#PlanDeRecuperación

Plan de Recuperación, Transformación y Resiliencia

GOBIERNO DE ESPAÑA
VICEPRESIDENCIA TERCERA DEL GOBIERNO
MINISTERIO PARA LA TRANSICIÓN ECOLÓGICA Y EL RETO DEMOGRÁFICO

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Bringing the complexity of organic chemistry to climate models with machine learning techniques

Camille Mouchel-Vallon, Alma Hodzic, John Schreck, Charlie Becker, Keeley Lawrence, Siyuan Wang, Jinkyul Choi, Daven Henze, David John Gagne

NSF | NCAR Operated by UCAR

PASC24, Zürich, 2024/06/05

Context

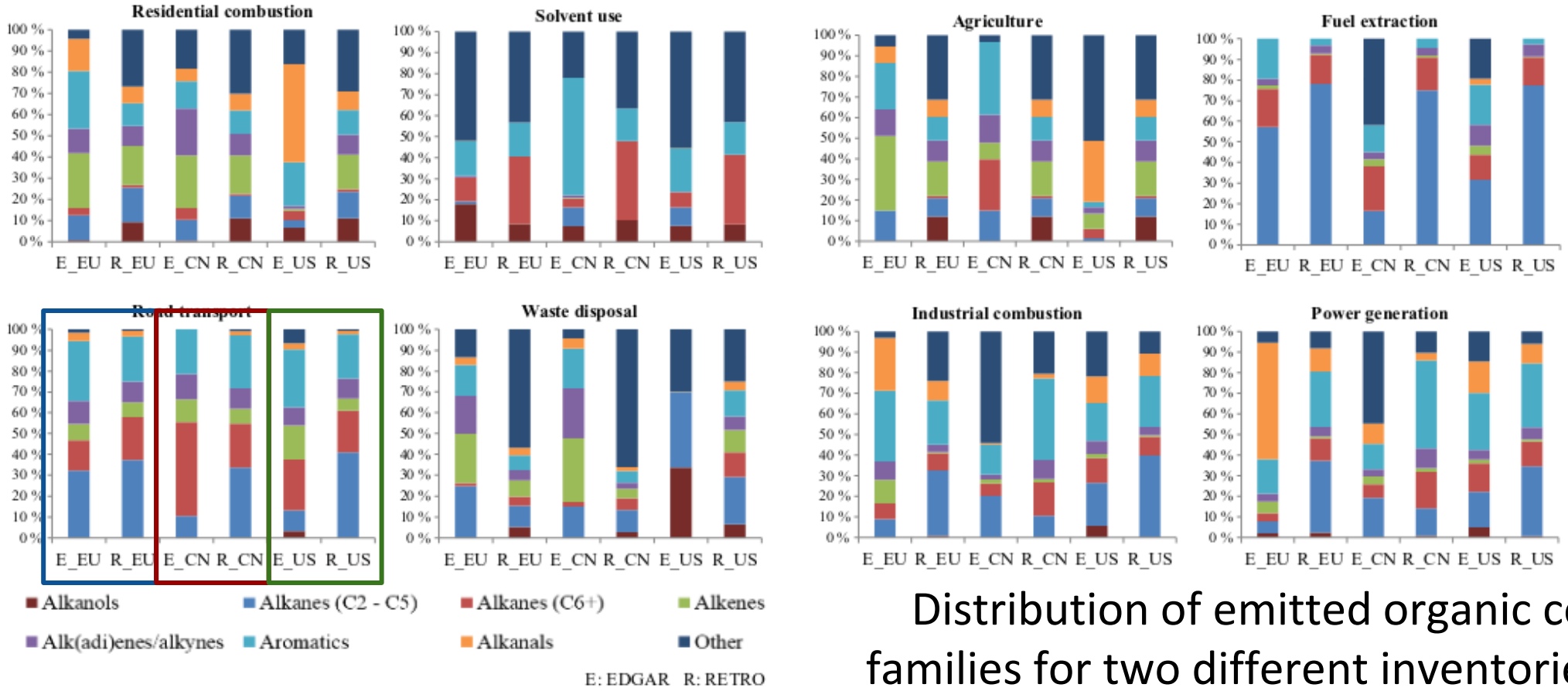
Plan de Recuperación, Transformación y Resiliencia

1st objective: A high resolution spatial (up to 2.5x2.5 km) and temporal (hourly) emissions system to compute emissions of **primary atmospheric pollutants** for Spain to be used in the national **air quality prediction** system.

Gaseous primary atmospheric pollutants
NO_x, CO, SO_x, NH_3, **NMVOCs**

Non-Methane Volatile Organic Compounds are emitted by human and natural activities and their chemical evolution in the atmosphere has an impact on air quality, health and climate

Primary organic compounds: many sources, high diversity, high uncertainty



Distribution of emitted organic compounds families for two different inventories in **Europe**, **China** and **USA**

(Huang et al., 2017)

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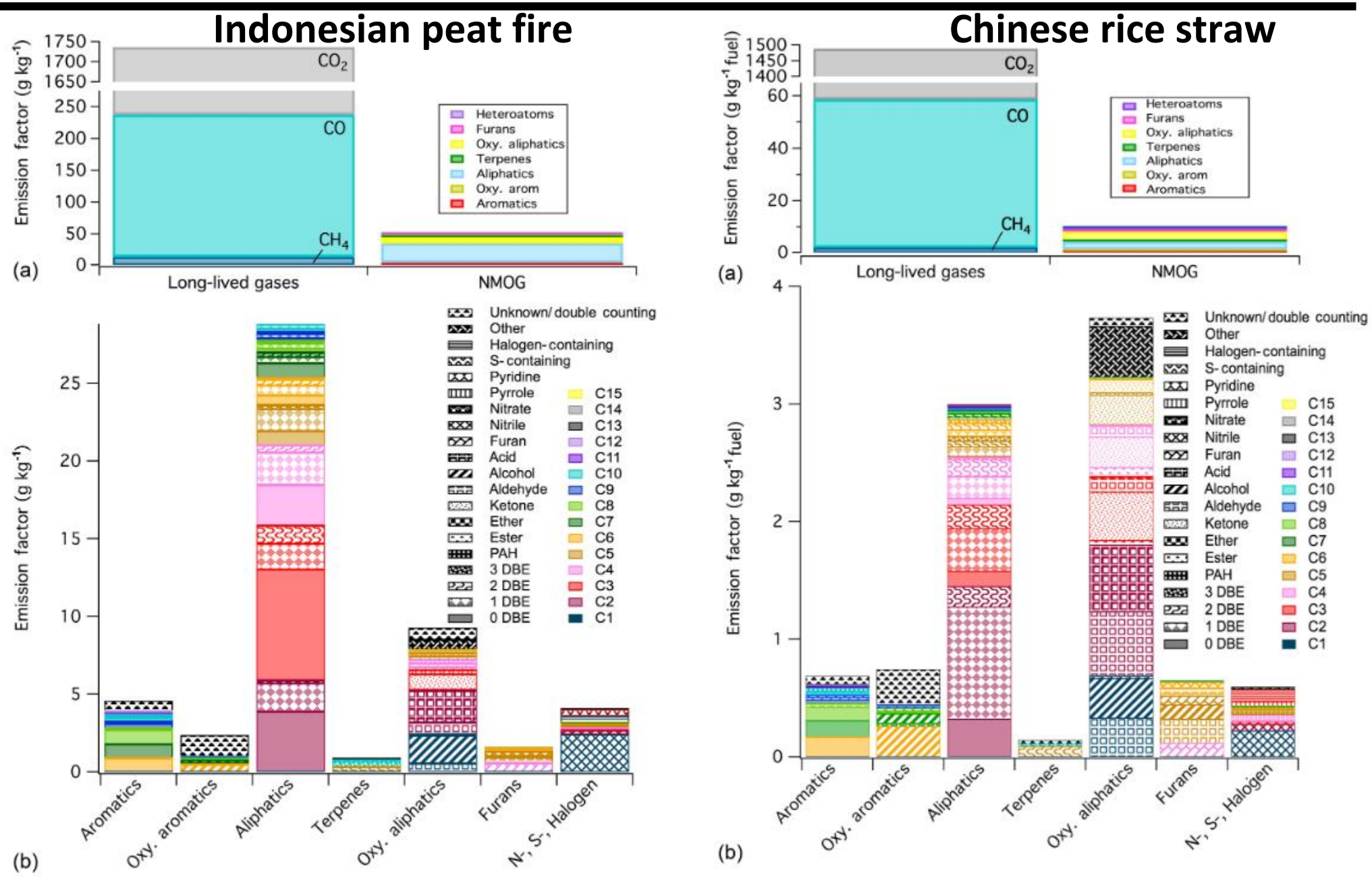
Primary organic compounds: many sources, high diversity, high uncertainty

Measured fire emissions composition (Hatch et al., 2017)



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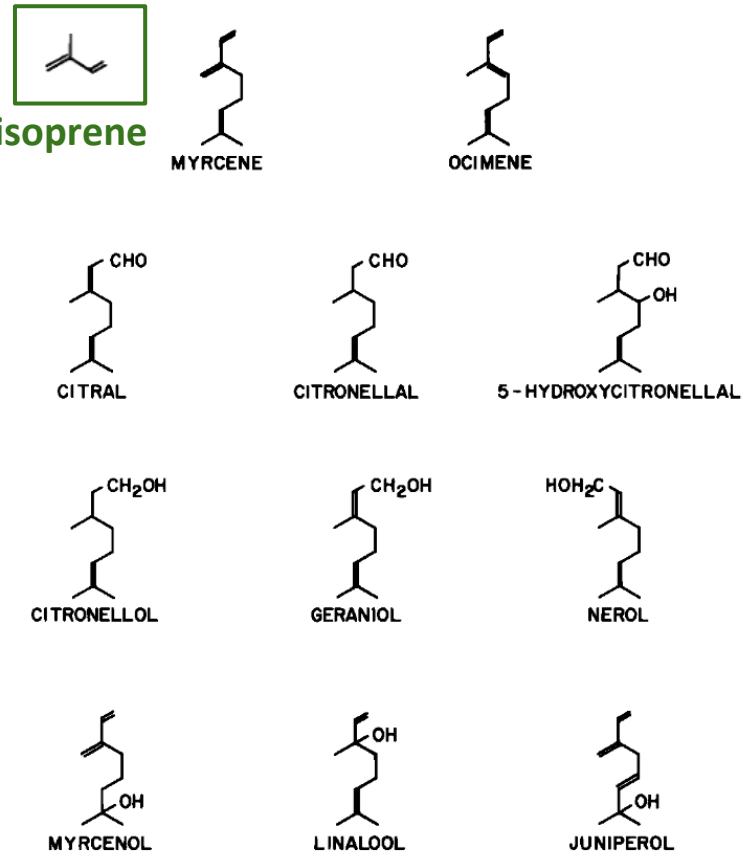
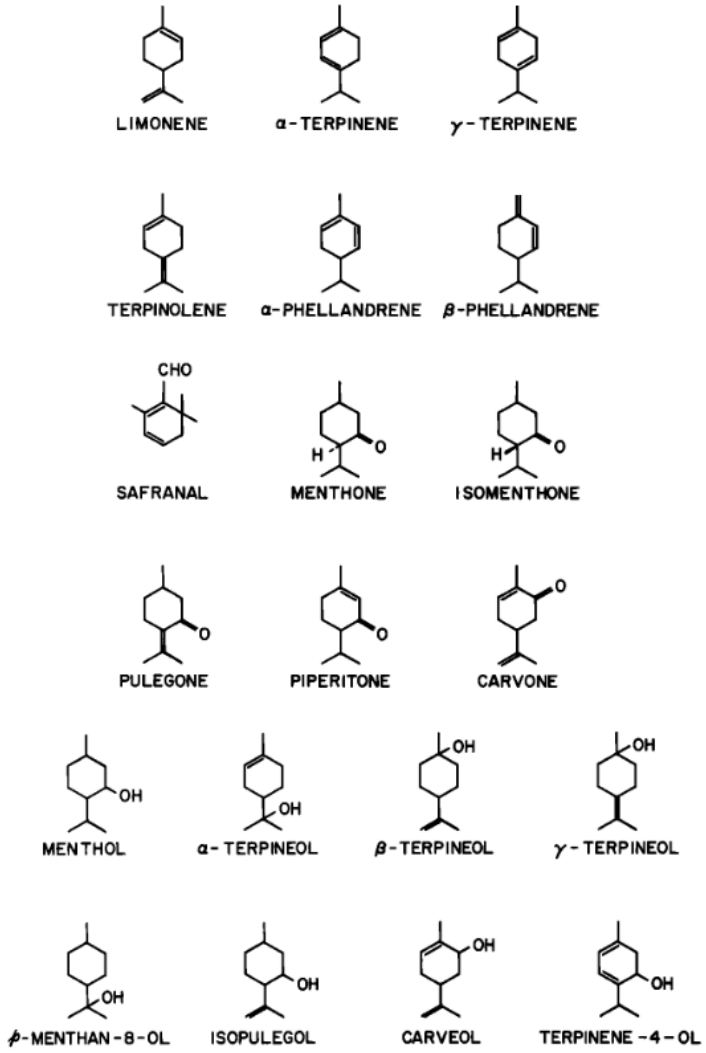
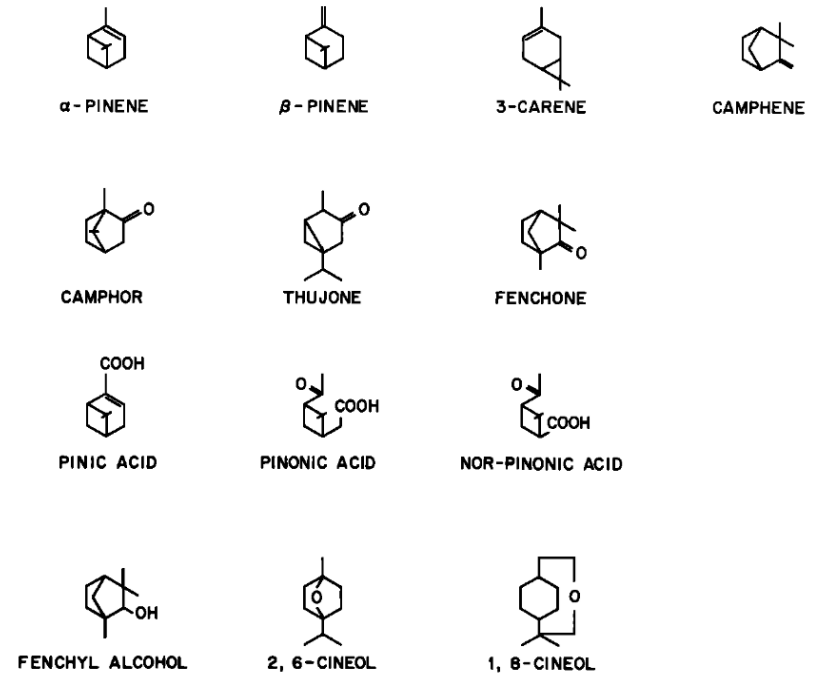
Primary organic compounds: many sources, high diversity, high uncertainty







