HYGEOS

Royal Netherlands Meteorological Institute
Ministry of Infrastructure and the Environment

ResearchConcepts Io

Atmosphere Monitoring

First representation of aerosol acidity in cycle 49R1 of the ECMWF IFS

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ICAP, 10/11/2023

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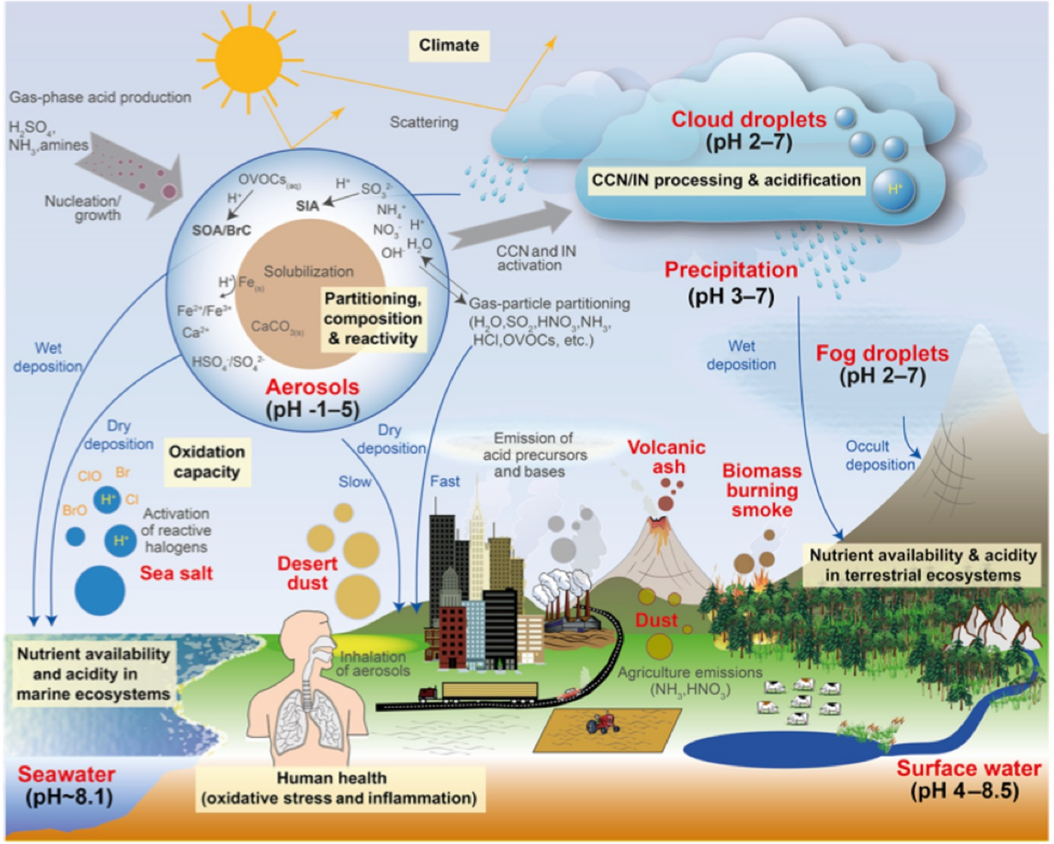
Aerosol acidity – why does it matter?

- Aerosol acidity is determined by the concentration of particulate cations and anions and the associated aerosol water content.
- Aerosol acidity is most often measured through potential hydrogen (pH)
- Transfer of gas-phase acids (HNO_3^-) and bases (NH_3) moderate the final acidity
- Aerosol acidity impacts cloud and precipitation pH (acid rain phenomenon).
- Cloud acidity in turn impact aqueous chemistry reactions (SO_2->$SO_4^=$; NH_3-> NH_4^+).
- Precipitation/cloud acidity also impacts wet deposition of SO_2 and NH_3.
- Precipitation acidity influences the pH in marine and terrestrial ecosystems.



Schematic of sources and receptors of aerosol and cloud droplet acidity, from Pye et al. (2020).

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IFS-AER in cycle 49R1- EQSAM4Clim

Cycle 49R1 is planned to become operational in October 2024. The cycle is frozen and evaluations are underway.

Cycle 49R1 will be used for next CAMS reanalysis.

Cycle 49R1 main aerosol related developments:

- New optical properties for sulfate
- New ageing scheme for OM/BC
- **Update and use of EQSAM4Clim for gas/particle partitioning**
- **Aerosol acidity from EQSAM4Clim used in aqueous chemistry and wet deposition**
- Common aerosol/chemistry wet and dry deposition
- Update of PM formula
- Desert dust developments:
  - Hydrophilic dust
  - Aspherical optics
- Simple representation of stratospheric sulphate

All results shown here are from IFS simulations **without data assimilation**

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IFS-AER in cycle 49R1 - EQSAM4Clim

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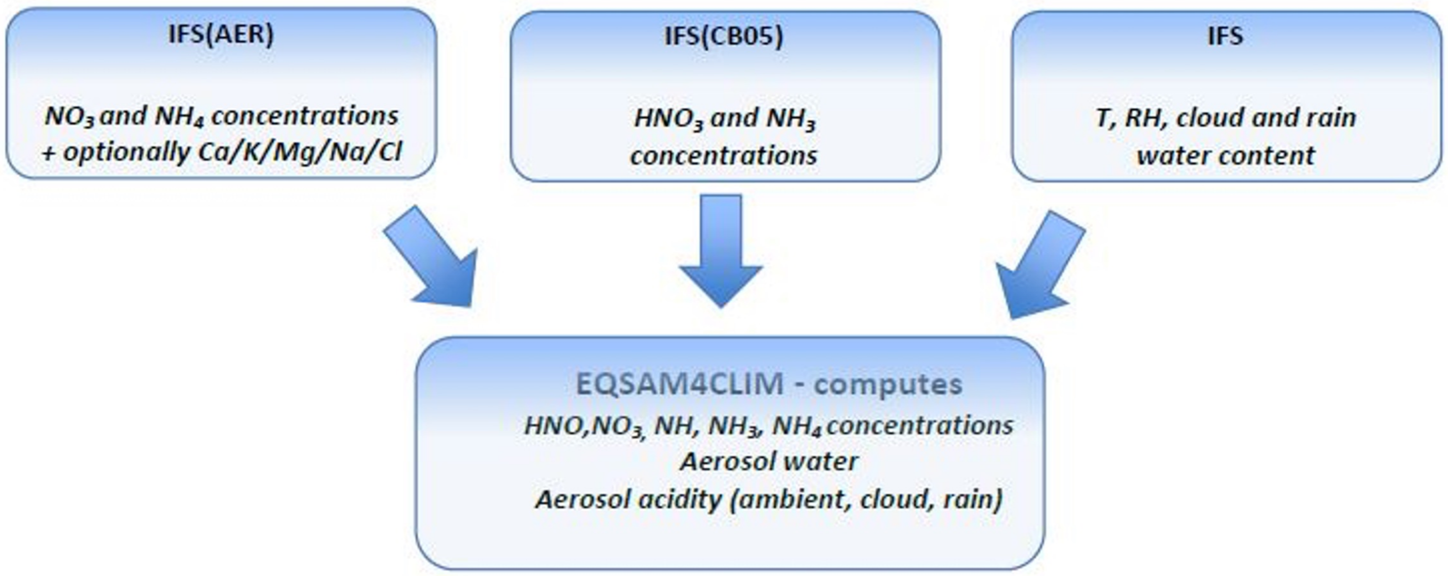
Cycle 49R1 main aerosol related developments:

