# Revisiting non-stomatal VOC dry deposition: Ecosystem-scale insights into humidity and physicochemical controls



# Revisiting non-stomatal VOC dry deposition:
## Ecosystem-scale insights into humidity and physicochemical controls



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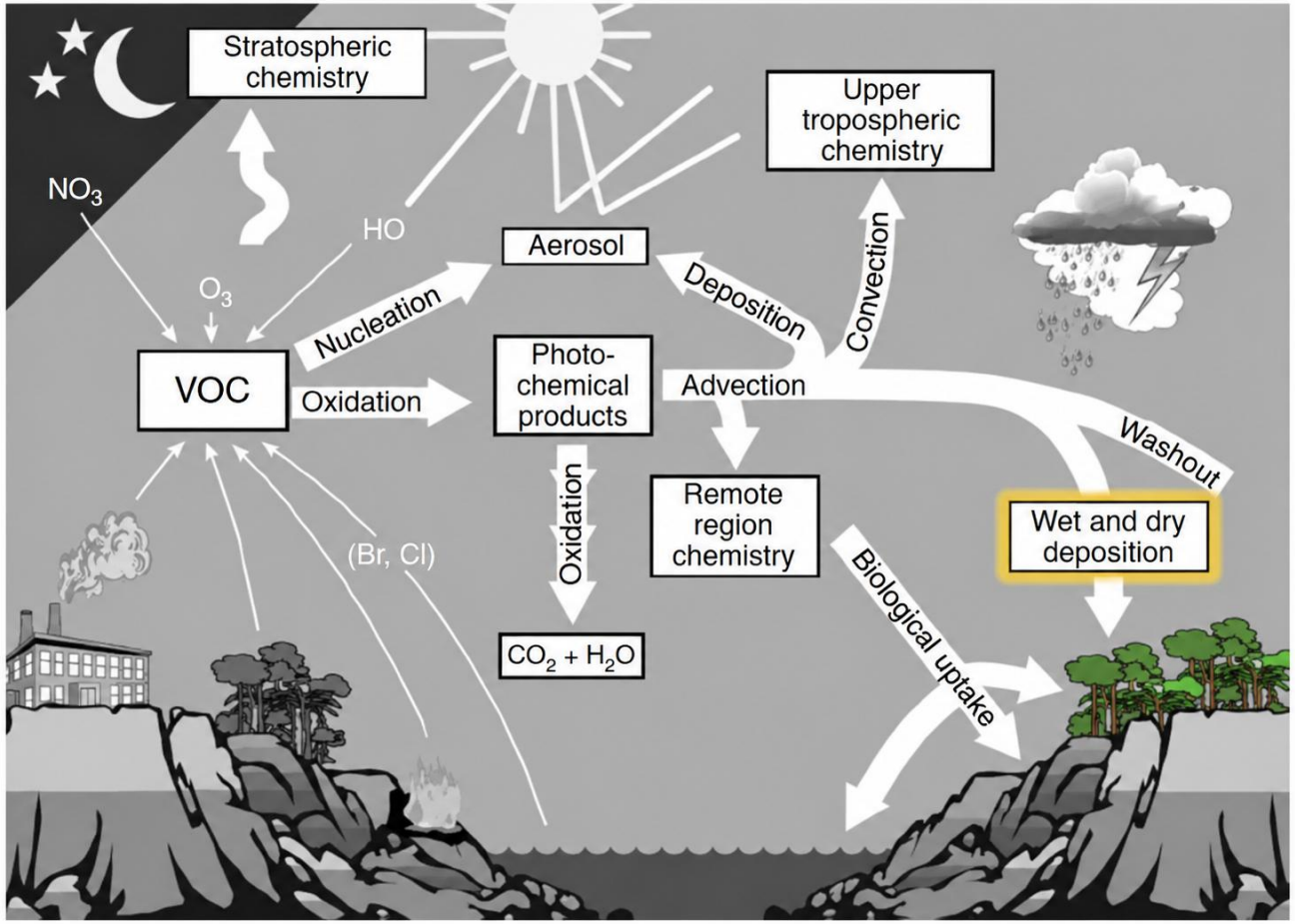
# Importance of dry deposition




Williams & Koppmann (2007)