Inventory of terpenoids emitted by plants
(Graedel, 1979)

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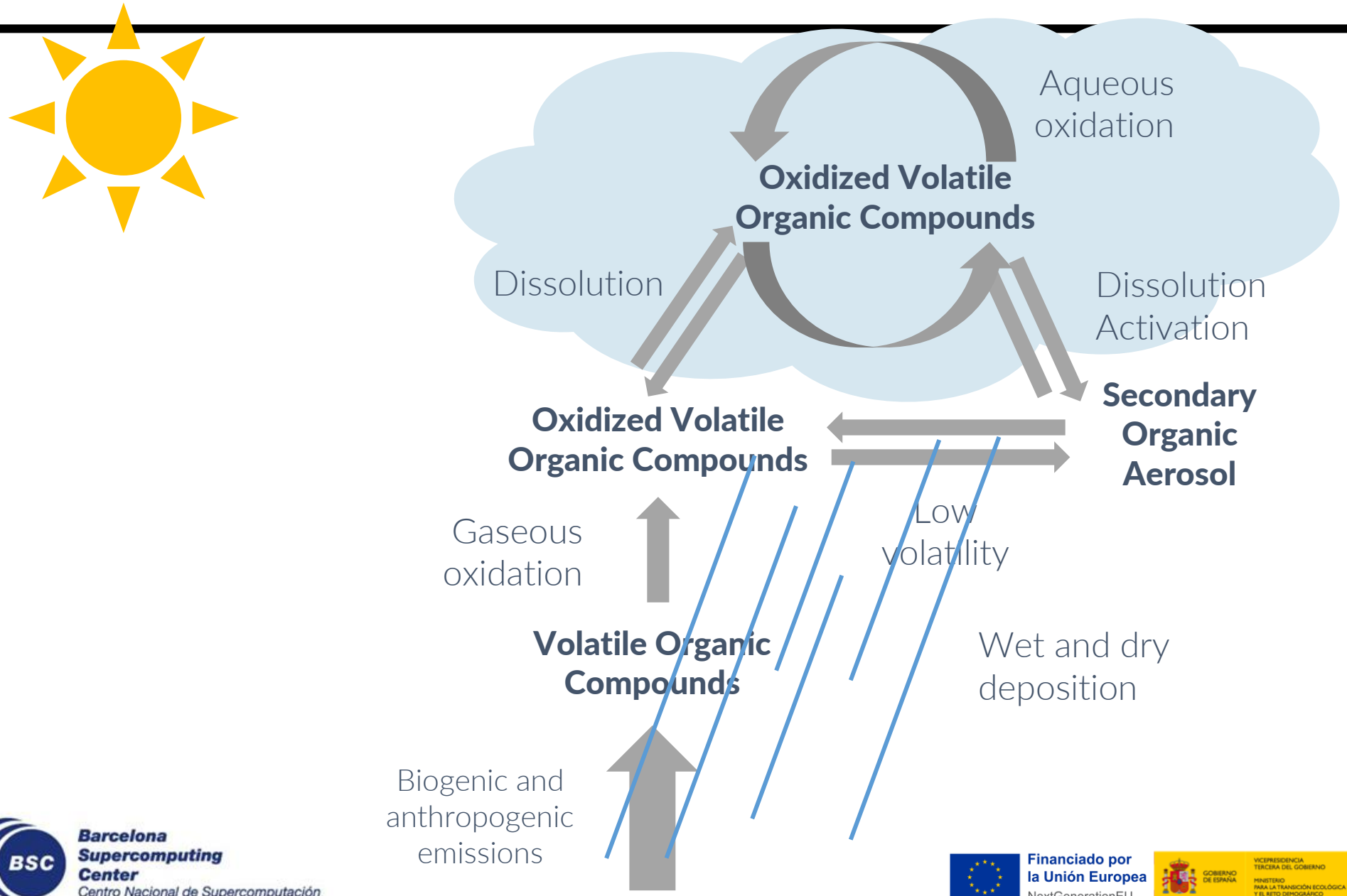
Atmospheric Evolution of Organics : formation of oxidized compounds



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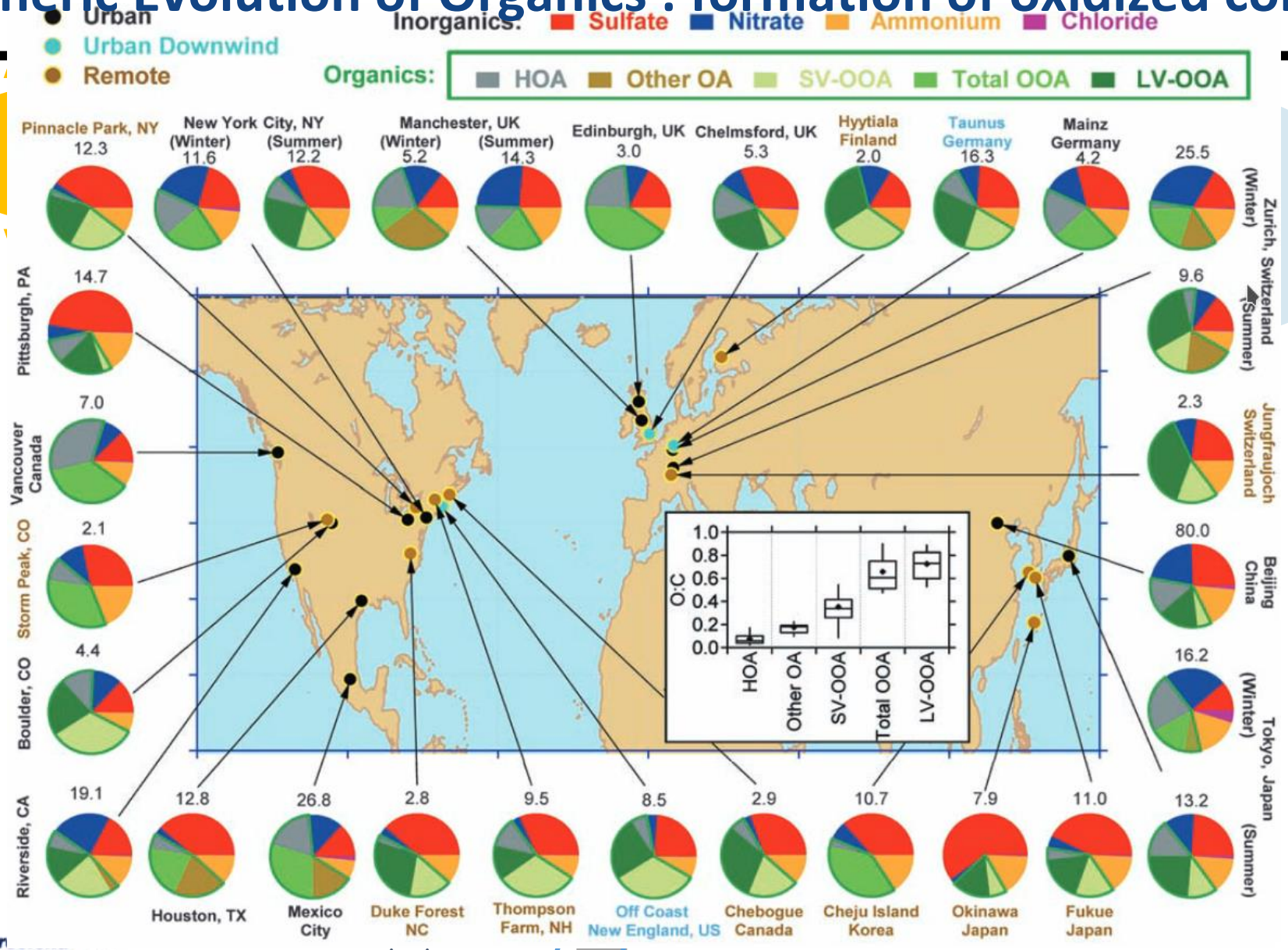
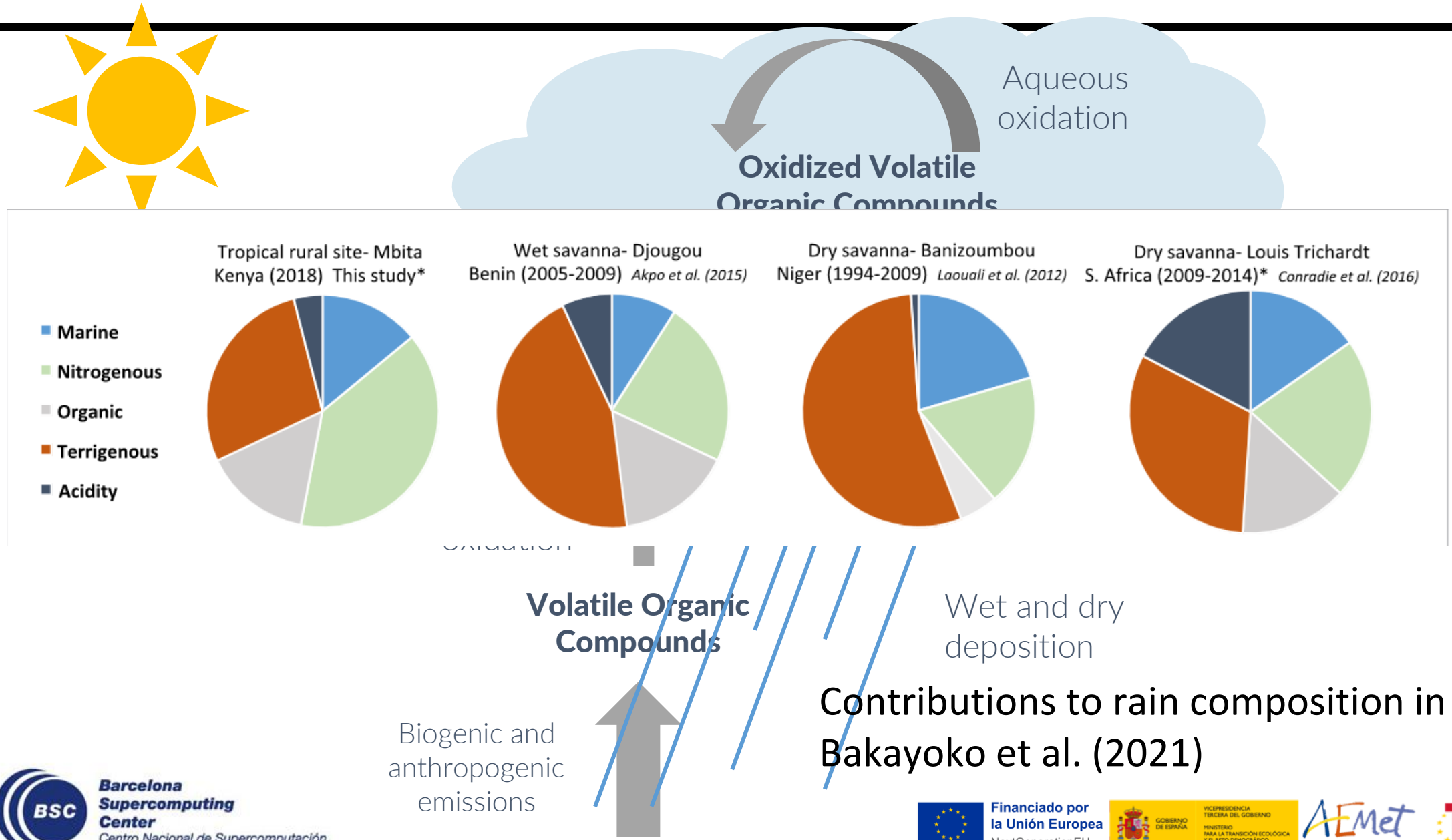


Measured PM1 composition Jimenez et al. (2009)

emissions

Atmospheric Evolution of Organics : formation of oxidized compounds



Contributions to rain composition in the tropics
Bakayoko et al. (2021)

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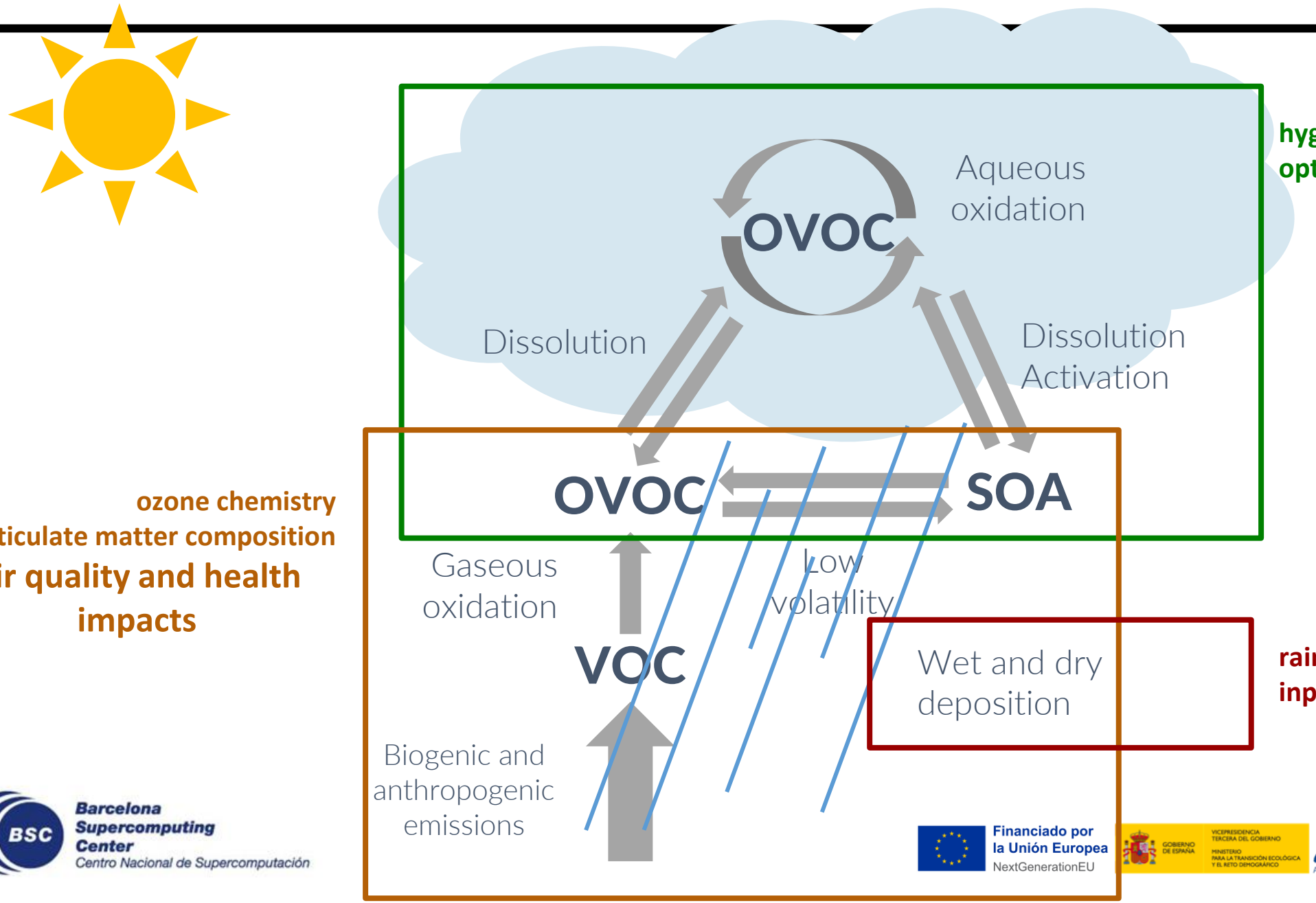
Impacts



