Process Systems Engineering for sustainable chemicals within planetary boundaries

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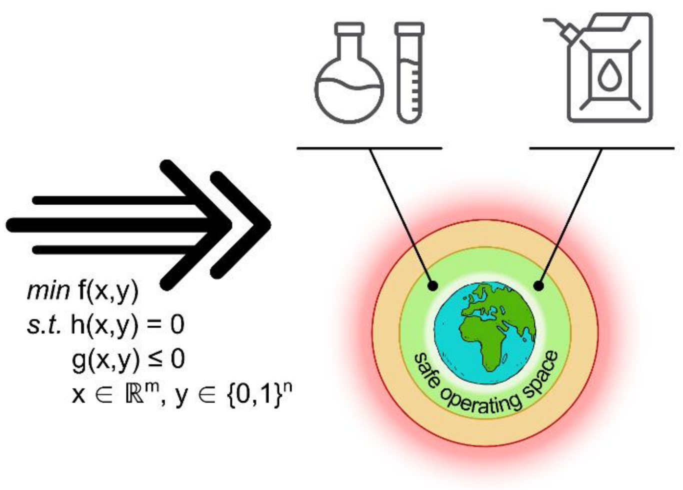
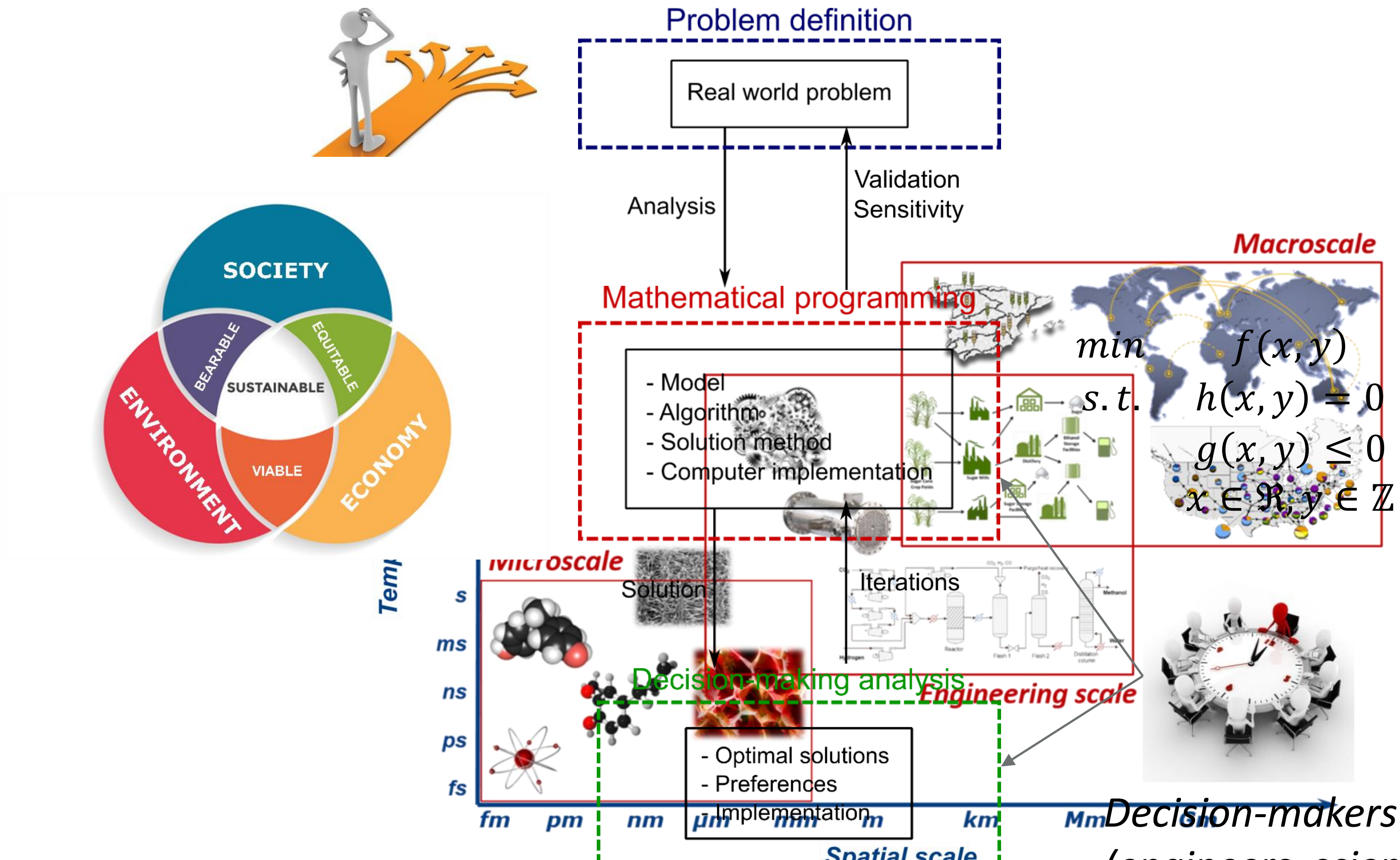


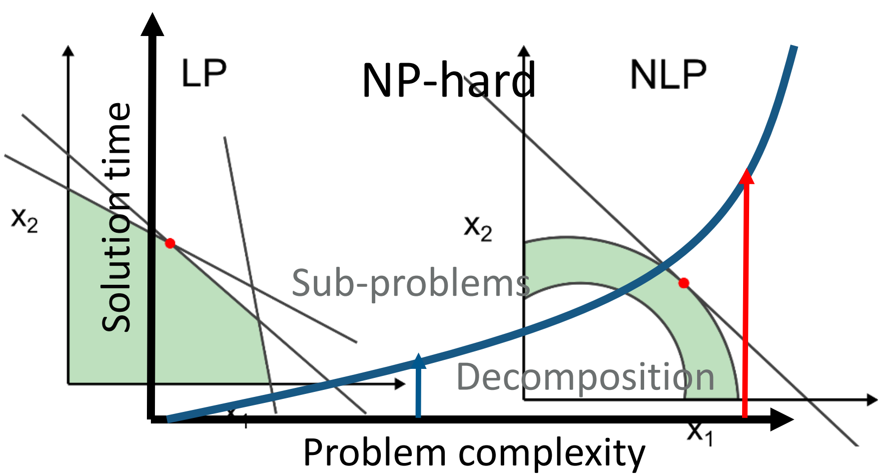
Sustainable decision-making process

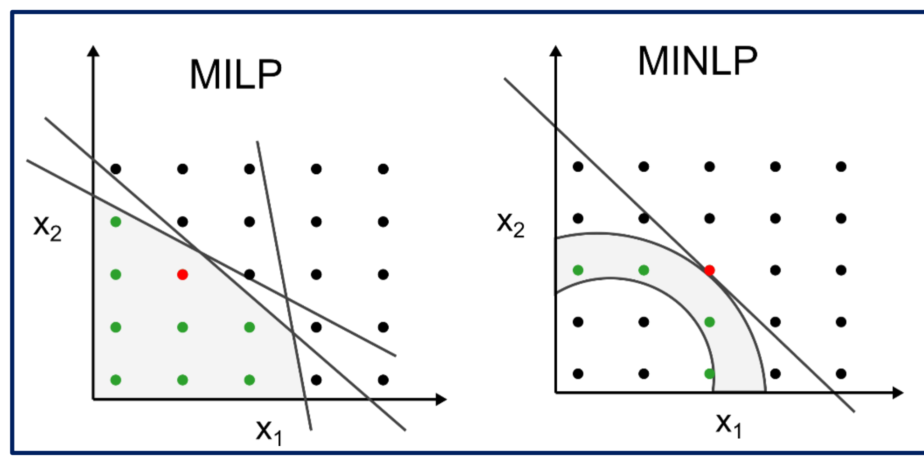


Decision-makers (engineers, scientists)

Mathematical programming

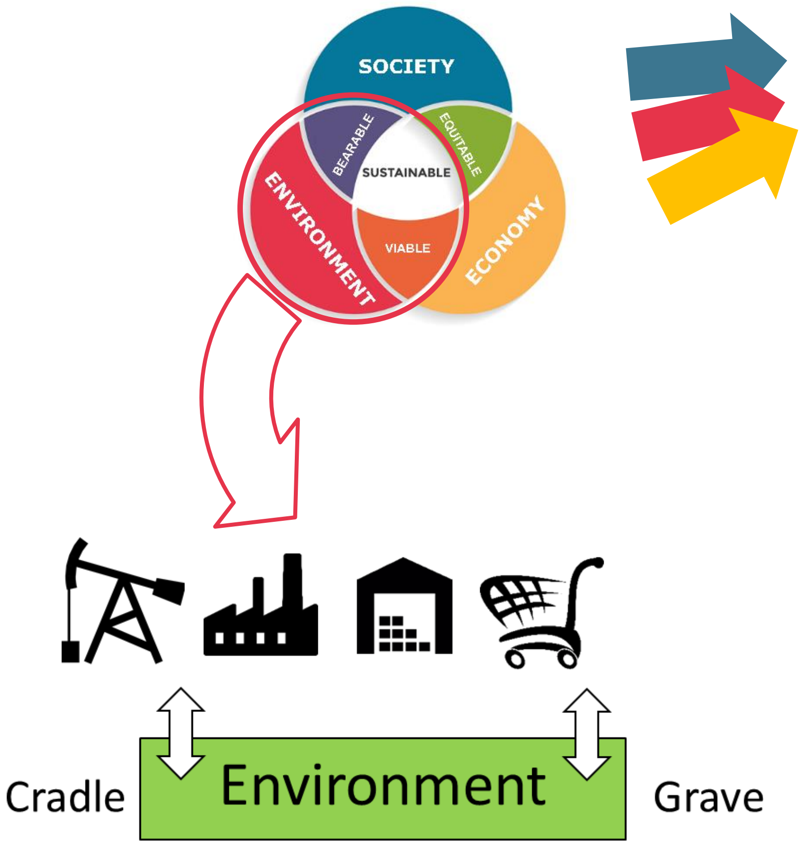
min	$f(x,y)$	**Objective function** (e.g., cost, environmental impact, safety)
$s.t.$	$h(x,y) = 0$	**Process equations** (e.g., mass & energy balances)
	$g(x,y) \leq 0$	**Specifications**
	$x \in \Re^n$	**Continuous variables** (e.g., pressures, temperatures, flows)
	$y \in \{0,1\}^m$	**Discrete variables** (logic decisions)





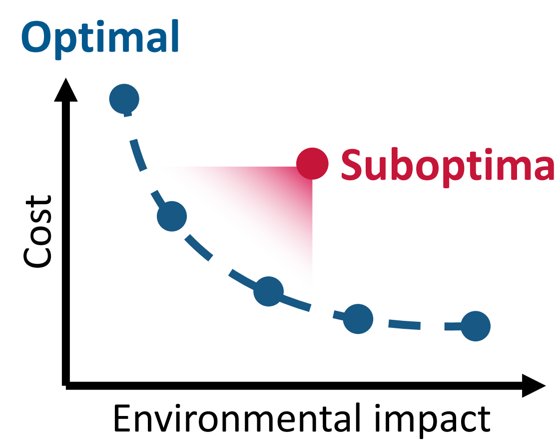
Multi-objective optimization

1. Several **objective functions**



$$
\begin{aligned}
\boxed{\min \quad f_1(x,y), \ldots, f_k(x,y)} \\
s.t. \qquad h(x,y) = 0 \\
g(x,y) \leq 0 \\
x \in \Re^n, y \in \{0,1\}^m
\end{aligned}
$$

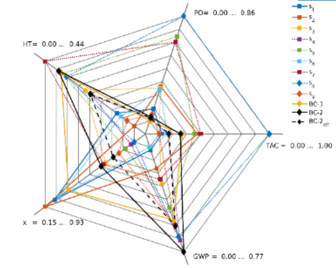
2. **Life cycle** sustainability assessment



3. **Pareto optimal** solutions

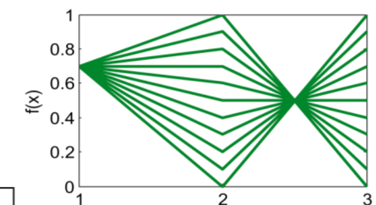
A. Azapagic, R. Clift, *Computers & Chemical Engineering* **23**, 1509-1526 (1999).
I. E. Grossmann, G. Guillén-Gosálbez, *Computers & Chemical Engineering* **34**, 1365-1376 (2010).

Complexity and solution algorithms

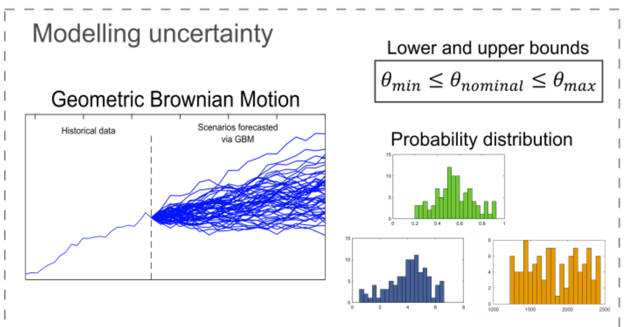


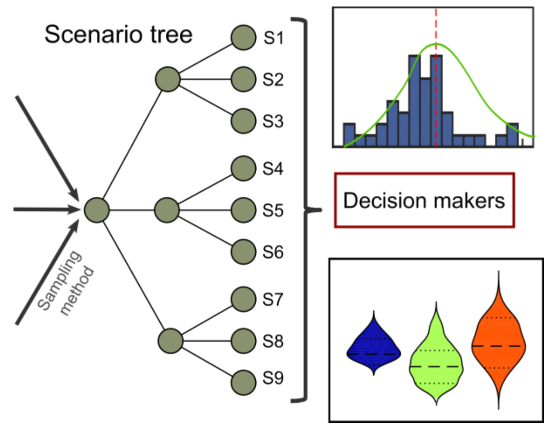
1. How to reduce the **number of objectives**?

$$\min \; f_1(x,y), f_2(x,y), f_3(x,y), f_4(x,y), f_5(x,y), f_6(x,y) \;\Rightarrow\; \min \; f_2(x,y), f_4(x,y)$$