- Dry deposition
  = important sink in global budgets

**The global nonmethane reactive organic carbon budget: A modeling perspective**

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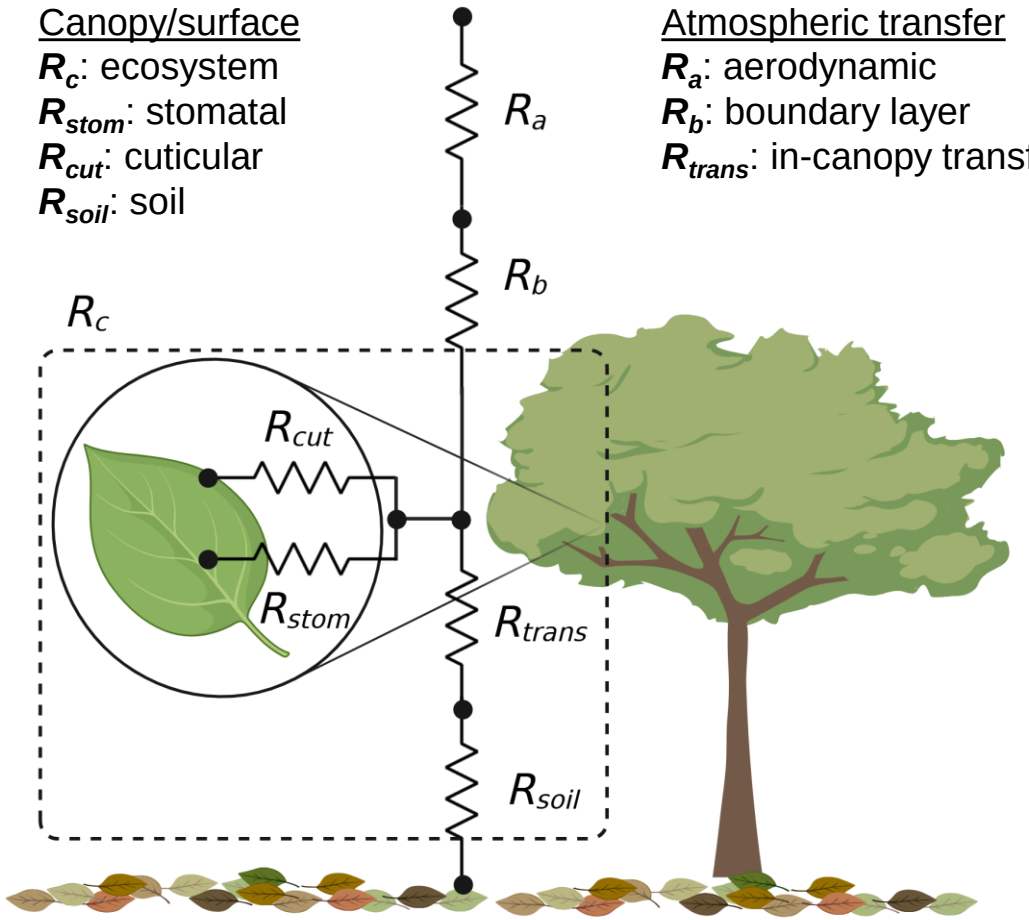
[1]Department of Civil and Environmental Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts, USA, [2]Office of Air Quality Planning and Standards, U.S. EPA, Research Triangle Park, North Carolina, USA

**Abstract** The cycling of reactive organic carbon (ROC) is central to tropospheric chemistry. We characterize the global tropospheric ROC budget as simulated with the GEOS-Chem model. We expand the standard simulation by including new emissions and gas-phase chemistry, an expansion of dry and wet removal, and a mass tracking of all ROC species to achieve carbon closure. The resulting global annual mean ROC burden is 16 Tg C, with sources from methane oxidation and direct emissions contributing 415 and 935 Tg C $yr^{-1}$. ROC is lost from the atmosphere via physical deposition (460 Tg C $yr^{-1}$) and oxidation to $CO/CO_2$ (875 Tg C $yr^{-1}$). Ketones, alkanes, alkenes, and aromatic hydrocarbons dominate the ROC burden, whereas aldehydes and isoprene dominate the ROC global mean surface OH reactivity. Simulated OH reactivities are between 0.8–1 $s^{-1}$, 3–14 $s^{-1}$, and 12–34 $s^{-1}$ over selected regions in the remote ocean, continental midlatitudes, and the tropics, respectively, and are consistent with observational constraints.

*Safieddine et al. (2017, GRL)*

- Dry deposition is greatest over forested ecosystems

# How do we usually model dry deposition?




$$v_d = \frac{-flux}{conc} = \frac{1}{R_a + R_b + R_c}$$

- Stomatal: relatively well constrained ($H_2O$ observations)
- Non-stomatal:

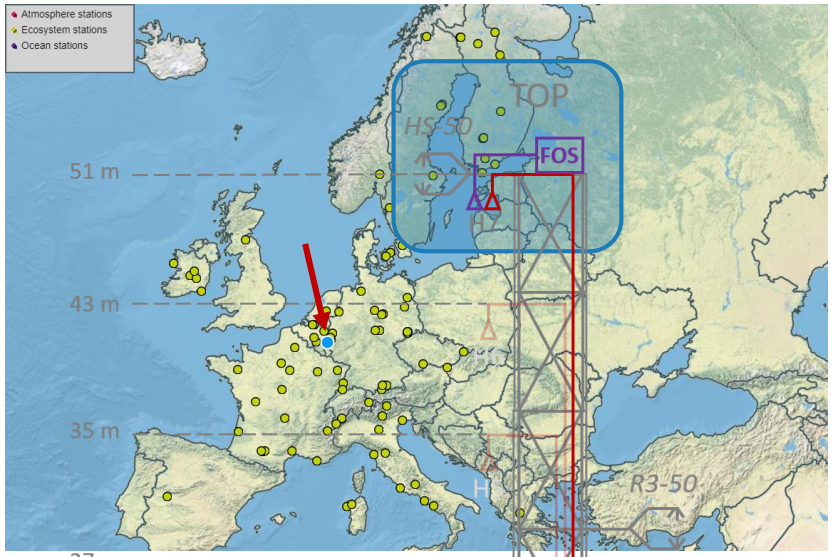
$$G_{cut,VOC_i} = \alpha_i G_{cut,SO_2} + \beta_i G_{cut,O_3}$$
$$G_{soil,VOC_i} = \alpha_i G_{soil,SO_2} + \beta_i G_{soil,O_3}$$

