Impact of pH computation from EQSAM4Clim on inorganic aerosols in the CAMS system

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EGU23-16085 - 28/04/2023

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PICO Presentation — EQSAM4Clim in the IFS

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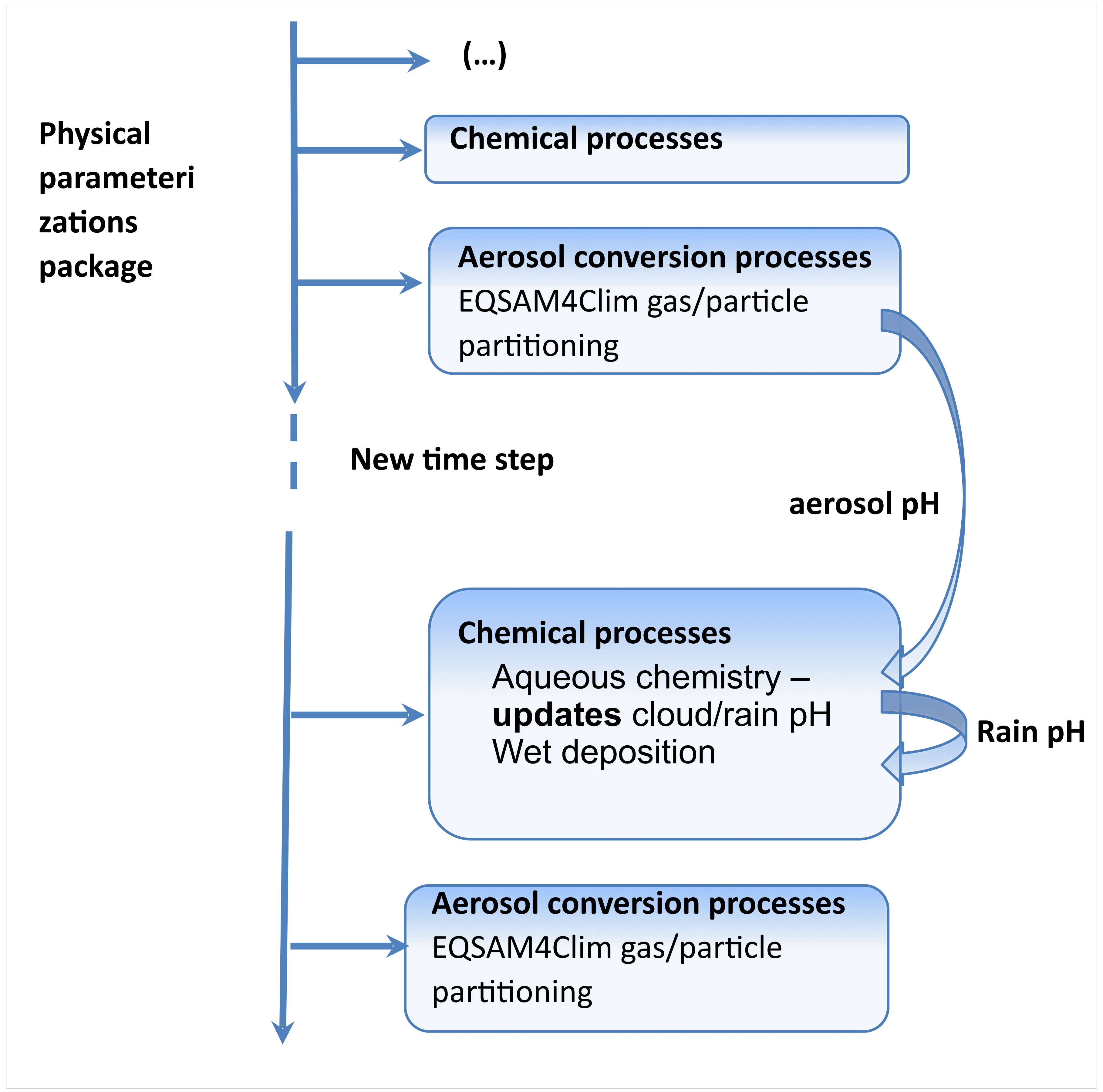

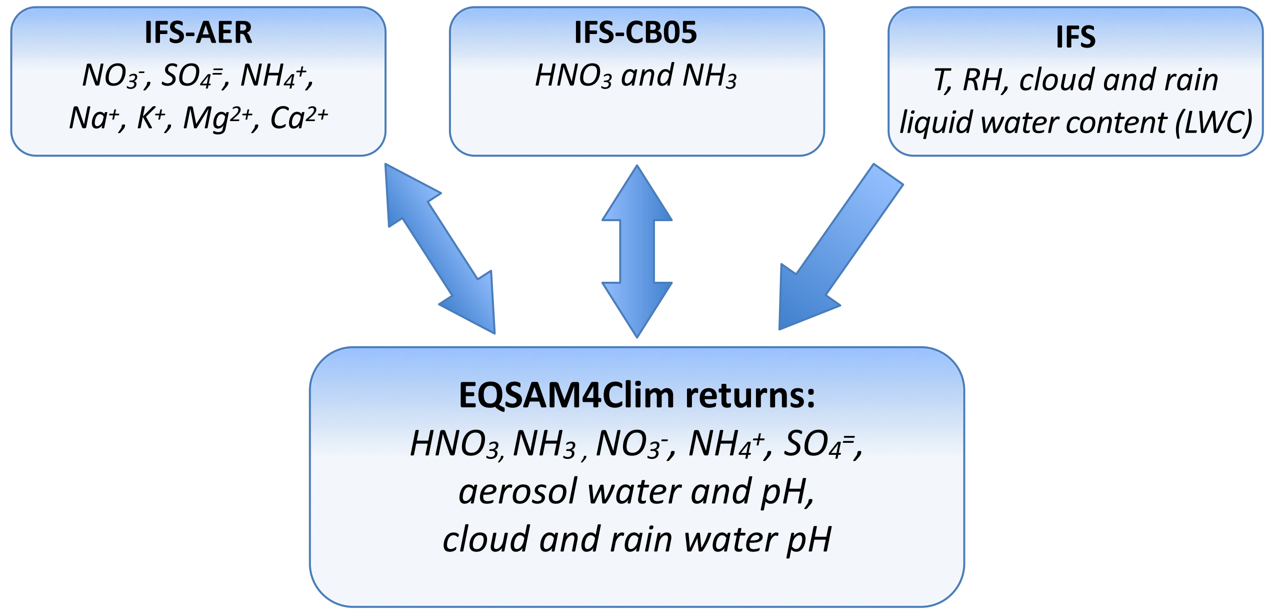

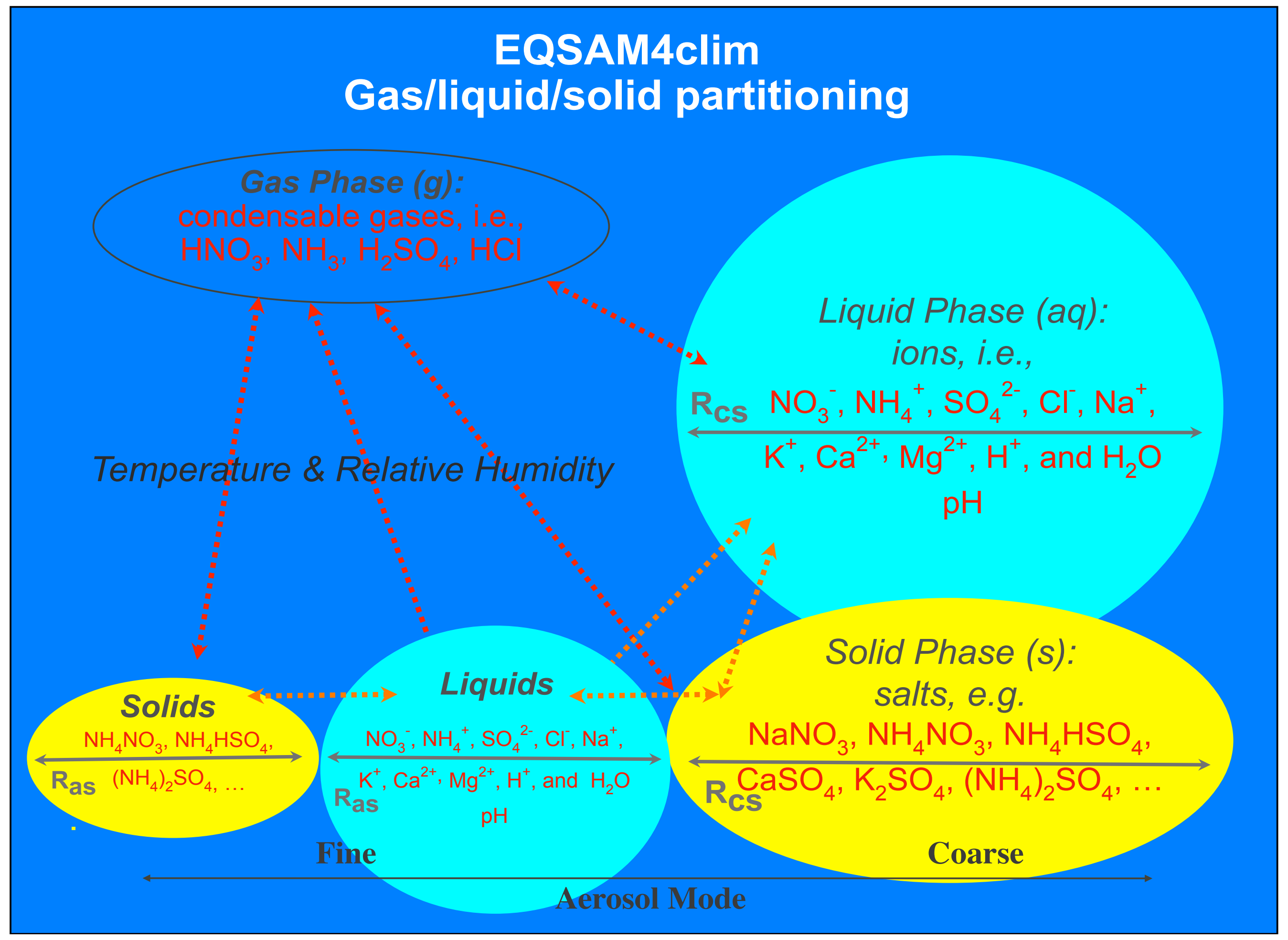

Left: Coupling of EQSAM4Clim with aqueous phase chemistry of the IFS.