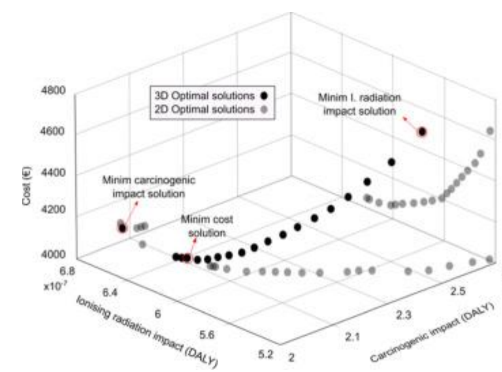


2. How to deal with **uncertainties**?

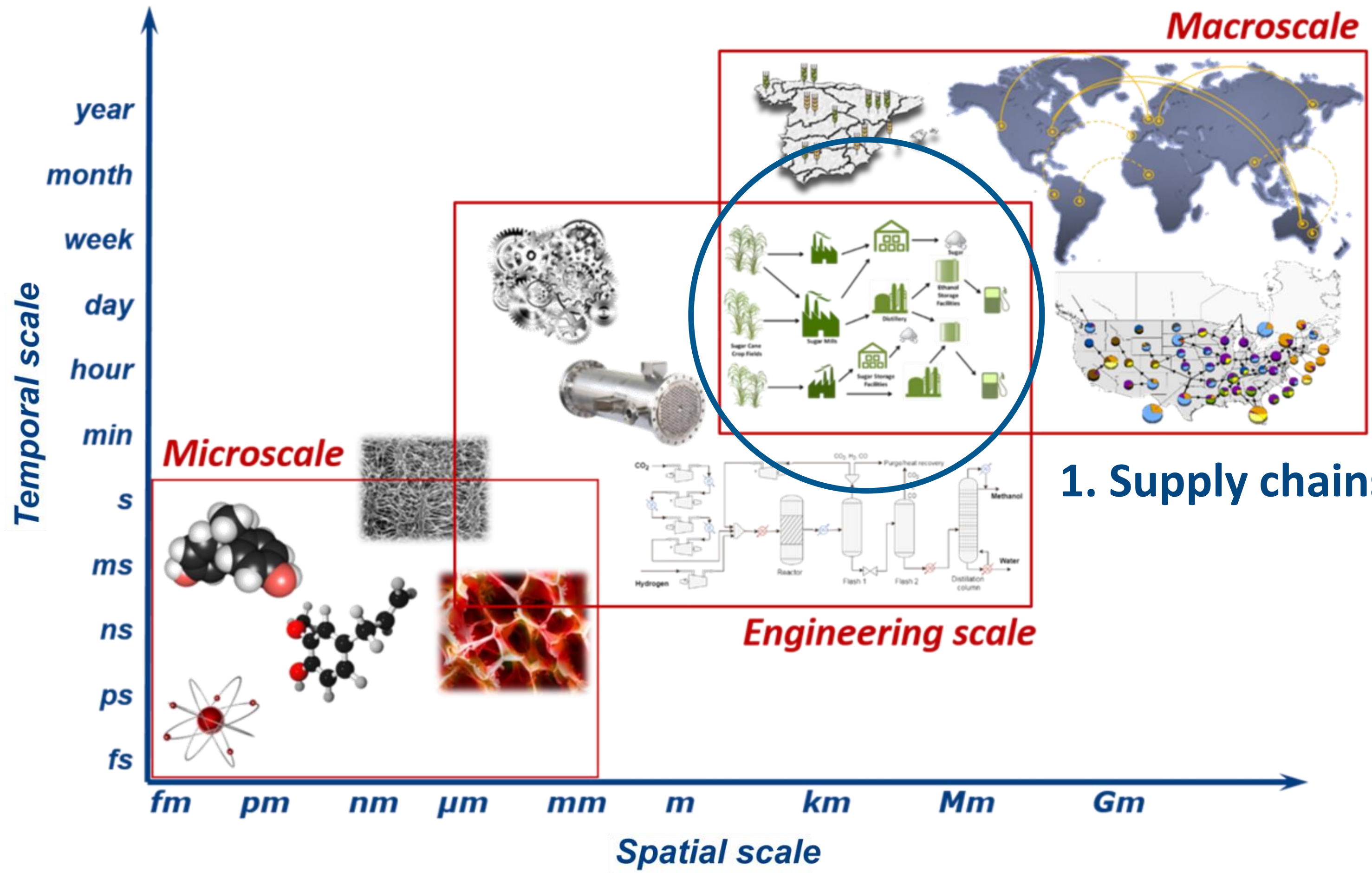




3. How to select **Pareto solutions**?

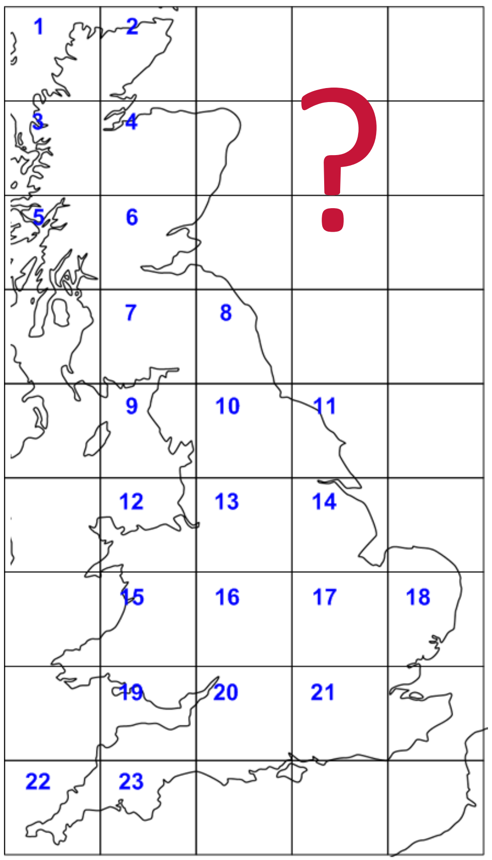


1. G. Guillén-Gosálbez, *Computers & Chemical Engineering* **35**, 1469-1477 (2011).
2. G. Guillén-Gosálbez, I. E. Grossmann, *AIChE Journal* **55**, 99-121 (2009).
3. P. Limleamthong, G. Guillén-Gosálbez, *Journal of Cleaner Production* **164**, 1602-1613 (2017).



1. Supply chains

Supply chains: Hydrogen Supply Chains



Given are:

- Hydrogen demand
- Available technologies and potential locations (i.e. grids)
- Investment and operating costs
- GHG emissions

The task is to determine:

- The optimal SC in terms of cost & environmental impact

Production

- CH_4 reforming
- Coal gasification
- Biomass gasification

Transportation

- Liquid H_2
- Compressed-gaseous H_2

Storage

- Liquid H_2
- Compressed gas

G. Guillén-Gosálbez, F. Mele, I. E. Grossmann, *AIChE Journal* **56**, 650-667 (2010).
N. Sabio *et al.*, *International Journal of Hydrogen Energy* **37**, 5385-5405 (2012).

$$\min_{x,X,N}\left(TDC(x,X,N), DAM(x,X,N)\right)$$
$$s.t. \quad g(x,X,N) \le 0$$
$$h(x,X,N) = 0$$
$$x \in \mathbb{R}, X \in \{0,1\}, N \in \mathbb{N}$$

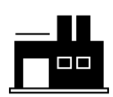
Multi-objective MILP

Mass balance equations
Capacity constraints
Objective function equations

1. Emissions and feedstock requirements: **LCI** from cradle to grave

$$LCI_b = \sum_i \sum_g \sum_p \sum_t PR_{igpt}\left(\omega_{bp}^{PR} + \omega_{bi}^{ST}\right) + \sum_i \sum_g \sum_{g' \neq g} \sum_{l \in LI_i} \sum_t Q_{igg'lt}\,\omega_b^{TR} \quad \forall b$$

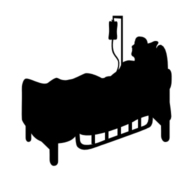
Production (raw materials, energy consumption and direct emissions)



Storage (compression of hydrogen)

Transportation tasks

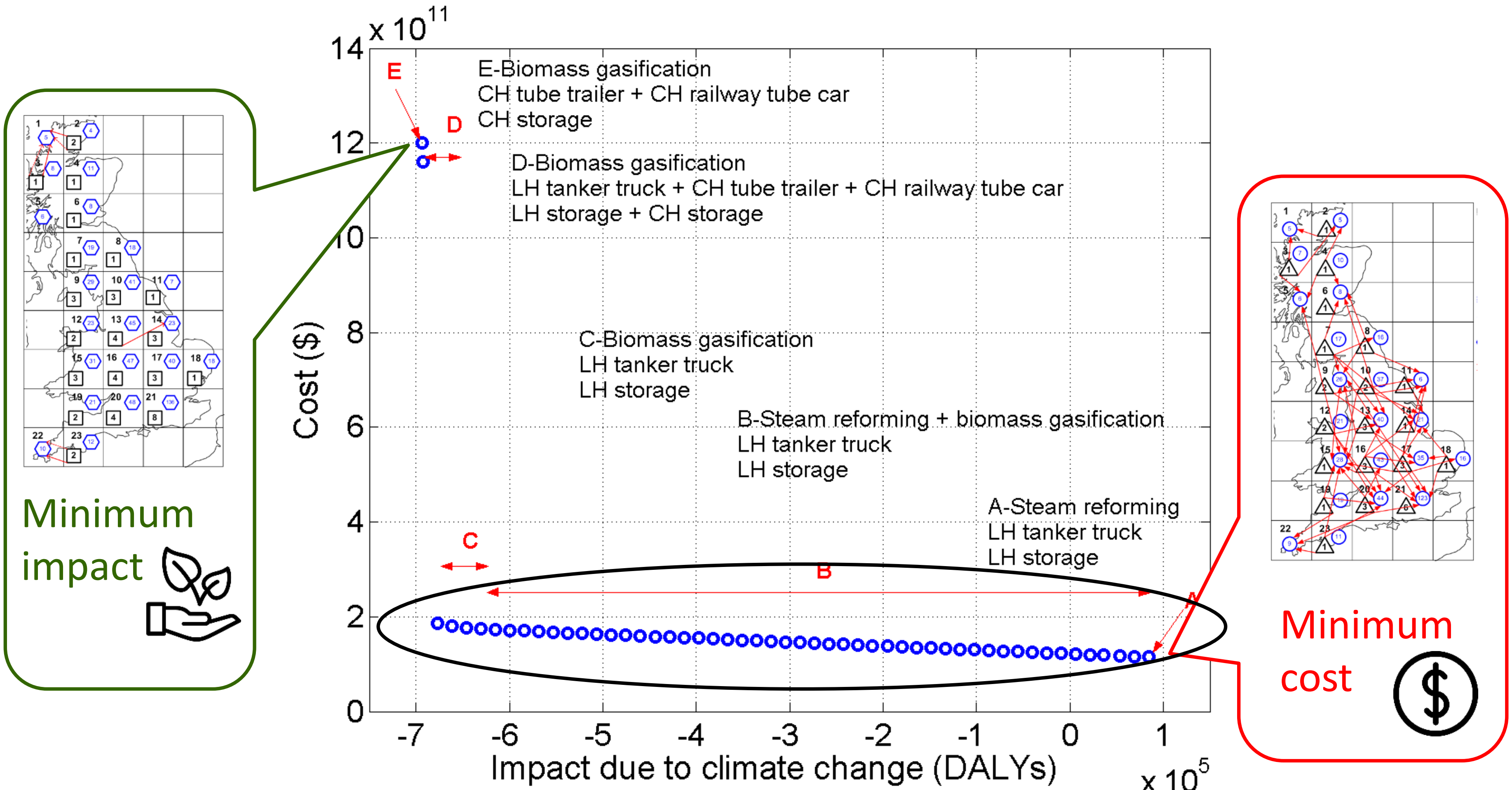
2. Translate LCI into damage (CML 2001, Recipe 2008, etc.): **Eco-indicator 99**



$$DAM = \sum_b \upsilon_b LCI_b$$

Damage factors translate life cycle inventory into impact

Binary 10,580
Discrete 940
Continuous 12,701
Equations 35,491
Bi-level decomposition + B&B (CPLEX)



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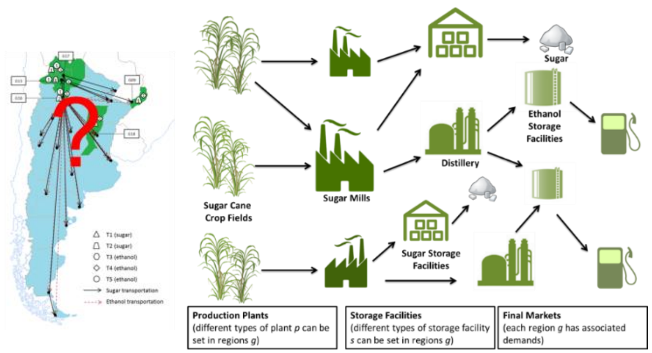
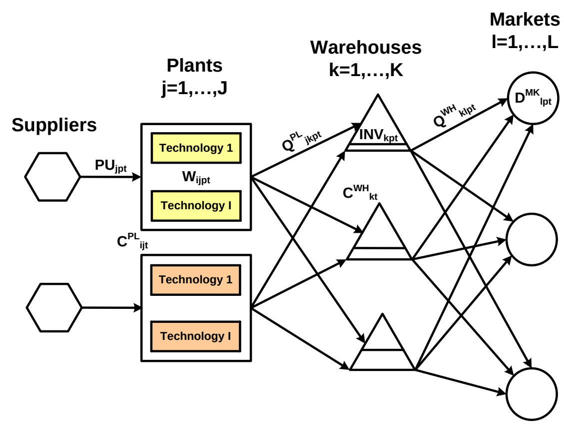
Other applications

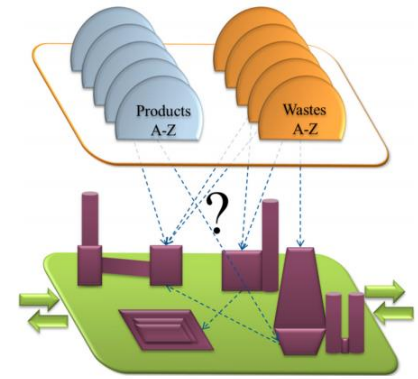
Biomass supply chains[1]



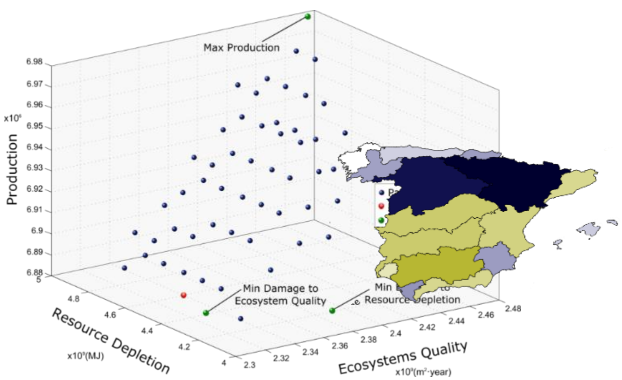
Petrochemicals[2]



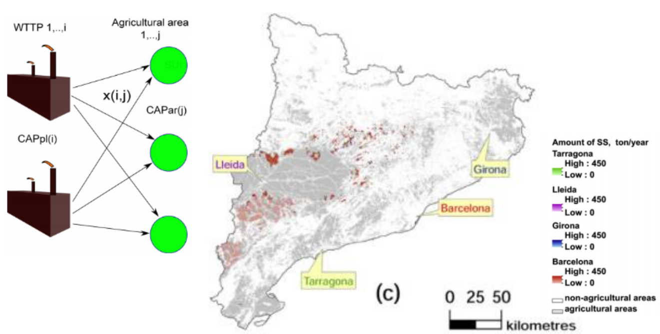
Waste management[3]



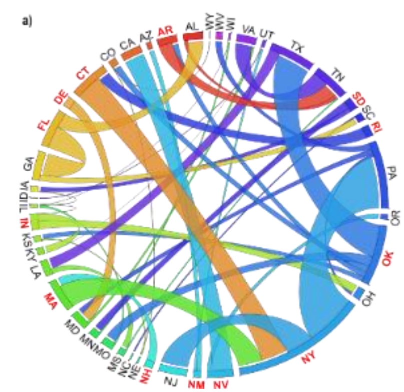
Agriculture planning[4]



Geographical Information Systems[5]



Input-output analysis[6]



1. F. Mele *et al.*, *Industrial & Engineering Chemistry Research* **50**, 4939-4958 (2011).
2. G. Guillén-Gosálbez, I. E. Grossmann, *AIChE Journal* **55**, 99-121 (2009).
3. C. Vadenbo, S. Hellweg, G. Guillén-Gosálbez, *Conservation and Recycling* **89**, 41-51 (2014).
4. A. Galán-Martín *et al.*, *Journal of Cleaner Production* **140**, 816-830 (2017).
5. P. Vaskan *et al.*, *Environmental Modelling & Software* **46**, 163-169 (2013).
6. D. Cortés-Borda *et al.*, *Energy Policy* **77**, 21-30 (2015).