- **Update and use of EQSAM4Clim for gas/particle partitioning**
- **Aerosol acidity from EQSAM4Clim used in aqueous chemistry and wet deposition**

EQSAM4Clim (Metzger et al 2016, 2018) is a simple and fast thermodynamical model based on EQSAM, implemented in the IFS in cycle 48R1, updated and used operationally in cycle 49R1. EQSAM4Clim is further developed and maintained by ResearchConcepts io GmbH



Schematic of the implementation of EQSAM4Clim in the IFS, showing inputs and outputs.

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IFS-AER in cycle 49R1 - EQSAM4Clim

The use of EQSAM4Clim led to a significant improvement of the representation of the nitrogen life cycle, by improving the representation of gas/particle partitioning

Table 2. Regional evaluation of the simulated surface concentration of selected aerosol and trace gases species. Mean bias, root mean square error (RMSE) and the Pearson correlation coefficient (R) are shown. Results are presented as CY48R1 / pre-CY49R1 / CY49R1_NOE4C. The values with the lowest biases are highlighted in bold. Values for the bias are given in $\mu g/m^3$. The evaluation is carried out against weekly values for CASTNET, three-daily values for AMoN, daily values for EMEP and yearly values for EANET.

species		USA (CASTNET/AMoN)	Europe (EMEP)	East Asia (EANET)
SO_4	mean Bias	**0.41** / 0.49 / 0.65	-0.51 / -0.42 / **-0.22**	**0.43** / 0.75 / 1.36
	RMSE	1.23 / **1.19** / 1.30	1.53 / 1.40 / **1.39**	**2.14** / 2.48 / 3.4
	R	0.52 / 0.52 / **0.54**	0.21 / 0.41 / **0.45**	**0.69** / 0.66 / 0.66
HNO_3	mean bias	-0.11 / 0.62 / **-0.07**	-0.12 / 0.36 / **-0.10**	
	RMSE	**0.50** / 0.87 / 0.50	**0.50** / 0.91 / 0.65	
	R	0.33 / **0.57** / 0.30	**0.21** / 0.20 / 0.20	
NO_3^-	mean bias	1.78 / **0.11** / 1.51	1.17 / **-0.08** / 0.6	3.19 / **-0.18** / 2.2
	RMSE	2.2 / **0.74** / 1.97	3.55 / **2.71** / 3.02	4.25 / **1.47** / 3.3
	R	0.31 / **0.65** / 0.24	0.19 / **0.32** / 0.21	**0.59** / 0.5 / 0.58
NH_3	mean bias	-0.35 / **-0.11** / -0.33	0.87 / **0.82** / 0.82	
	RMSE	1.77 / **1.72** / 1.76	1.84 / **1.67** / 1.79	
	R	0.44 / **0.46** / 0.44	0.45 / **0.57** / 0.48	
NH_4^+	mean bias	0.66 / **0.19** / 0.69	0.27 / **-0.05** / 0.33	1.16 / **0.63** /1.42
	RMSE	0.90 / **0.43** / 0.92	0.88 / **0.69** / 0.95	2.03 / **1.4** / 2.5
	R	0.32 / **0.45** / 0.30	0.40 / **0.49** / 0.38	0.43 / **0.45** / 0.41

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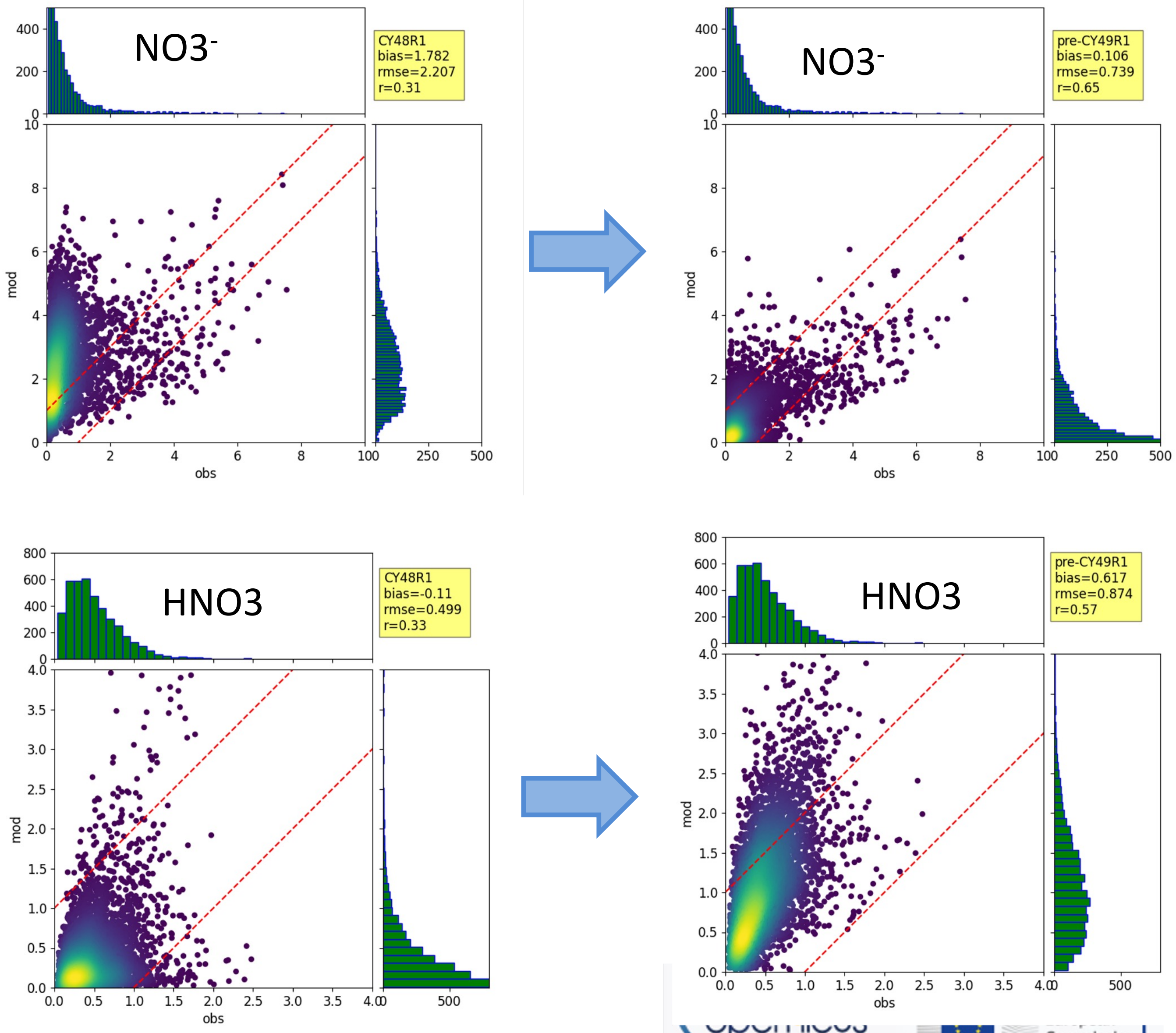
IFS-AER in cycle 49R1 - EQSAM4Clim