Right. Tracer interface of IFS with EQSAM4Clim (top).

T + RH dependent gas/liquid/solid aerosol partitioning within EQSAM4Clim (bottom, right).

Developments shown are to be included in cycle 49R1 (ie not in the incoming 48R1).

$$[H^+] = 2[SO_4^{2-}] + [HSO_4^-] + [NO_3^-] + [Cl^-] - 2[Ca^{2+}] - 2[Mg^{2+}] - [Na^+] - [K^+] - [NH_4^+]$$

$$pH_{aerosol} = -\log_{10}(H^+/H_2O_{aerosol})$$

$$pH_{cloud} = -\log_{10}(H^+/H_2O_{cloud})$$

$$pH_{rain} = -\log_{10}(H^+/H_2O_{rain})$$

Left: The pH formula is used with different LWC to calculate the pH for the aerosol phase, cloud and rain water of the IFS.

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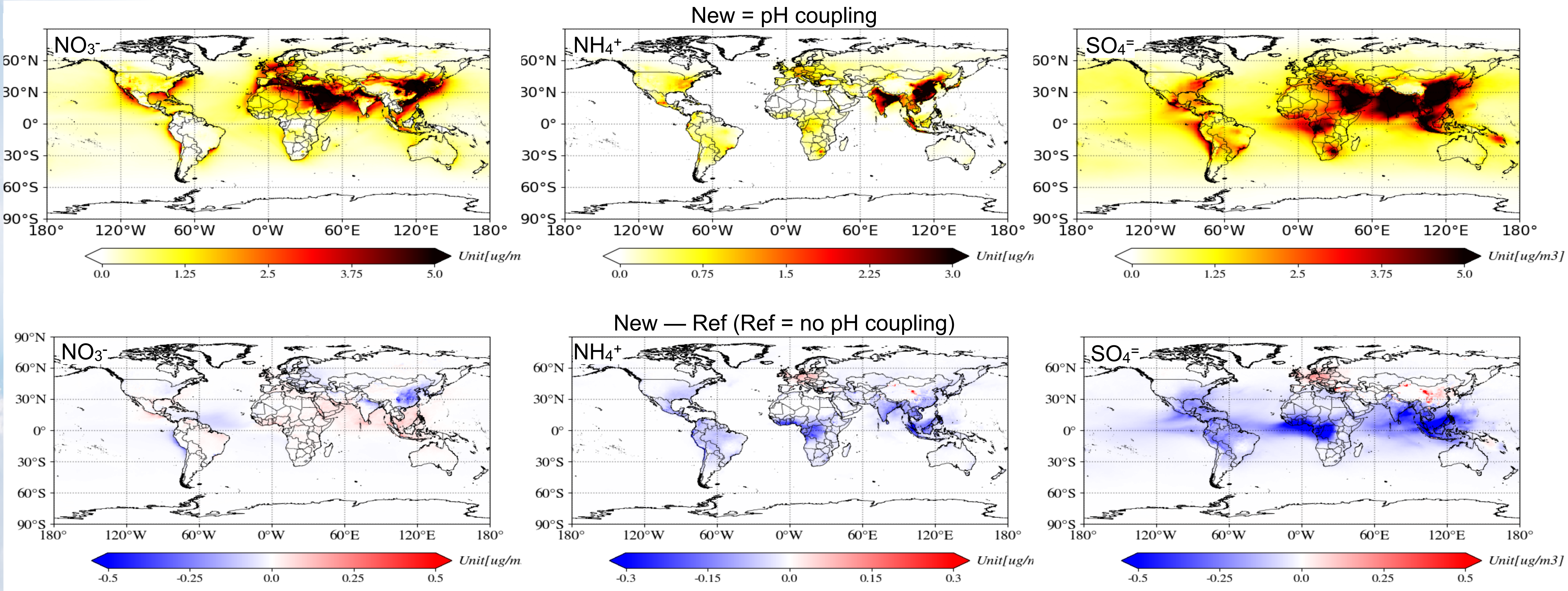
Effect of pH coupling — 2019 (surface avg)



- Strongest effect of pH coupling from EQSAM4clim with aqueous phase chemistry is found for sulphate aerosol because of the pH dependency of the SO_2 oxidation. Interestingly, the coupling can yield both more and less acidic aqueous solutions.
- But also nitrate and ammonium are affected. Ammonium directly through aqueous phase chemistry, and nitrate indirectly through gas/liquid/solid partitioning which also depends on the presence of sulphate aerosol.
- For annual means, the effect is generally less than 10%, but regionally with a different sign.

Effect of pH coupling — US 2019 (surface avg)

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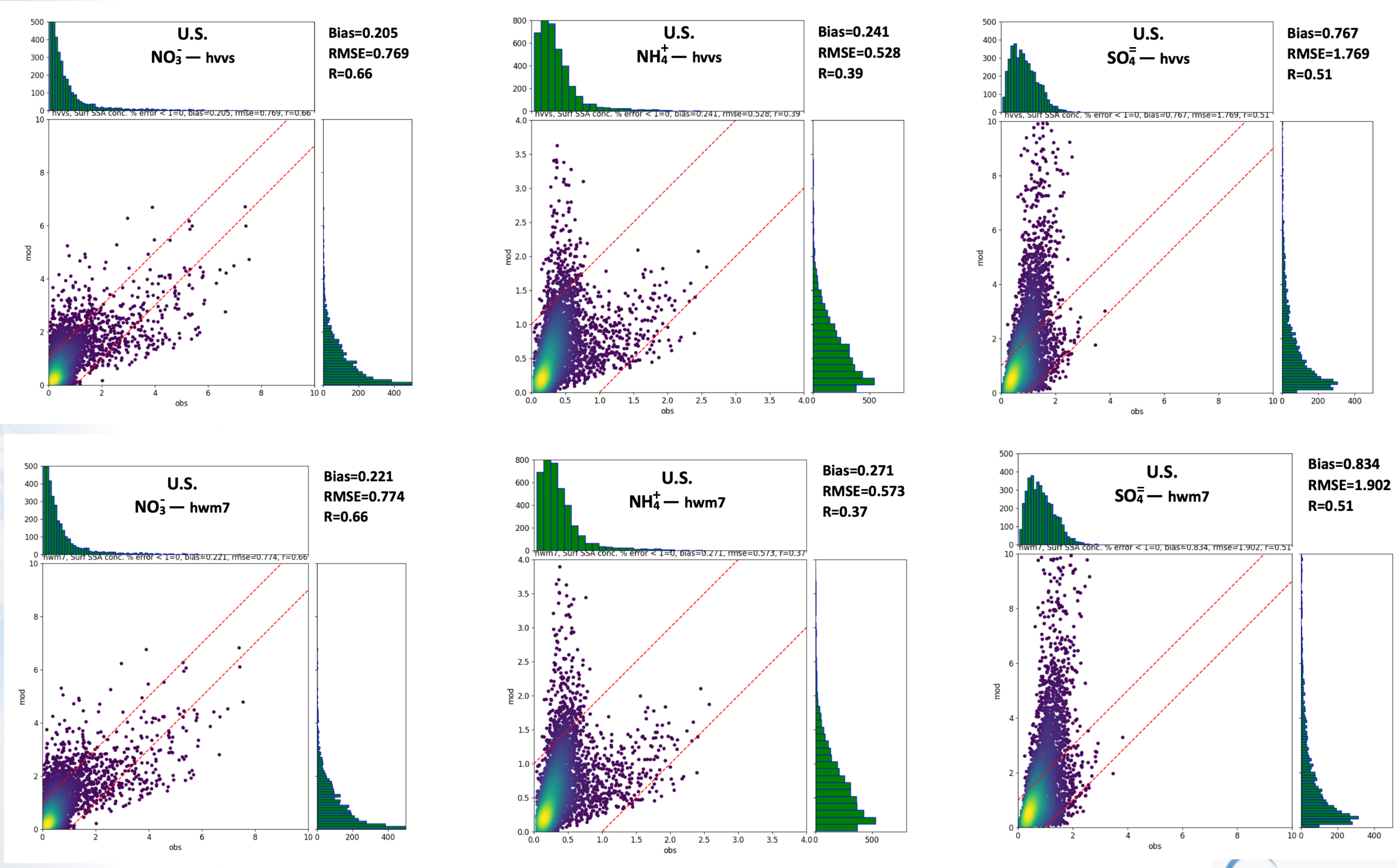

Top:

New = pH coupling of EQSAM4Clim with aqueous phase chemistry of the IFS.

Bottom:

Ref = no pH coupling and for annual mean slightly higher bias.