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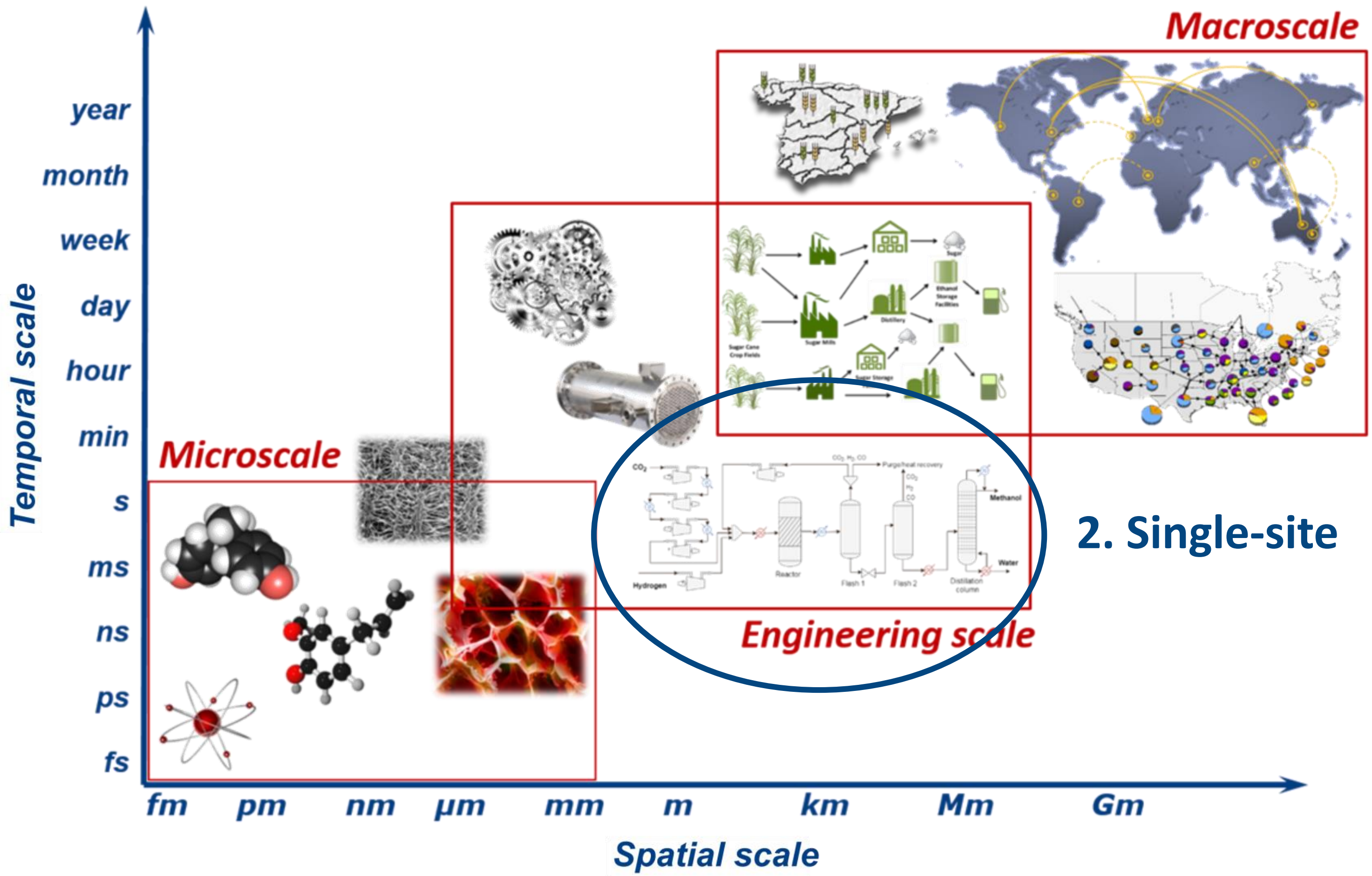
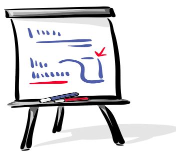


2. Single-site

Hydro-dealkylation of toluene (HDA) process



Given are:

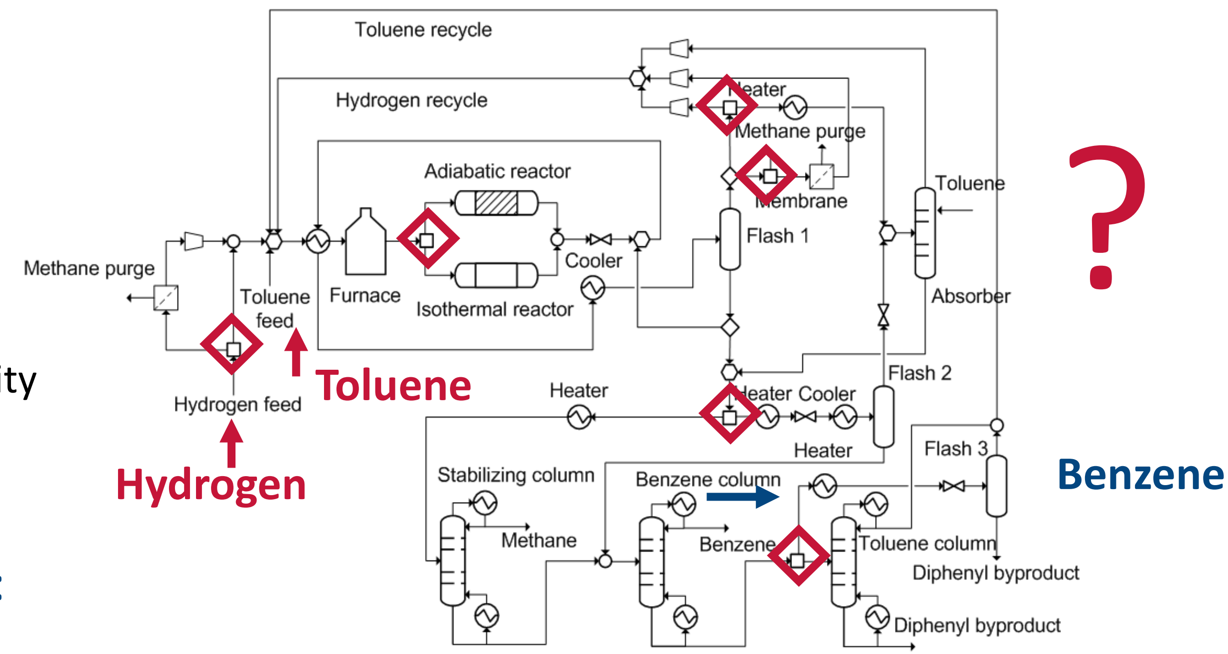
- Product demands and quality
- Cost data
- Environmental LCA data

The task is to determine:

- Topology
- Operating conditions

So as to:

- Minimize the total cost
- Minimize the environmental LCA impact



□ Single choice stream splitter

◇ Multiple choice stream splitter

○ Single choice stream mixer

⬡ Multiple choice stream mixer

$$\text{toluene} + \text{hydrogen} \rightarrow \text{benzene} + \text{methane}$$

$$2\text{benzene} \leftrightarrow \text{diphenyl} + \text{hydrogen}$$

G. Guillén-Gosálbez, J. A. Caballero, L. Jiménez, *Industrial & Engineering Chemistry Research* **47**, 777-789 (2008).

Superstructure optimization using MINLP: HDA process

$$\begin{aligned} min \quad & (TC(x,y), ECO_{99}(x,y)) \\ s.t. \quad & h(x,y) = 0 \\ & g(x,y) \leq 0 \\ & x \in \Re^n, y \in \{0,1\}^m \end{aligned}$$

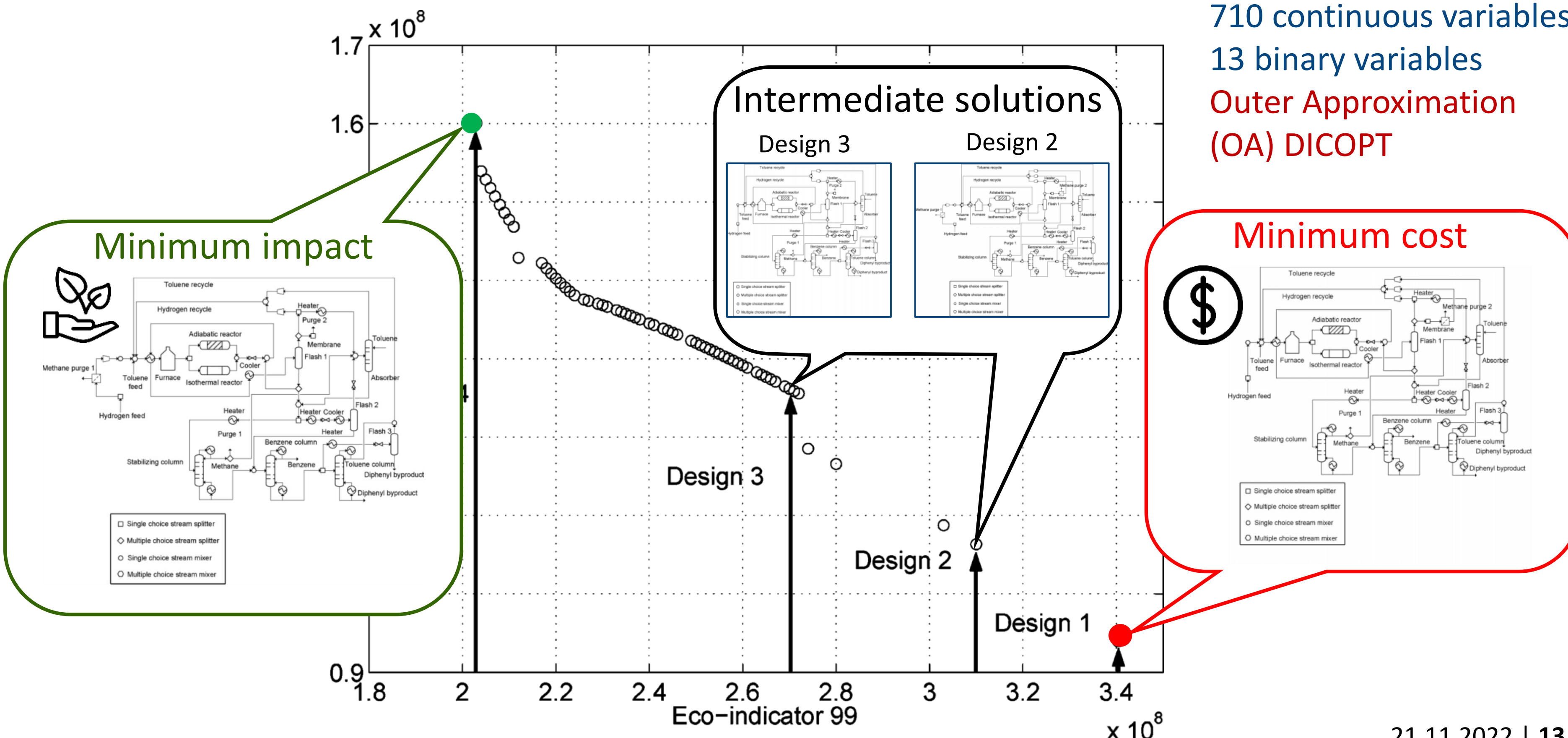
Multi-objective mixed-integer nonlinear program (MINLP)

Mass & energy balances constraints

Thermodynamic constraints

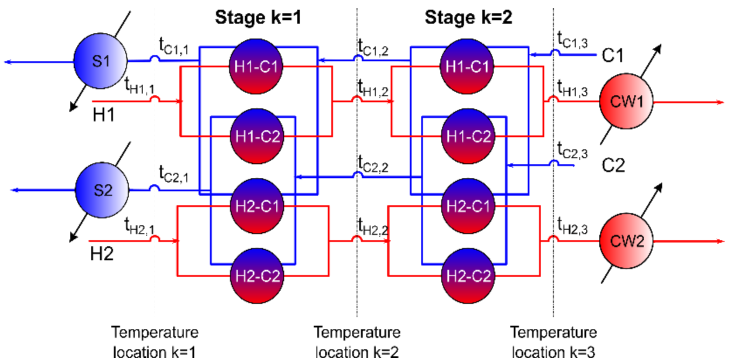
Short-cut methods **LCA**

MINLP
724 constraints
710 continuous variables
13 binary variables
Outer Approximation
(OA) DICOPT

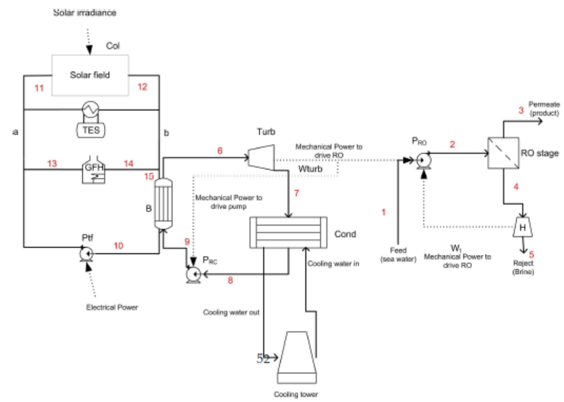


Other applications

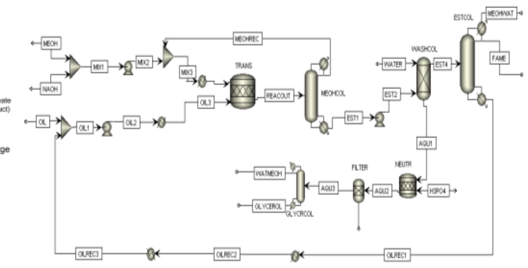
Heat exchangers[1]



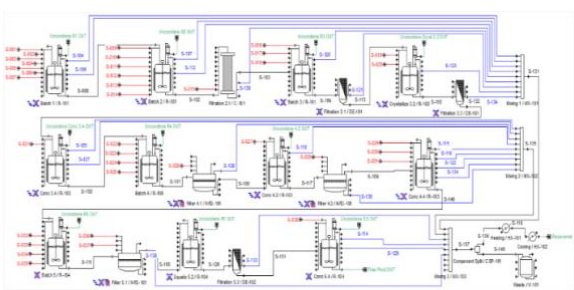
Thermodynamic cycles[2]



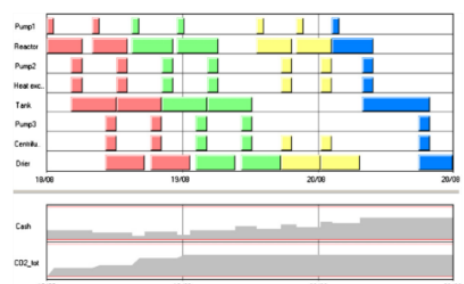
Biofuels[3]



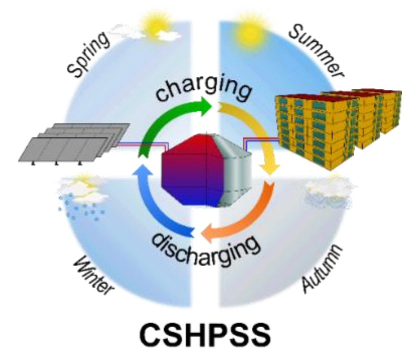
Pharmaceutical processes[4]



Scheduling[5]



Buildings[6]



1. P. Vaskan, G. Guillén-Gosálbez, L. Jiménez, *Applied Energy* **98**, 149-161 (2012).
2. R. Salcedo *et al.*, *Desalination* **286**, 358-371 (2012).
3. R. Brunet *et al.*, *Chemical Engineering Research & Design* **93**, 203-212 (2015).
4. R. Brunet, G. Guillén-Gosálbez, L. Jiménez, *Journal of Cleaner Production* **76**, 55-63 (2014).
5. M. J. Arbiza *et al.*, *Journal of Cleaner Production* **16**, 233-244 (2008).
6. V. Tulus *et al.*, *Applied Energy* **181**, 549–561 (2016).