The use of EQSAM4Clim leads to a significant improvement of the representation of the nitrogen life cycle, by improving the representation of gas/particle partitioning for both oxidised and reduced nitrogen

Comparison of simulated surface concentration of HNO3/NO3 vs weekly CASTNET obs in 2019



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IFS-AER in cycle 49R1 - EQSAM4Clim

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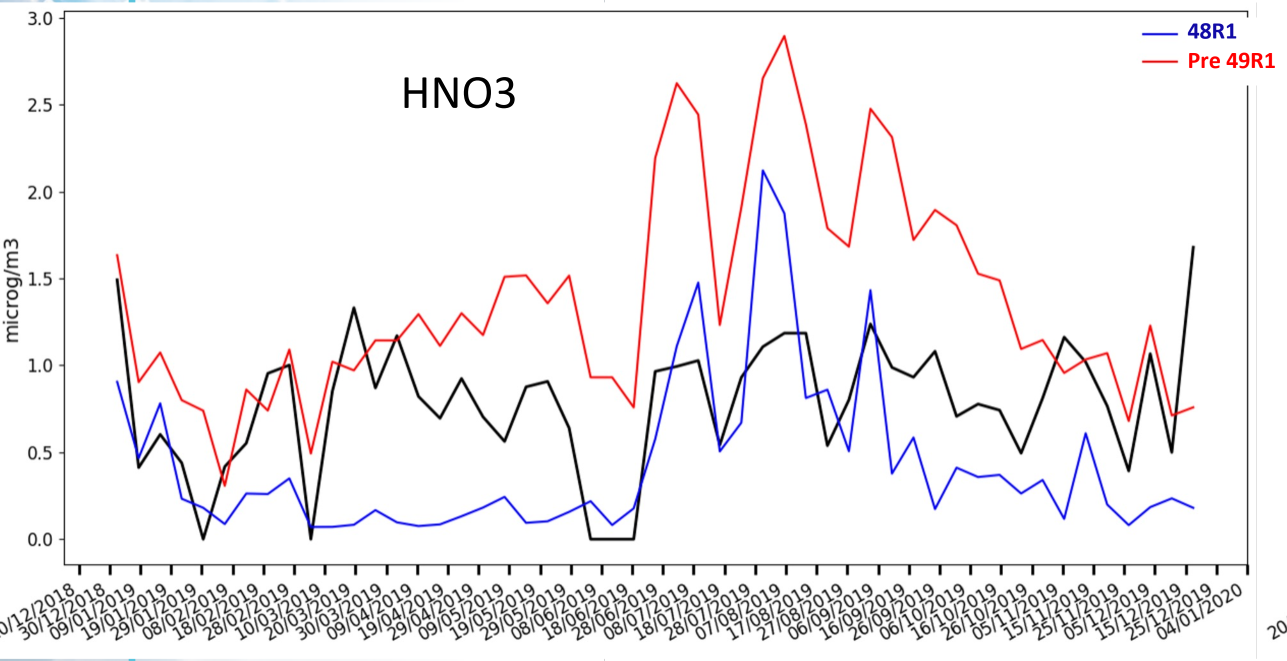
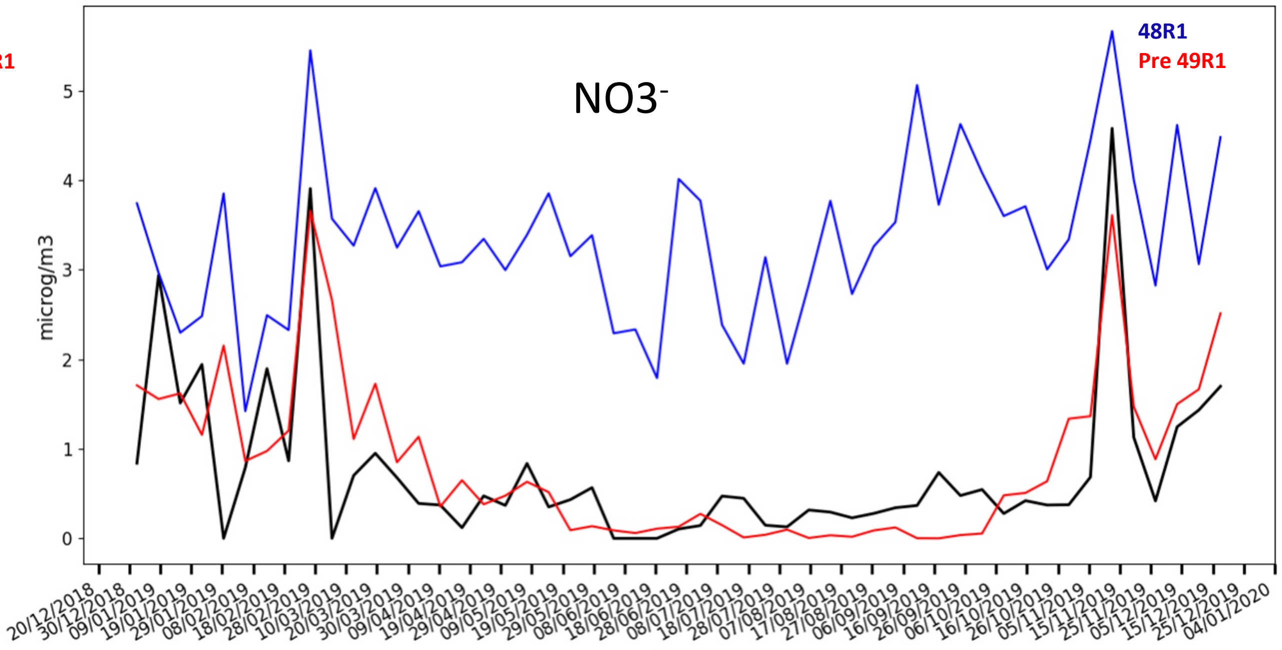
The use of EQSAM4Clim led to a significant improvement of the representation of the nitrogen life cycle, by improving the representation of gas/particle partitioning.

Comparison of simulated surface concentration of HNO_3/NO_3^- vs weekly CASTNET observations in 2019, at Mackville (Kentucky).