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Effect of pH coupling — EU 2019 (surface avg)

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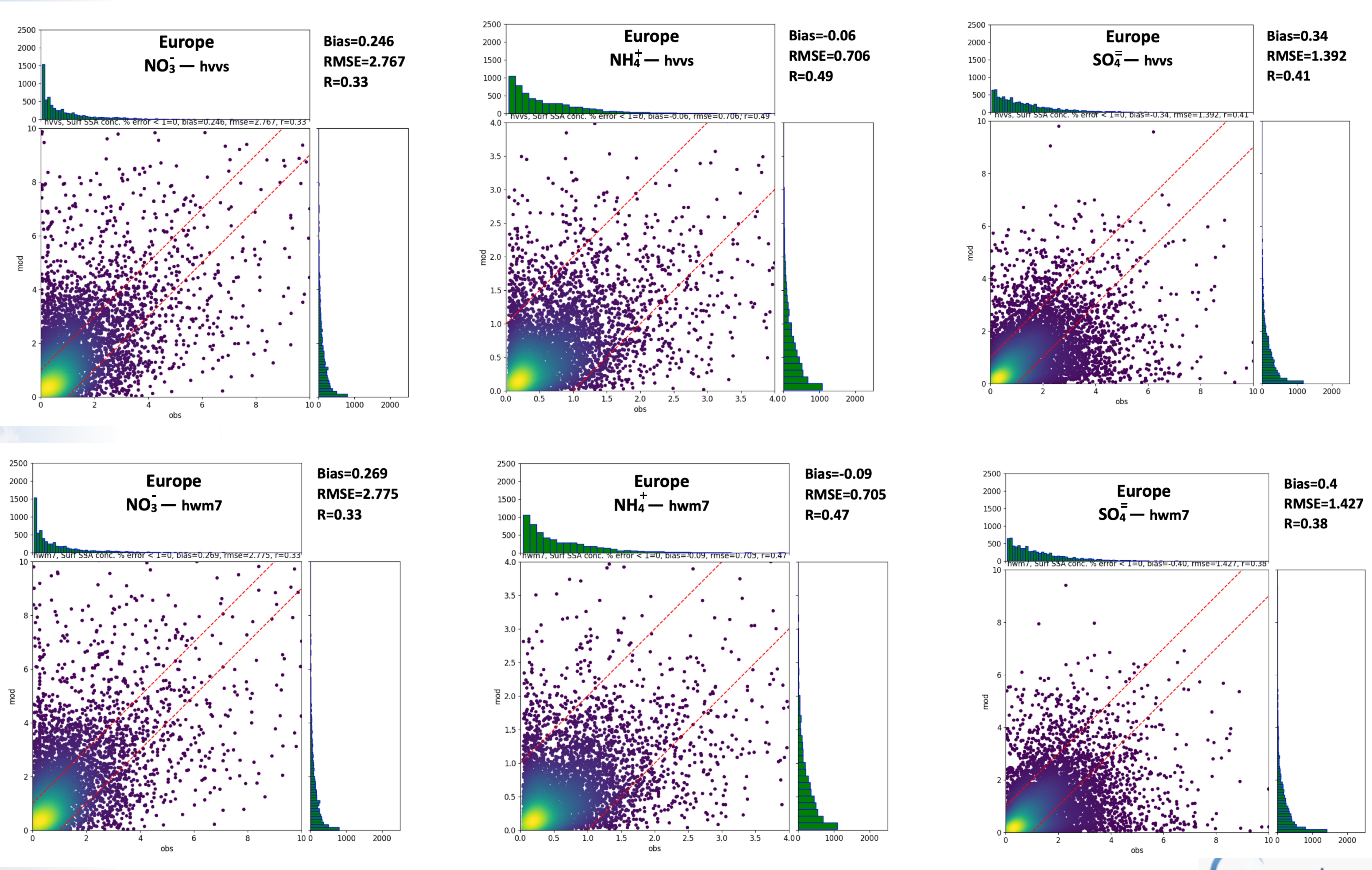

Top:

New = pH coupling of EQSAM4Clim with aqueous phase chemistry of the IFS.

Bottom:

Ref = no pH coupling shows for annual mean slightly higher bias.

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Take Home Message

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- Overall, there is an impact of pH coupling of EQSAM4Clim with aqueous phase chemistry on the IFS aerosol properties (AOD, PM1, PM2.5, NO_3^-, NH_4^+, and $SO_4^=$),
  although the effect is much smaller compared to the potential impact of emission changes on the gas/aerosol partitioning and conversion (particularly of NH_3 and SO_2).
- The pH coupling impacts the aqueous solution to be regionally more or less acidic, which in turn impacts the oxidation efficiency of the aerosol precursor gases.
- Changes of ammonium and sulphate through aqueous phase chemistry subsequently impacts aerosol nitrate through the gas/liquid/solid aerosol partitioning, which also depends on the presence of mineral cations.
- For annual means, the effect is less than 10%, but for a model time-step the effect can be significantly larger, and regionally it can have a different sign.
- The impact on PM and AOD is generally small with a regionally mixed improvement.

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PICO — Supplemental Material: Motivation

- The reason for testing/using the EQSAM4clim is its ability to parameterise the gas/liquid/solid aerosol partitioning and associated aerosol water uptake sufficiently fast and accurate (i.e., noise free) for NWP:
  - Aerosol water parameterization: long-term evaluation and importance, 10.5194/acp-18-16747-2018,
  - Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components, EMEP report 2019, ISSN 1504-6192 (on-line),
  - Comparing the ISORROPIA and EQSAM Aerosol Thermodynamic Options in CAMx, Springer Book chapter 2020: 10.1007/978-3-030-22055-6_16).
- The current situation in IFS is that the (EQSAM based) Hauglustaine scheme (https://doi.org/10.5194/acp-14-11031-2014) is used (before EQSAM was used in the IFS: https://doi.org/10.5194/gmd-8-975-2015 — EQSAM: Gas/aerosol partitioning: 1. A computationally efficient model, JGR 2001: 10.1029/2001JD001102).
- Compared to the previous schemes, EQSAM4clim improves the IFS aerosol tracer ammonium and nitrate, as its concept basically allows to (better) consider mineral cations (i.e., here: calcium, magnesium, potassium), which is important for the whole gas/liquid/solid aerosol partitioning. Besides the importance for aerosol water, mincer cations are especially crucial for ammonium nitrate and ammonium chloride (Importance of mineral cations and organics in gas-aerosol partitioning of reactive nitrogen compounds: Case study based on MINOS results, 10.5194/acp-6-2549-2006).
- Note that other IFS aerosol species do not undergo gas/aerosol partitioning as ammonium is the only volatile cation (considered).
- Finally, the pH is largely determined by ammonium and sulfate chemistry, so the gas/liquid/solid aerosol partitioning can become important for the subsequent oxidation of SO2 and NH3 in the aqueous phase chemistry, which in turn alters the gas/liquid/solid aerosol partitioning of ammonium nitrate (and ammonium chloride) and the associated water uptake.
- Next step, once the water uptake (aerosol water) is fully coupled in the IFS (upcoming task), EQSAM4clim could also impact all aerosol tracer transport and aerosol radiative feedback due to the impact of the gas/liquid/solid aerosol partitioning and aerosol water on the aerosol dry and wet radius.

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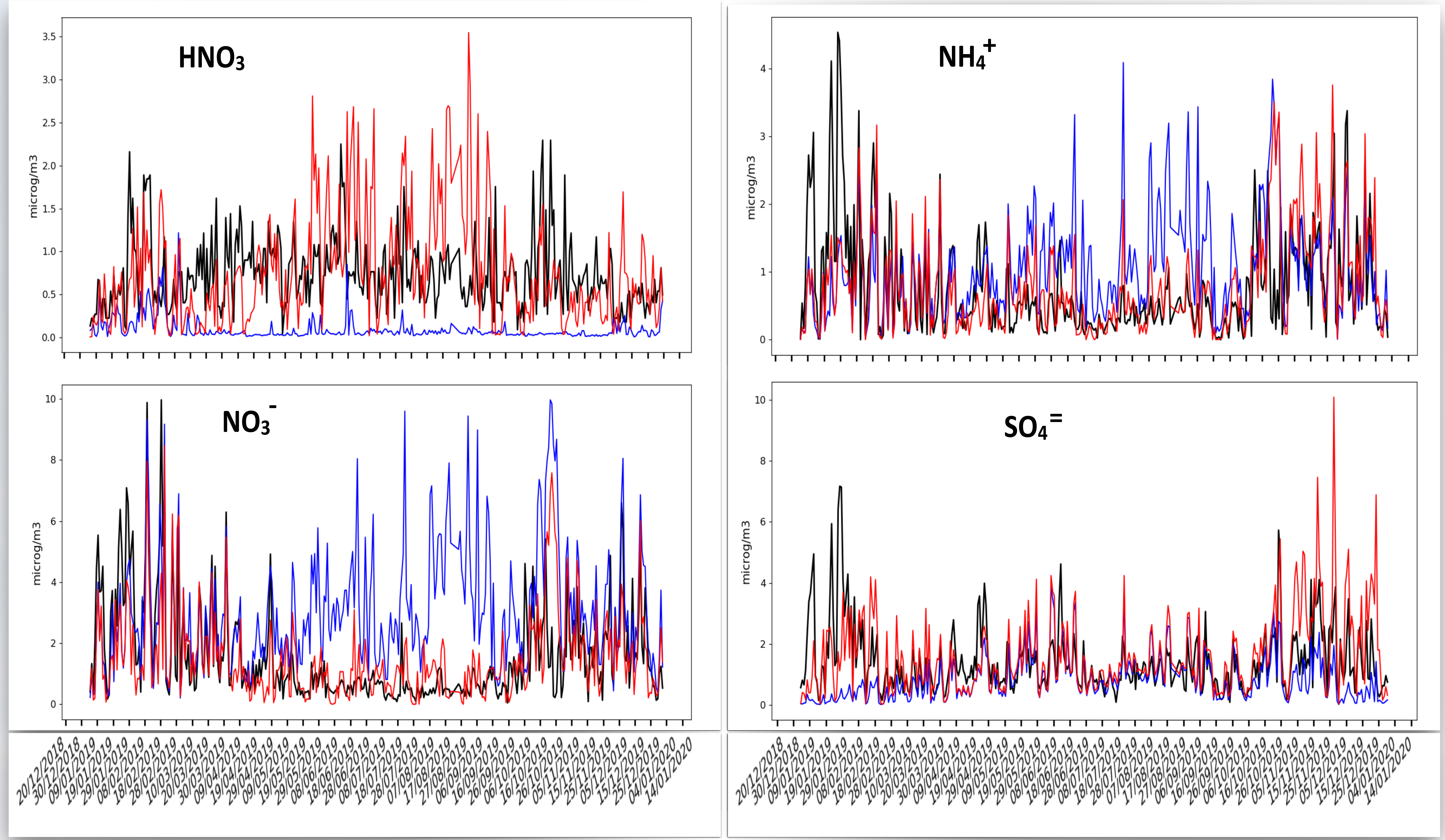
PICO — Supplemental Material: Additional Results



– Weekly gas+aerosol surface concentrations of 2019 in ug/m3 over the Diabla Gora (Poland) EMEP site. IFS experiments shown are reference without EQSAM4clim (blue) and with plus pH coupling (red) vs EMEP observations (black) for top to bottom, HNO_3, NH_4^+, NO_3^-, $SO_4^=$.