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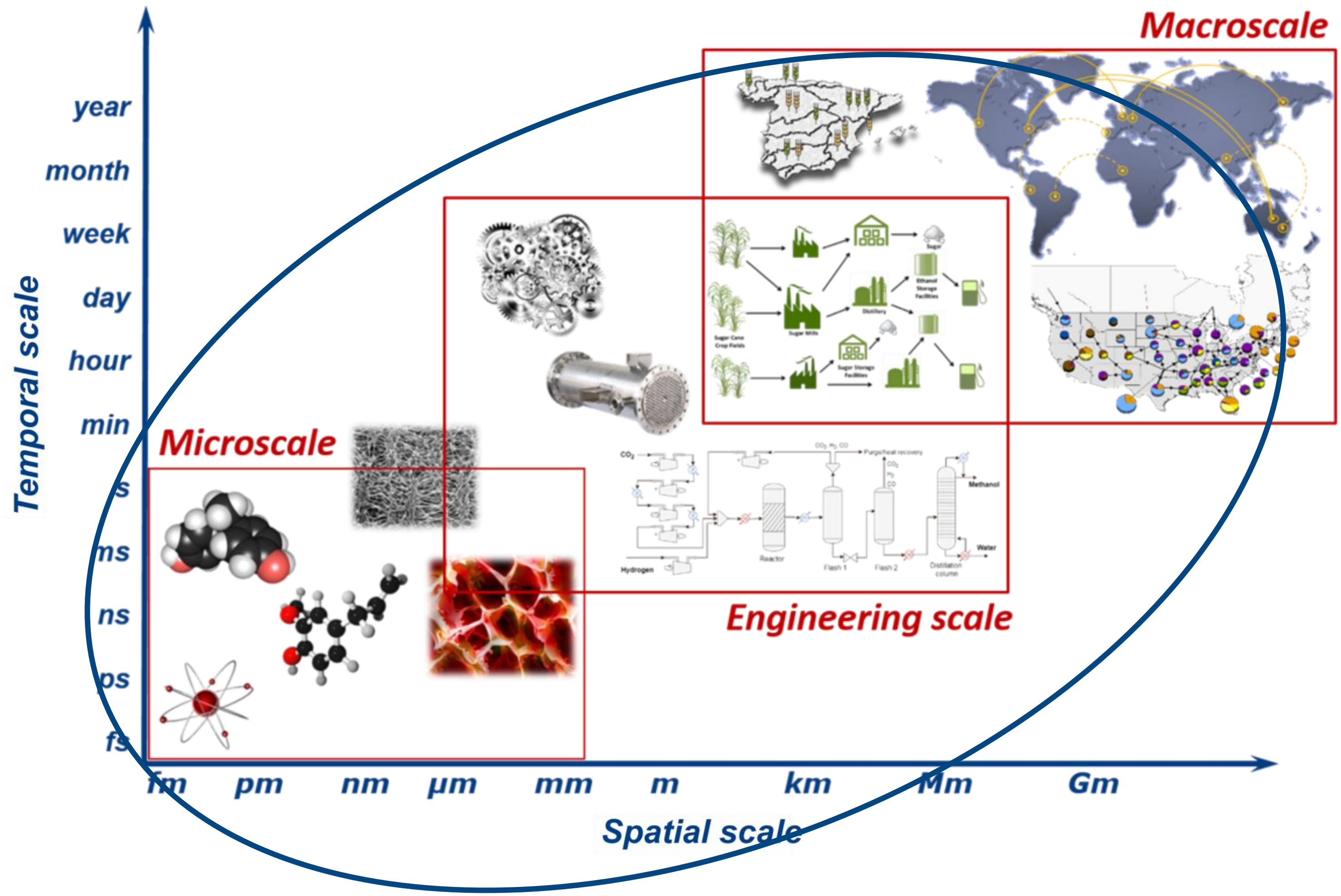
PSE tools for a more sustainable chemical industry

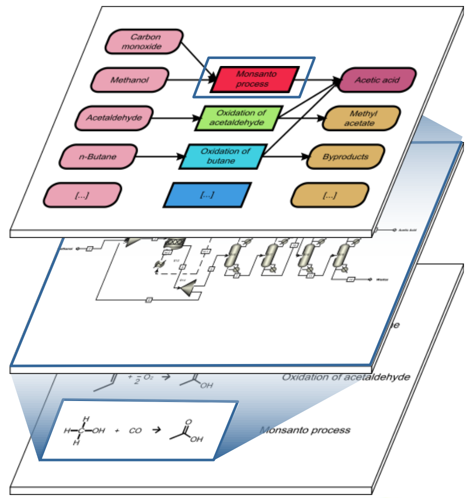


Impact quantification based on process modelling

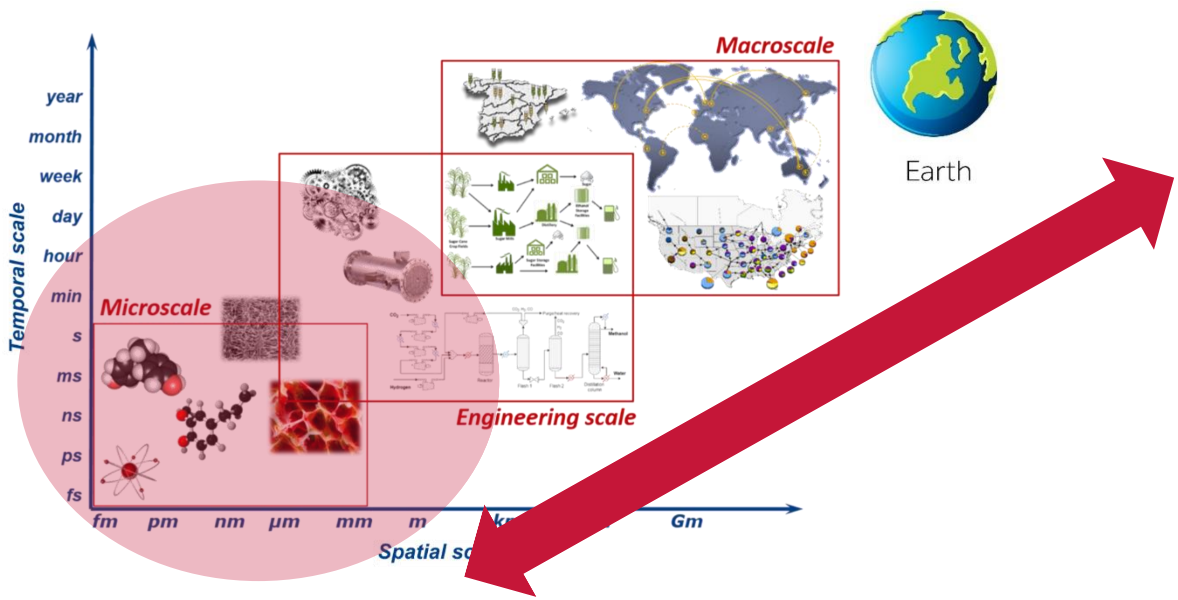
3. Sector level/LCA

2. Process flowsheets

1. Chemical routes

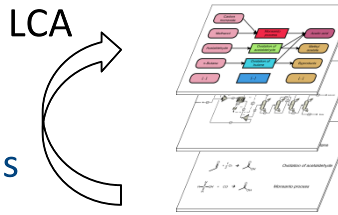


A proper assessment of chemical routes requires a **multi-scale approach**



Streamlined LCA methods for chemicals

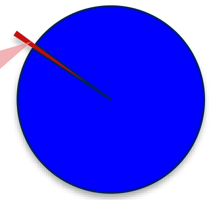
1 → 3 Streamlined LCA methods

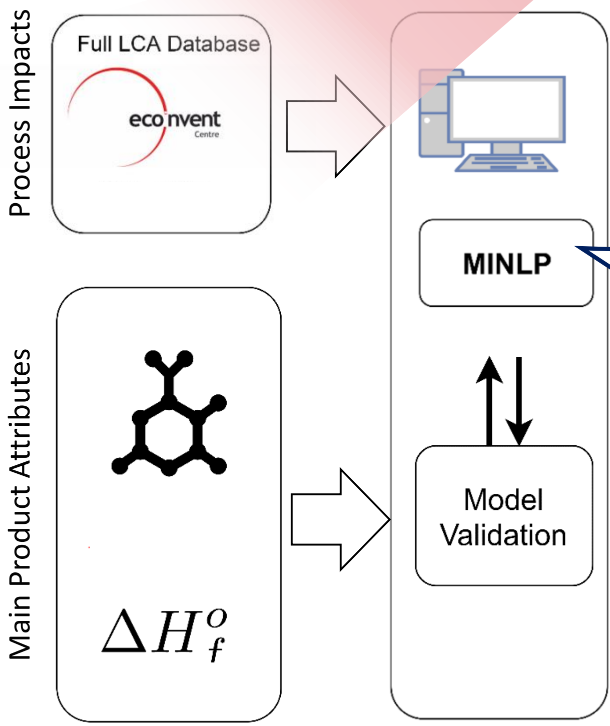


~1 Million registered chemicals

LCA data of only a few thousand chemicals

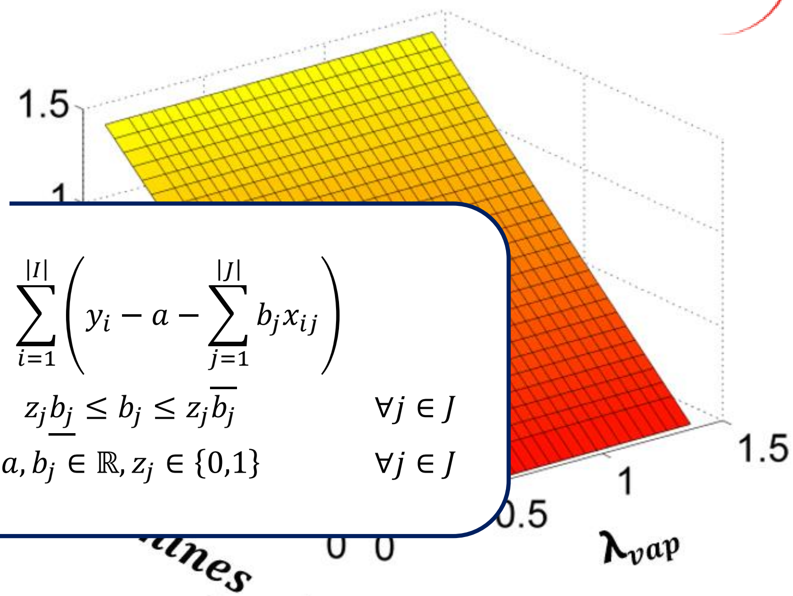
Automatic Data Regression + Feature Selection





$$\begin{aligned} &Min \quad \sum_{i=1}^{|I|}\left(y_i - a - \sum_{j=1}^{|J|} b_j x_{ij}\right) \\ &s.t. \quad z_j \underline{b_j} \le b_j \le z_j \overline{b_j} \qquad \forall j \in J \\ &\qquad a, b_j \in \mathbb{R}, z_j \in \{0,1\} \qquad \forall j \in J \end{aligned}$$

MIQCP
200 continuous
32 binary
315 equations
CPLEX

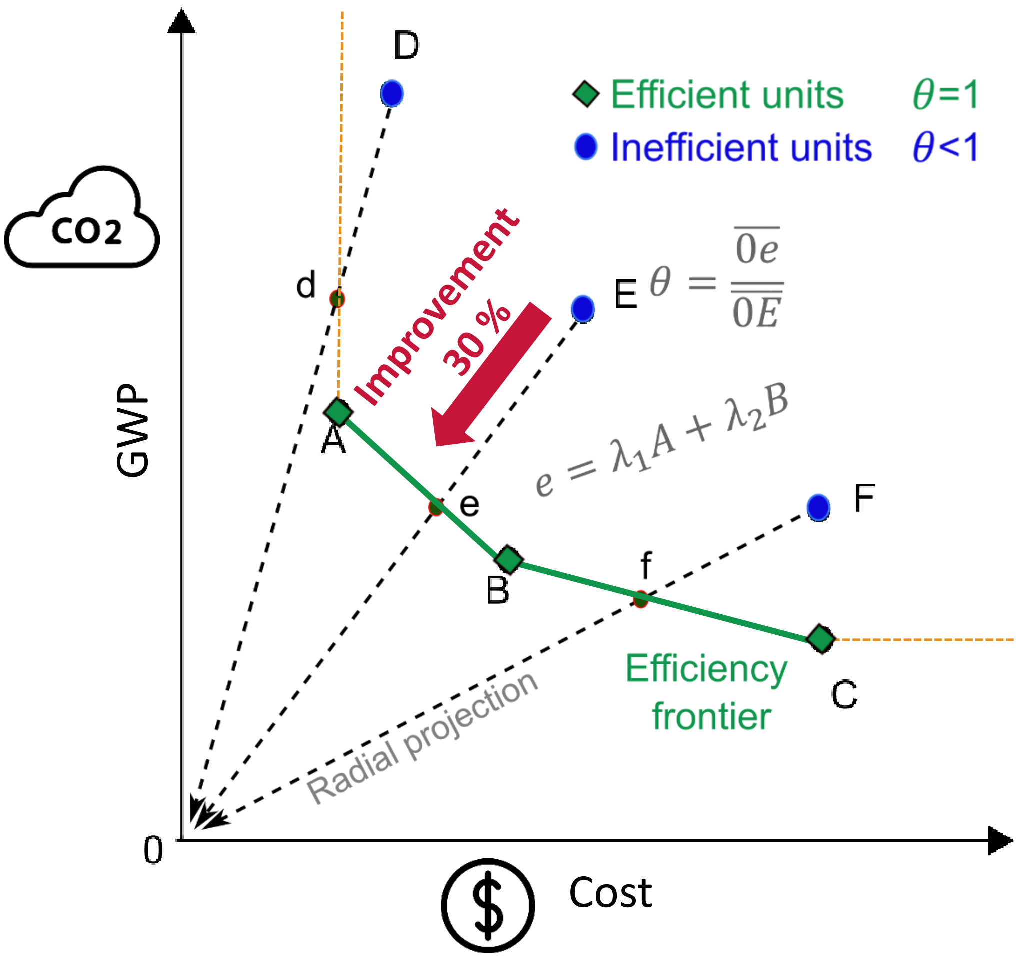


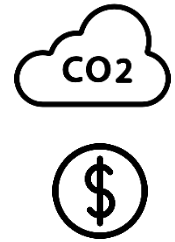
$$GWP = 75.2 + 0.786 \cdot amines + 3.1 \cdot 10^{-8} \cdot \lambda_{vap}$$

~30% error

G Wernet *et al.*, *Environmental Science & Technology* **42**, 6717-6722 (2008).
R. Calvo-Serrano *et al.*, *Computers & Chemical Engineering* **108**, 179-193 (2018).

Screening via data envelopment analysis





?

How to improve?