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IFS-AER in cycle 49R1 - EQSAM4Clim

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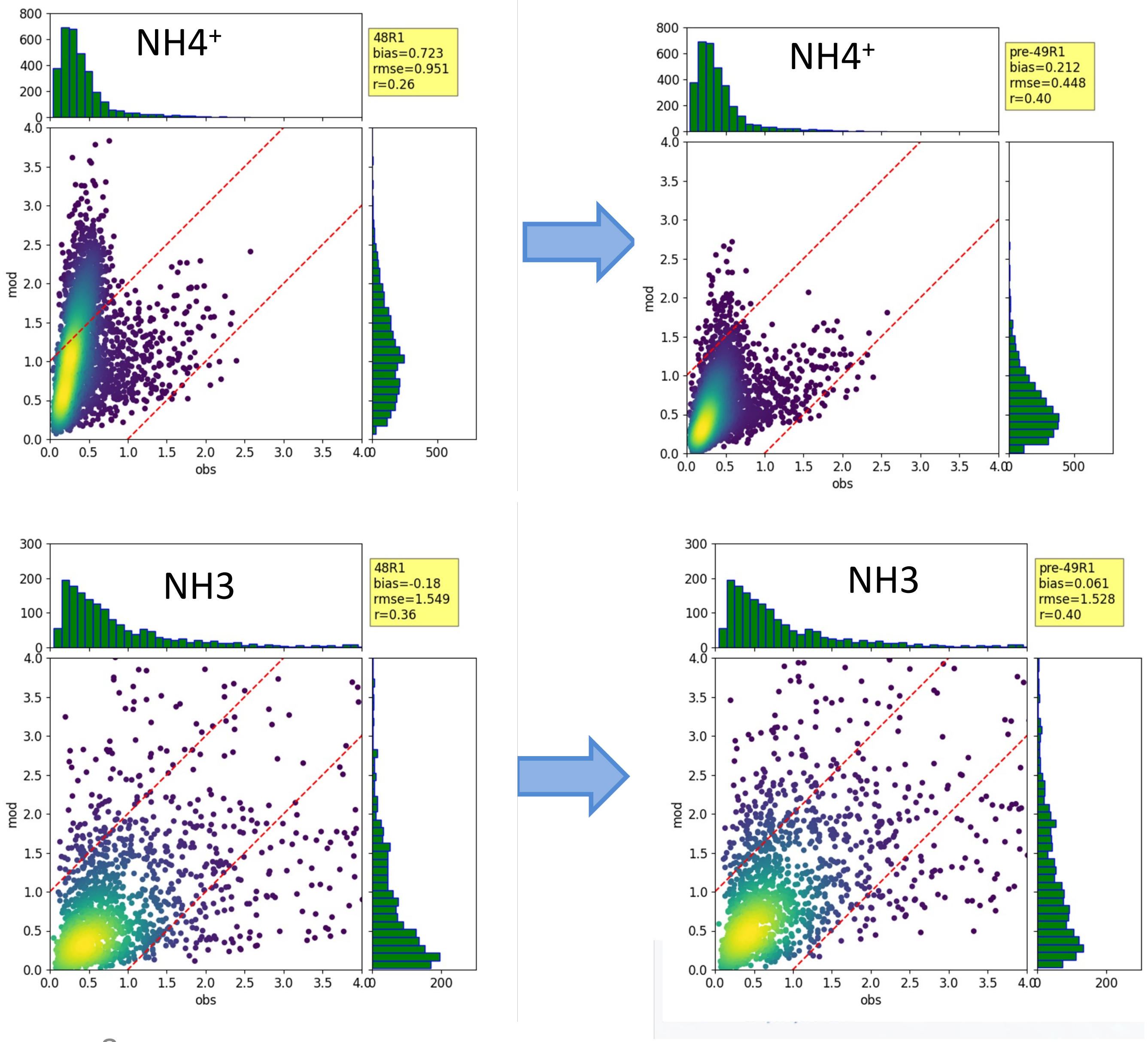
Comparison of simulated surface concentration of NH_3/NH_4^+ vs AMoN/CASTNET obs in 2019



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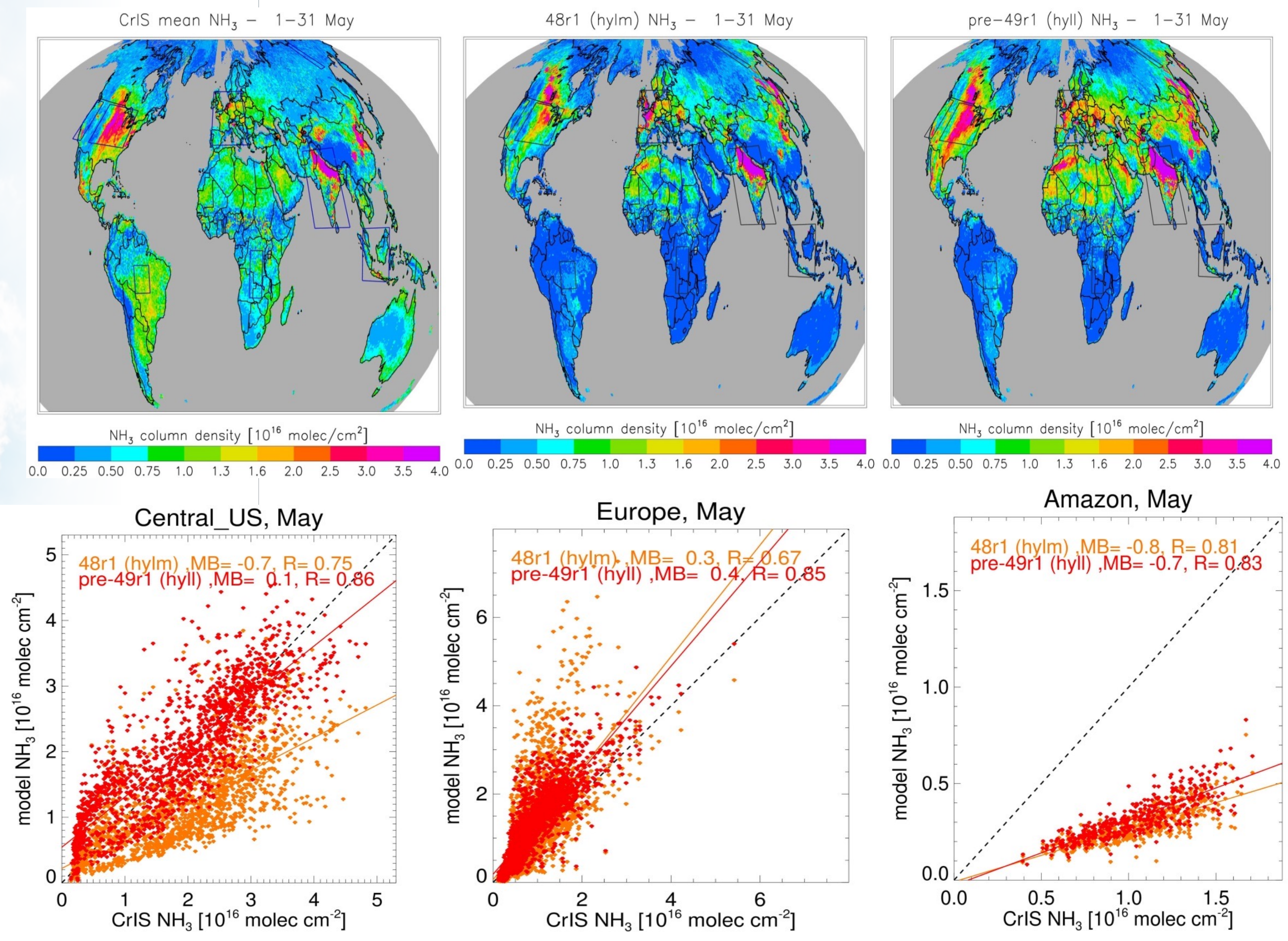
IFS-AER in cycle 49R1 - EQSAM4Clim

The use of EQSAM4Clim leads to a significant improvement of the representation of the nitrogen life cycle, by improving the representation of gas/particle partitioning for both oxidised and reduced nitrogen.