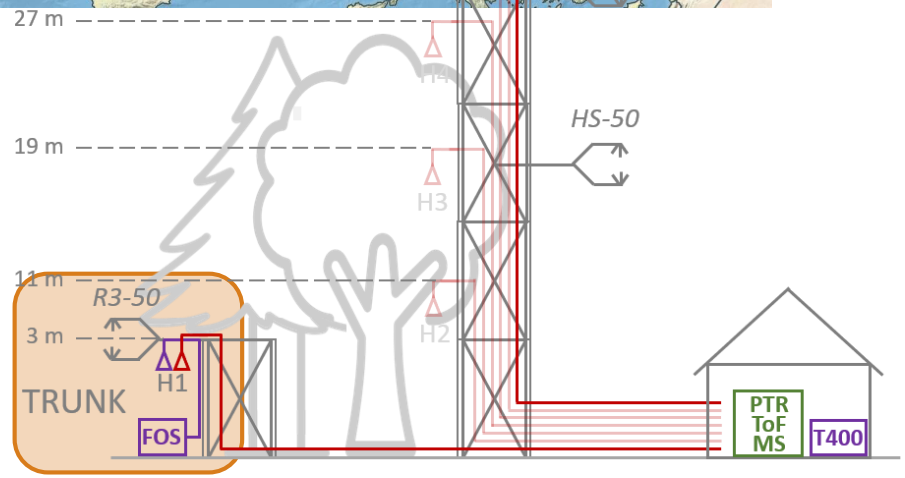
  - With $\alpha_i$ = solubility $\beta_i$ = reactivity
  - With $G_{cut/soil,SO_2/O_3}$ cst or dynamic: $f(RH, u_*, LAI)$

Questions

1. Do VOCs behave like $SO_2$ and $O_3$?
2. Are we capturing the relevant environmental responses?
3. Are solubility and reactivity the only mechanisms of surface deposition?

# VOC and $O_3$ measurements and processing




## Vielsalm station

- Mixed temperate forest (beech, douglas fir, Norway spruce, silver fir), rural environment
- ICOS network (Belgian Ardenne region)

ICOS Integrated Carbon Observation System

## Measurements

- 2022 – 2023 – 2024, from Spring to Autumn
- PTR-ToF-MS for VOCs, T400 and FOS for $O_3$
- Fluxes and concentrations above the canopy (TOP) and above the soil level (TRUNK) (+ concentration profile)
- 10 VOCs with significant deposition

*Dumont et al. (2026, ESSD)*

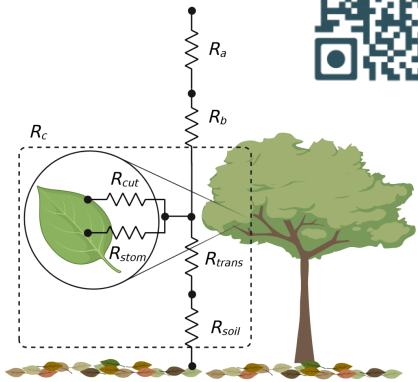
## Flux partitioning

- TRUNK measurements:

$$G_{c,trunk} = G_{soil}$$

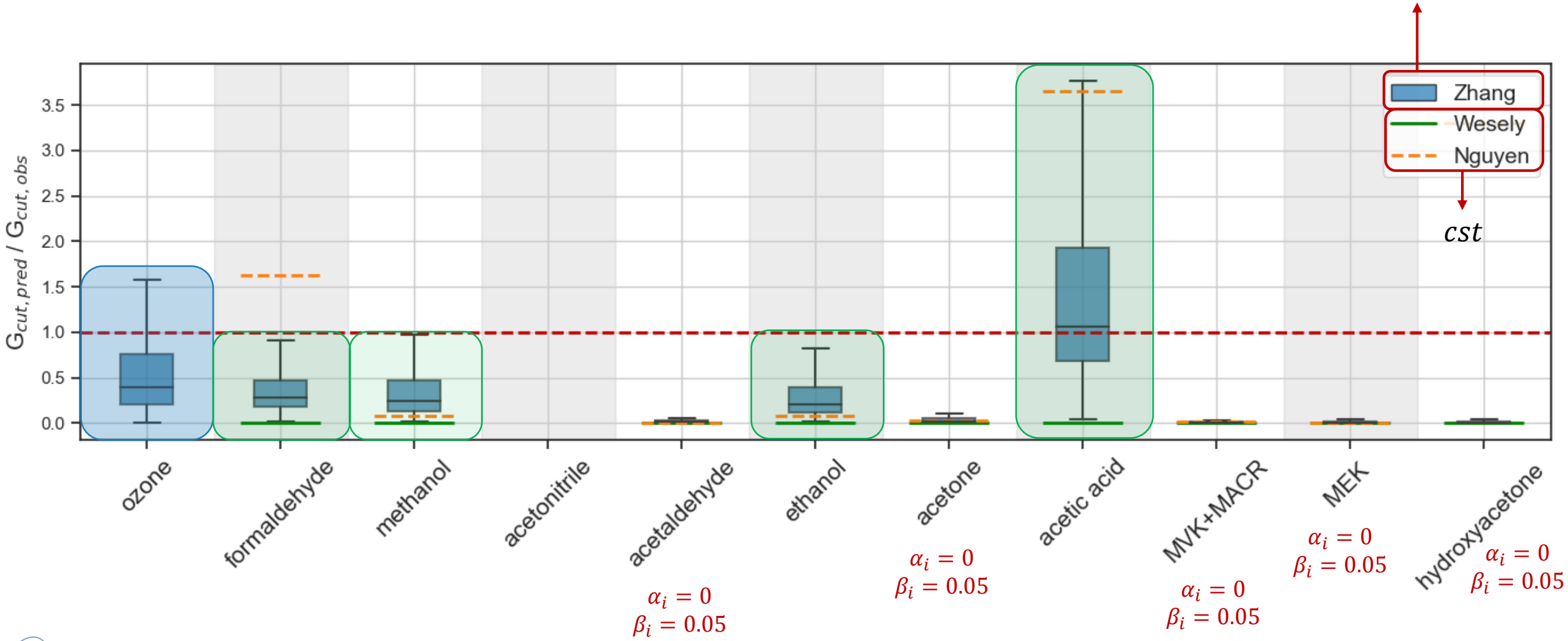
- TOP measurements:

$$G_{c,top} = G_{stom} + G_{cut} + G_{soil}$$



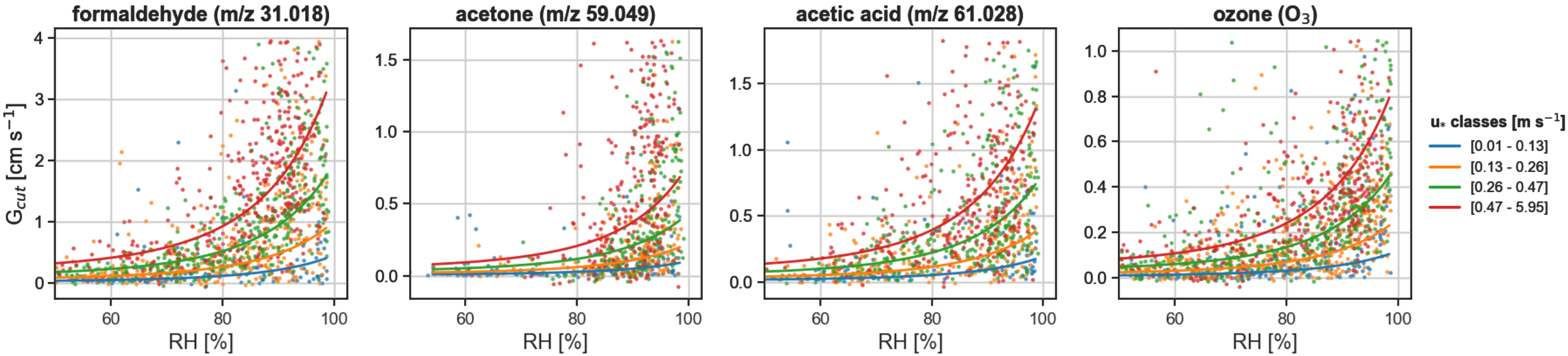
# Evaluation of existing models

$$G_{cut,VOC_i} = G_{cut0,VOC_i} \cdot f(RH, u_*, LAI)$$
with $G_{cut0,VOC_i} = \alpha_i G_{cut0,SO_2} + \beta_i G_{cut0,O_3}$




*Zhang et al. (2003, ACP)*
*Nguyen et al. (2015, PNAS)*
*Wesely (1989, Atmos. Environ.)*

# Towards new parametrizations

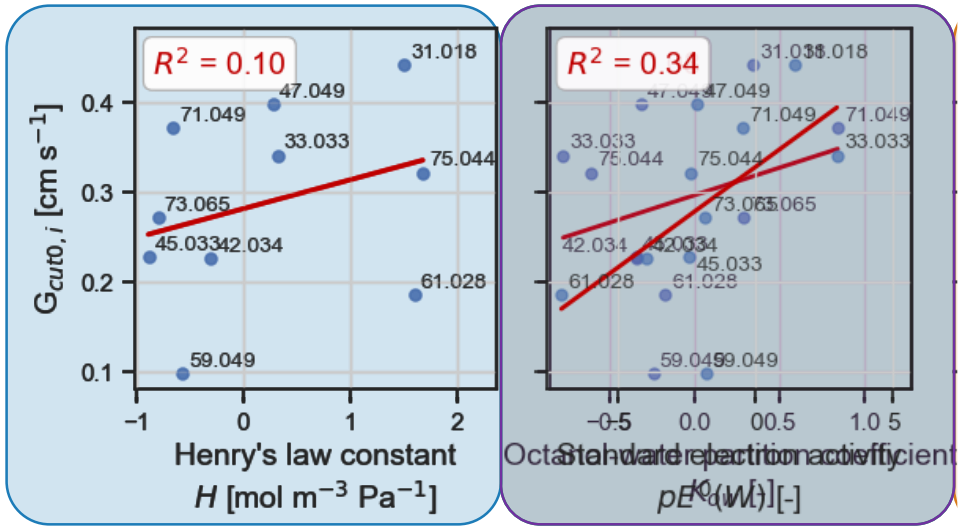



$$G_{cut,VOC_i} = G_{cut0,VOC_i} \cdot e^{b_{cut}\left(RH^3 - RH_{ref}^3\right)} \cdot \left(\frac{u_*}{u_{*,ref}}\right) \cdot LAI^{1/4}$$