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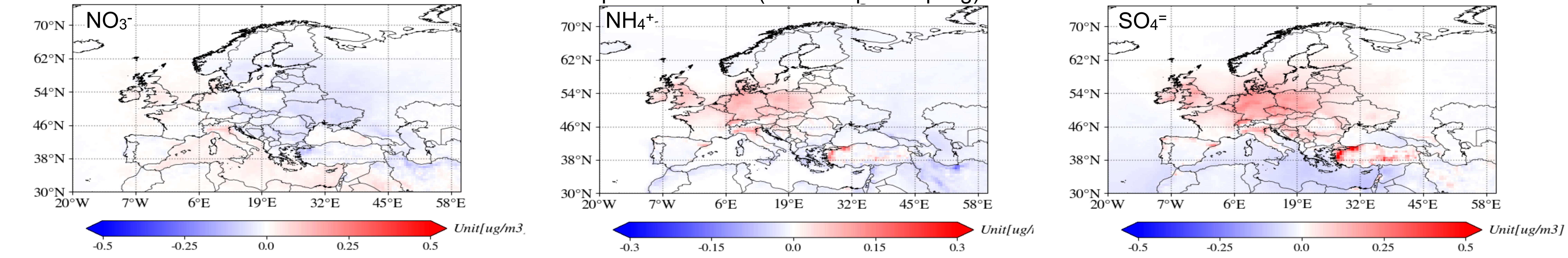
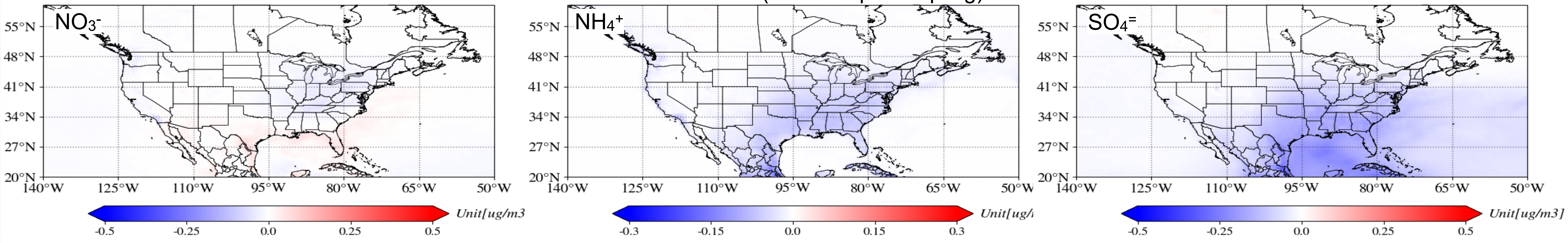
Effect of pH coupling — 2019 (surface avg)

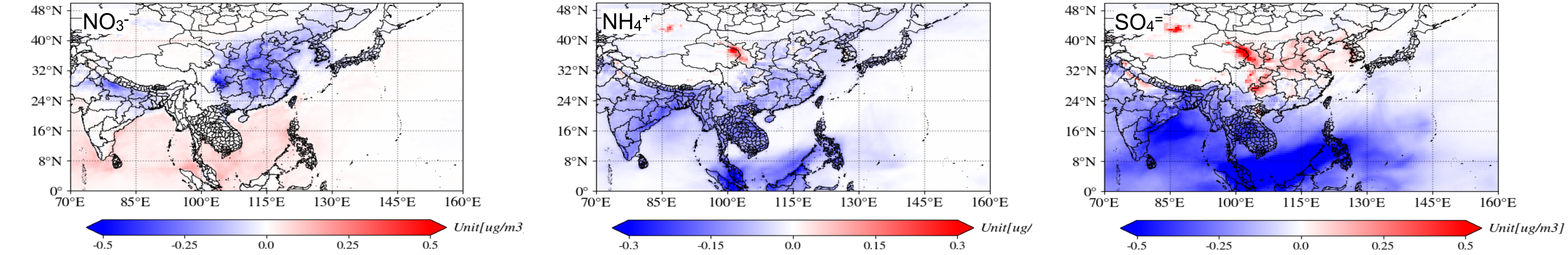
Europe: New — Ref (Ref = no pH coupling)



US: New — Ref (Ref = no pH coupling)



Asia: New — Ref (Ref = no pH coupling)



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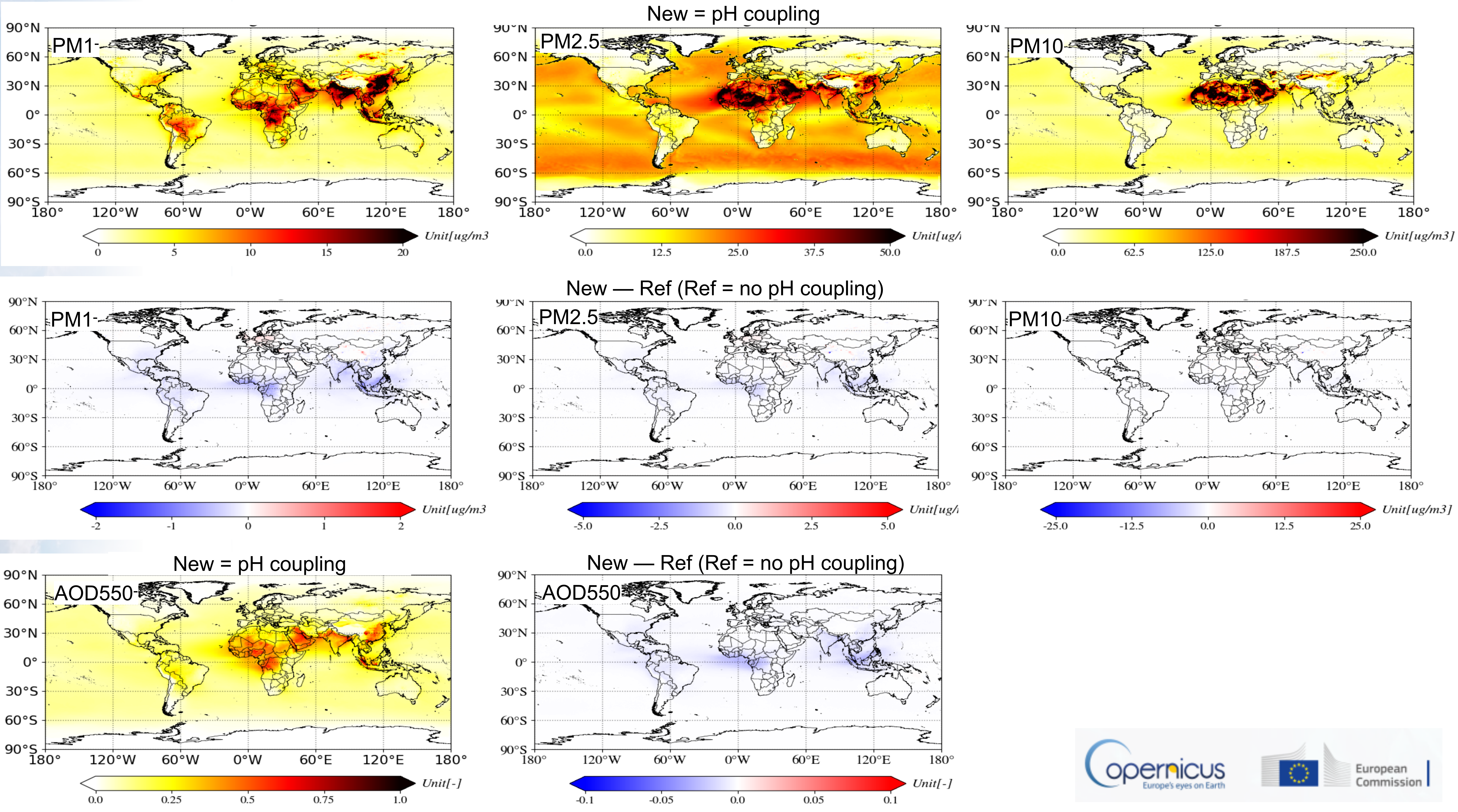
Effect of pH coupling — 2019 (surface avg)



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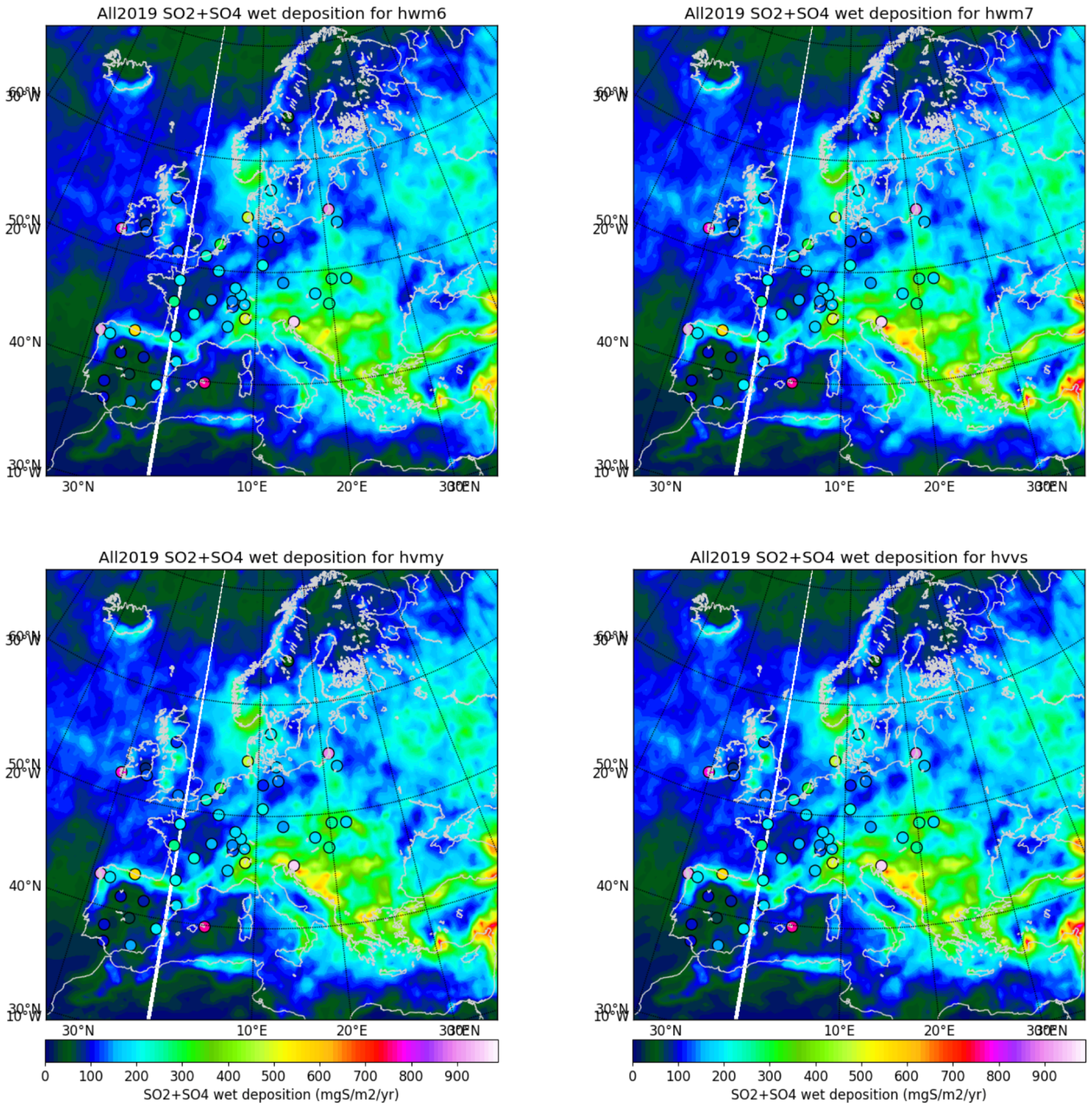
Sensitivity results I: Wet deposition — Europe