hygroscopicity
optical properties
Climate impacts

ozone chemistry
particulate matter composition
Air quality and health impacts

rain composition
inputs to soils
Ecosystems impacts

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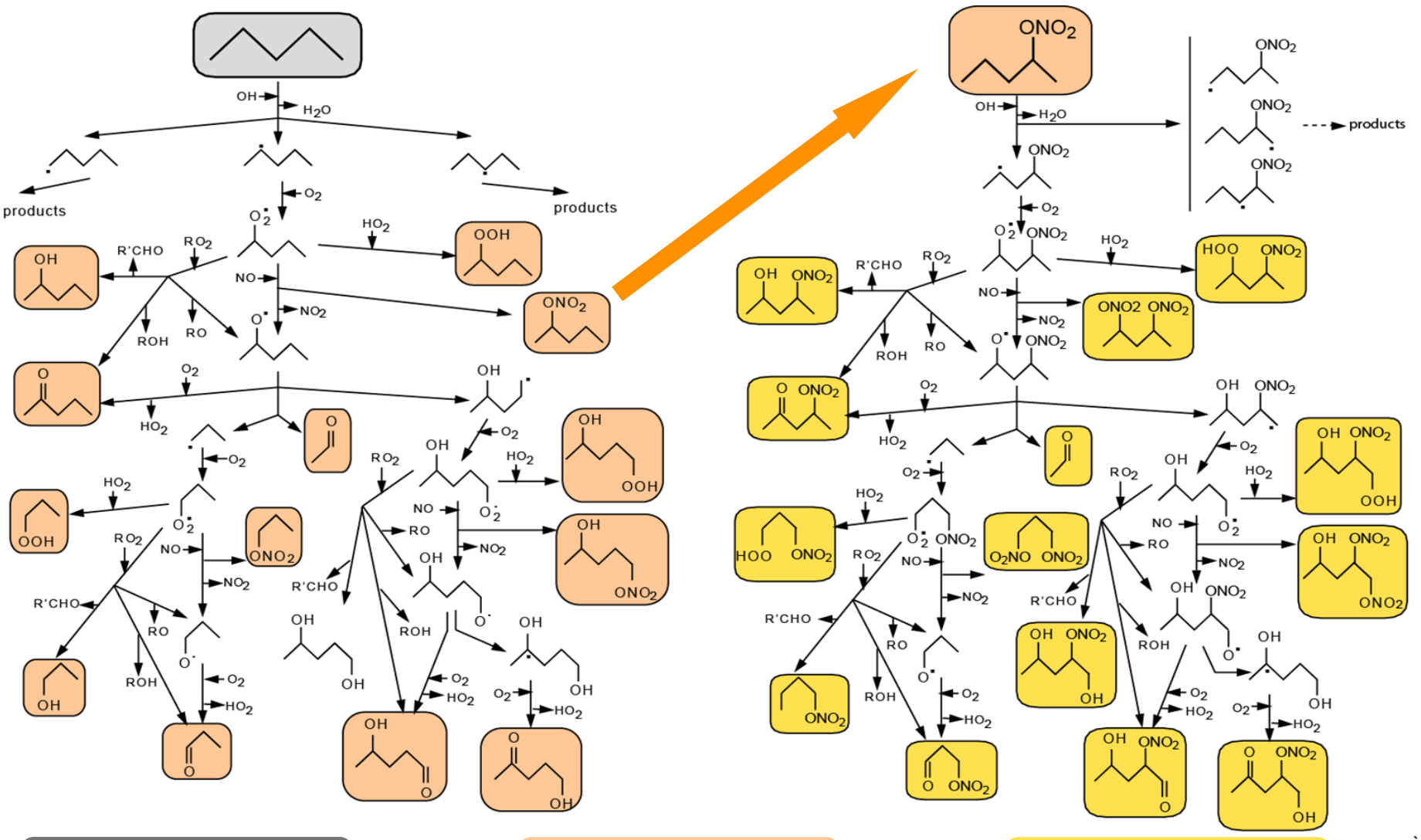
Progressive and complex oxidation of organics



Oxidized Volatile Organic Compounds

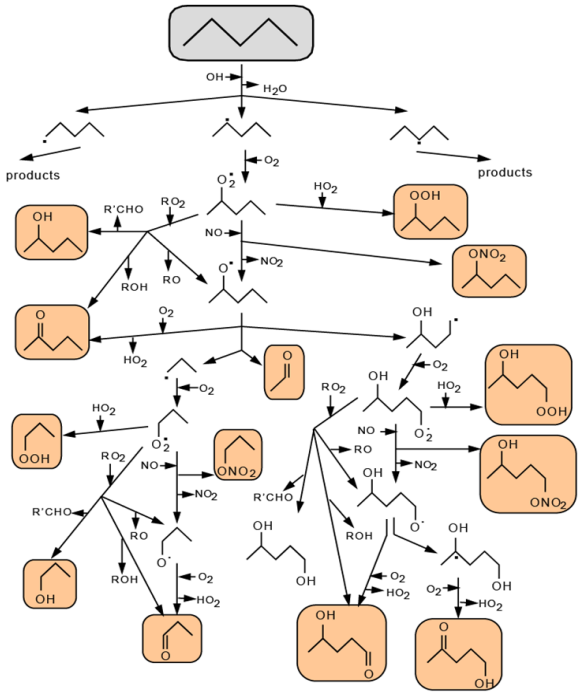
Gaseous oxidation

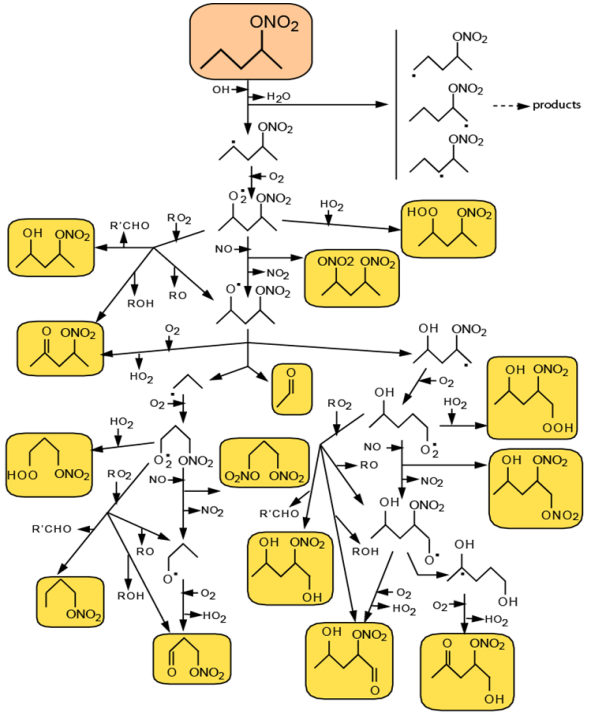
Volatile Organic Compounds

How to bring this complexity to air quality models?

Parent Hydrocarbon → **1st Generation Species** → **2nd Generation Species** → CO_2

How I Learned to Stop Worrying and Love the Complexity





Master Chemical Mechanism (Jenkin et al., 1997; 15yrs 2-3people ?)

- 143 precursors from C_1 to C_{12}
- 17000 reactions of 6700 species
- Simplified!

CLEPS (Mouchel-Vallon et al., 2017; 2yrs 2people)

- Cloud oxidation of C_1-C_4 products from isoprene oxidation
- 1315 reactions involving 717 chemical species
- Simplified!

Handwritten detailed mechanisms: high potential for errors and time consuming (creation and maintenance)

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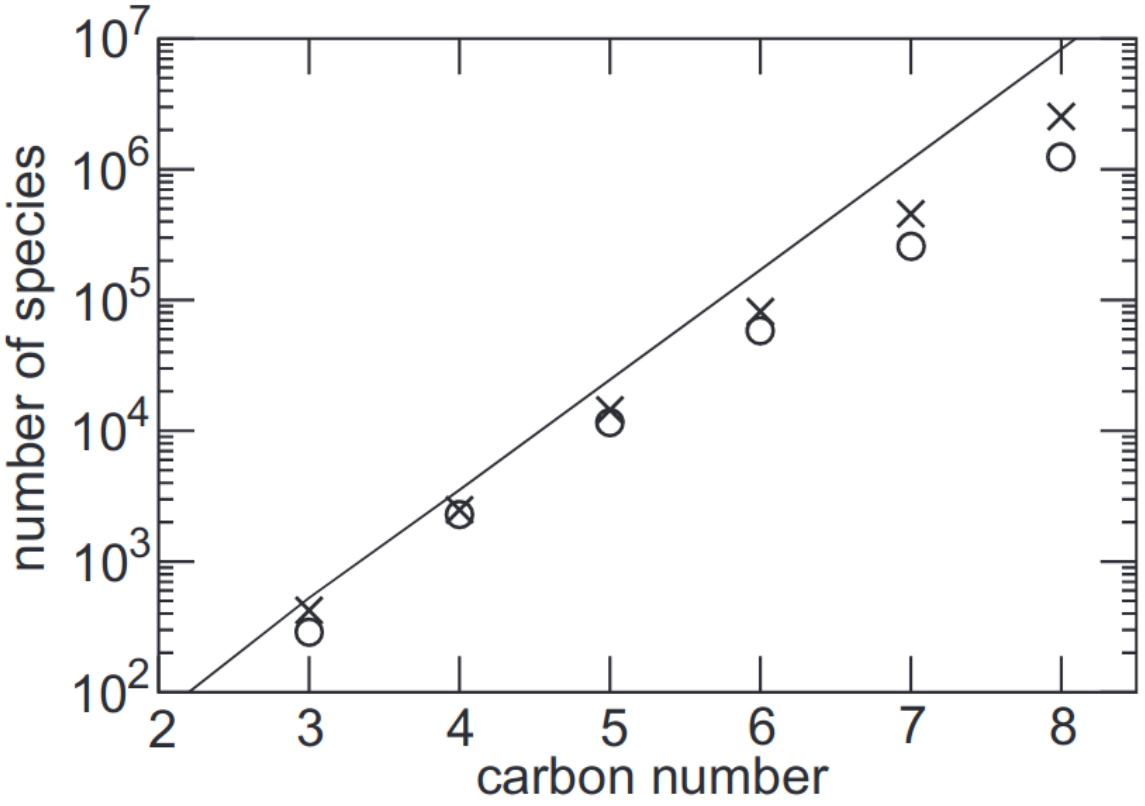

Potential number of distinct species in a full oxidation mechanism as a function of the size of the precursor

Aumont et al. (2005)

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We need to go faster

Writing the chemical mechanism

Solving the ODEs in a (3D) model

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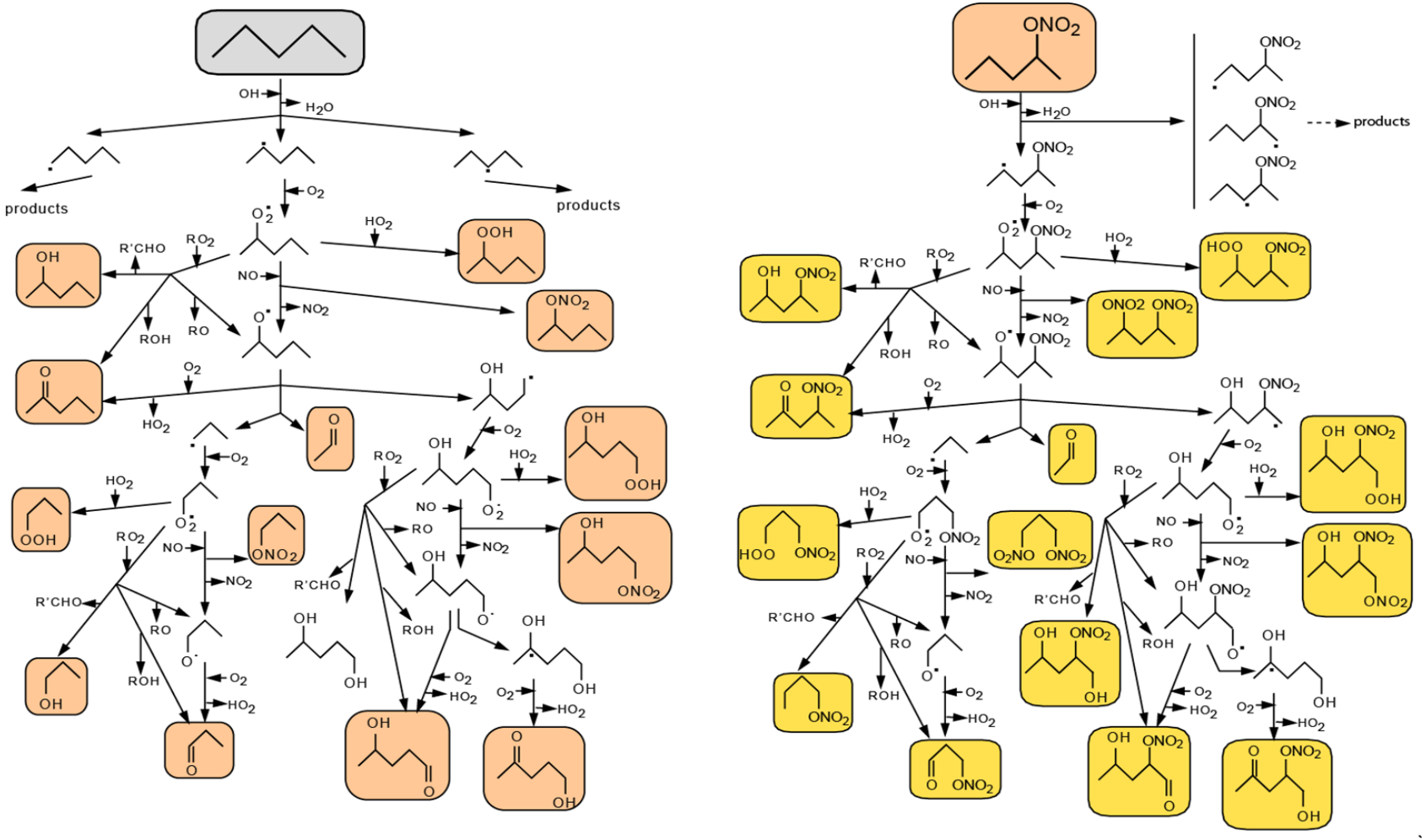
Automating boring things