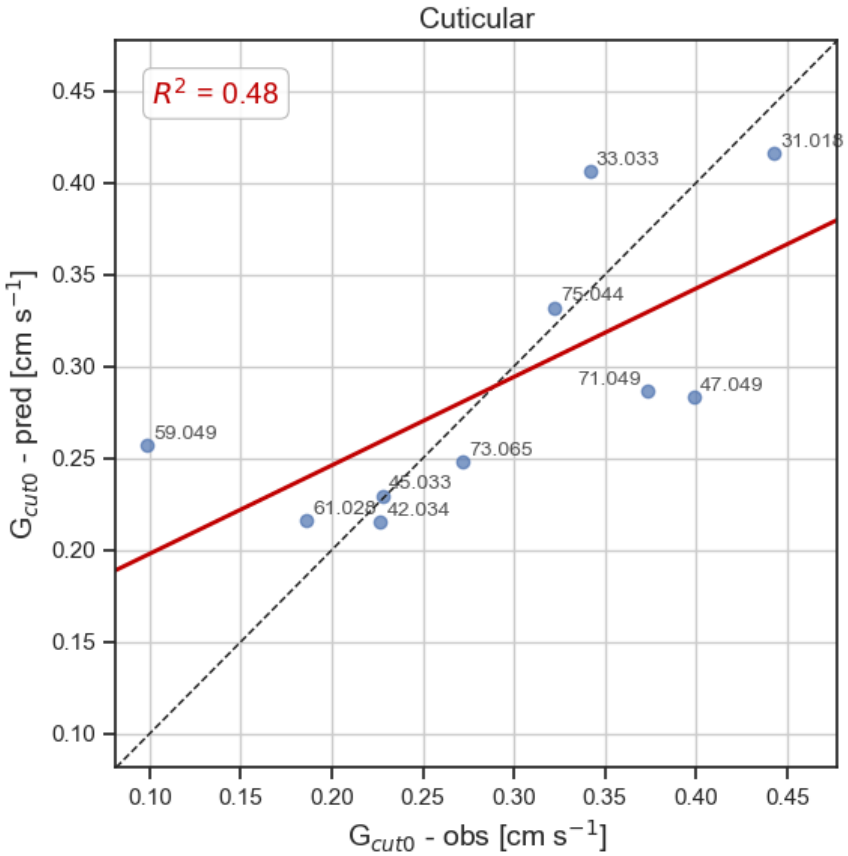
$$G_{cut,VOC_i} = g(physico) \cdot f(dynamic)$$

- Same drivers as in Zhang, but greater sensitivity to $RH$
- Normalization: $RH_{ref} = 80\%$, $u_{*,ref} = 0.3\ m\ s^{-1}$
- Comparable sensitivity to $RH$ across VOCs, $b_{cut} = 2.7 \cdot 10^{-6}$
- Base conductance = compound-specific

# Physicochemical relationships




$$G_{cut0,VOC_i} = f[H, pE^0(W)]$$




GAS / WATER — Dissolution into water films and hydrophilic regions present on surfaces

GAS / PARTICLE — Partitioning from the gas phase onto atmospheric particles

GAS / LIPID — Dissolution into waxy leaf layers or cellular membranes

$GAS_{ox}$ / $GAS_{red}$ — Consumption through redox reactions with cellular metabolites/structures

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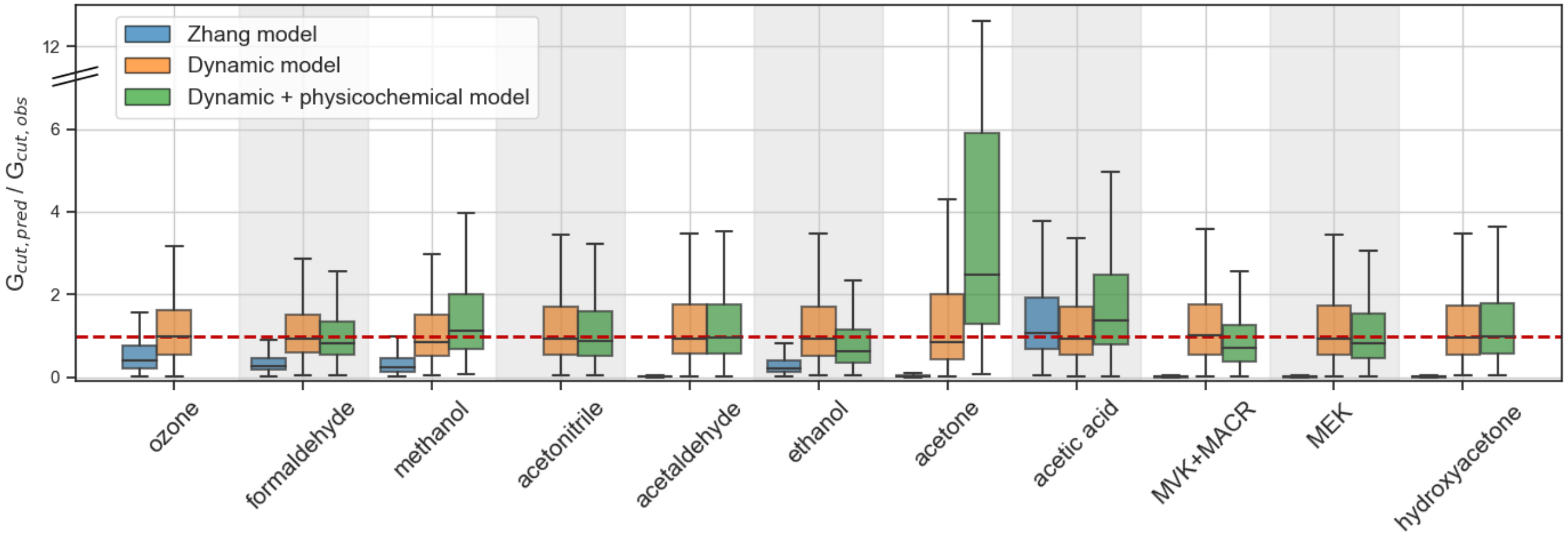
# Model robustness

$$G_{cut,VOC_i} = G_{cut0,VOC_i} \cdot f(RH, u_*, LAI)$$

- with $G_{cut0,VOC_i}$ coming from fitted observations
- with $G_{cut0,VOC_i}$ coming from physicochemical model (LOOCV)