A

B

C

D

E

F

$$Z = min\, \theta_o - \varepsilon \left(\sum_{r \in R} S_r^+ + \sum_{i \in I} S_i^- \right)$$

$$s.t. \quad \sum_{j \in J} \lambda_j \mathrm{x_{ij}} + S_i^- = \theta_o \mathrm{x_{io}} \qquad \forall\, i \in I$$

$$\sum_{j \in J} \lambda_j \mathrm{y_{rj}} - S_r^+ = \mathrm{y_{ro}} \qquad \forall\, r \in R$$

$$\lambda_j, S_i^-, S_r^+ \geq 0 \qquad \forall j \in J, i \in I, r \in R$$

P. Limleamthong, G. Guillén-Gosálbez, *Industrial & Engineering Chemistry Research* **57**, 9866-9878 (2018).

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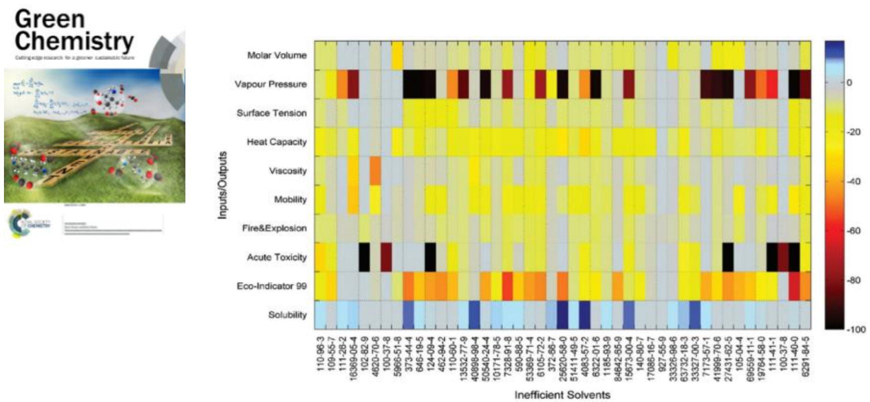
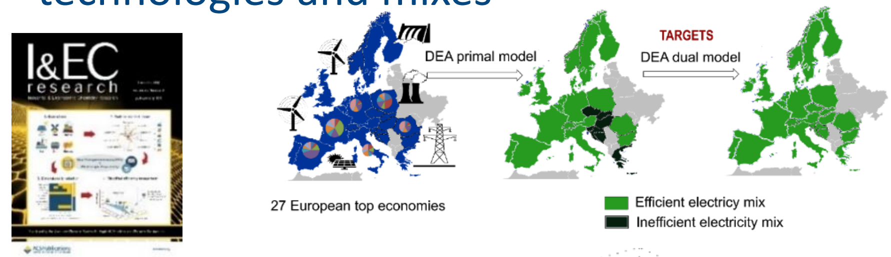
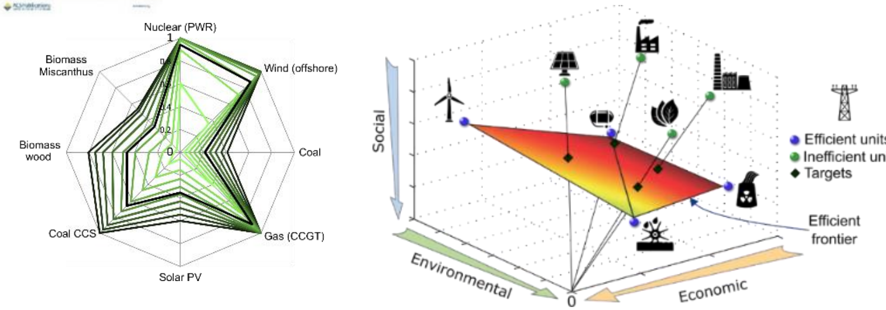
Screening of chemicals for CO_2 capture[1]

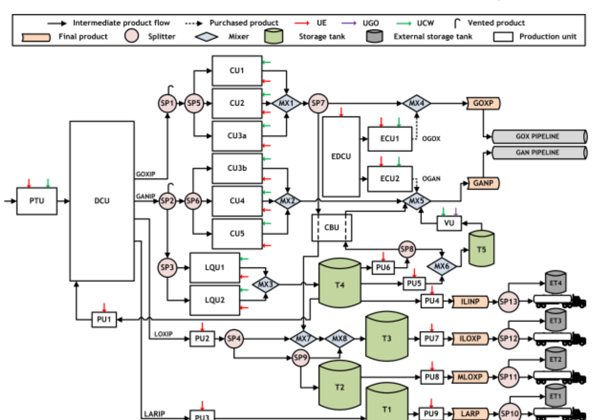


Eco-efficiency assessment of energy technologies and mixes[2]

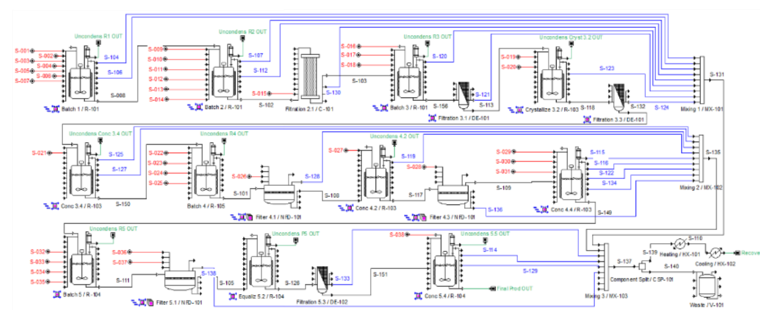




Efficiency assessment of industrial gases facilities[3]

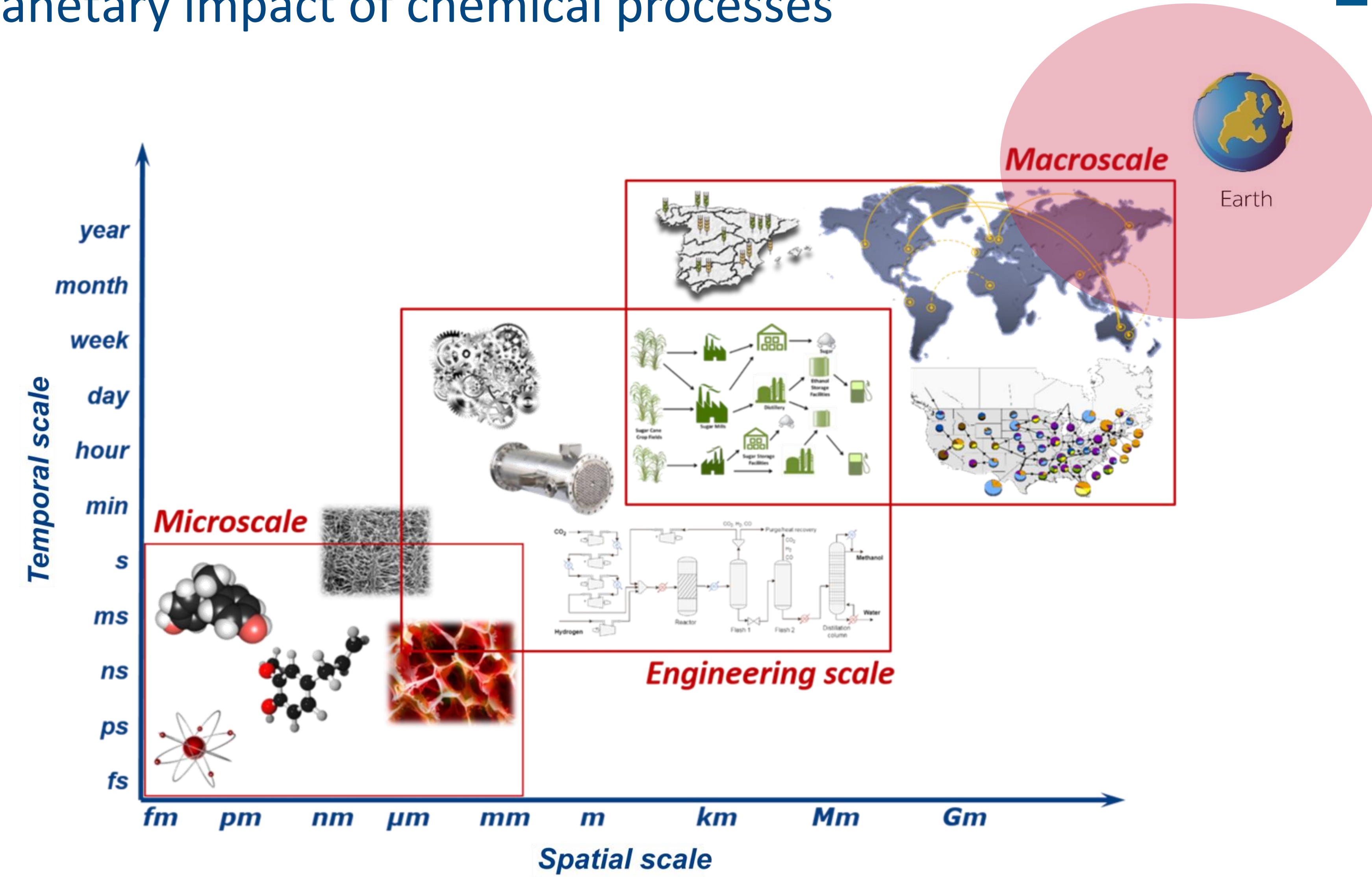


Eco-efficiency assessment of pharma processes[4]



1. P. Limleamthong *et al.*, *Green Chemistry* **18**, 6468-6481 (2016).
2. A. Ewertowska *et al.*, *Journal of Cleaner Production* **116**, 13-22 (2016).
3. D. Fernández *et al.*, *Applied Energy* **212**, 1563-1577 (2018).
4. A. Mio *et al.*, *Industrial & Engineering Chemistry Research* **57**, 7946-7960 (2018).

Planetary impact of chemical processes



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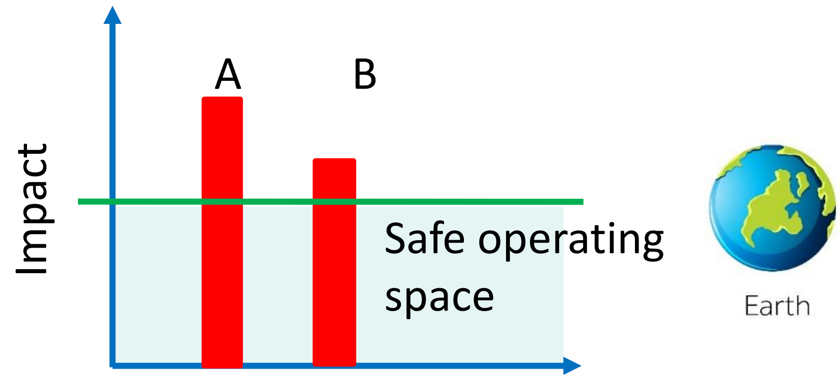
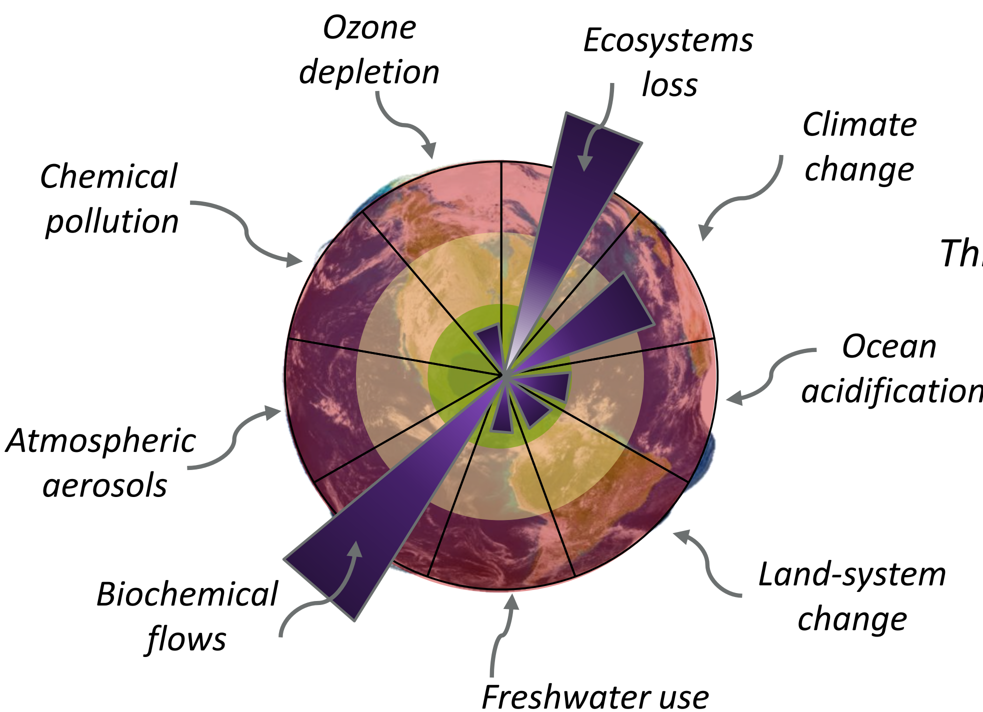
How to quantify the impact?

Current metrics hard to interpret



Planetary boundaries (PBs) defined on nine **Earth-system processes** key for resilience*



Three impact levels:

- *Safe op. space (SOS)*
- *Uncertain zone*
- *High-risk zone*

J. Rockström *et al., Nature* **461**, 472-475 (2009).

Computing the planetary impact of industrial systems

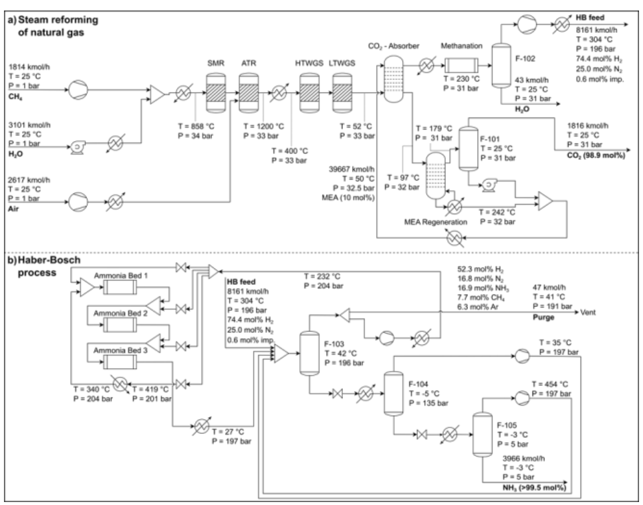
Evaluate the impact of a chemical process relative to the Earth's ecological limits:

1. Downscale the PBs to the industrial system

Process

$$\mathrm{SOS}_p \cdot \frac{\mathrm{GVA}^{Process}}{\mathrm{GVA}^{Total}} = \mathrm{SoSOS}_p \quad \forall p$$

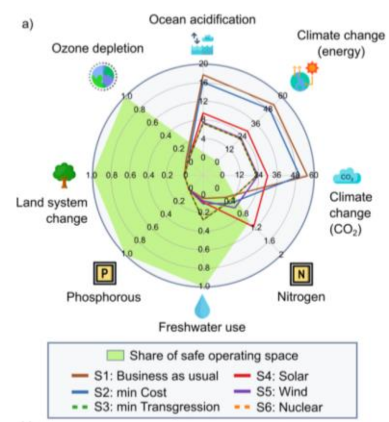
2. Link emissions and waste (life cycle inventory entries) to the associated planetary impact



$$\mathrm{IMP}_p = \sum_i \mathrm{CF}_{ip}\mathrm{LCI}_i \quad \forall p$$

$$\mathrm{Trangression_p} = \frac{\mathrm{IMP}_p}{\mathrm{SoSOS}_p}$$

$$\mathrm{Trangression_p} \begin{Bmatrix} < 1 \rightarrow \textit{Sustainable} \\ > 1 \rightarrow \textit{Unsustainable} \end{Bmatrix} \quad \forall p$$