Exp id / name	**pH computation and use**
hwm6 / NoCouple	EQSAM4Clim, but no pH coupling
hwm7 / AqRate	hwm6 + updated aqueous chemistry rates, pH threshold 4.5
hwm9 / AqRate-L	hwm7 + lower pH threshold for aqueous chemistry (4 instead of 4.5)
hvmy / AqRate+AqPhase	hwm9 + pH coupling only for aqueous chemistry
hvvs / FullCouple	hwm9 + full pH coupling



Wet deposition of SO_2 + $SO_4^{=}$ for 2019 in mgS/m2/yr as compared to EMEP. Experiments shown are hwm6 (ul), hwm7 (ur), hymy (ll), and hvvs (lr).

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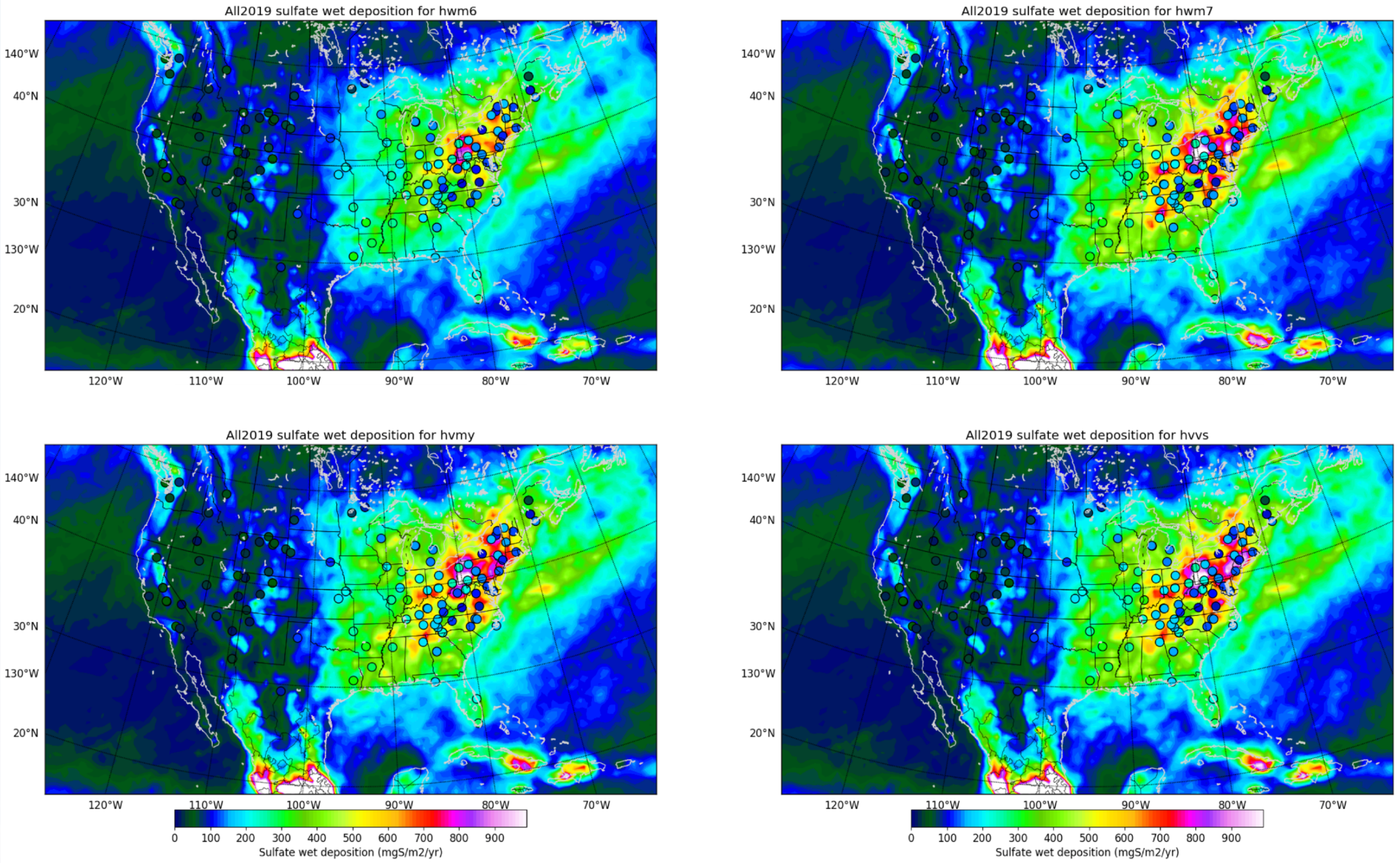
Sensitivity results I: Wet deposition — US

Exp id / name
hwm6 / NoCouple
hwm7 / AqRate
hwm9 / AqRate-L
hvmy / AqRate+AqPhase
hvvs / FullCouple



Wet deposition of SO_2 + $SO_4^=$ for 2019 in mgS/m2/yr as compared to CASTNET. Experiments shown are hwm6 (ul), hwm7 (ur), hymy (ll), and hvvs (lr).

IFS aerosol ammonium vs CASTNET 2019

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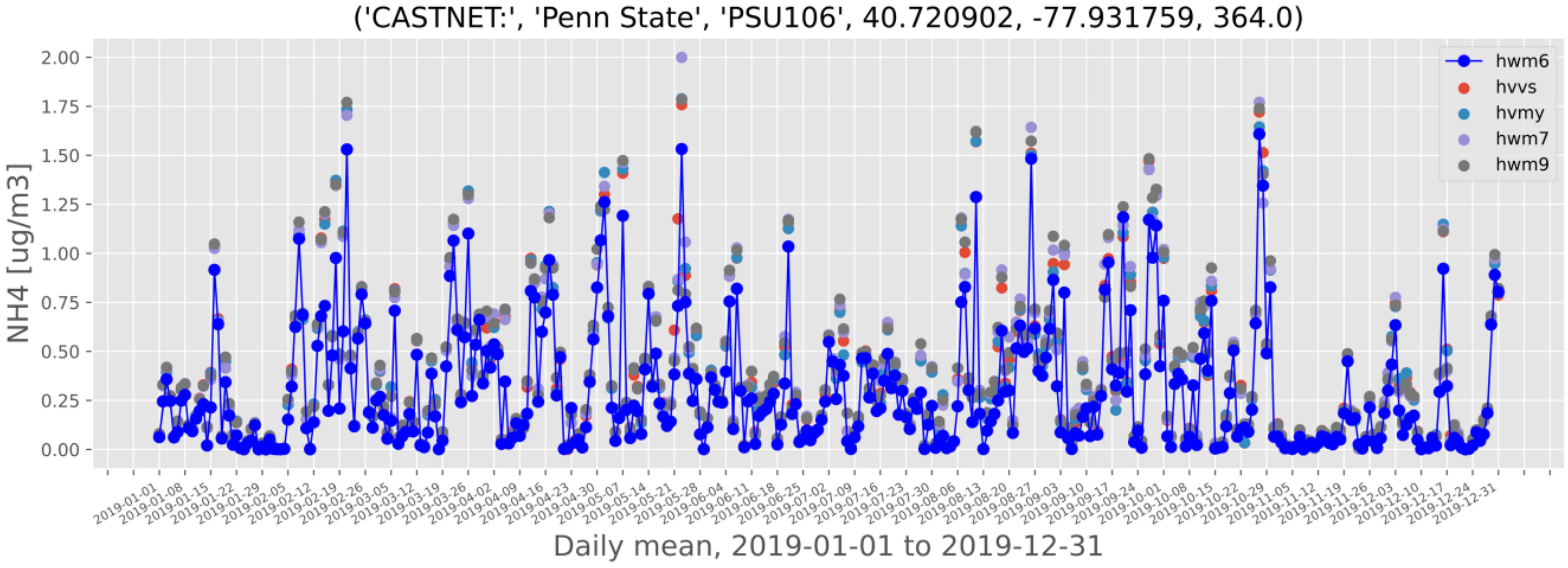
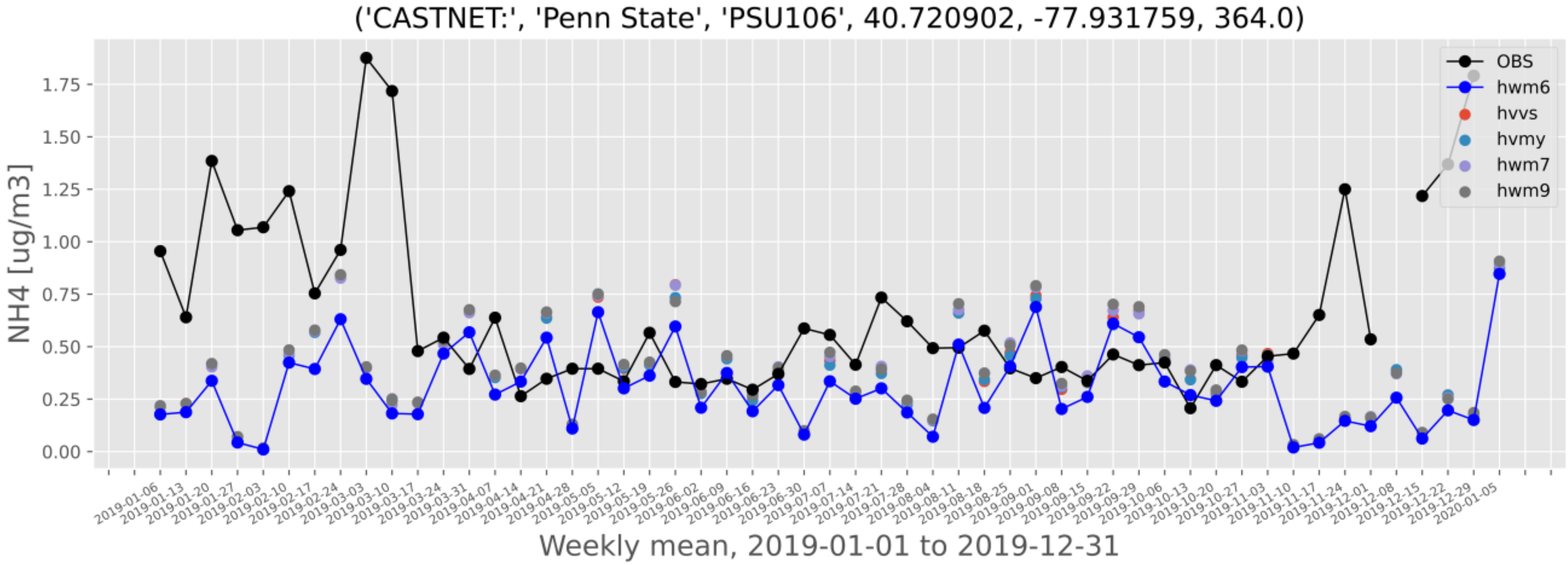
Exp id / name
hwm6 / NoCouple
hwm7 / AqRate
hwm9 / AqRate-L
hvmy / AqRate+AqPhase
hvvs / FullCouple





