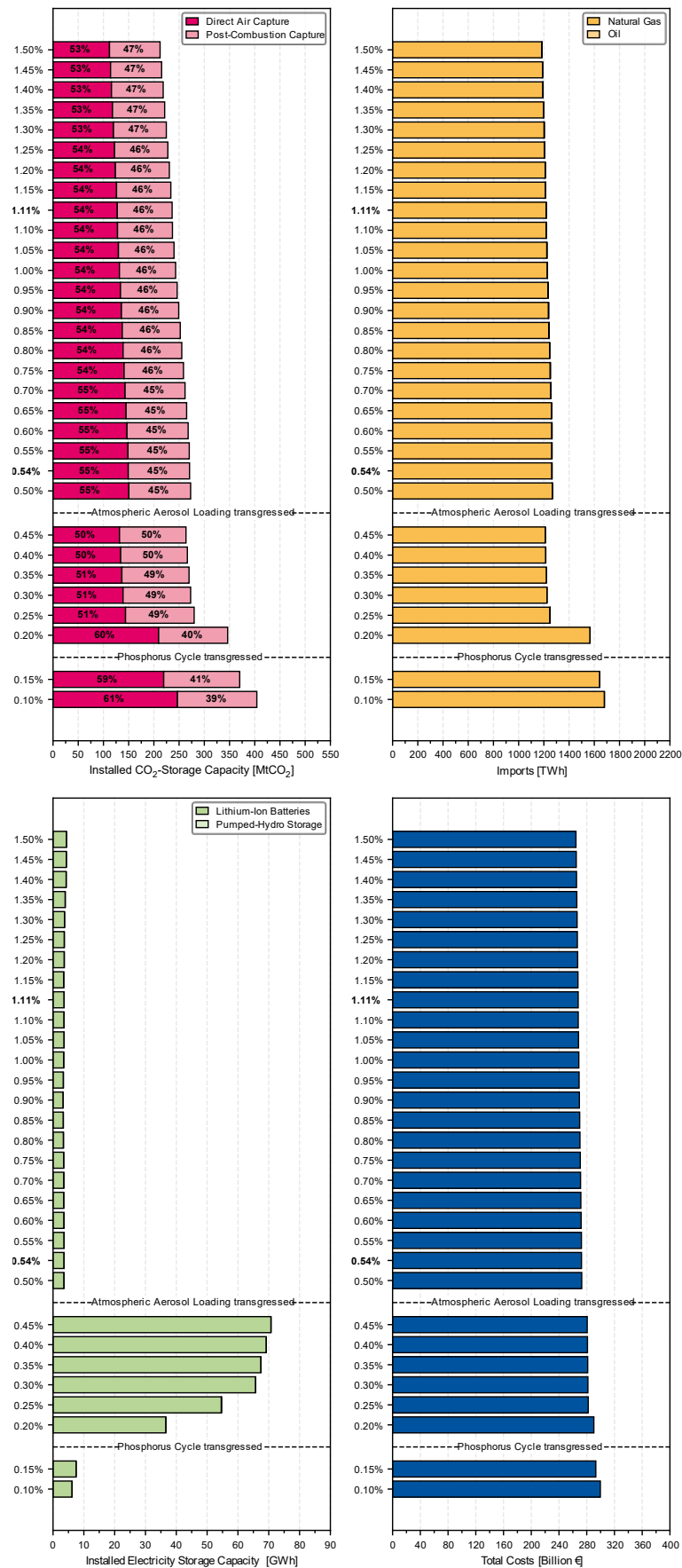


Figure S30: SSP2-PkBudg650 scenario: Capacities for CO₂ storage, energy imports and electricity storage with technology contributions, and total system costs for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