M. W. Ryberg *et al.*, *Ecological Indicators* **88**, 250-262 (2018).

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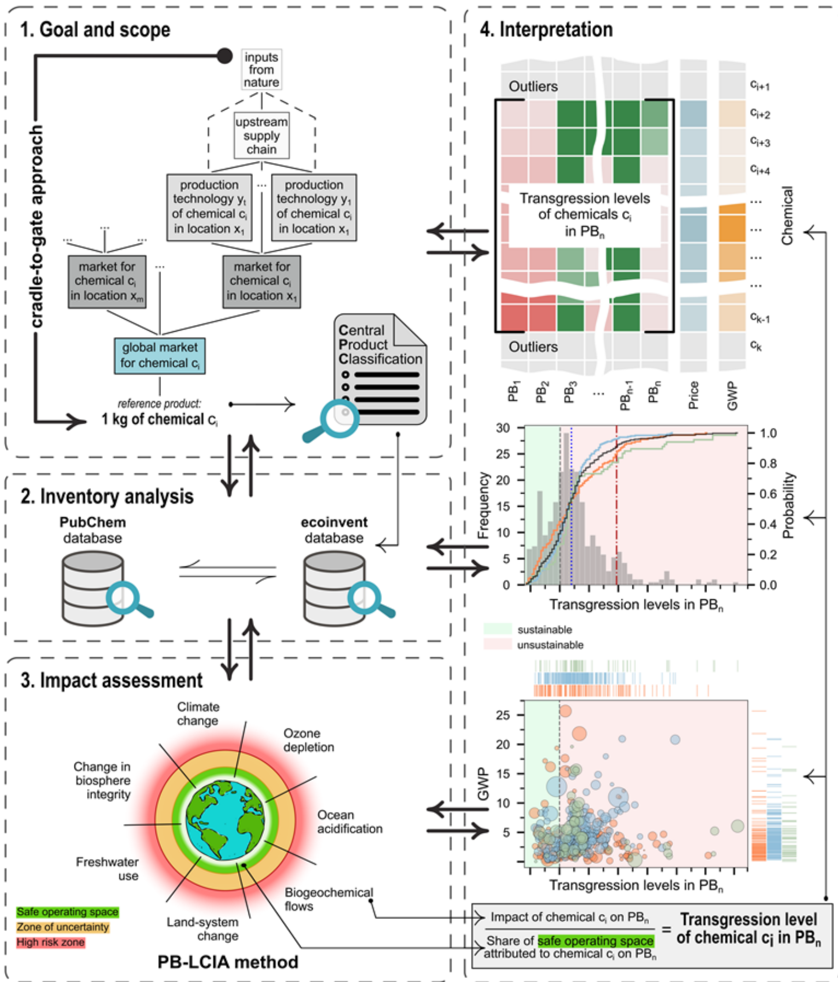
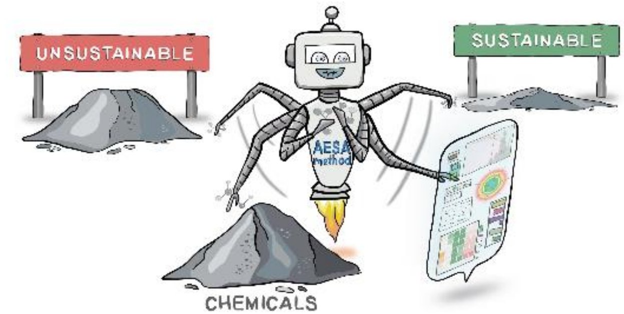
Are current fossil-based chemicals sustainable?





- **492 Chemicals** (organic, inorganic, petroleum-derived, fertilizers, pesticides, plastics and rubber, pharmaceutical products, and perfumes)
- **9 Planetary boundaries**

V. Tulus, J. Pérez-Ramírez, G. Guillén-Gosálbez, *Green Chemistry* **23**, 9881-9893 (2021).

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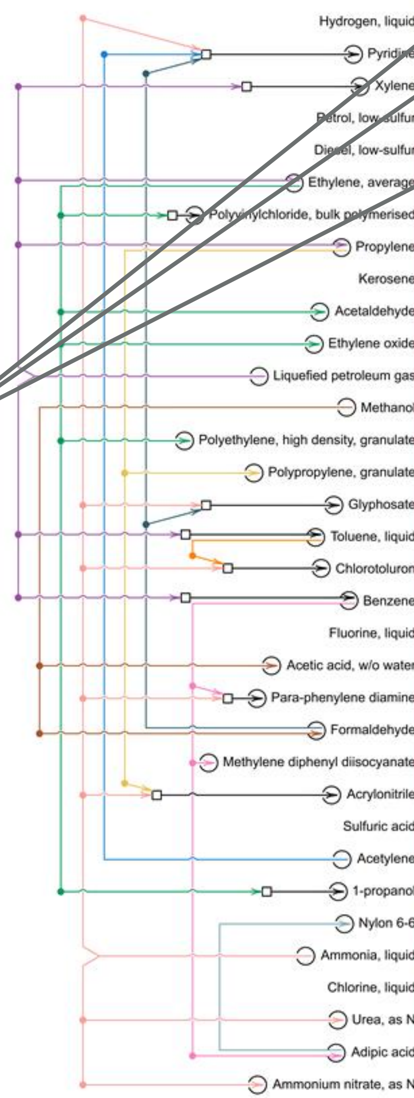
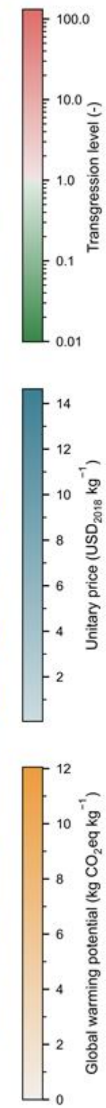
Planetary footprints of widespread chemicals

- **99.4%** are unsustainable

- **Carbon-related PBs** are more heavily transgressed



	CC - CO_2 conc.	CC - Energy imb.	SOD	OA	BGC flows - P	BGC flows - N	LSC	FWU	CBI - BII loss	Price	GWP
Hydrogen, liquid	13.2	12.8	≈ 0	4.19	≈ 0	0.16	≈ 0	≈ 0	1.02	3.85	2.37
Pyridine	13.6	13.1	≈ 0	4.35	≈ 0	0.03	≈ 0	0.02	0.93	14.6	8.15
Xylene	15.1	14.5	≈ 0	4.82	≈ 0	0.01	≈ 0	≈ 0	1.10	2.55	1.75
Petrol, low-sulfur	20.3	19.4	0.04	6.49	≈ 0	0.03	≈ 0	0.02	1.30	1.02	0.80
Diesel, low-sulfur	24.6	23.6	0.07	7.85	≈ 0	0.05	≈ 0	0.04	1.62	0.57	0.56
Ethylene, average	29.8	28.6	≈ 0	9.50	≈ 0	0.02	≈ 0	≈ 0	2.14	1.08	1.46
Polyvinylchloride, bulk polymerised	30.5	29.0	0.02	9.71	0.02	0.13	≈ 0	0.04	1.85	1.85	2.17
Propylene	30.4	29.1	≈ 0	9.68	≈ 0	0.02	≈ 0	≈ 0	2.21	1.08	1.50
Kerosene	31.8	30.4	0.09	10.1	≈ 0	0.06	0.01	0.05	2.07	0.43	0.53
Acetaldehyde	37.4	36.2	0.02	11.9	≈ 0	0.10	≈ 0	0.06	2.71	1.16	1.85
Ethylene oxide	38.2	37.0	0.02	12.2	≈ 0	0.09	≈ 0	0.03	2.66	1.33	2.15
Liquefied petroleum gas	43.3	41.4	0.10	13.8	≈ 0	0.10	0.01	0.06	2.77	0.40	0.67
Methanol	43.8	42.6	0.05	14.0	≈ 0	0.05	≈ 0	0.10	3.52	0.32	0.67
Polyethylene, high density, granulate	44.7	42.7	≈ 0	14.3	≈ 0	0.03	≈ 0	0.02	3.16	1.06	2.09
Polypropylene, granulate	45.7	43.5	≈ 0	14.6	0.25	0.73	≈ 0	0.02	3.10	1.09	2.12
Glyphosate	47.0	45.7	0.05	15.0	11.5	0.63	≈ 0	0.11	3.35	6.03	11.2
Toluene, liquid	48.2	46.3	≈ 0	15.4	≈ 0	0.05	≈ 0	≈ 0	3.55	0.73	1.61
Chlorotoluron	45.0	46.8	0.70	14.4	0.01	14.6	≈ 0	0.06	3.61	3.36	6.86
Benzene	51.7	49.6	0.01	16.5	≈ 0	0.06	≈ 0	0.06	3.97	0.88	2.09
Fluorine, liquid	52.5	51.9	0.06	16.8	≈ 0	0.17	≈ 0	0.10	3.64	5.78	12.1
Acetic acid, w/o water	56.6	55.3	0.07	18.0	≈ 0	0.21	≈ 0	0.11	4.33	0.69	1.70
Para-phenylene diamine	59.5	62.2	1.08	19.0	0.02	47.5	≈ 0	0.10	4.73	4.31	11.9
Formaldehyde	64.5	62.5	0.07	20.6	≈ 0	0.23	≈ 0	0.12	4.95	0.34	0.99
Methylene diphenyl diisocyanate	59.9	64.4	1.39	19.1	0.02	4.37	≈ 0	0.11	5.07	2.55	7.47
Acrylonitrile	71.9	68.7	0.02	22.9	≈ 0	0.12	≈ 0	0.11	4.81	1.02	3.02
Sulfuric acid	81.3	79.0	0.13	25.9	0.03	0.90	≈ 0	0.28	6.86	0.04	0.15
Acetylene	90.5	89.6	0.09	28.9	≈ 0	0.22	≈ 0	0.16	6.10	1.58	5.67
1-propanol	94.0	91.0	0.07	30.0	≈ 0	0.26	≈ 0	0.12	6.79	1.19	4.53
Nylon 6-6	101.2	98.1	0.29	32.3	1.18	105.5	≈ 0	≈ 0	7.17	1.85	8.34
Ammonia, liquid	109.2	104.0	0.08	34.8	≈ 0	0.15	≈ 0	0.03	6.72	0.50	2.11
Chlorine, liquid	120.7	119.1	0.23	38.5	0.08	4.10	≈ 0	0.21	8.37	0.21	1.02
Urea, as N	131.6	125.7	0.10	41.9	0.01	0.37	≈ 0	0.08	8.43	0.66	3.35
Adipic acid	77.6	147.5	15.4	24.7	≈ 0	0.19	≈ 0	0.12	15.5	1.55	13.7
Ammonium nitrate, as N	133.6	237.0	23.0	42.6	0.03	0.70	≈ 0	0.09	23.4	0.59	7.99



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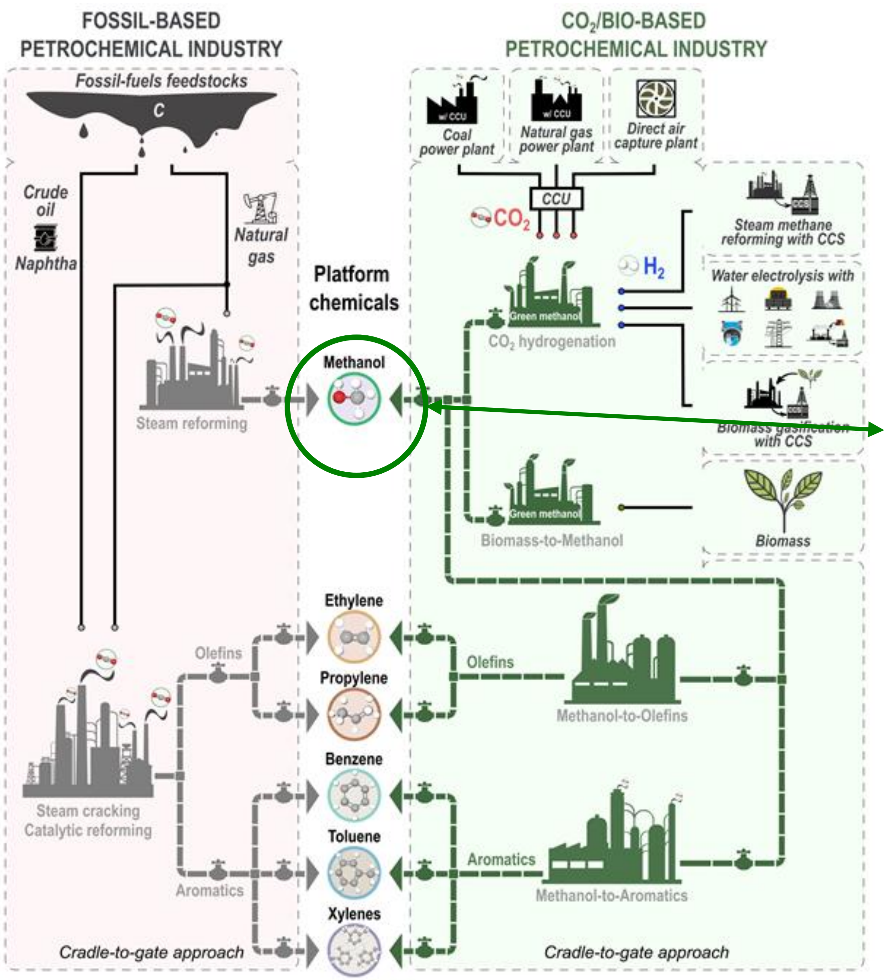
Could renewable-carbon help to operate within the PBs?

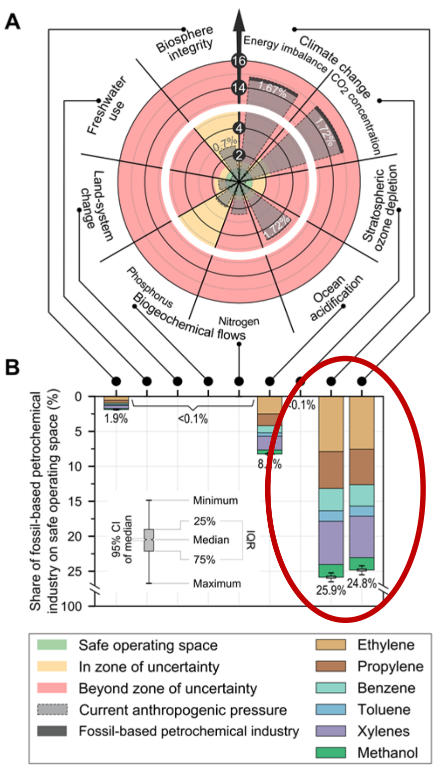


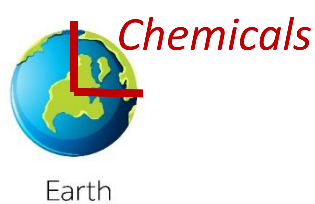
Green methanol could be used as a precursor for olefins and aromatics

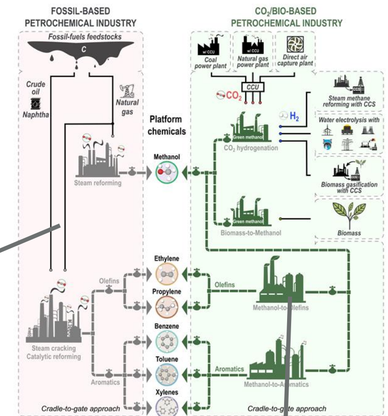
Á. Galán-Martín *et al., One Earth* **4**, 565-583 (2021).
A. González-Garay *et al., Energy & Environmental Science* **12**, 3425-3436 (2019).
A. Kätelhön *et al., PNAS* **116**, 11187-11194 (2019).

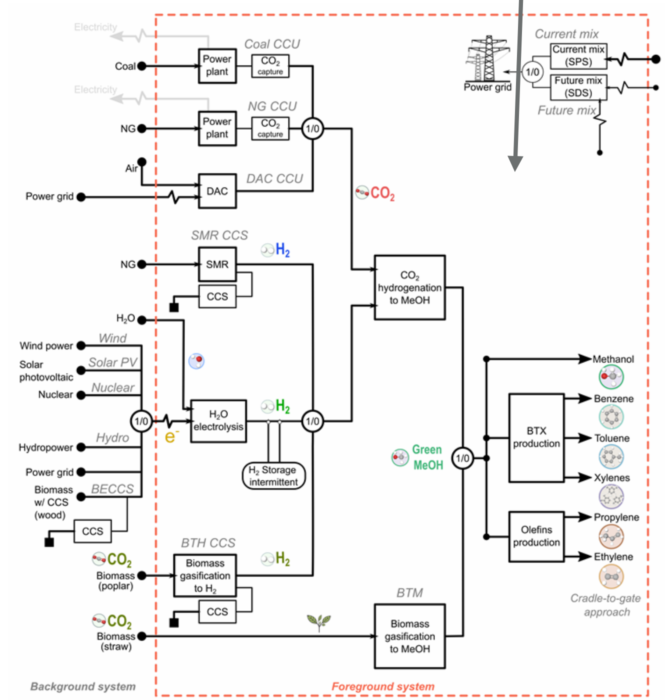
Would a green methanol chemical industry be sustainable?