Systematic and repeated oxidation steps

Generator for Explicit Chemistry and Kinetics of Organics in the Atmosphere (GECKO-A, Aumont et al., 2005)

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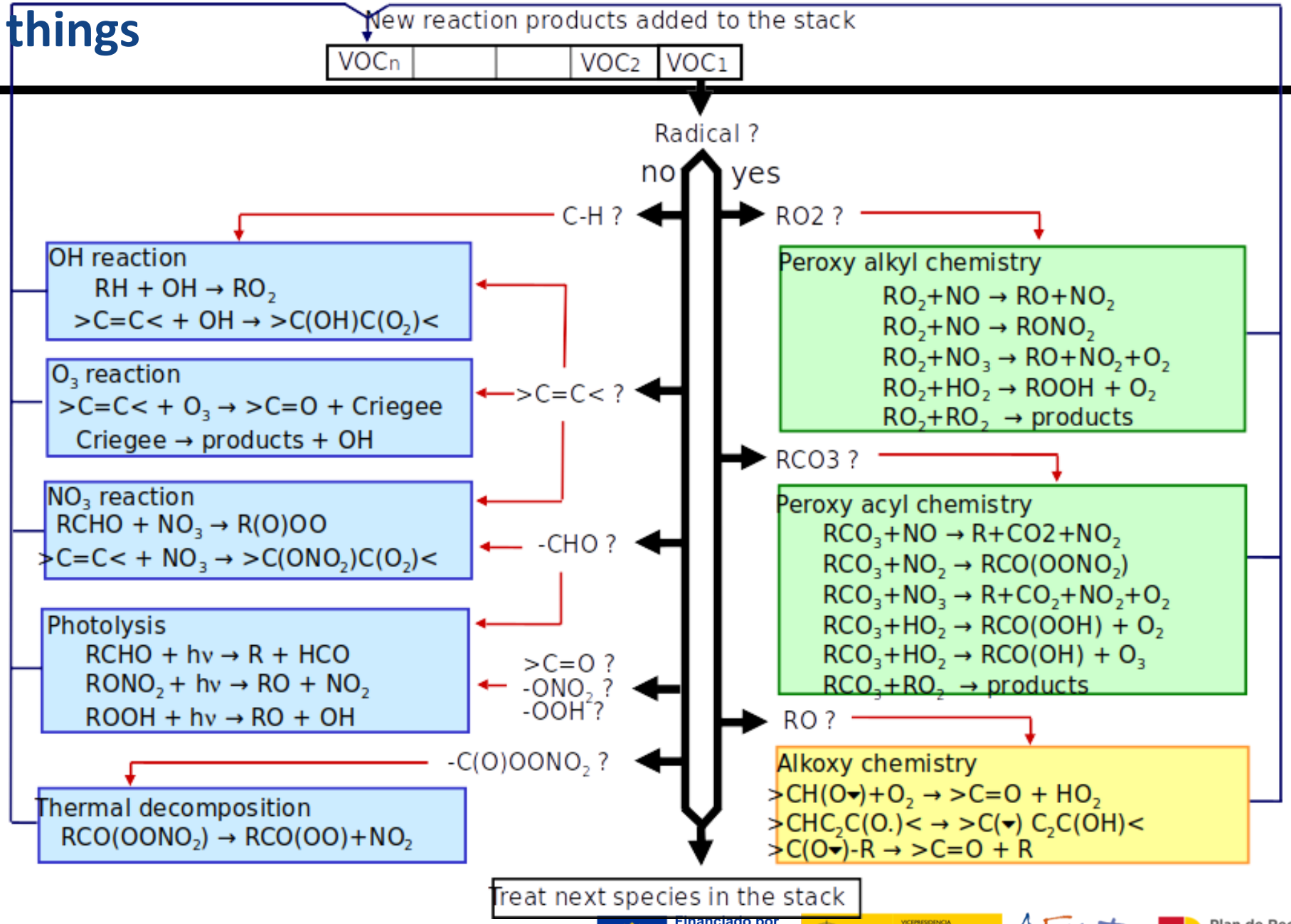
Automating boring things

Aumont et al., 2005



Accelerating things

Writing the chemical mechanism

GECKO-A

12 biogenic, 53 anthropogenic precursors

23 million reactions involving 4.4 million species

(Mouchel-Vallon et al., 2020)

Solving the ODEs in a (3D) model

Simulating 2 days in 2 grid cells

≈ 36 hours on 16 cores

Machine learning?

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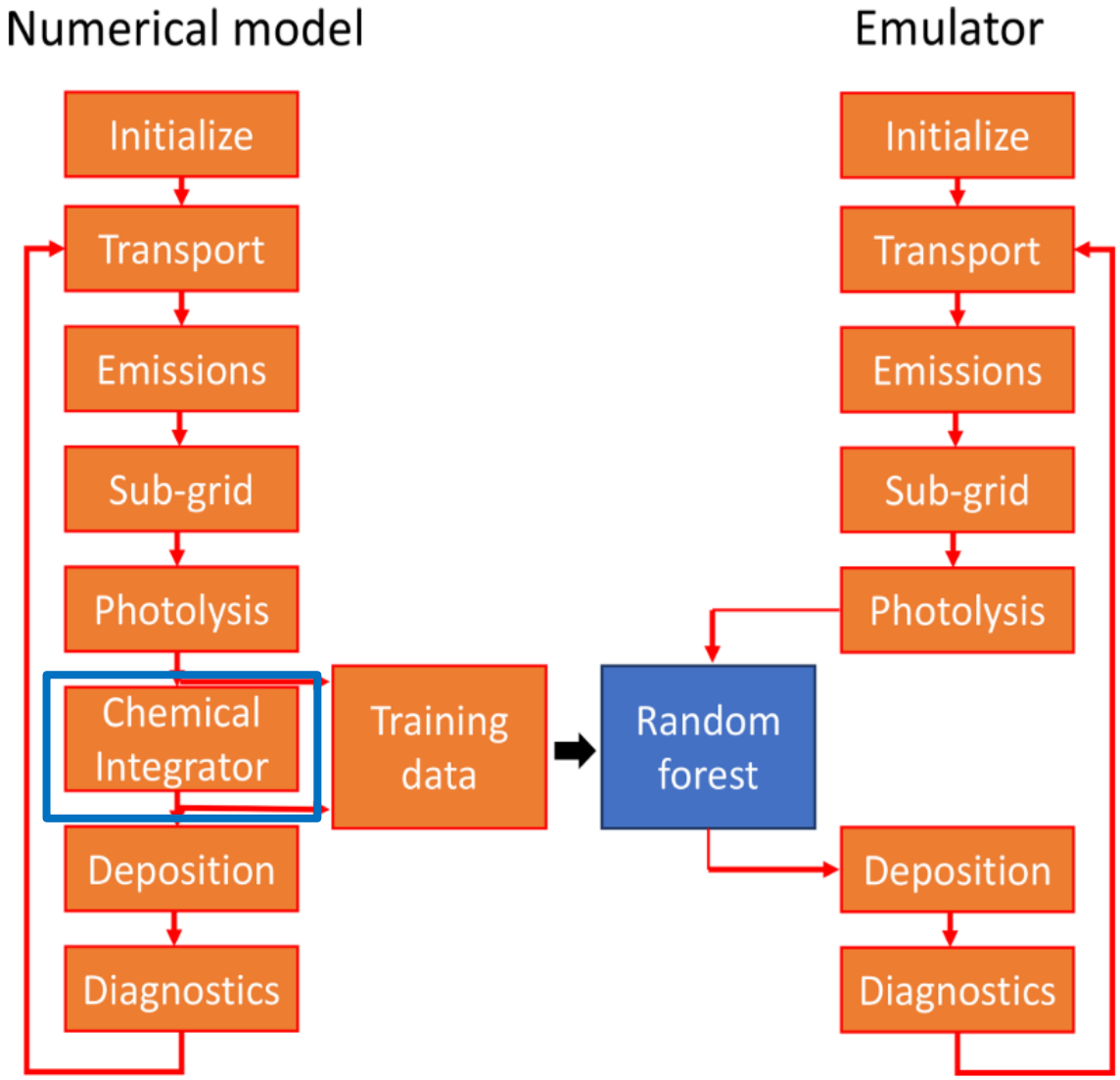
Emulating atmospheric chemistry



Computationally expensive

Use GECKO-A 0D explicit simulations as training dataset

Emulate the behavior of the detailed model with machine learning

- **Neural Networks (NN)**
- **Random Forest (RF)**

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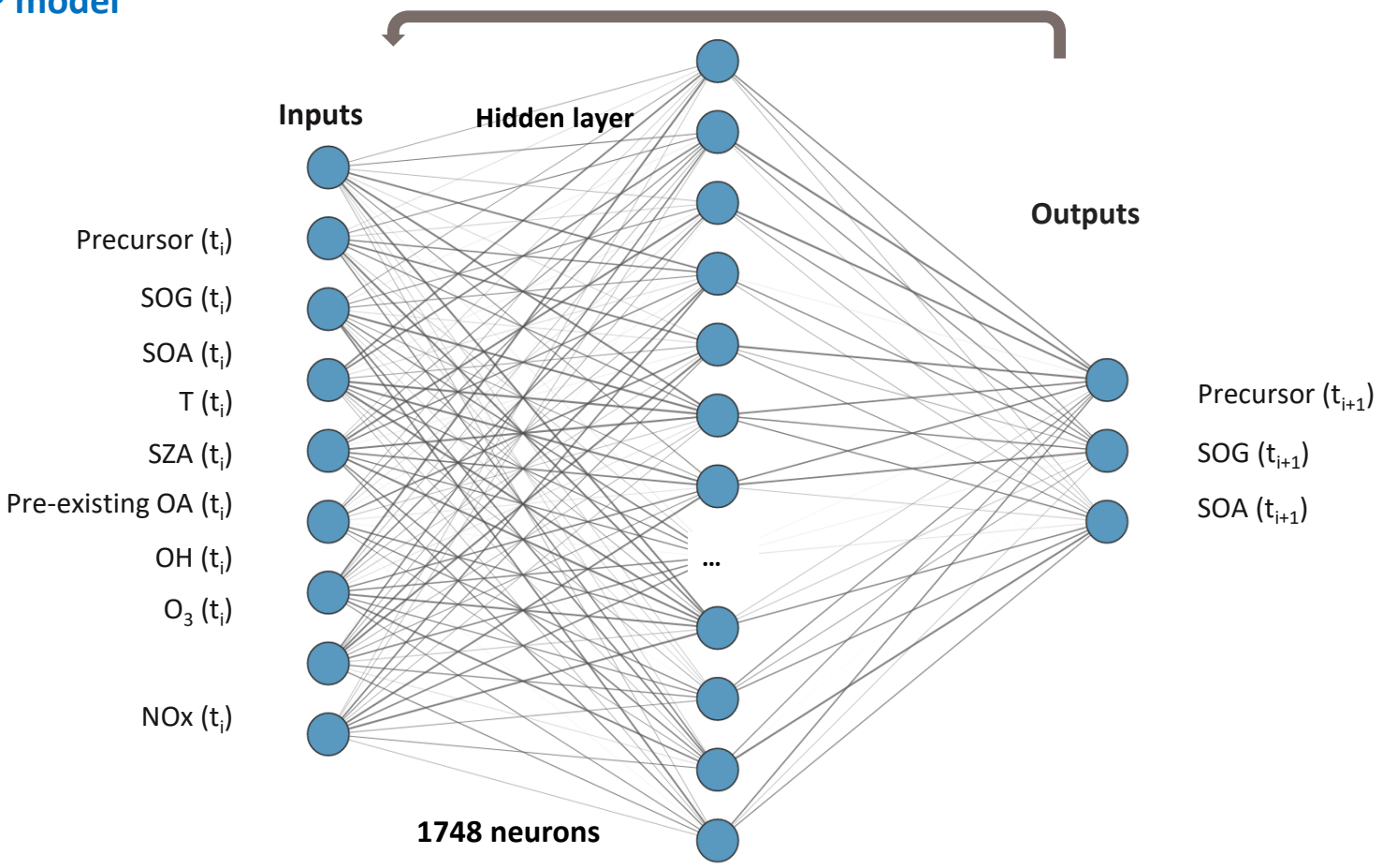
NN approach for predicting time-series of concentrations