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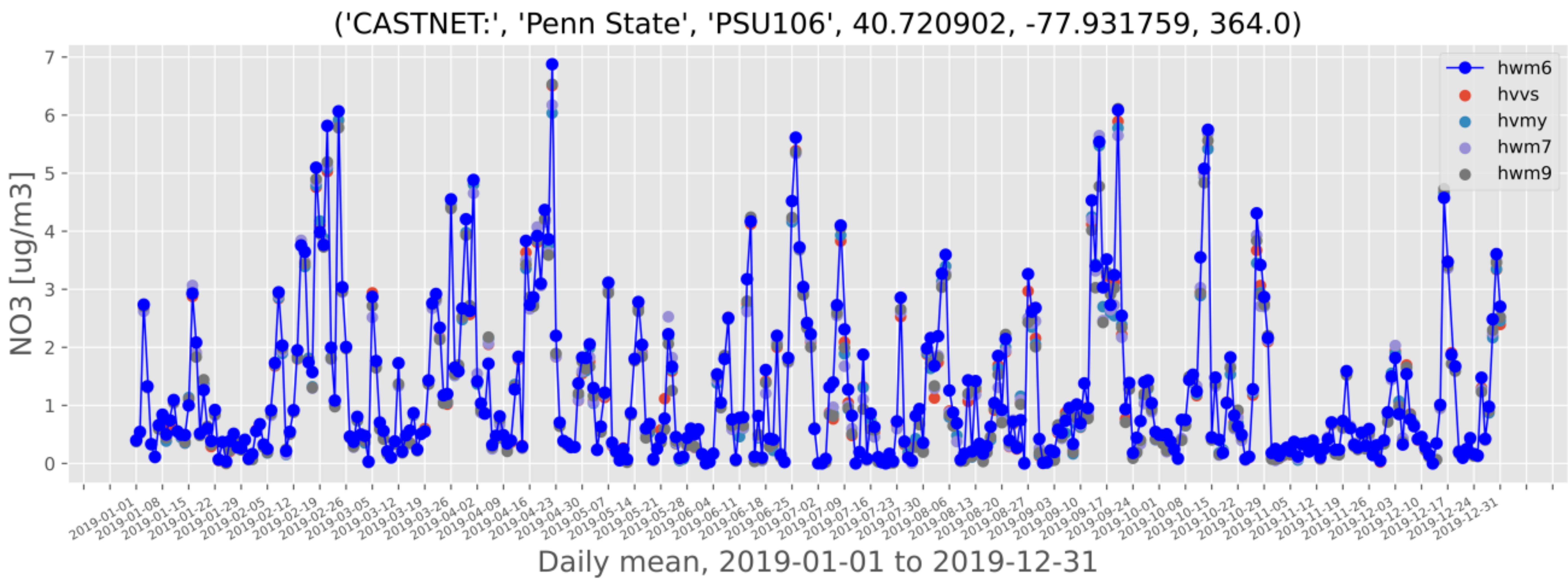
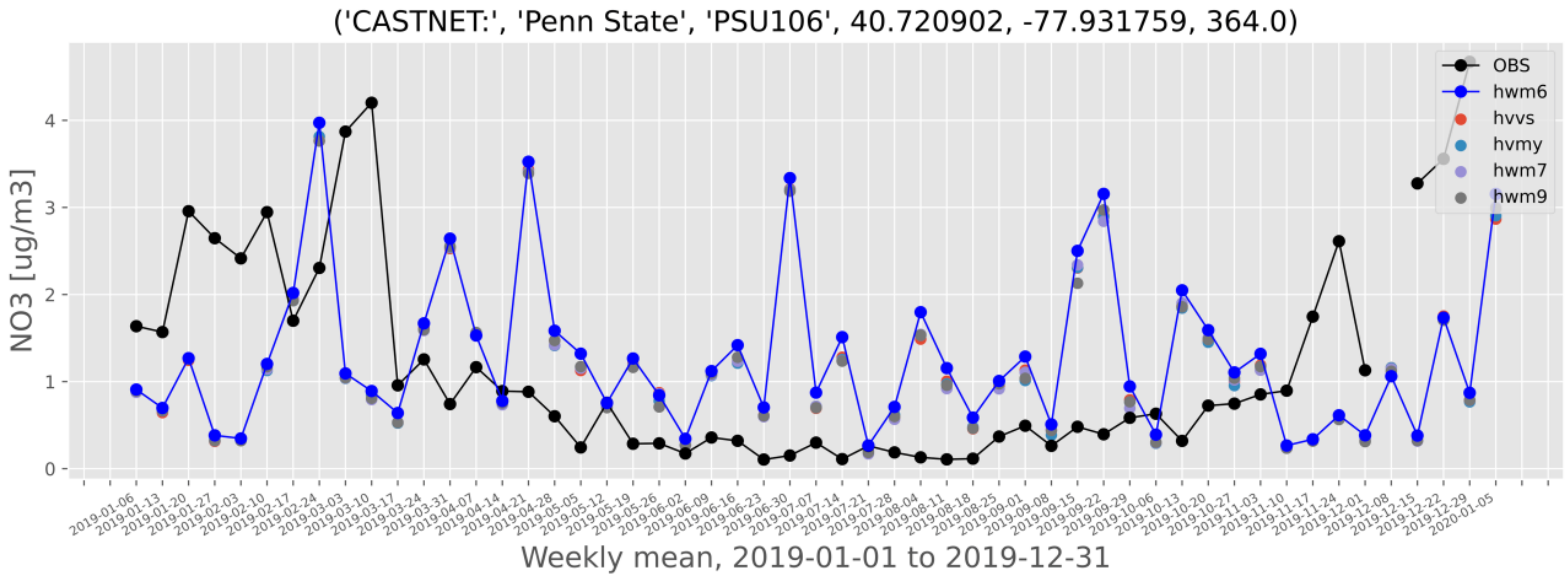
IFS aerosol nitrate vs CASTNET 2019

Exp id / name
hwm6 / NoCouple
hwm7 / AqRate
hwm9 / AqRate-L
hvmy / AqRate+AqPhase
hvvs / FullCouple