# Take-home messages

1. Existing models **underestimate** surface conductances for the VOCs investigated
2. **Humidity** and **turbulence** control surface conductances
3. Reference conductances scale with VOC **reactivity** and **solubility**, rather than lipid affinity or volatility

# Limitations and perspectives

1. **Mechanisms**
   Integrate biological mechanisms (biological consumption)
2. **Generality**
   Validate the new parameterizations across VOCs and ecosystem types
3. **Atmospheric relevance**
   Assess their impact in atmospheric models

# Acknowledgement

The authors wish to acknowledge the Belgian Federal Science Policy Office for their support to the ACTRIS-BE project and the BIRA/ULiège FEDtWIN project BERTRAC as well as the Public Service of Wallonia for their support to the ICOS-Wallonia project.

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# Evaluation of existing models

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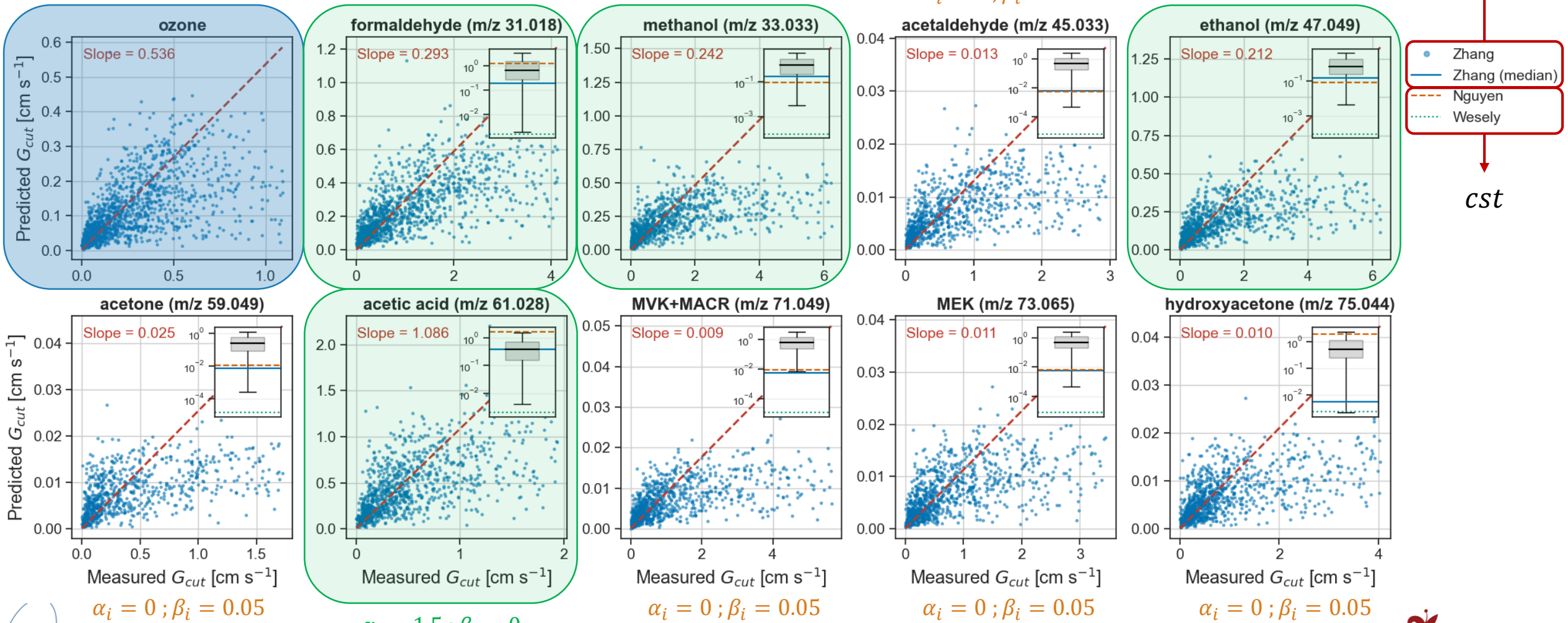
$$\text{with} G_{cut0,VOC_i} = \alpha_i G_{cut0,SO_2} + \beta_i G_{cut0,O_3}$$