Evaluation of simulated NH3 burden versus CrIS retriveals for May 2018



IFS-AER in cycle 49R1 - EQSAM4Clim

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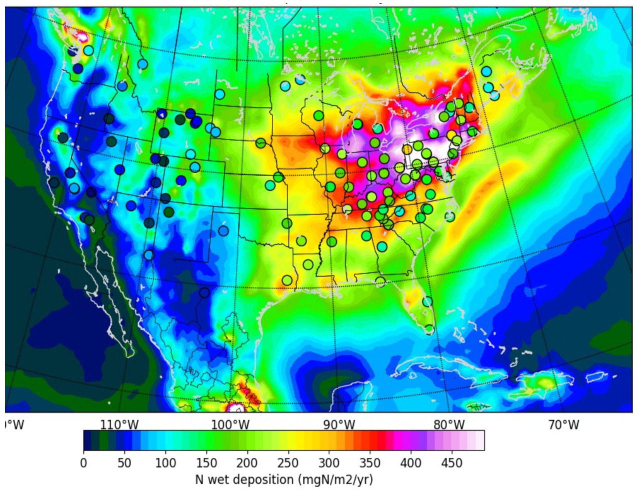
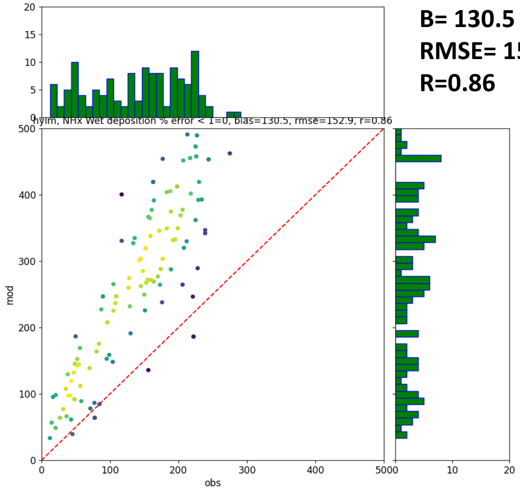
The use of EQSAM4Clim leads to a significant improvement of the representation of the nitrogen life cycle, by improving the representation of gas/particle partitioning for both oxidised and reduced nitrogen.
Evaluation of simulated HNO3+NO3 wet deposition versus CASTNET obs for 2018