- Fossil chemicals consume **25% of the planet**
- 54 alternative scenarios based on green methanol









A. Galán-Martín *et al., One Earth* **4**, 565-583 (2021).

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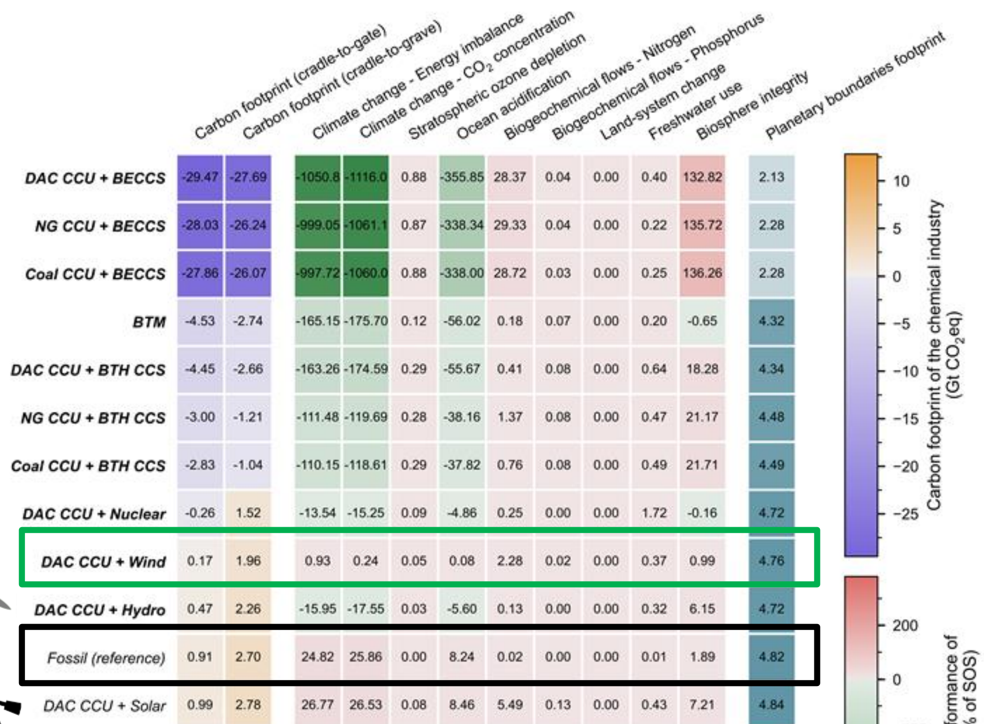
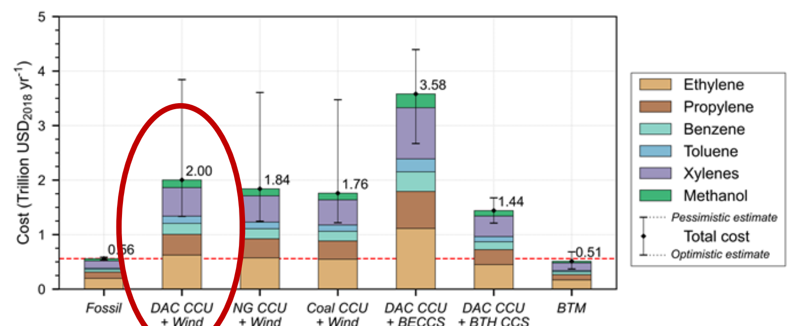
Assessing the chemical sector's impact based on the PBs

- From **25% (fossil carbon)** to **1% (renewable carbon)** of the planet
- Renewable carbon routes are too expensive: **4-fold** increase in cost



	Carbon footprint (cradle-to-gate)	Carbon footprint (cradle-to-grave)	Climate change - Energy imbalance	Climate change - CO_2 concentration	Stratospheric ozone depletion	Ocean acidification	Biogeochemical flows - Nitrogen	Biogeochemical flows - Phosphorus	Land-system change	Freshwater use	Biosphere integrity	Planetary boundaries footprint
DAC CCU + BECCS	-29.47	-27.69	-1050.8	-1116.0	0.88	-355.85	28.37	0.04	0.00	0.40	132.82	2.13
NG CCU + BECCS	-28.03	-26.24	-999.05	-1061.1	0.87	-338.34	29.33	0.04	0.00	0.22	135.72	2.28
Coal CCU + BECCS	-27.86	-26.07	-997.72	-1060.0	0.88	-338.00	28.72	0.03	0.00	0.25	136.26	2.28
BTM	-4.53	-2.74	-165.15	-175.70	0.12	-56.02	0.18	0.07	0.00	0.20	-0.65	4.32
DAC CCU + BTH CCS	-4.45	-2.66	-163.26	-174.59	0.29	-55.67	0.41	0.08	0.00	0.64	18.28	4.34
NG CCU + BTH CCS	-3.00	-1.21	-111.48	-119.69	0.28	-38.16	1.37	0.08	0.00	0.47	21.17	4.48
Coal CCU + BTH CCS	-2.83	-1.04	-110.15	-118.61	0.29	-37.82	0.76	0.08	0.00	0.49	21.71	4.49
DAC CCU + Nuclear	-0.26	1.52	-13.54	-15.25	0.09	-4.86	0.25	0.00	0.00	1.72	-0.16	4.72
DAC CCU + Wind	0.17	1.96	0.93	0.24	0.05	0.08	2.28	0.02	0.00	0.37	0.99	4.76
DAC CCU + Hydro	0.47	2.26	-15.95	-17.55	0.03	-5.60	0.13	0.00	0.00	0.32	6.15	4.72
Fossil (reference)	0.91	2.70	24.82	25.86	0.00	8.24	0.02	0.00	0.00	0.01	1.89	4.82
DAC CCU + Solar	0.99	2.78	26.77	26.53	0.08	8.46	5.49	0.13	0.00	0.43	7.21	4.84



Earth

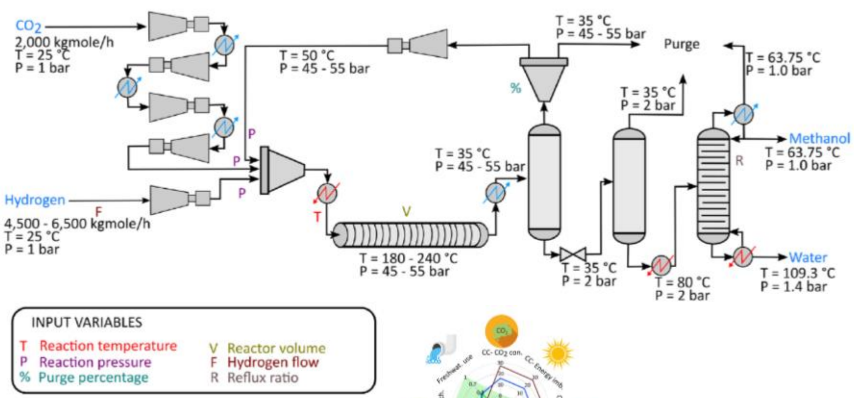
Chemicals 25%

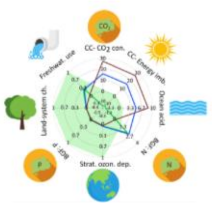
Earth

Chemicals 1%

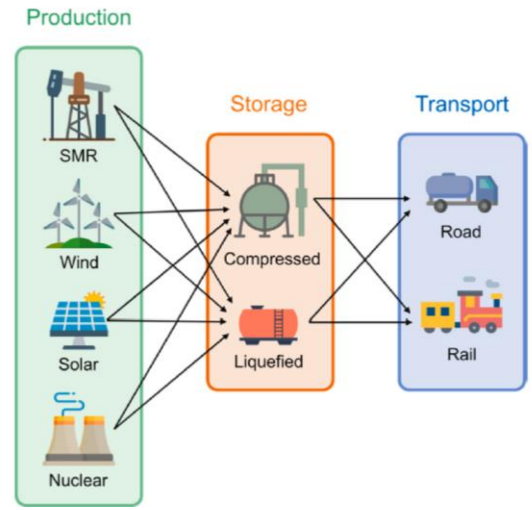
Other applications

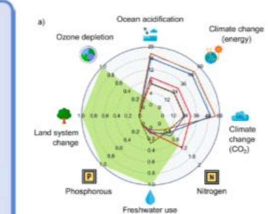
Process design[1]



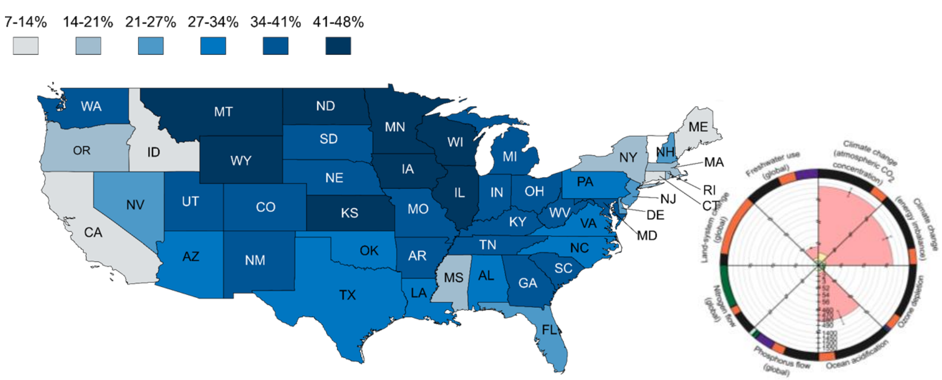


Supply chain design[2]

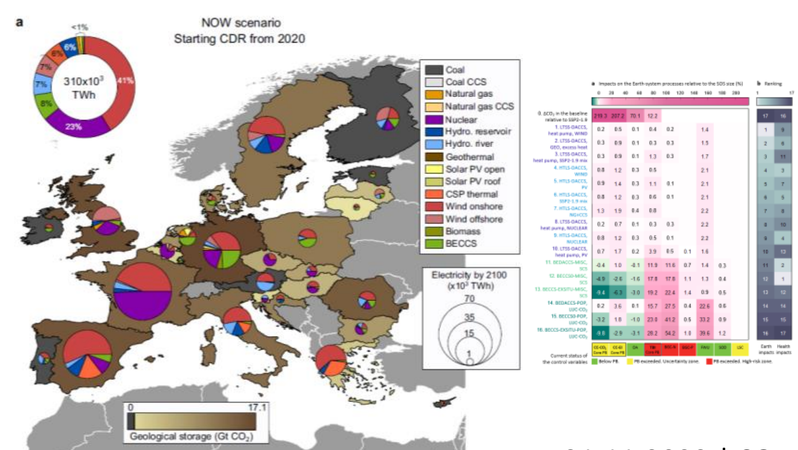




Power systems[3]



Negative emissions technologies[4,5]



1. D. Vázquez, G. Guillén-Gosálbez, *Chemical Engineering Science* **246**, 116891 (2021).
2. M. Ehrenstein *et al.*, *Applied Energy* **276**, 115486 (2020).
3. M. Algunaibet *et al.*, *Energy & Environmental Science* **12**, 1890-1900 (2019).
4. S. Cobo *et al.*, *Nature Communications* **13**, 1-11 (2022).
5. Á. Galán-Martín *et al.*, *Nature Communications* **12**, 1-12 (2021).

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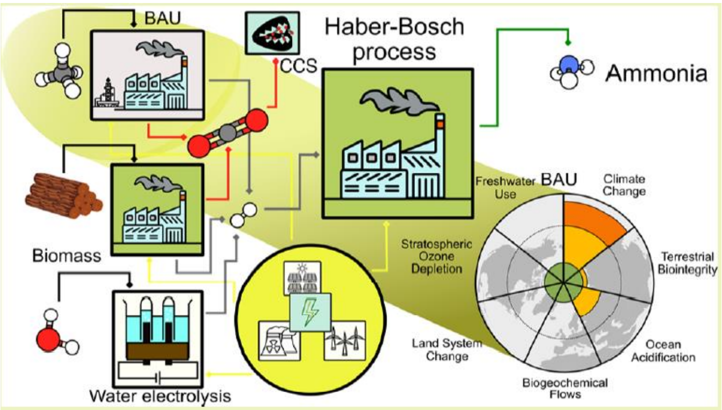
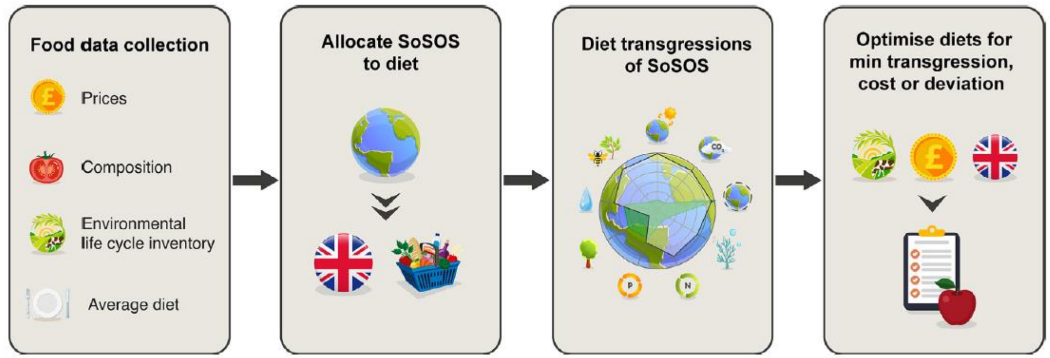
Other applications

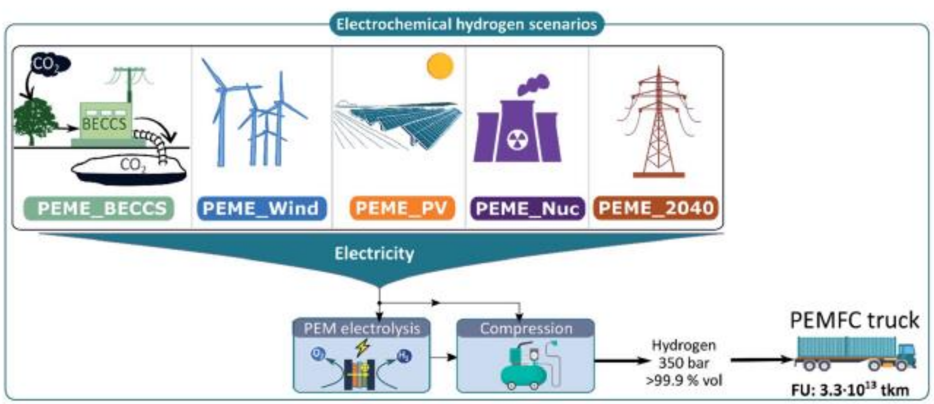
Process design[1]



Diets[2]



E-fuels[3]



1. SC D'Angelo *et al.*, *ACS Sustainable Chemistry & Engineering* **9**, 9740-9749 (2021).
2. E. Lucas, M. Guo, G. Guillén-Gosálbez, *Sustainable Production and Consumption* **28**, 877-892 (2021).
3. A. Valente *et al.*, *Sustainable Energy & Fuels* **5**, 4637-4649 (2021).