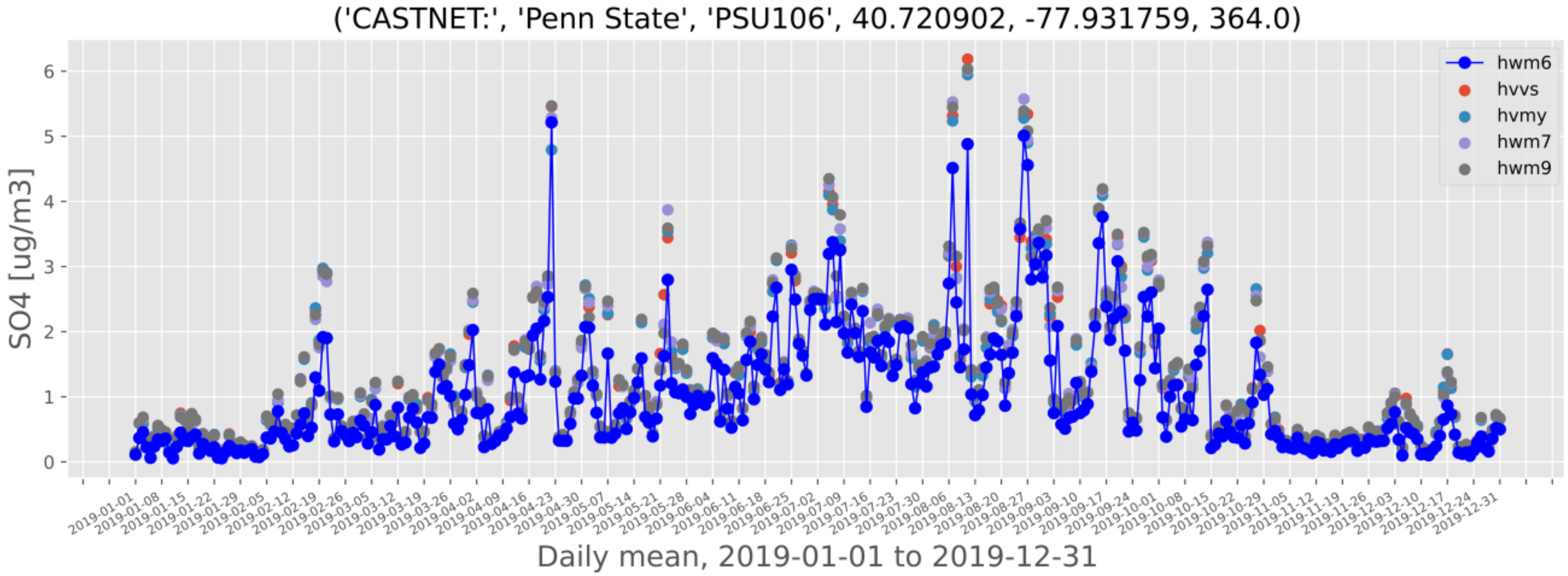


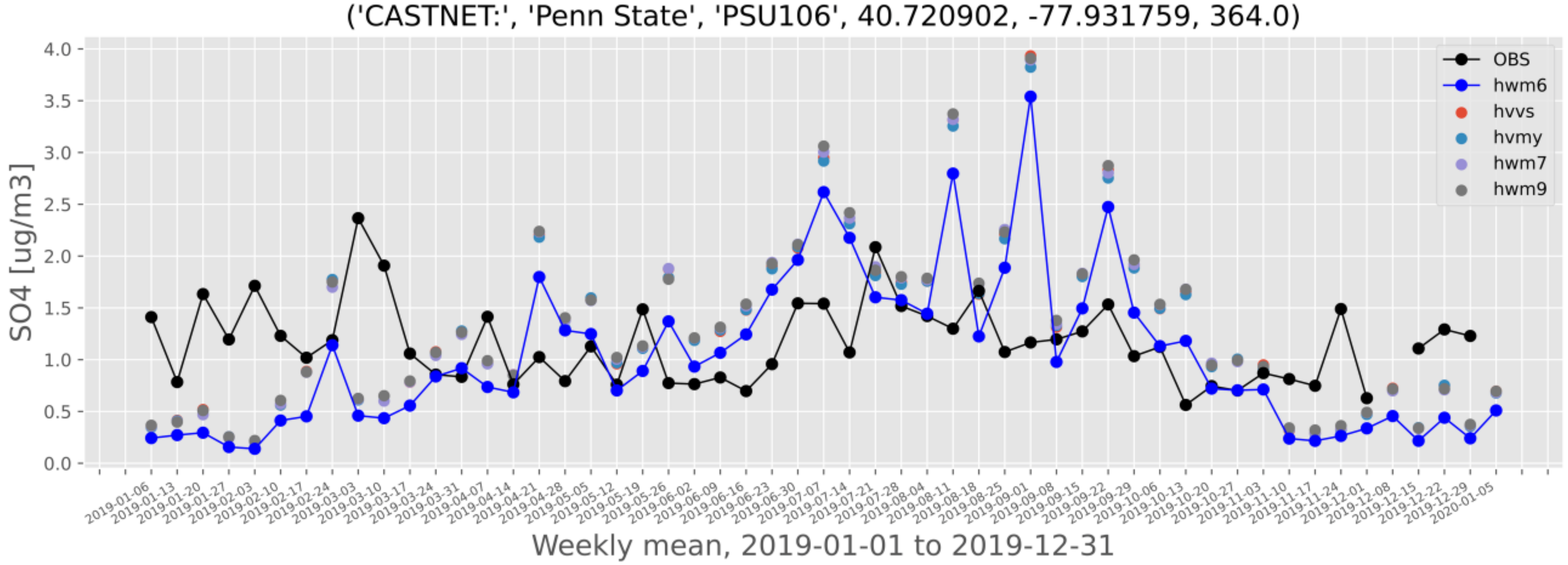


IFS aerosol sulphate vs CASTNET 2019

Atmosphere Monitoring

Exp id / name
hwm6 / NoCouple
hwm7 / AqRate
hwm9 / AqRate-L
hvmy / AqRate+AqPhase
hvvs / FullCouple





Sensitivity results: PM2.5 — US / China

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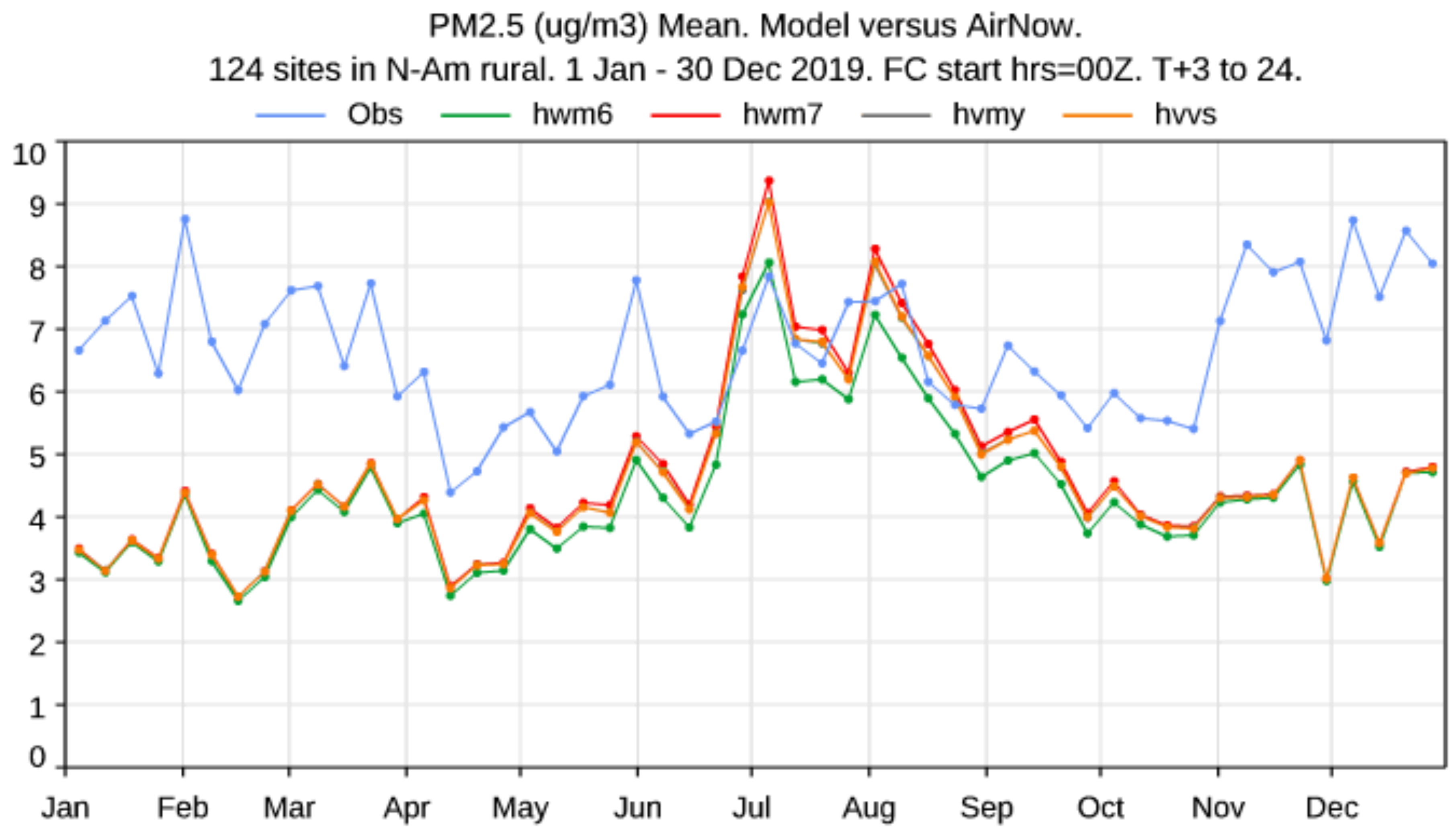
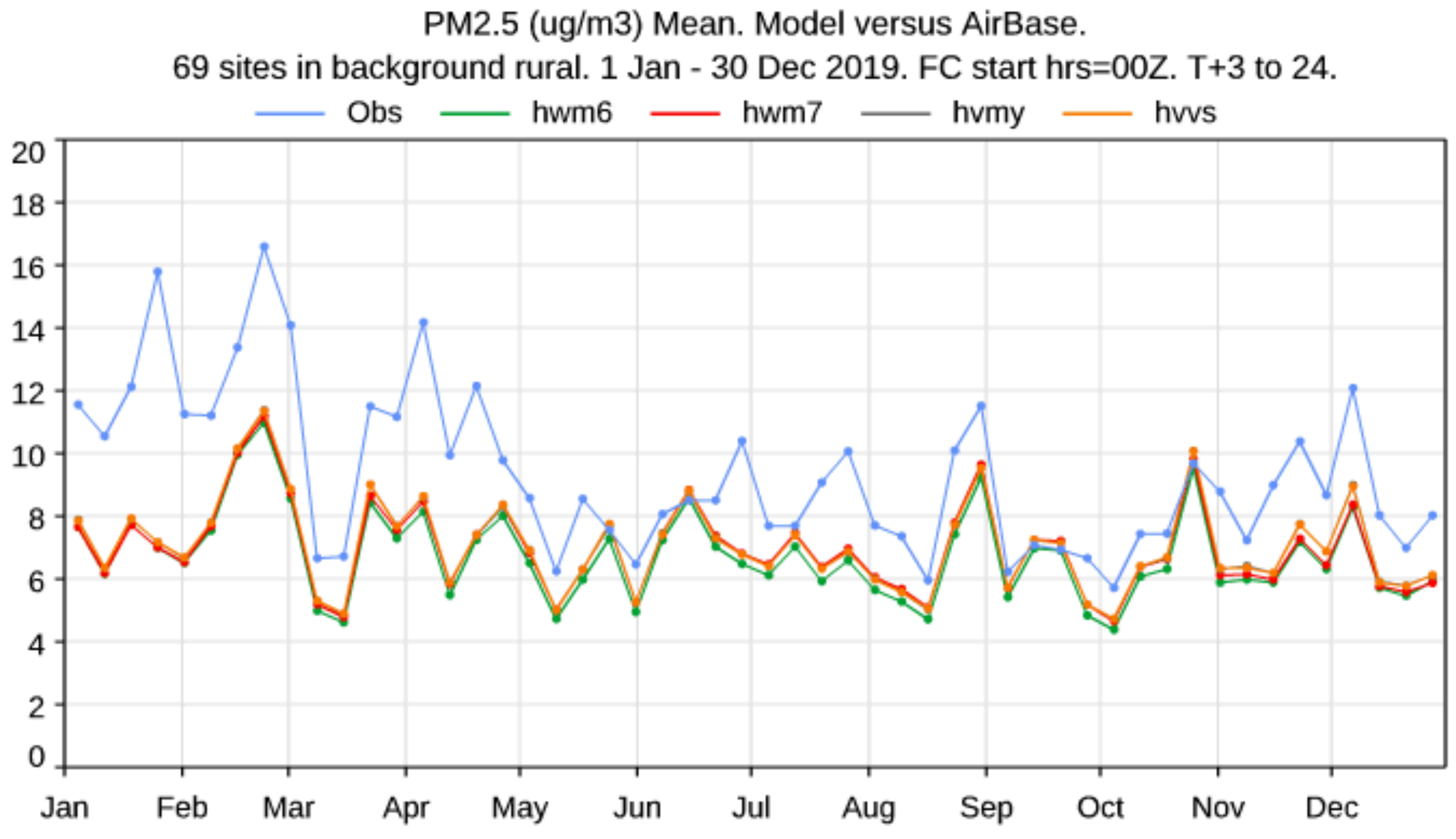
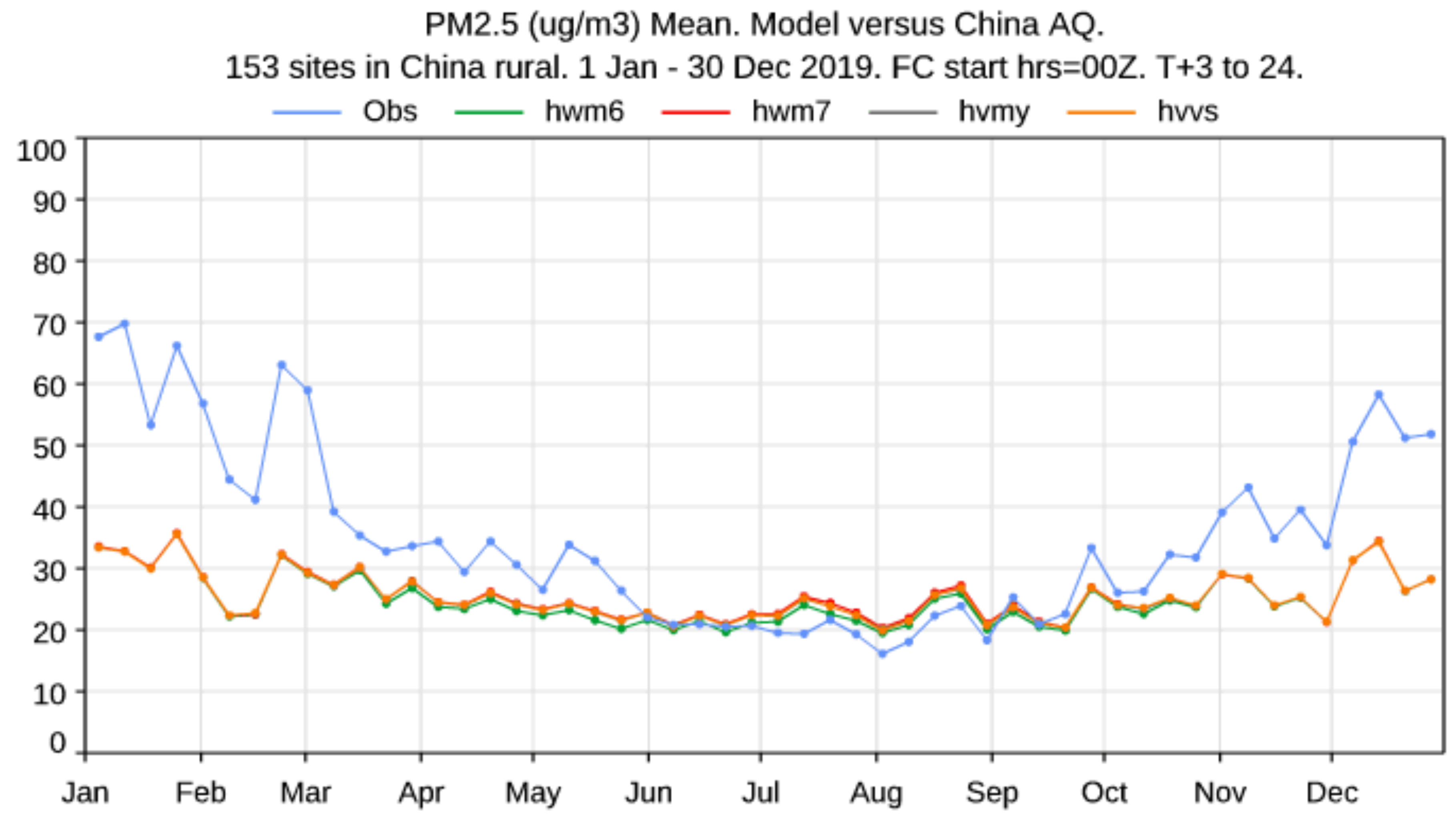
Exp id / name
hwm6 / NoCouple
hwm7 / AqRate
hwm9 / AqRate-L
hvmy / AqRate+AqPhase
hvvs / FullCouple