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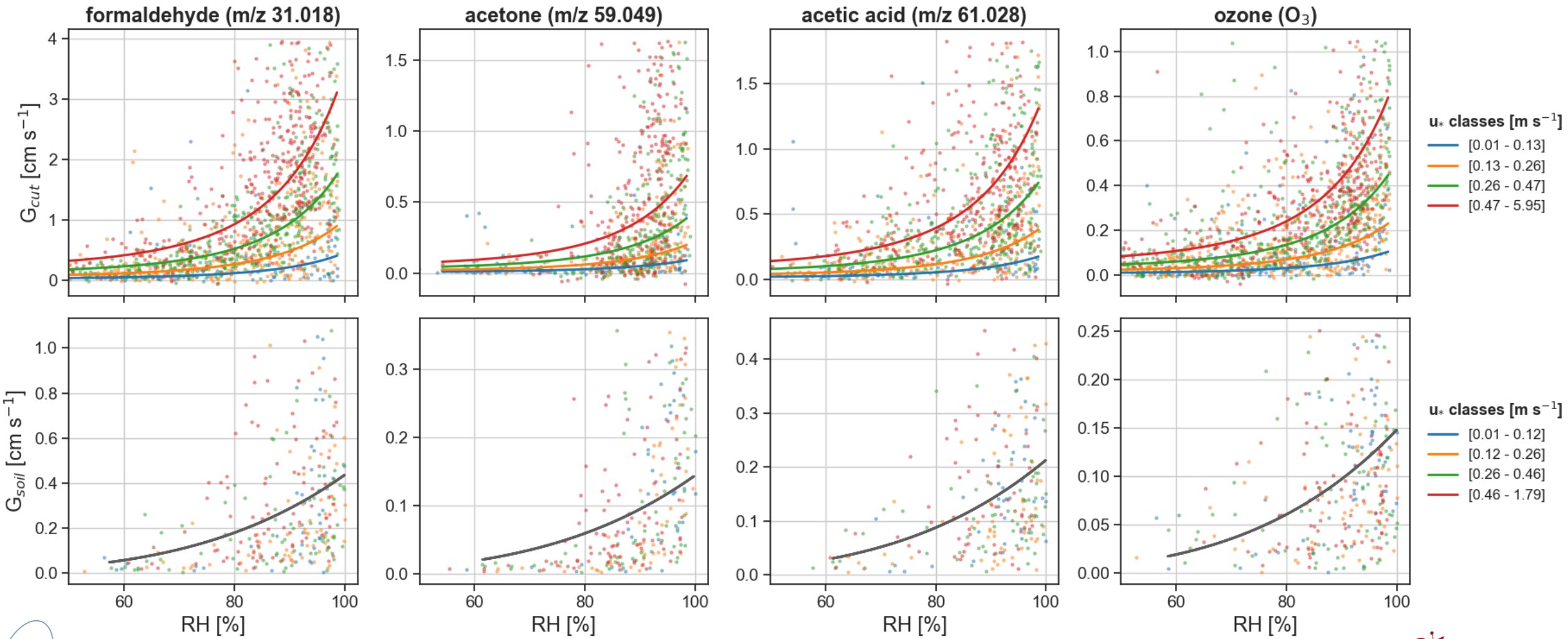
# New parametrizations



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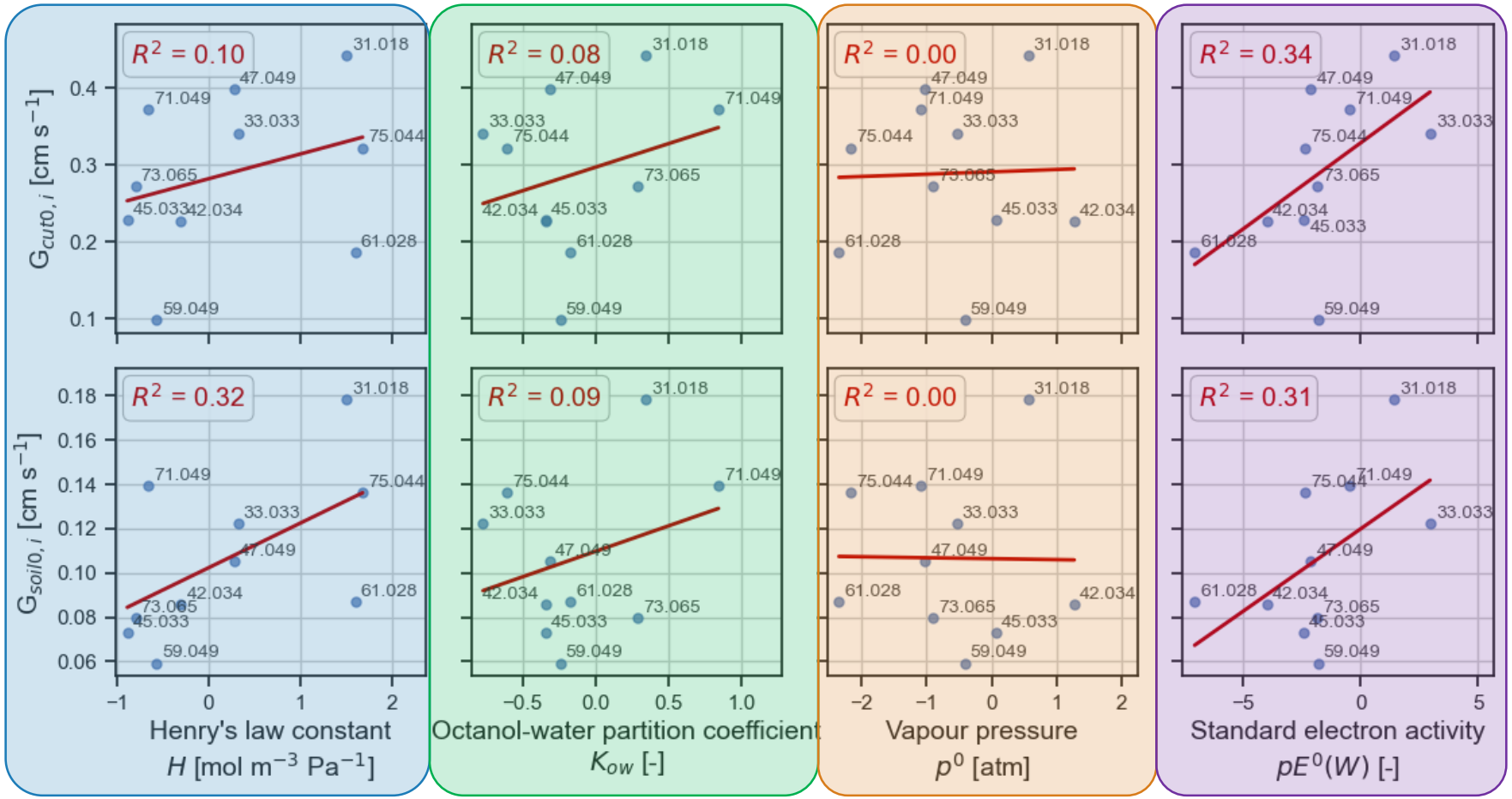