2.13. Energy system designs for the SSP2-NPi scenario

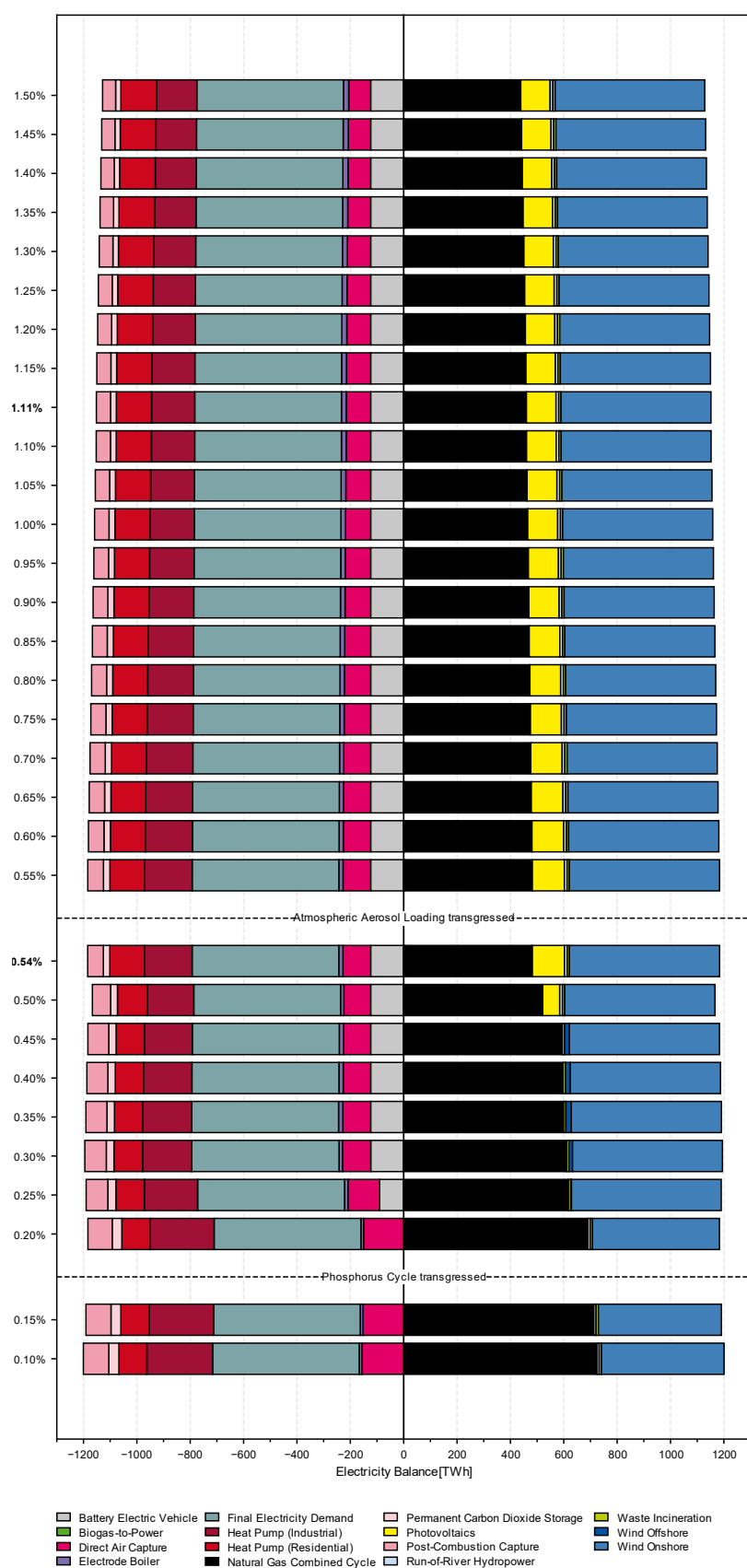


Figure S31: SSP2-NPi scenario: Electricity balance for a varying share of safe operating space with technology contribution to electricity generation and electricity consumption. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