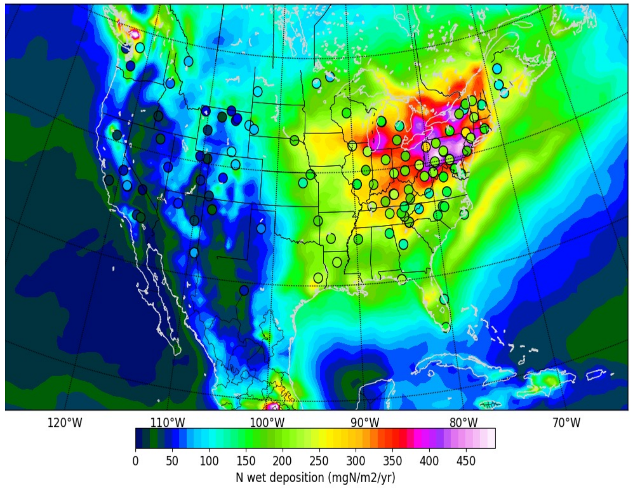
48R1

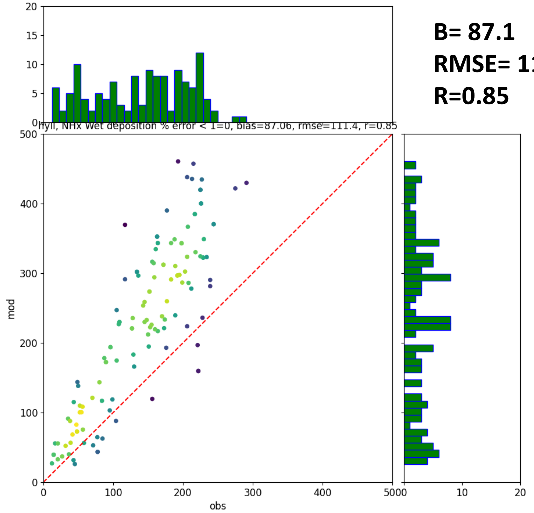




B= 130.5
RMSE= 152.9
R=0.86

Pre 49R1





B= 87.1
RMSE= 111.4
R=0.85

IFS-AER in cycle 49R1 - EQSAM4Clim

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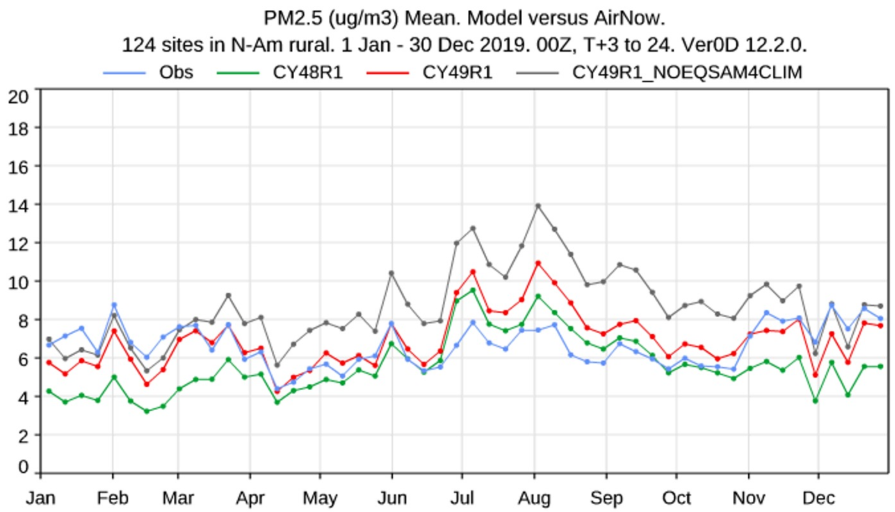
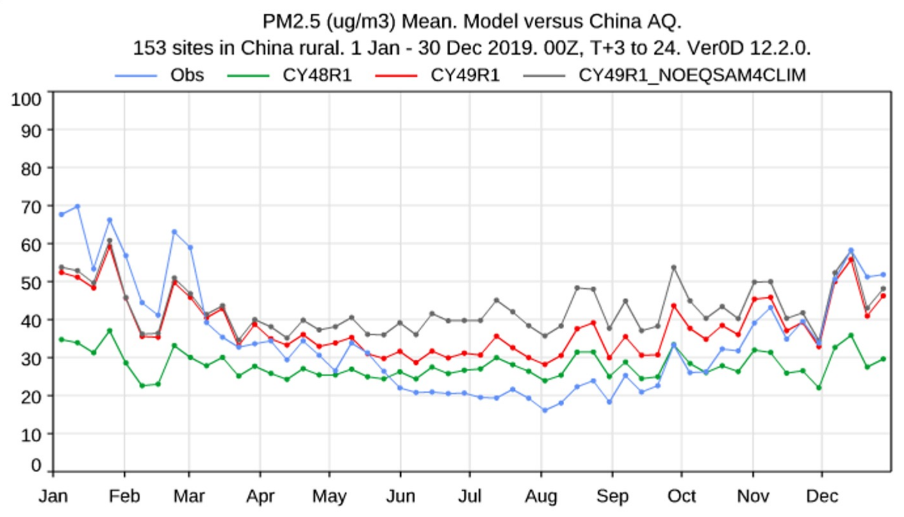
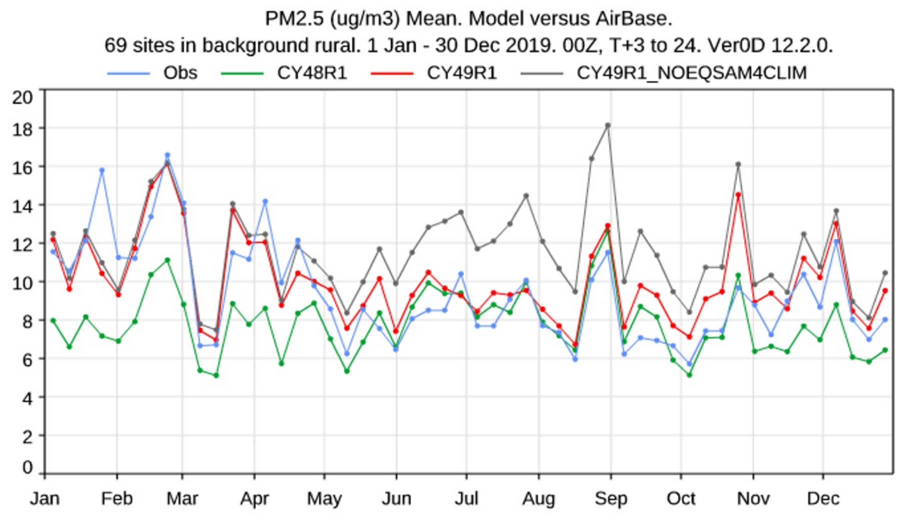
The use of EQSAM4Clim led to a significant improvement of the representation of the nitrogen life cycle, by improving the representation of gas/particle partitioning.

Test of pre 49R1 with and without EQSAM4CLIM shows that EQ4Clim has a positive impact on simulated PM2.5







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Aerosol pH in the IFS

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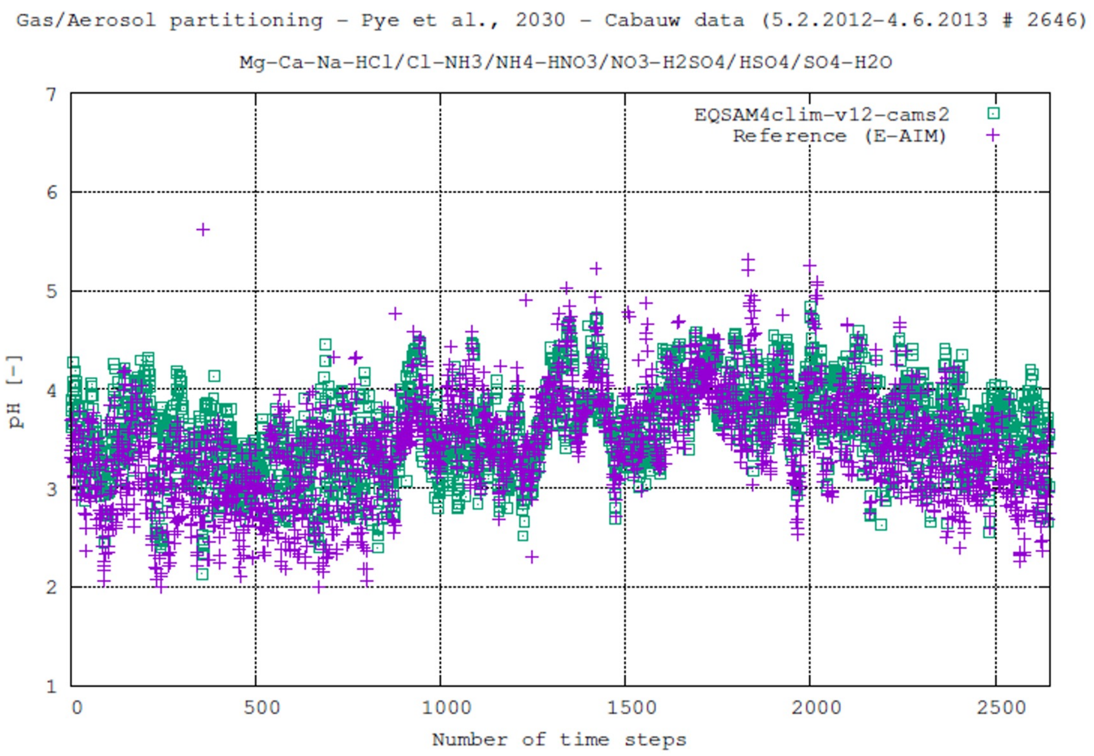
EQSAM4Clim computes aerosol pH – but how well? Problem: aerosol pH is (very) seldom measured.

Box model comparison using data from super sites that provide all input of EQSAM4Clim : cations/anions concentration, meteorological input. Here for Cabauw (Netherlands). E-AIM, a complex thermodynamical model, provides reference data.