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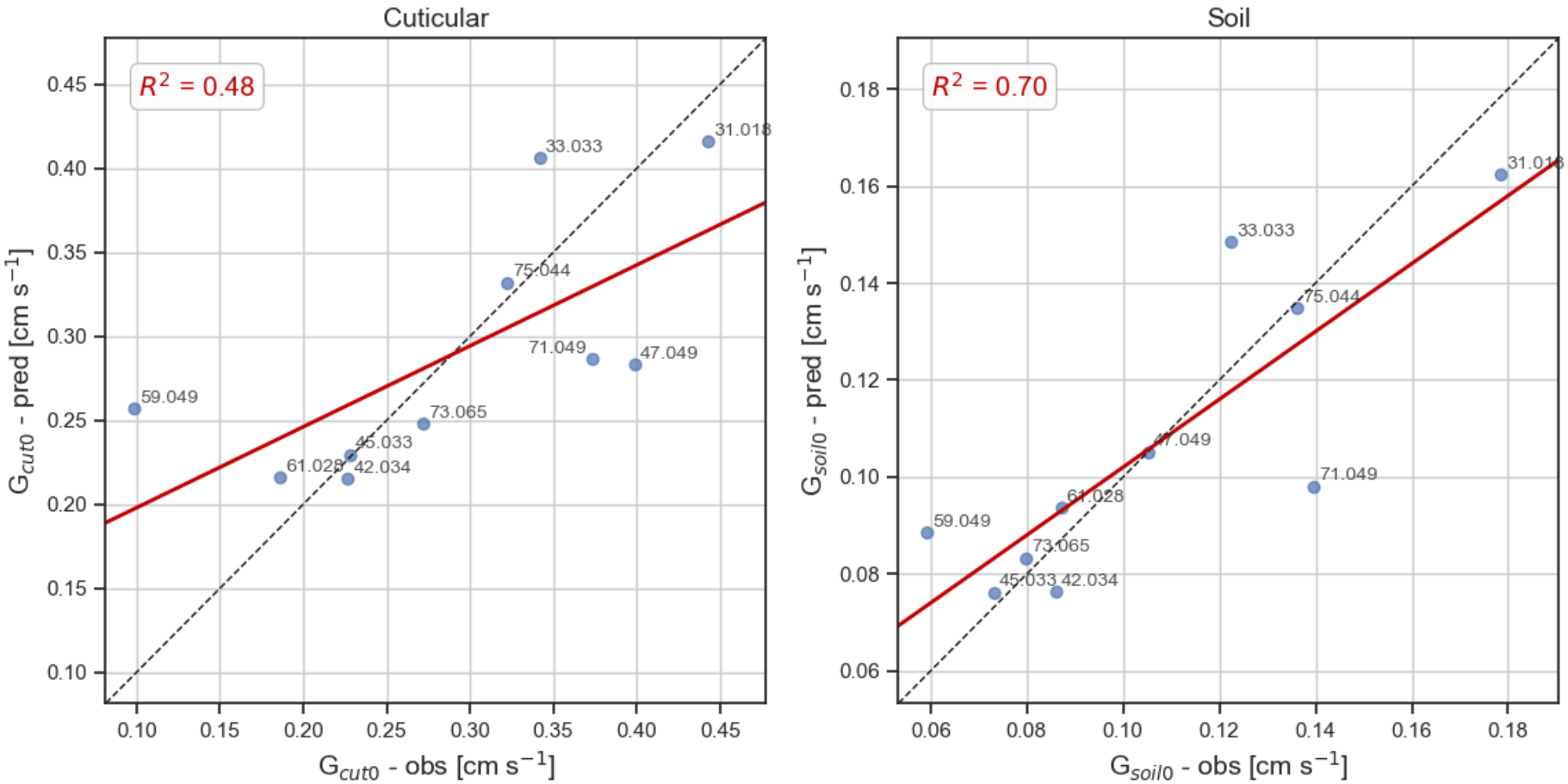
# Multilinear model




$$G_{cut0,VOC_i} = \beta_{cut0} + \beta_{cut1} \cdot H_{VOC_i} + \beta_{cut2} \cdot pE^0\,(W)_{VOC_i}$$
$$G_{soil0,VOC_i} = \beta_{soil0} + \beta_{soil1} \cdot H_{VOC_i} + \beta_{soil2} \cdot pE^0\,(W)_{VOC_i}$$

# Model robustness

- Train: $G_{cut0,i}$ estimated from fitted $(RH; u_*)$ curves
- Val: $G_{cut0,i}$ estimated from physicochemical properties with leave-one-out