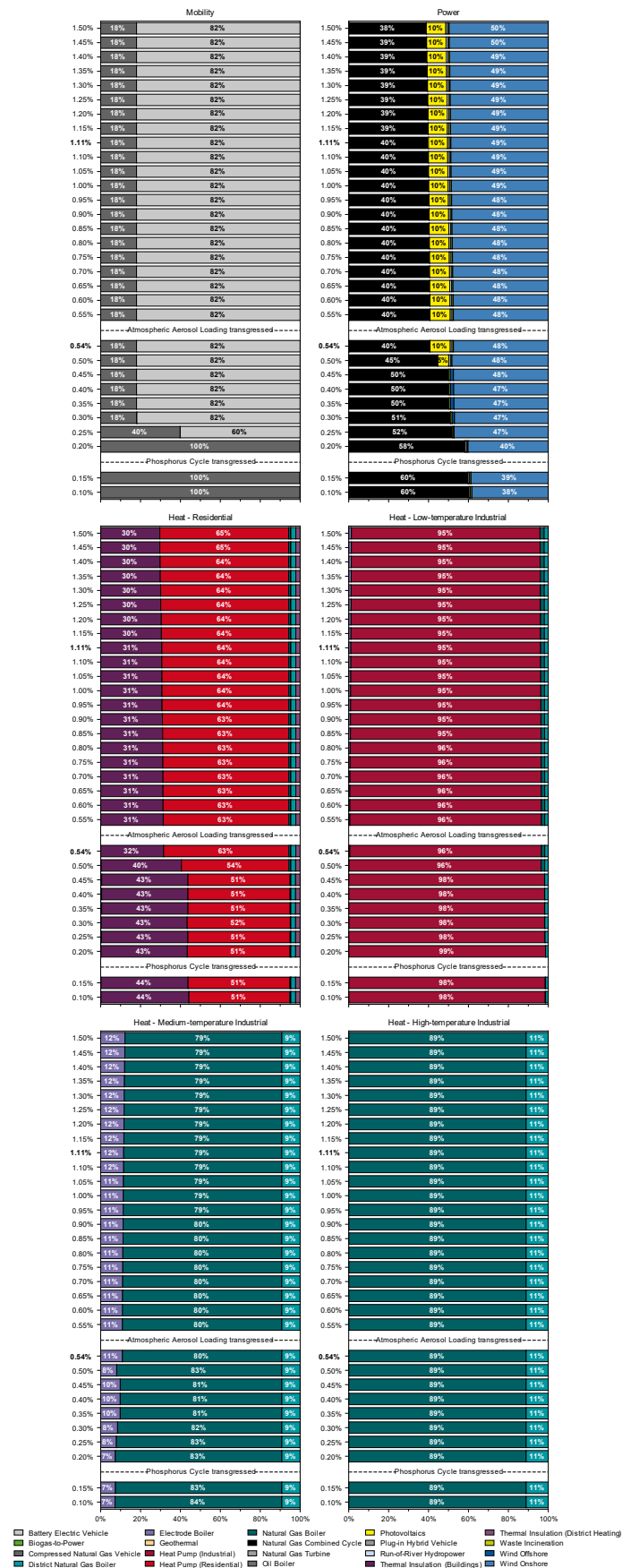


Figure S32: SSP2-NPi scenario: Contributions of technologies serving final energy demand of the mobility, power, residential and industrial heat (low-temperature, medium-temperature, high-temperature) sectors for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