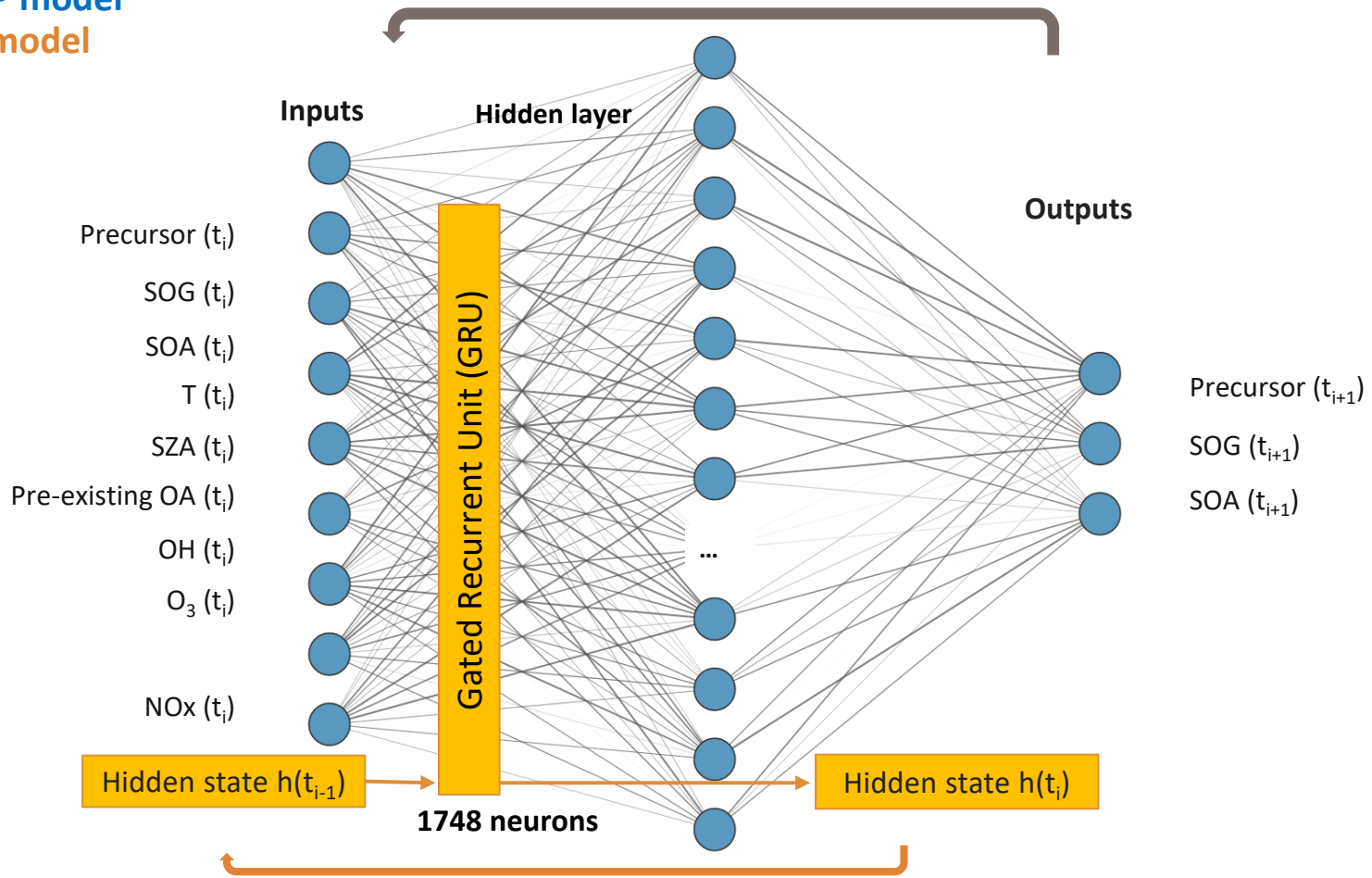
1. Multi-layer perceptron MLP model



Schreck et al. JAMES 2022: Neural network emulation of the formation of OA based on the explicit GECKO-A chemistry model

NN approach for predicting time-series of concentrations

1. Multi-layer perceptron MLP model
2. Gated-recurrent unit GRU model



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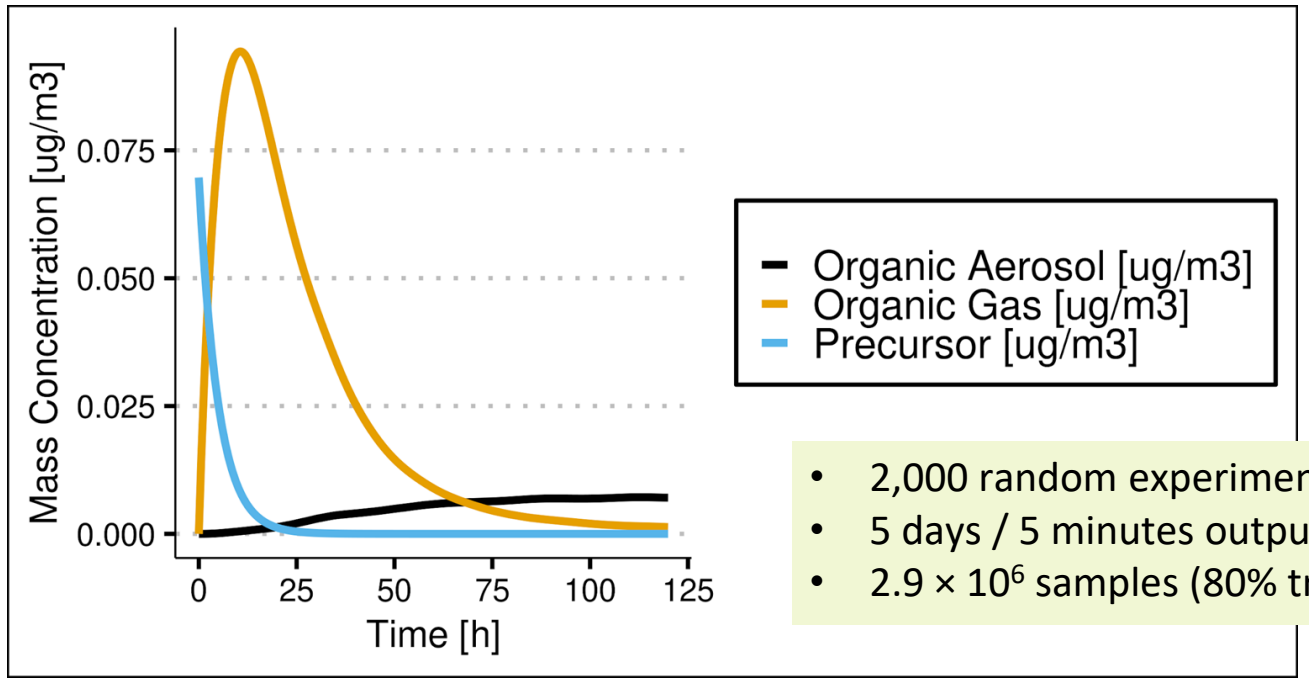
Generate Neural Network training dataset with GECKO-A

- 3 precursors: **toluene**, **dodecane**, **a-pinene**
- Random environmental conditions
- No diurnal variations
- Initial precursor amounts of 10 ppt, 0.1 and 1 ppb

Temperature	240 - 320 K	Uniform
Solar zenith angle (SZA)	0-90 degrees	Uniform
Pre-existing aerosols	0.01-10 $\mu g/m^3$	Logarithmic
Ozone	1 - 150 ppb	Uniform
NOx	0.01-10 ppb	Logarithmic
OH	10^1–10^6 molecules/cm^3	Uniform



- 2,000 random experiments for each precursor
- 5 days / 5 minutes output
- 2.9×10^6 samples (80% training, 20% validation)

Schreck et al., JAMES, 2022

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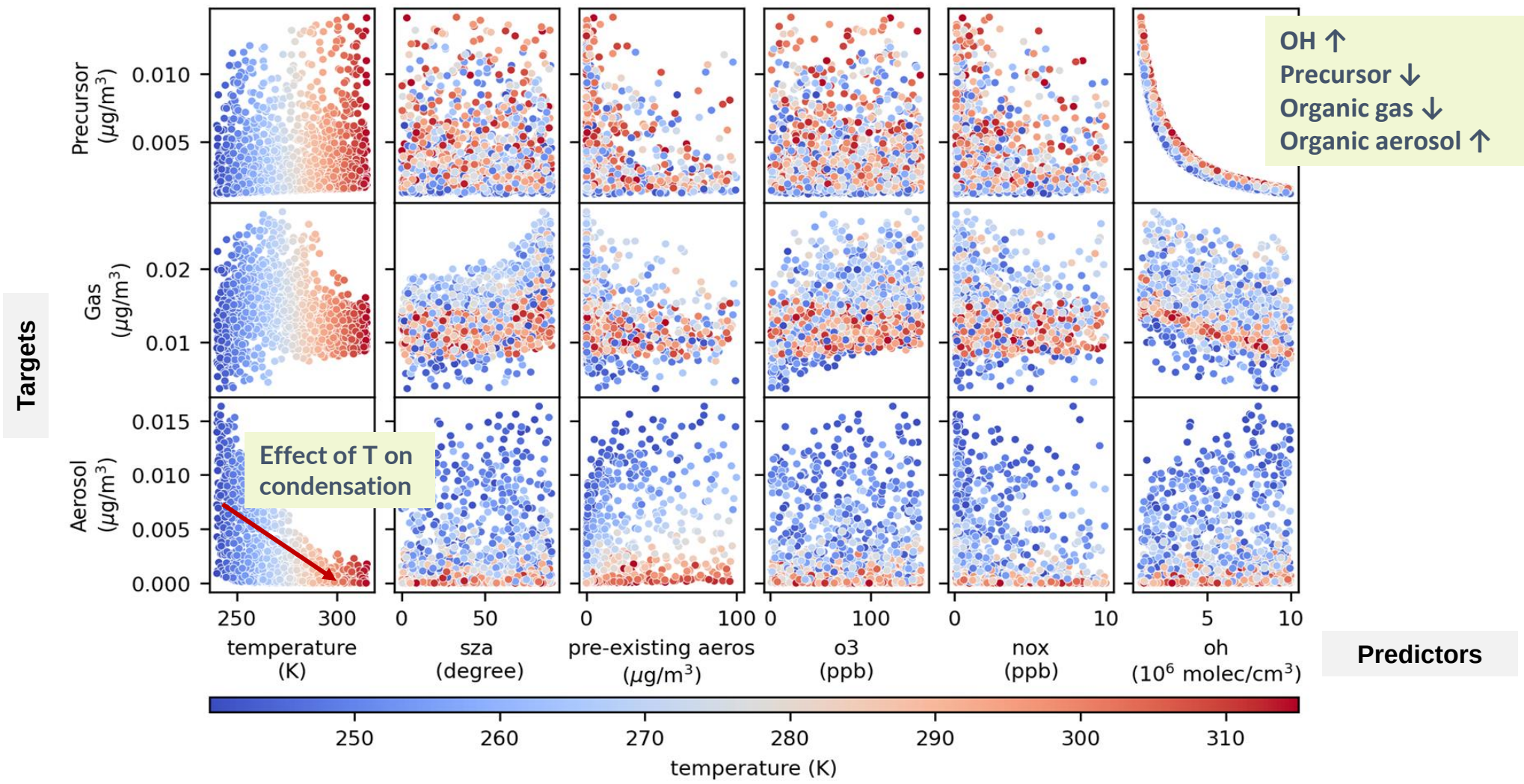
Neural Network training dataset



Schreck et al., JAMES, 2022

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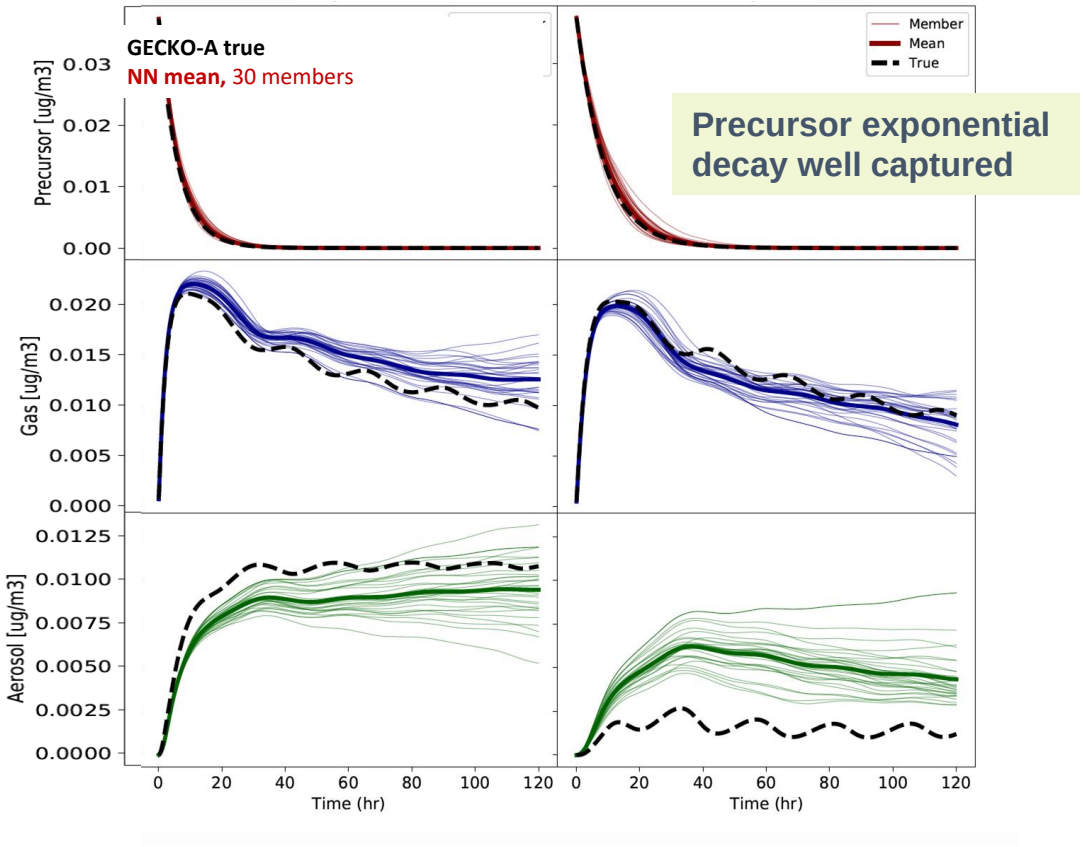
Multi-Layer Perceptron vs. Gated-Recurrent Unit network

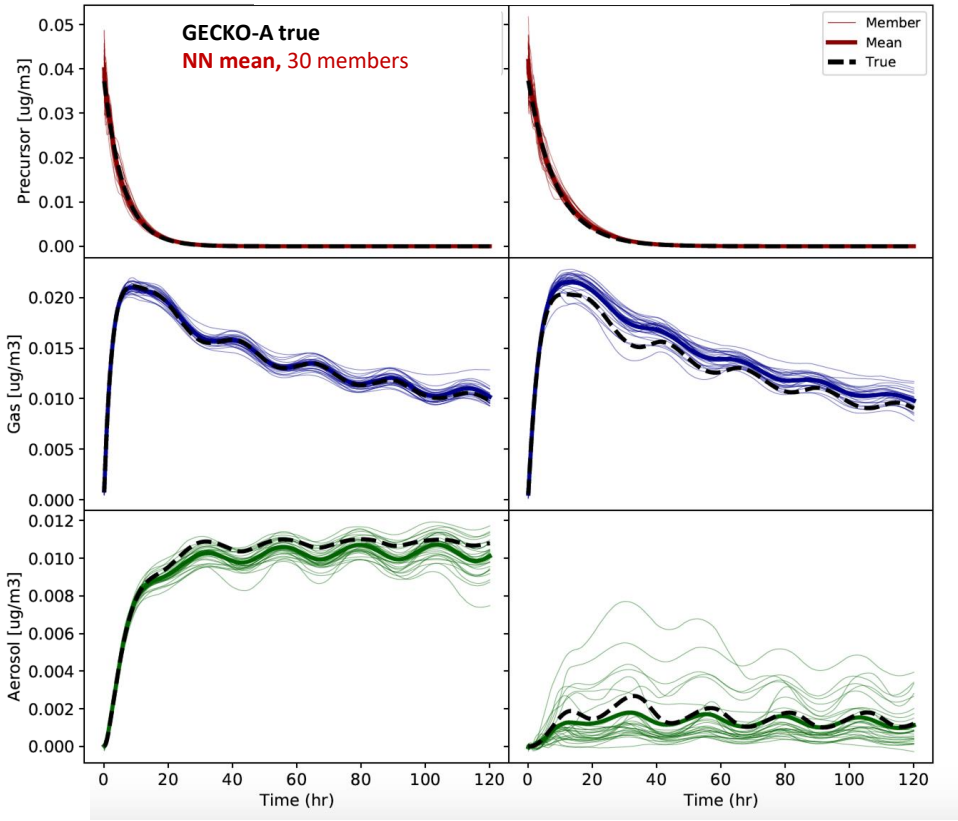
30-ensemble member predictions for Toluene