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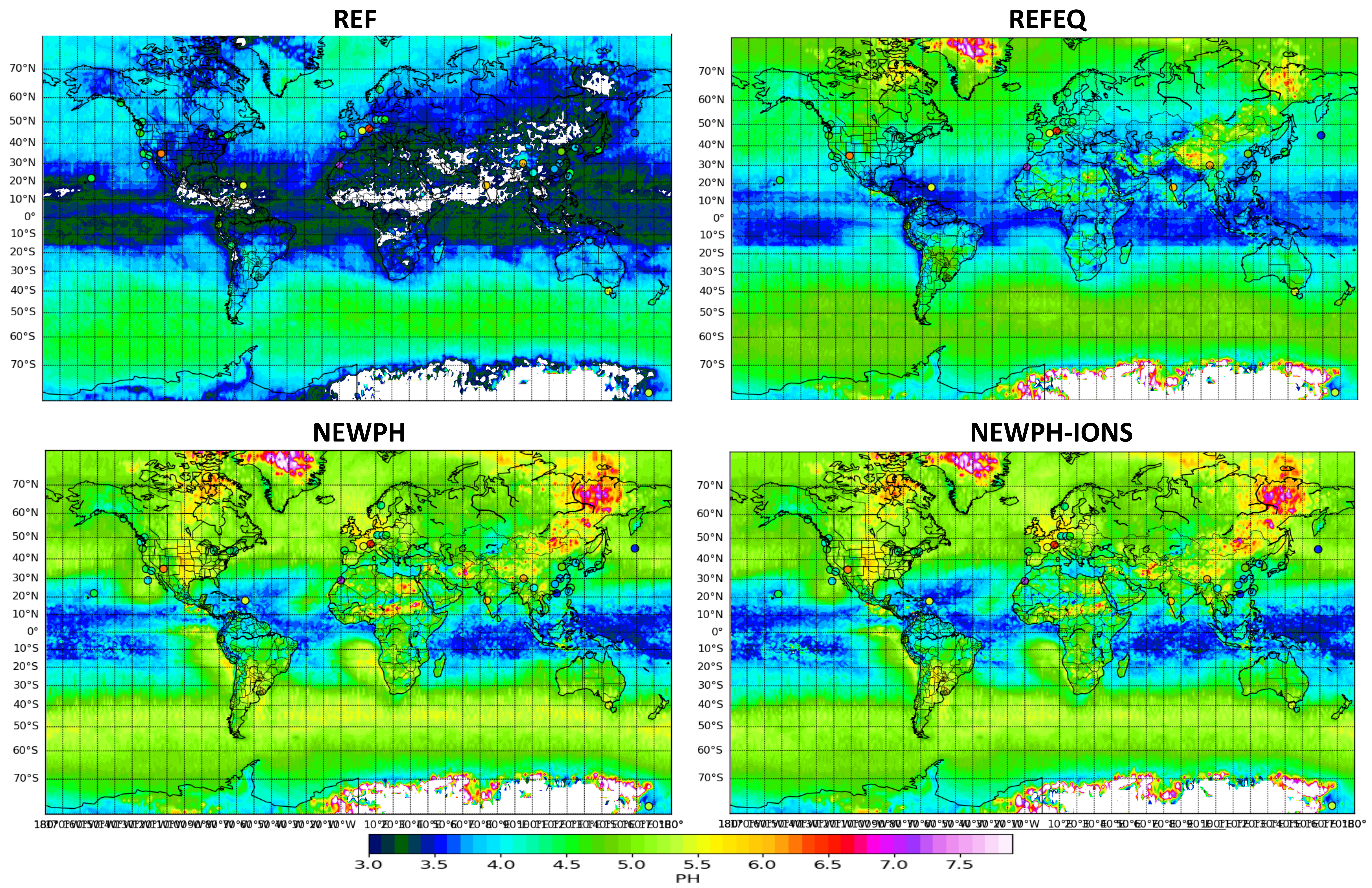
Sensitivity results II: cloud water pH (2019)

Experiment	pH computation
REF	CY47R3 single value for precip pH in wet deposition).
REFEQ	Oper (CY47R3 single value for precip pH in wet deposition), with EQSAM4Clim.
NEWPH	pH from EQSAM4Clim, updated in TM5 wetchem. More cations and anions input to EQSAM4Clim.
NEWPH-IONS	pH from EQSAM4Clim, updated in TM5 wetchem. Higher LWC threshold for aqueous chemistry.



– The global distribution of simulated cloud water pH, weighted by cloud water content and averaged between the surface and 700 hPa during 2019. Experiments shown are: REF (top left), REFEQ (top right), NEWPH (bottom left), NEWPH-IONS (bottom right). Note, averaging is experimental — it can lead to too low cloud pH.

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