From S. Metzger

Aerosol acidity in the IFS

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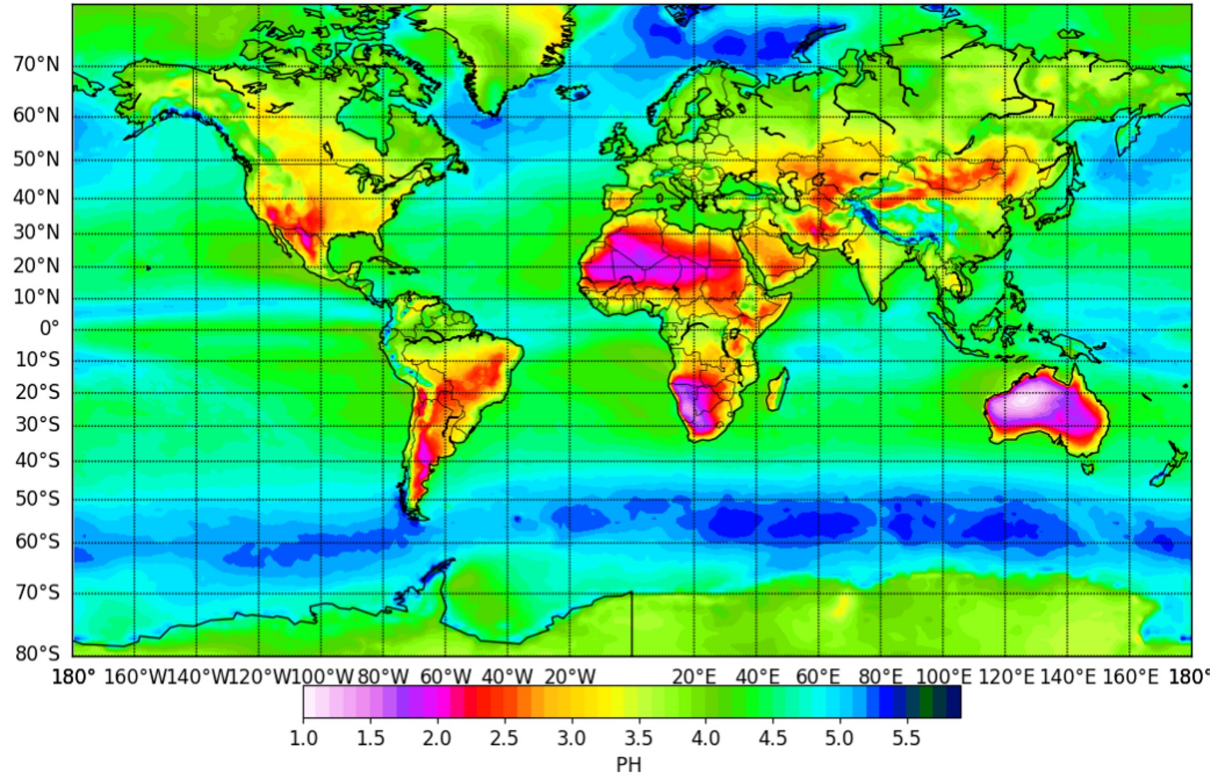
New diagnostic output of the IFS in cycle 49R1:

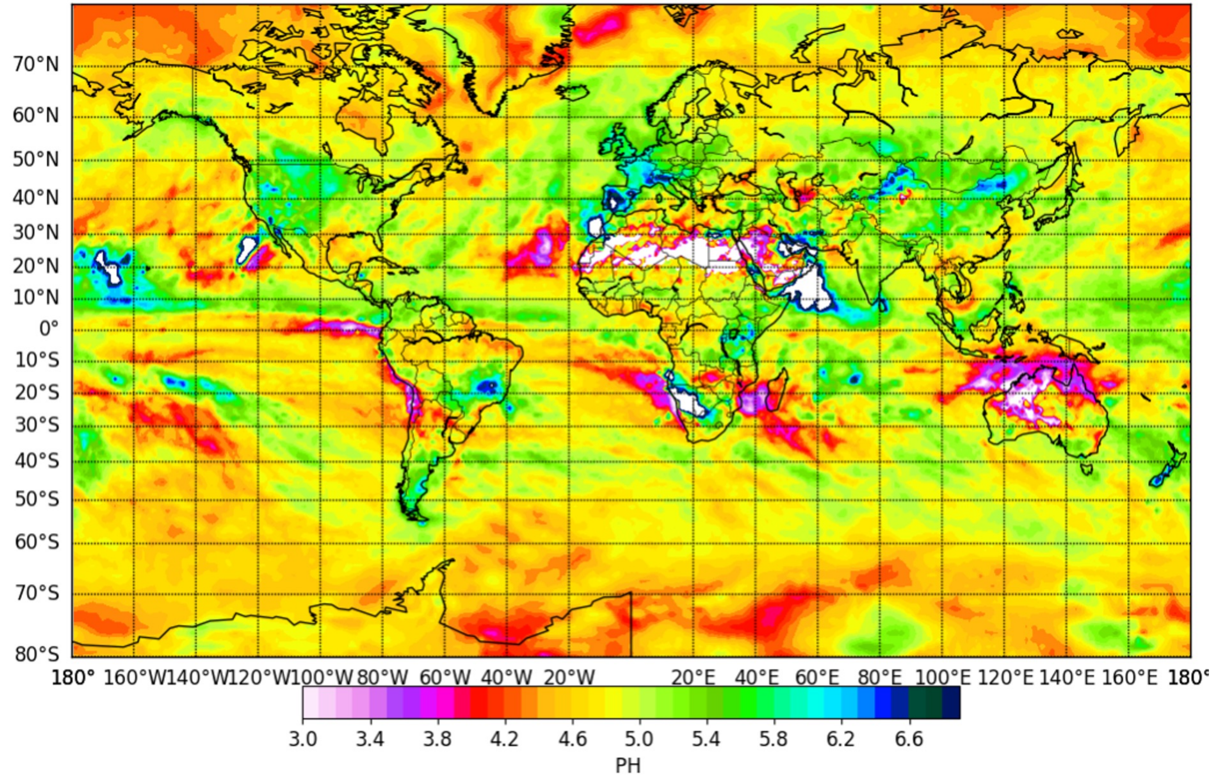
- Aerosol pH at surface
- 3D cloud and rain pH