Multi-layer perceptron



Precursor exponential decay well captured

Gated-recurrent unit



⇒GRU performs better but is challenging to implement in a 3D model

Schreck et al., JAMES, 2022

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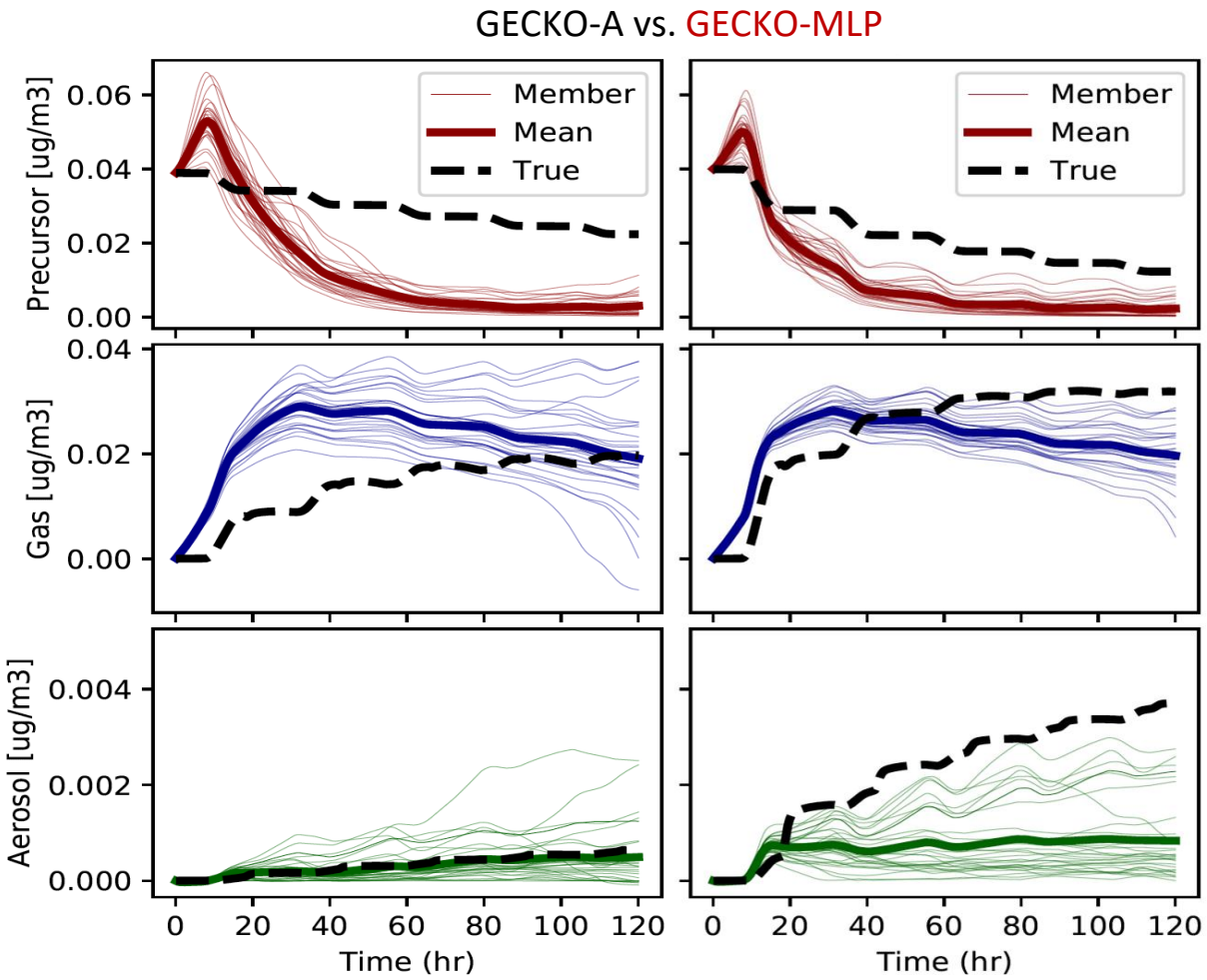
Application in a box model for diurnally varying conditions



⇒ Neural Networks trained on datasets built under constant conditions cannot reproduce realistic diurnal cycles

⇒ Raises the question of the representativeness of the training data sets

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Computational gain GECKO-A vs. GECKO-NN

- For Toluene, GECKO-NN is 4×10^2 times faster than GECKO-A on CPU, and 10^4 times faster on GPU

Model	Toluene		Dodecane		α-pinene	
GECKO-A	0.9 s	1	71 s	1	220 s	1
MLP CPU	2.1 µs	430	0.8 µs	8.88×10^4	1.6 µs	1.38×10^5
MLP GPU	0.08 µs	11250	0.07 µs	1.01×10^6	0.08 µs	2.75×10^6
GRU CPU	3.1 µs	290	3.2 µs	2.22×10^4	3.3 µs	6.67×10^4
GRU GPU	0.38 µs	2368	0.38 µs	1.87×10^5	0.38 µs	5.79×10^5

Ratio of GECKO-A / GECKO-NN

Timing per 5min timestep

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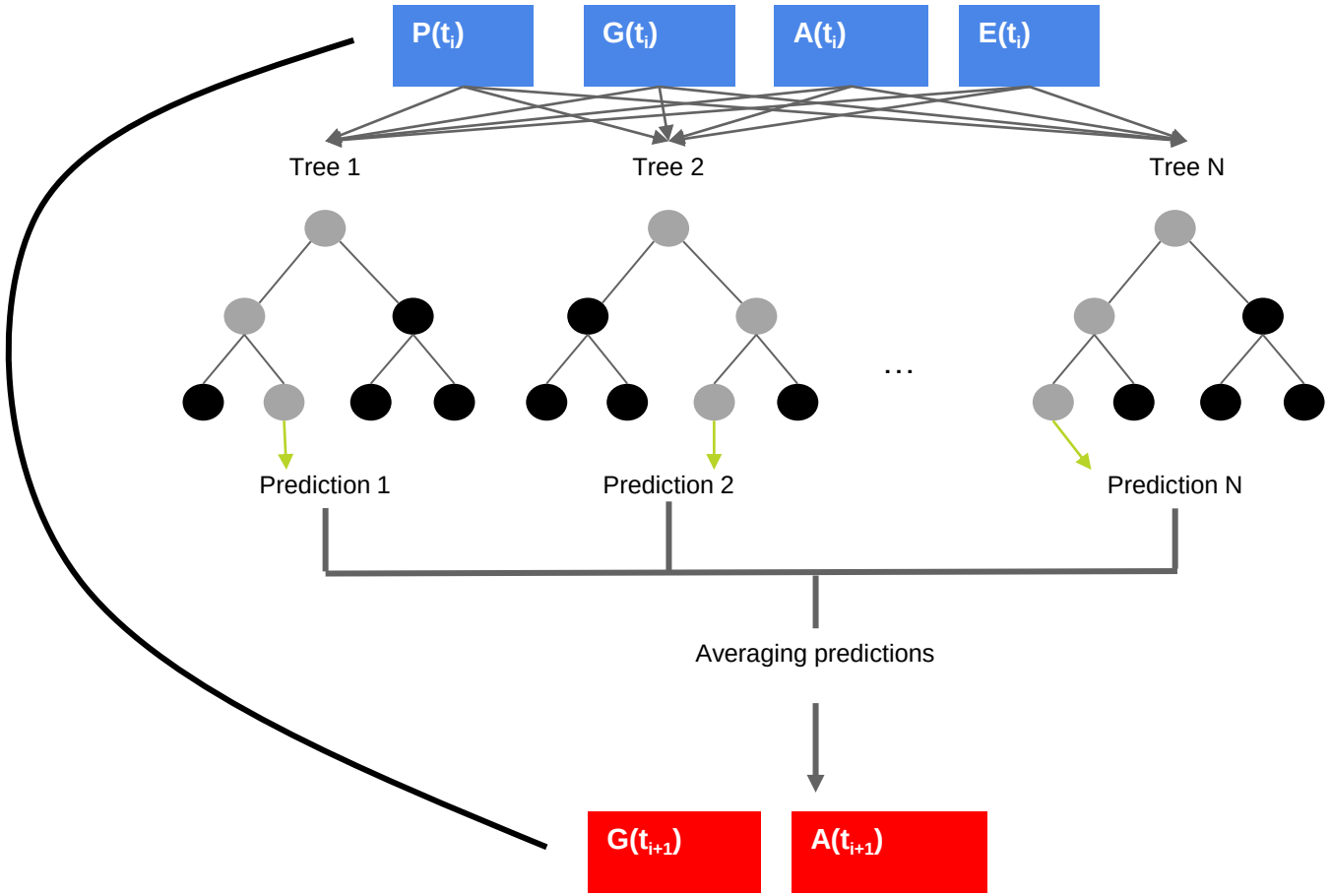
Random Forest approach



Inputs :

- Precursor concentration
- Gas organic concentration
- Aerosol organic concentration
- Temperature
- Solar zenith angle
- Pre-existing aerosol
- O_3
- NOx
- OH

$P(t_i)$ $G(t_i)$ $A(t_i)$ $E(t_i)$

Output :

- Gas organic concentration
- Aerosol organic concentration

$G(t_{i+1})$ $A(t_{i+1})$

Mouchel-Vallon & Hodzic, JGR, 2023:
Toward emulating an explicit organic chemistry mechanism with random forest models.

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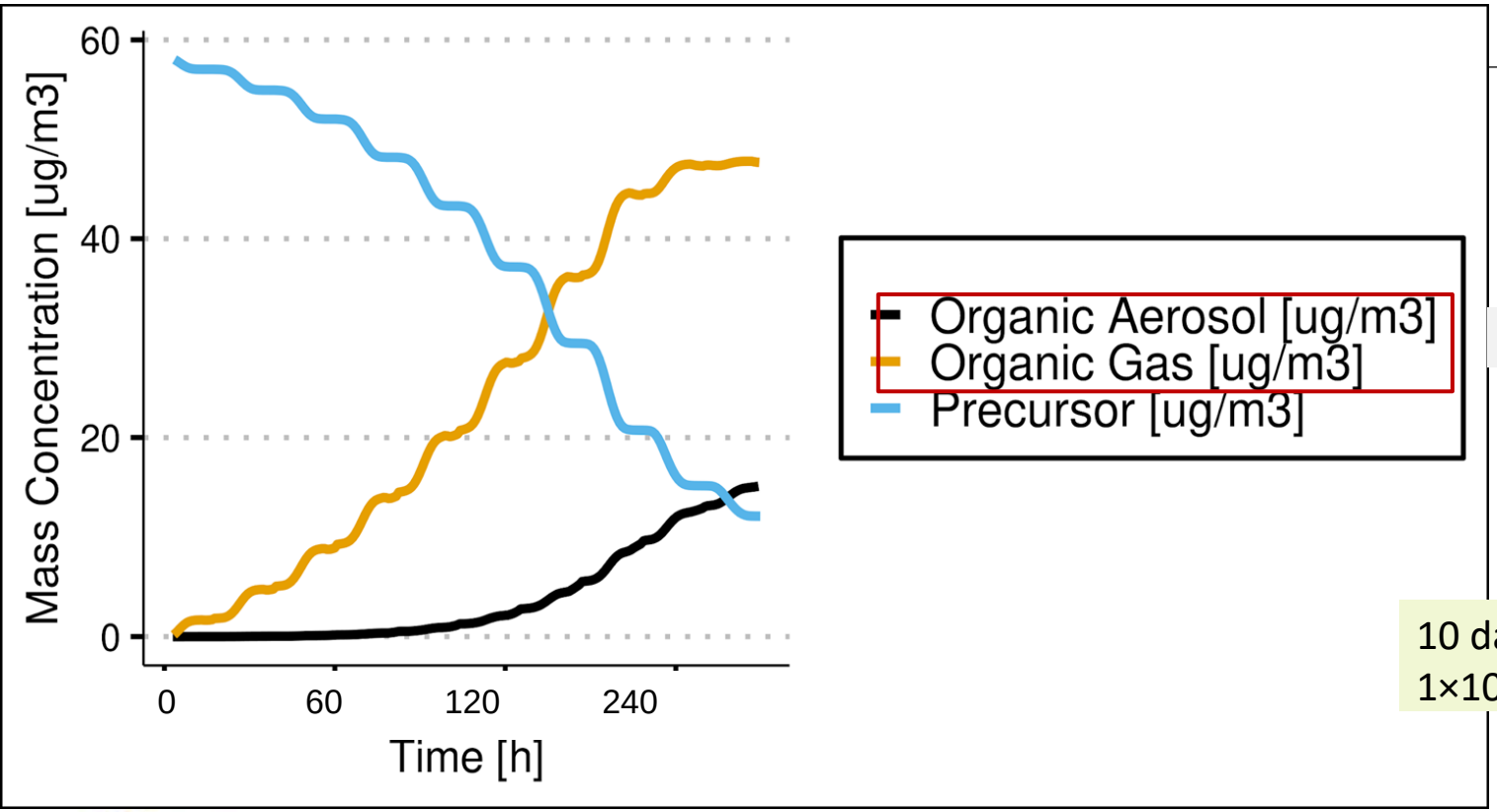
Random Forest: Realistic training dataset

- 2 precursors: **toluene**, **dodecane**
- Random environmental conditions
- With diurnal variations
- Random initial precursor concentration

Parameter	Range	Parameter	Range
Latitude [°]	80S–80N	Relative humidity [%]	3–102
Temperature [K]	216–313	Atmospheric pressure [atm]	0.5–1.02
Preexisting aerosol seed [μg m^{-3}]	0.03–340	Initial NO_x [ppb]	10^{-4}–42
Initial precursor [ppb]	0–16	Initial CO	33–1012
Initial O_3 [ppb]	1–100	NO Emission [molec cm^{-2} s^{-1}]	10^{7}–10^{9}