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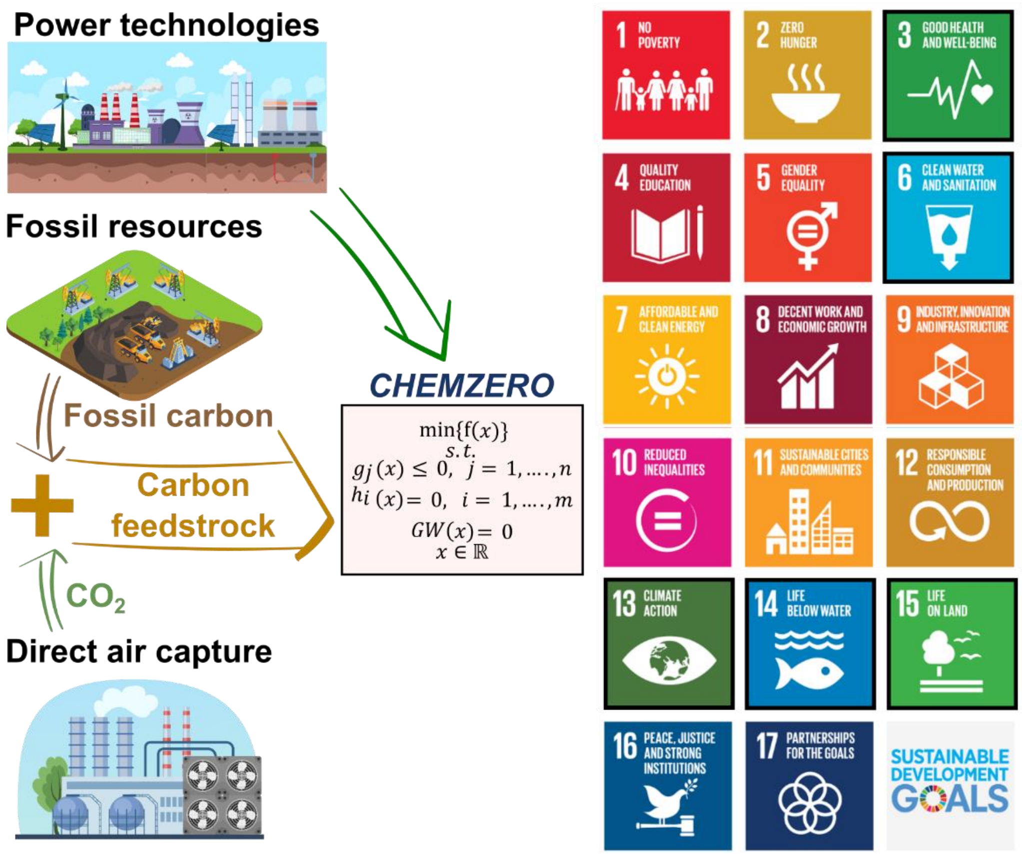
Future steps: Sustainable Development Goals optimization

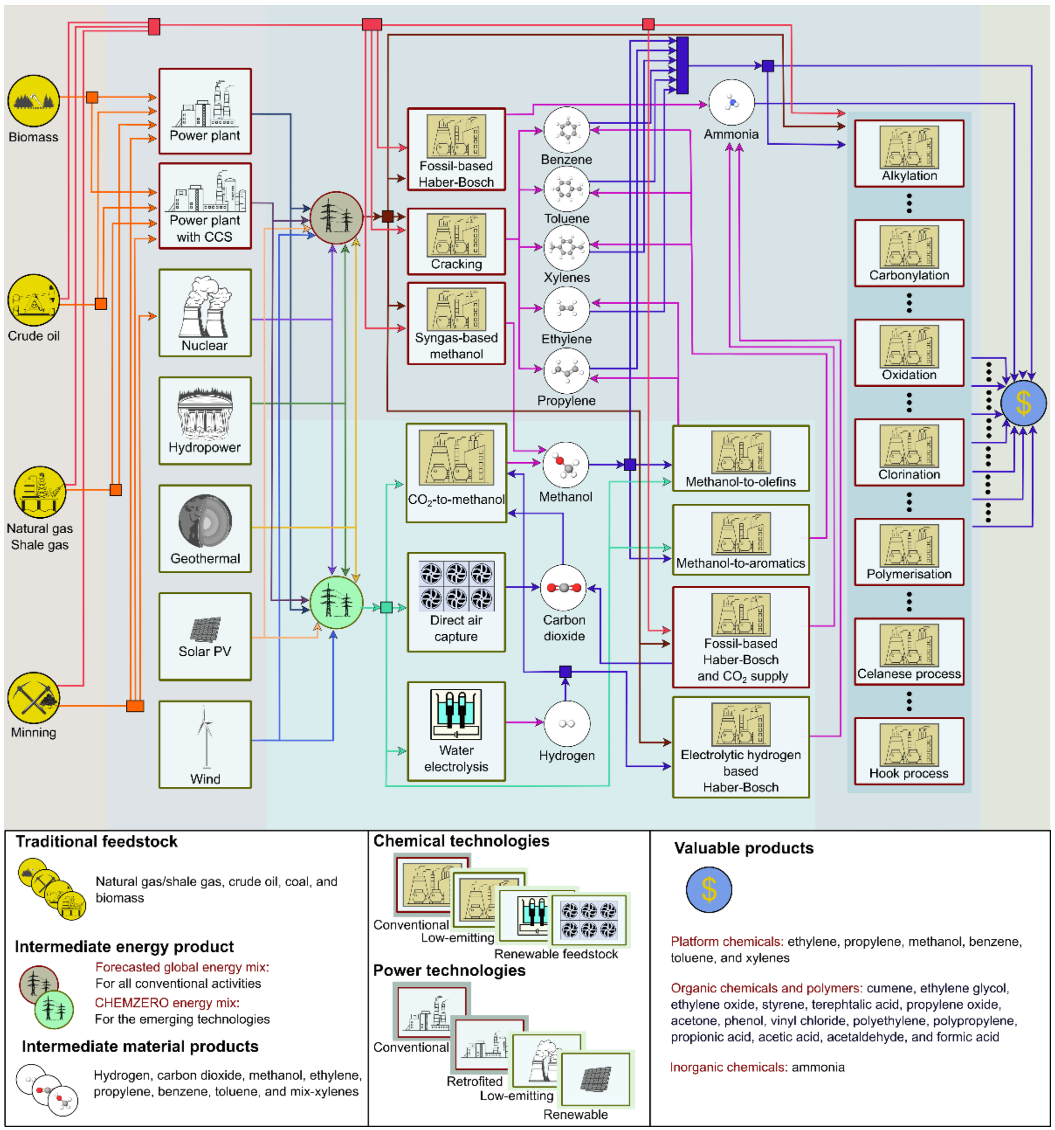
Carbon neutral chemical industry under the lens of five SDGs



2030 Agenda for Sustainable Development

I. Ioannou *et al., Energy & Environmental Science*, accepted (2022).

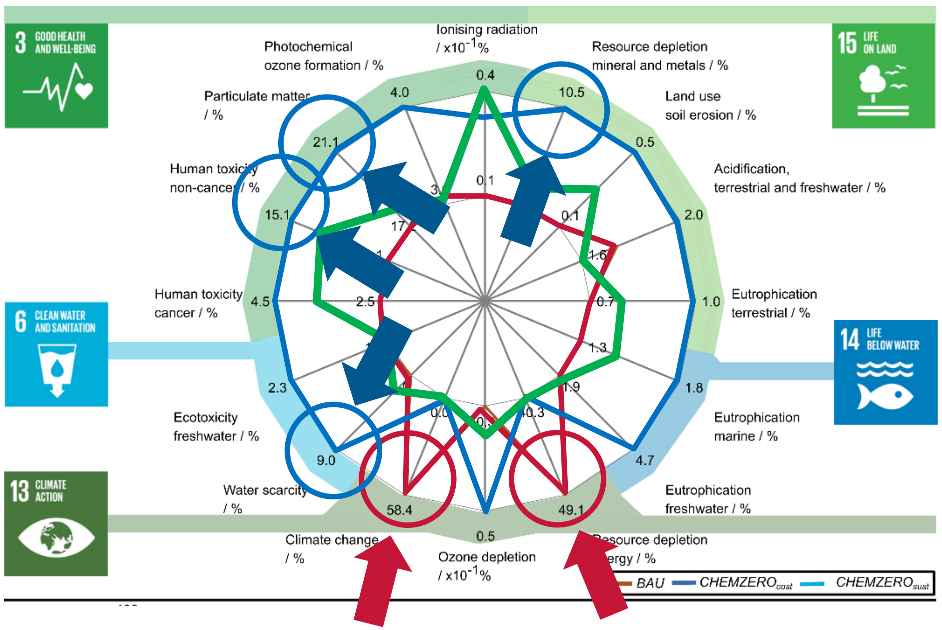
Future steps: Sustainable Development Goals optimization



- **Superstructure** of technologies encompassing thousands of alternatives

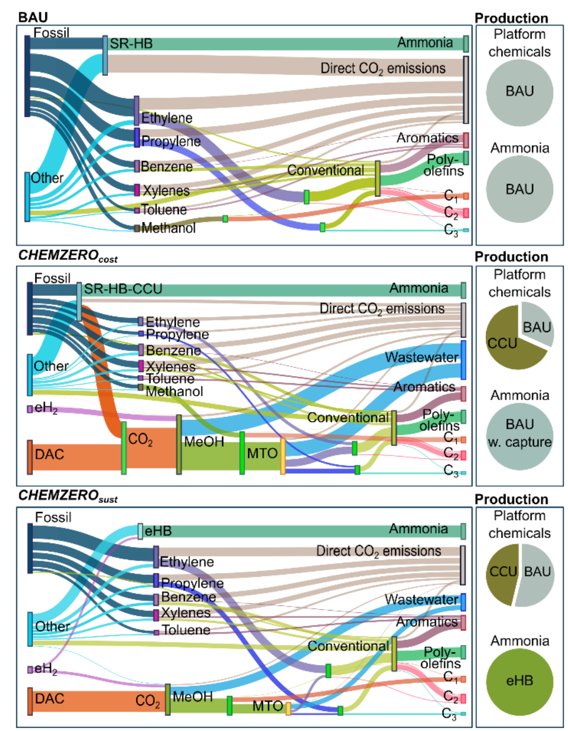
$$\min_x \quad f(x) = 0 \quad \textit{Objective function}$$
$$\text{s.t.} \quad h(x) = 0 \quad \textit{Mass \& energy balances}$$
$$g(x) \leq 0 \quad \textit{Capacity constraints}$$

Future steps: Sustainable Development Goals optimization



- **Fossil chemicals**
- **Carbon neutral at minimum cost leads to <u>burden-shifting</u>**
- **Carbon neutral optimizing SDGs performance**

- **Hybrid production** patterns are required to produce chemicals sustainably



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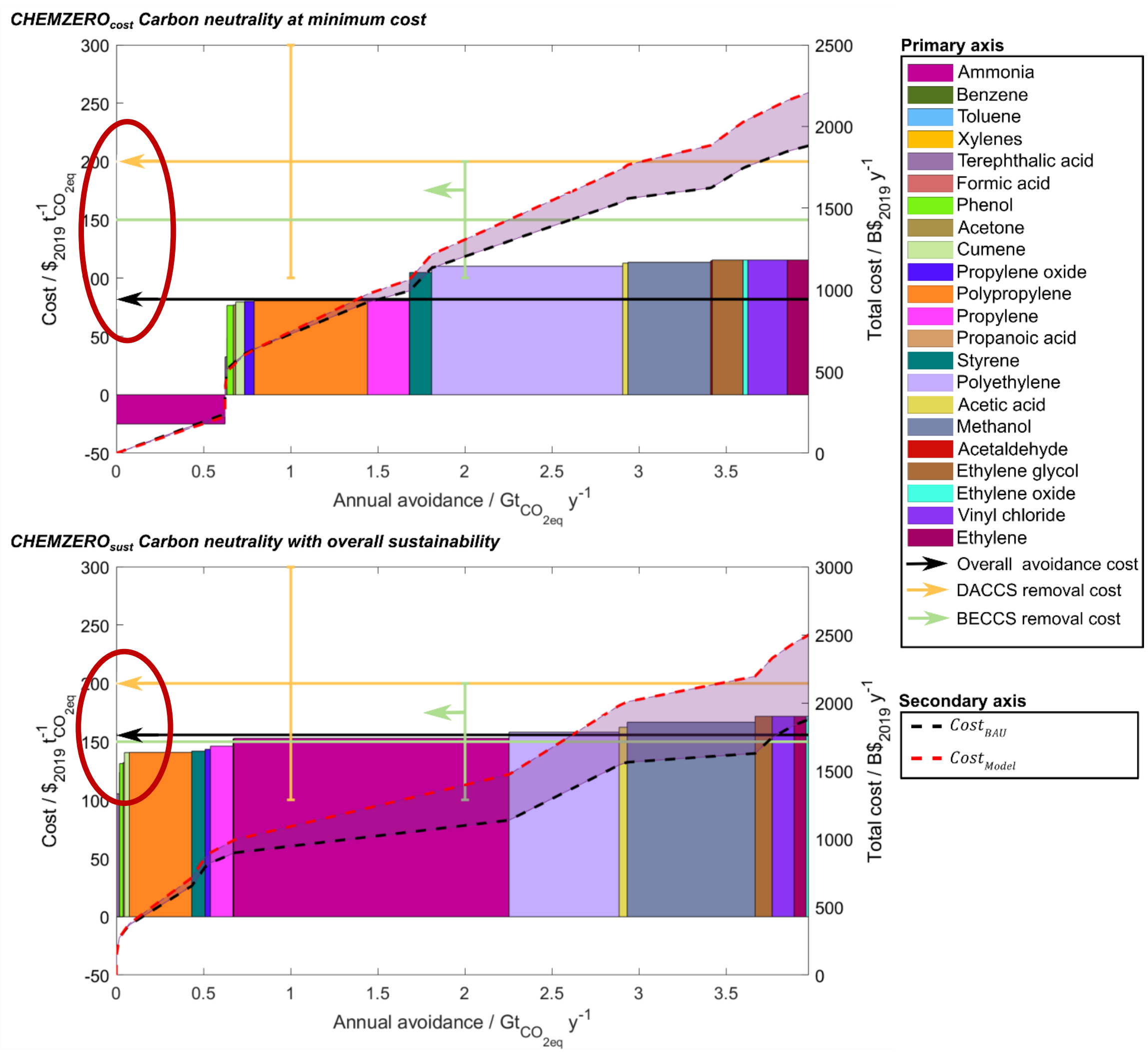
Future steps: Sustainable Development Goals optimization



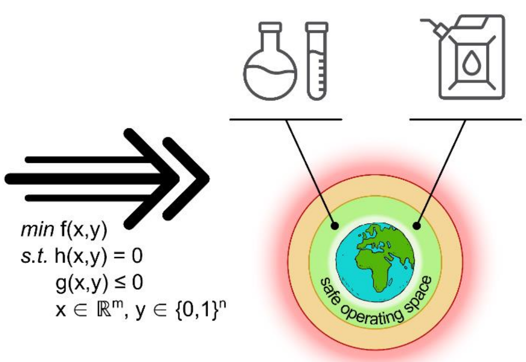
- **Mitigation** cheaper than carbon removal

Conclusions

- Process Systems Engineering can be applied to a wide range of sustainability problems
- Main applications at the multi-site and single-site levels
- Combining systems thinking, optimization and life cycle thinking leads to sustainability gains
- Future directions: planetary boundaries optimization

Take-home message

PSE concepts and tools provide a general framework to address a wide variety of relevant problems related to our sustainable development



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Process Systems Engineering for sustainable chemicals within planetary boundaries

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Department of Chemistry and Applied Biosciences