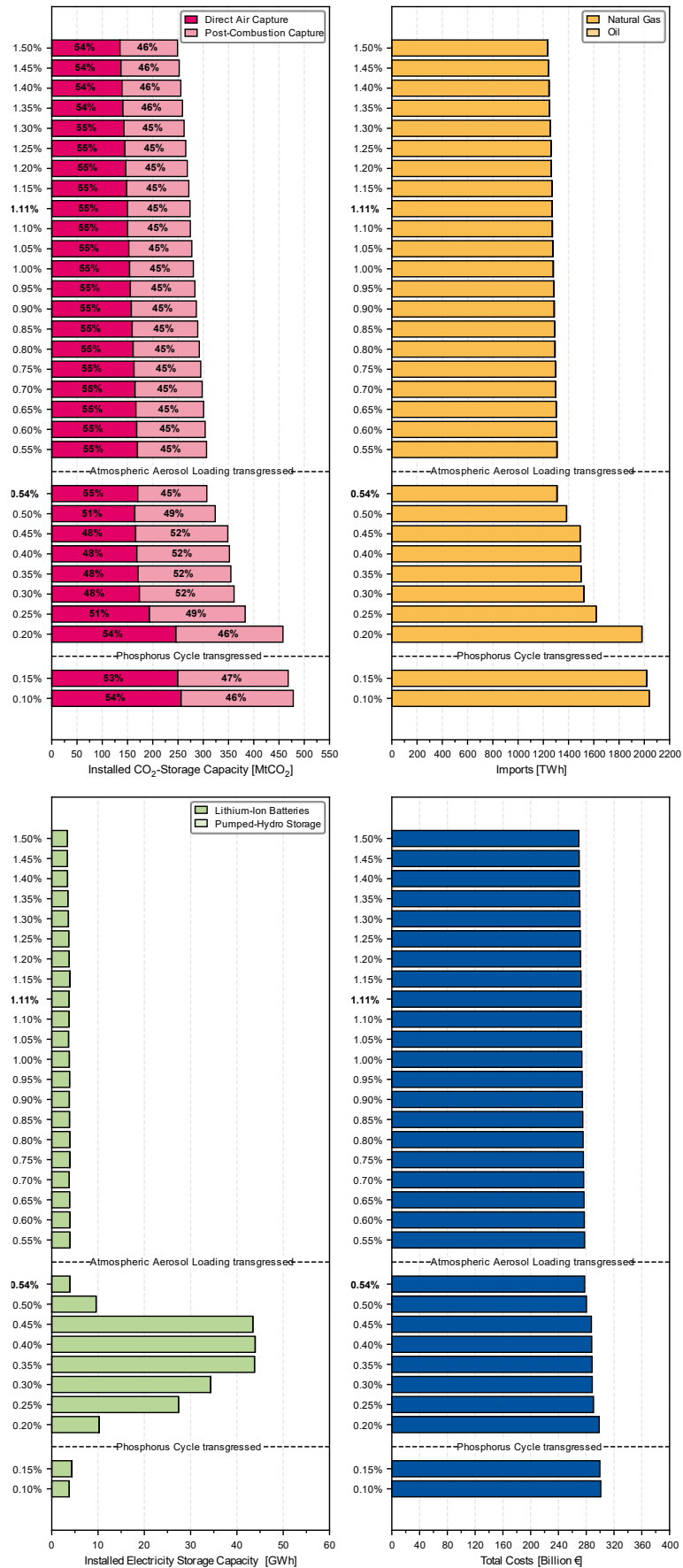


Figure S33: SSP2-NPi scenario: Capacities for CO₂ storage, energy imports and electricity storage with technology contributions, and total system costs for a varying share of safe operating space. Results for a share of safe operating space based on total final consumption expenditure (0.54 %) and total gross value added (1.11 %) are marked in bold.

2.14. Sensitivity analysis for CO₂ storage capacity constraint

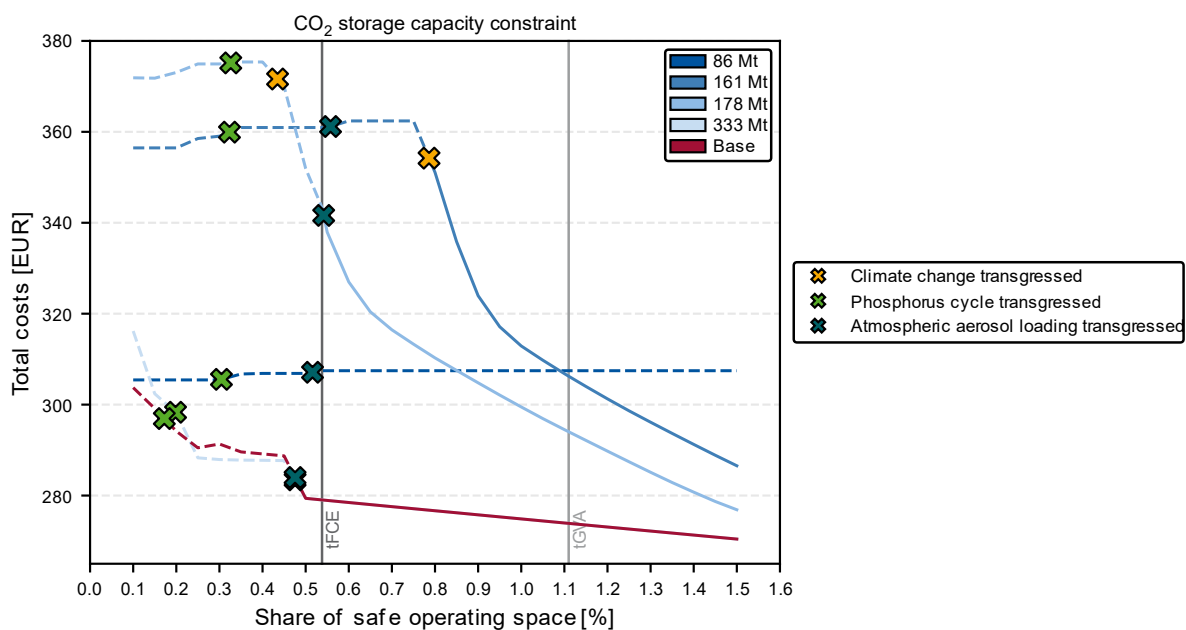


Figure S34: Sensitivity analysis for CO₂ storage capacity constraints based on applying the allocation factor based on total final consumption expenditure to global CO₂ storage capacity projections from the IPCC¹⁶ (161 Mt CO₂ annually) and Zhang et al.¹⁷ (86 Mt CO₂ annually) and the allocation factor based on total gross value added to global CO₂ storage capacity projections from the IPCC (333 Mt CO₂ annually) and Zhang et al. (178 Mt CO₂ annually).

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