From 3D model tropospheric ranges



Only 2 targets

10 days / 5 min output
1×10^6 samples/precursor

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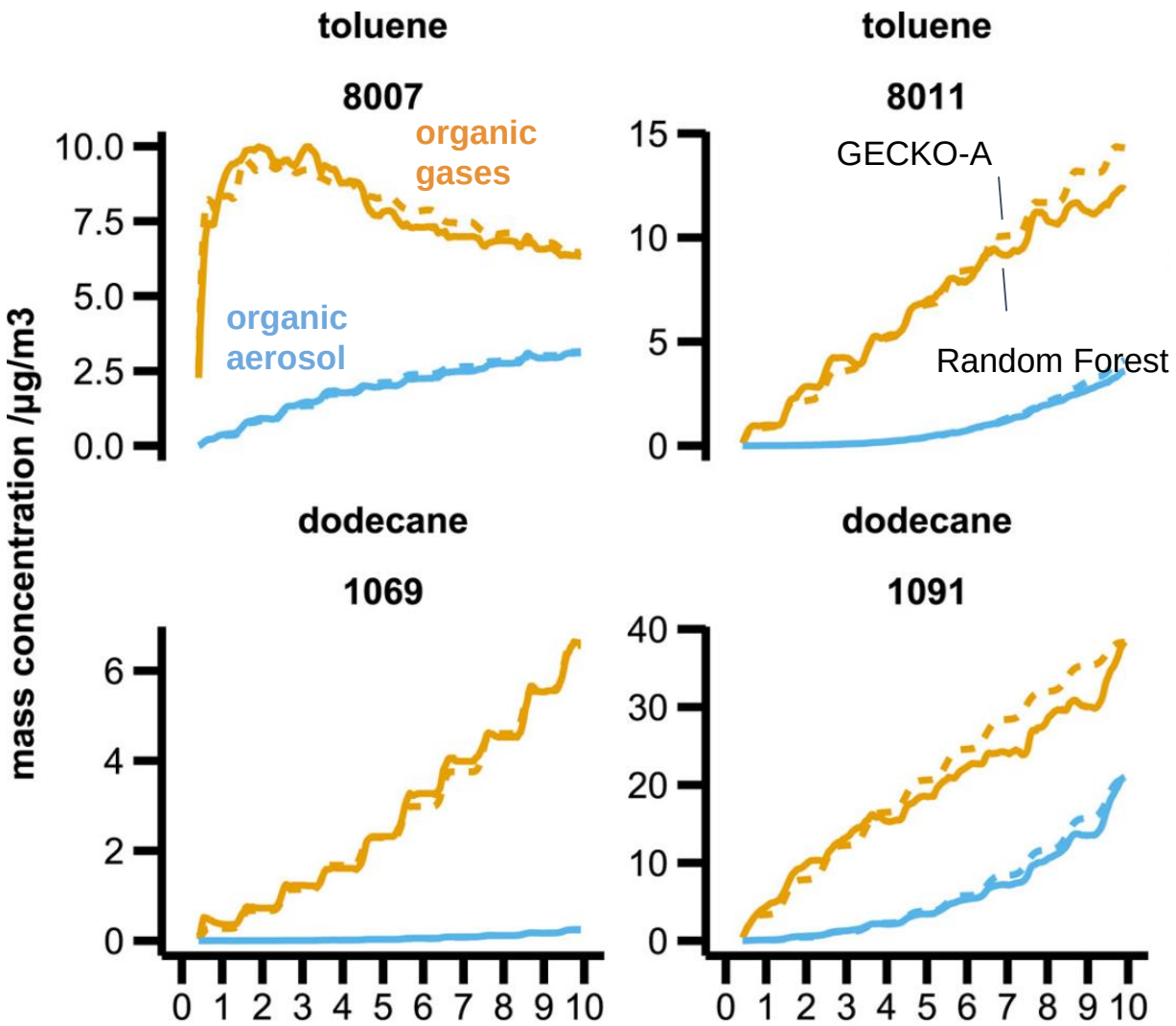
Random Forest results: examples



RF perform similarly to NN
The training set allows reproducing the diurnal cycles

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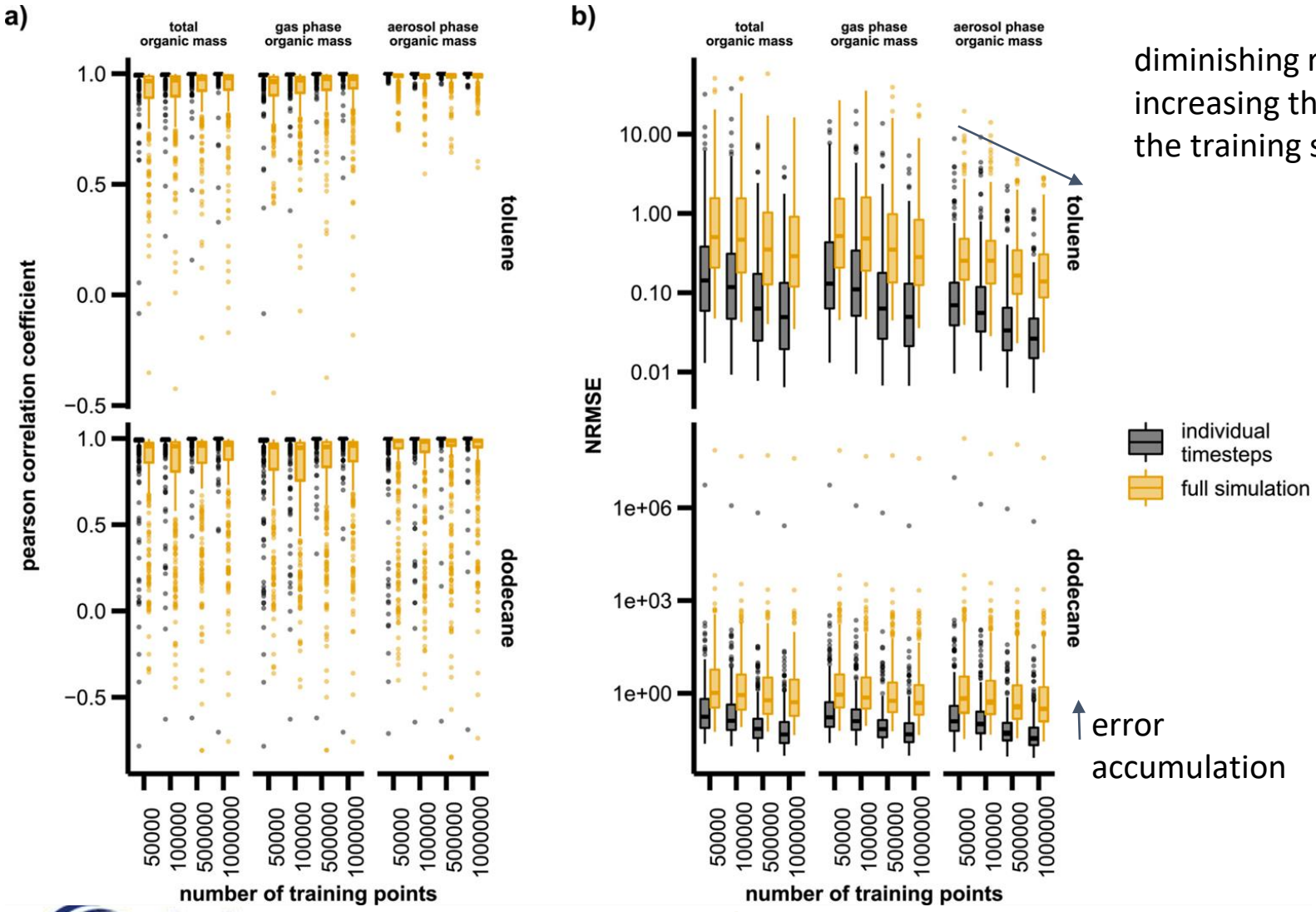
Random Forest results: performance



diminishing returns of increasing the size of the training sample

error accumulation

Increasing the number of training samples improves performance

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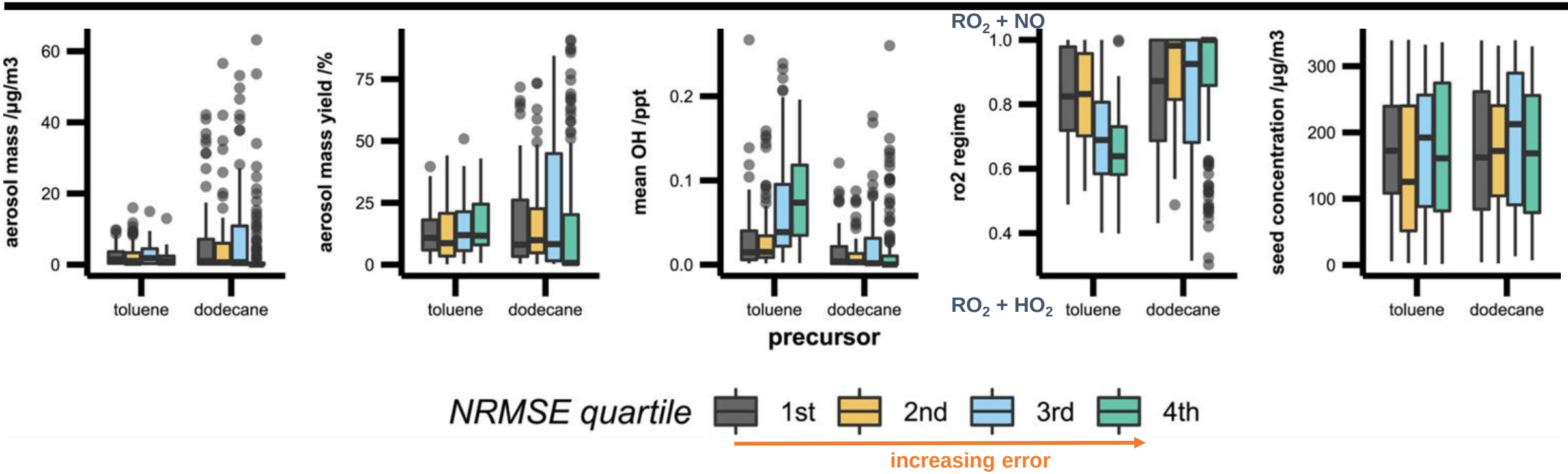
Random Forest results: errors distribution



Little sensitivity of errors to the aerosol quantity to predict (seed, yield, mass)
Errors sensitive to NOx regime and OH mixing ratios: underrepresented regimes in training set

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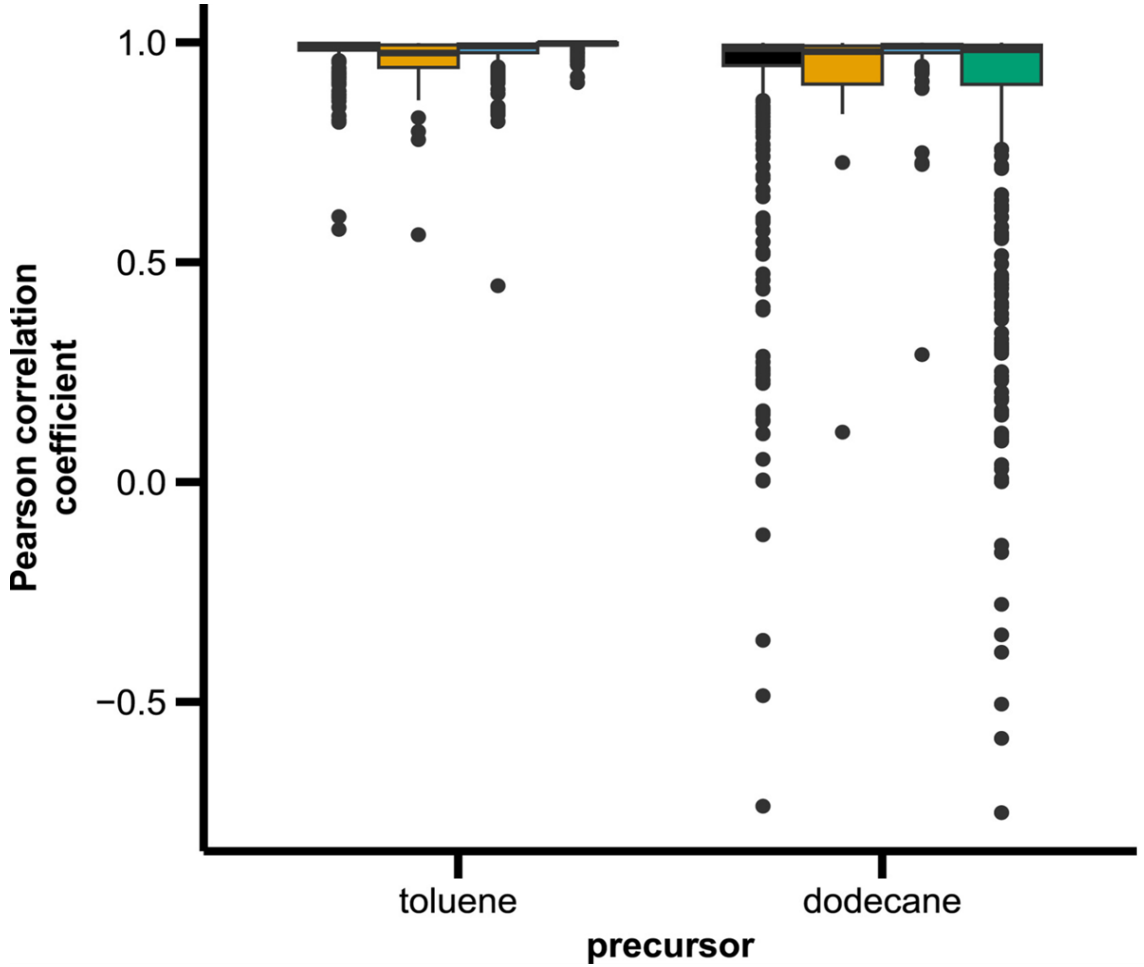
Random Forest results: specializing random forests

Do performances improve if the range of chemical regimes to predict is reduced?



random forest

- original
- low nox
- mid nox
- high nox

# training points	
toluene	dodecane
1×10^5	7×10^4
6×10^5	3×10^5
4×10^5	7×10^5

Specialization requires **more** or **better constructed** training data

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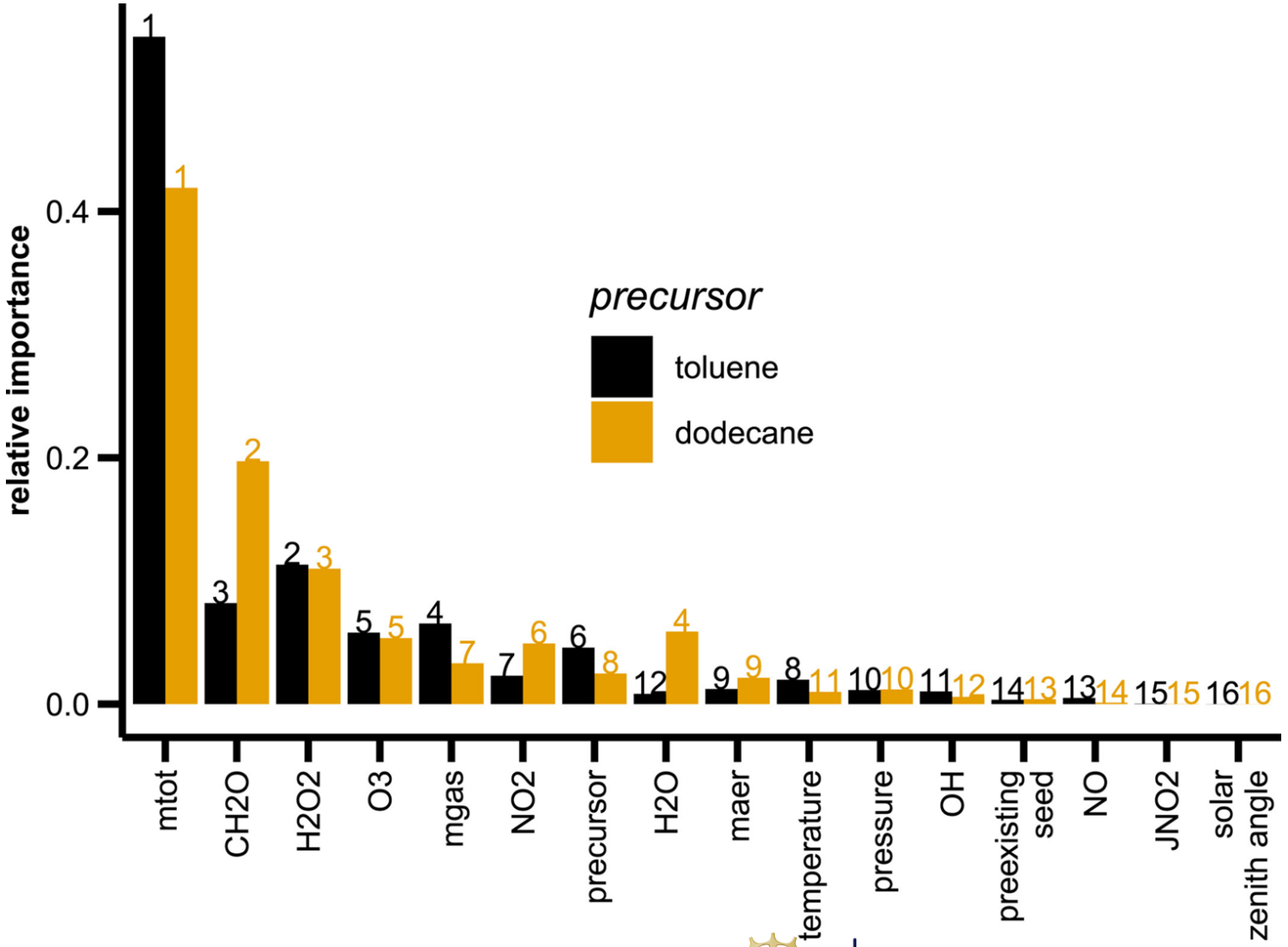
Random Forest results: predictors relative importance



The importance of predictors (mean decrease impurity) is similar for **both** species

Distribution of importance and predictive ability among **correlated variables**

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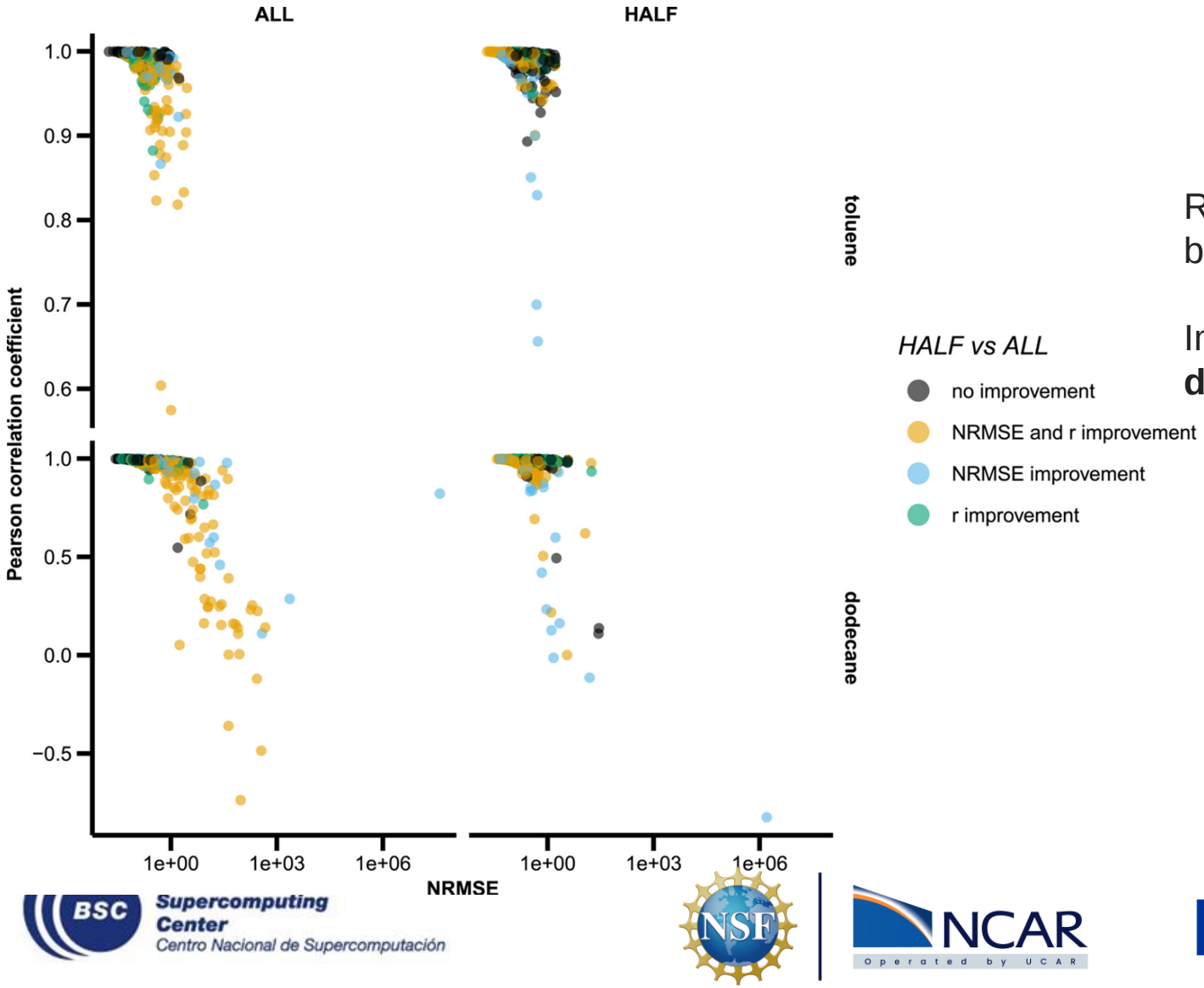
Random Forest results: reducing the number of predictors

Do performances improve if the number of predictor is reduced?



Reducing the number of (**correlated?**) predictors is beneficial for the worst performing simulations

Importance of **selecting predictors** and **dimensionality reduction**

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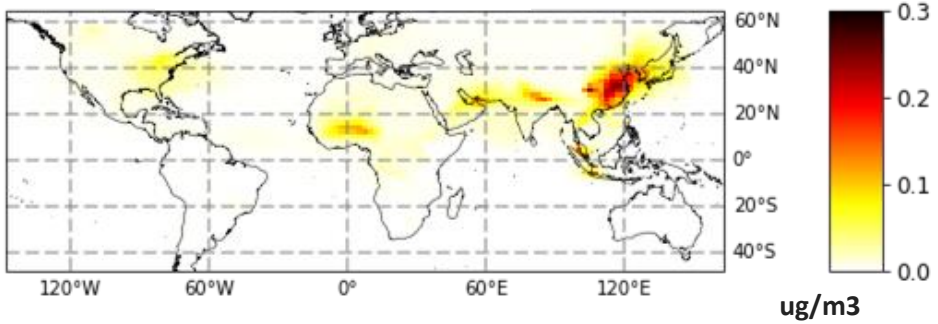
First tests in a global model: VBS vs. GECKO-NN

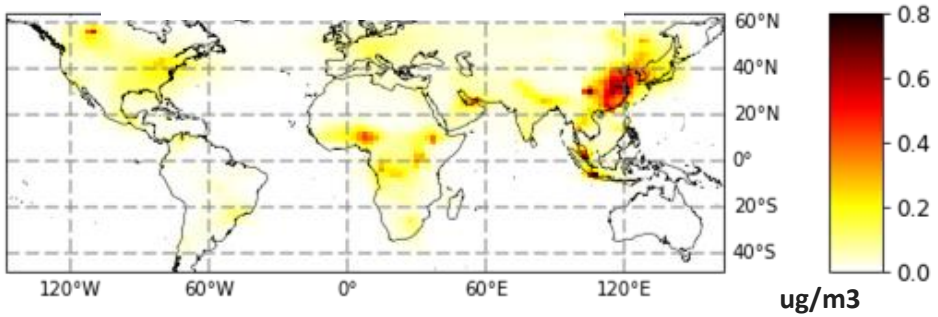
Implementing GECKO-NN in Geos-Chem

GEOS-Chem Monthly average Toluene SOA (May 2016)

GECKO-NN-MLP



VBS Hodzic et al., 2016



GECKO-NN simulations are

- stable over several months
- within a factor of 2 of the VBS parametrization for Toluene-SOA.

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Conclusions and Outlook

- It is possible to emulate the behavior of detailed atmospheric chemistry models with machine learning
- Long term stability can be achieved for recurrent neural networks with GRU
- The training dataset must be carefully constructed to cover all environmental conditions
- Random forests can perform similarly to NN+GRU
- Predictors selection is crucial

Current and future works at BSC

- Bring the complexity of organic chemistry to air quality models, built on the development of detailed Spanish emissions
- Explore ML use, with lessons learned from this work: start again from the basics and systematic exploration by (i) progressively increasing chemical complexity (from toy mechanisms to GECKO-A complexity) and (ii) testing multiple families of ML techniques (RF, NN, GraphNet …)
- Implementation in the MONARCH air quality model

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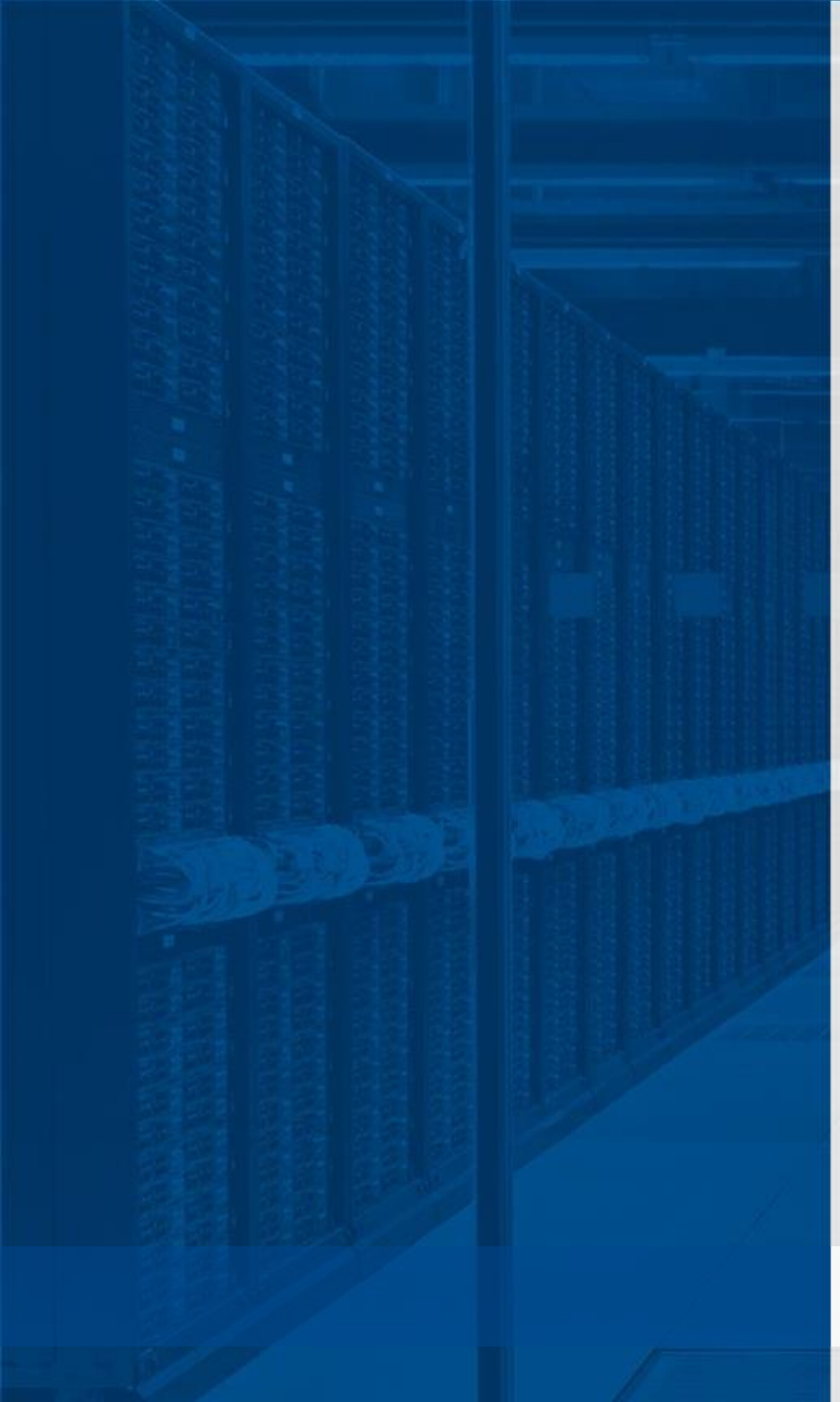

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Thank you for your attention

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