Aerosol pH at surface is influenced by aerosol composition **and** aerosol water

Cloud/Rain pH more impacted by cloud/rain water than aerosol composition



2019 Surface aerosol pH



2019 surface precipitation weighted rain pH

Evaluation of aerosol acidity products

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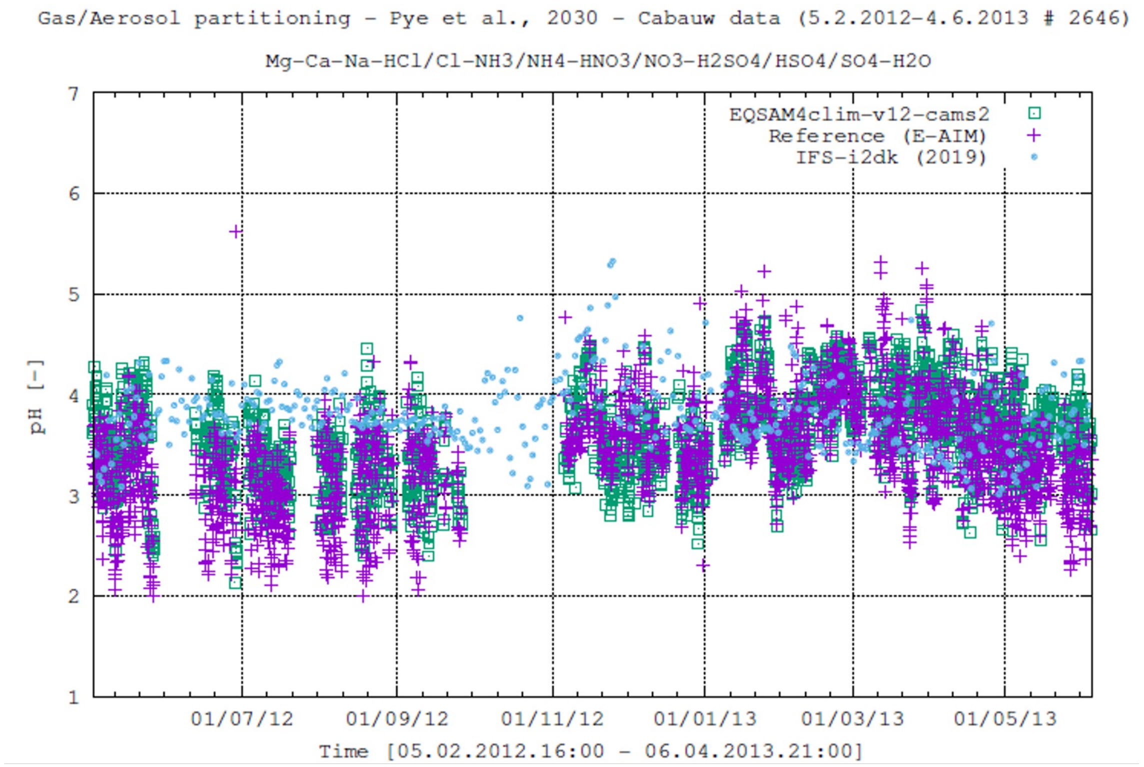
Evaluation of aerosol pH not easy because of lacking observations.

Use of super-site data: here, Cabauw.

IFS simulated aerosol pH is often biased, missing variability



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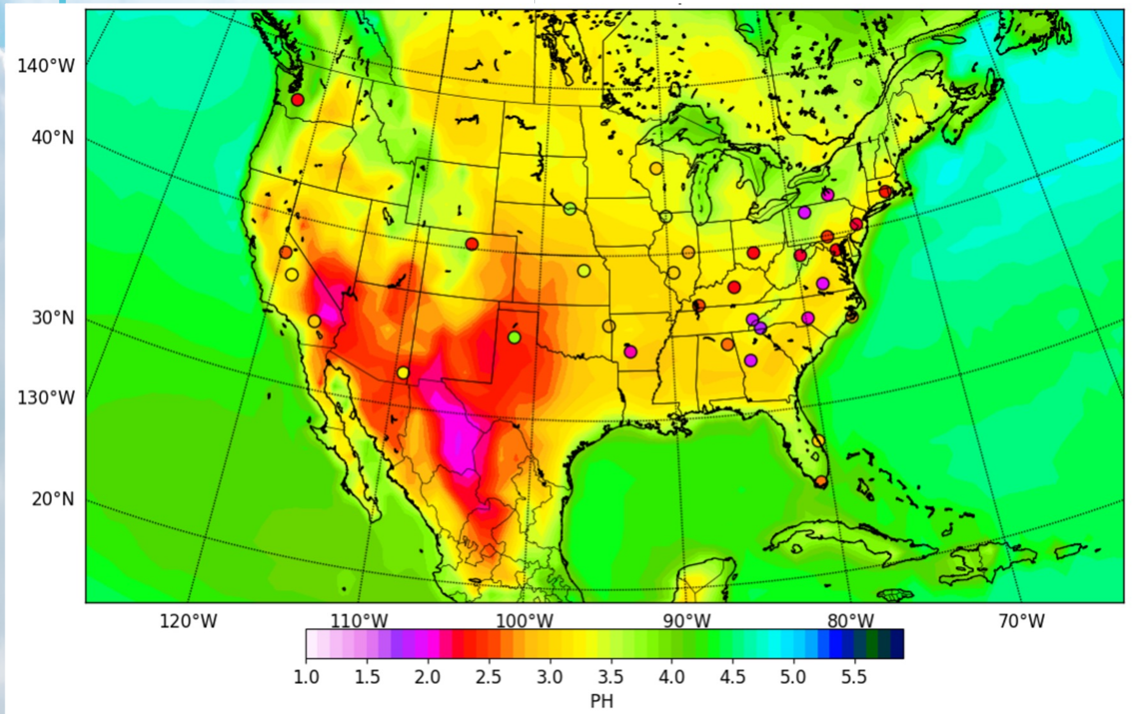
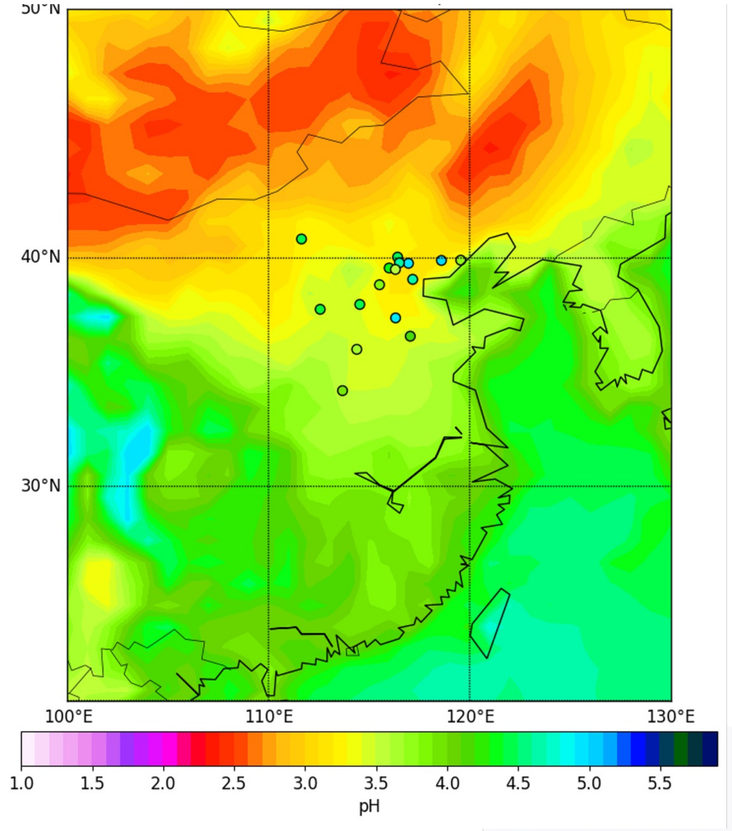
Evaluation of aerosol acidity products

Evaluation of aerosol pH not easy because of lacking observations.

Use of 2011 (US)/2017 (China) surface aerosol pH computed by ISORROPIA2, using CASTNET/AMoN/Chinese stations input, from Zhang et al. (2021, ACP), compared against 2019 simulated values

In general, high bias over U.S. (not acidic enough) and low bias over China (too acidic)





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Evaluation of aerosol acidity products

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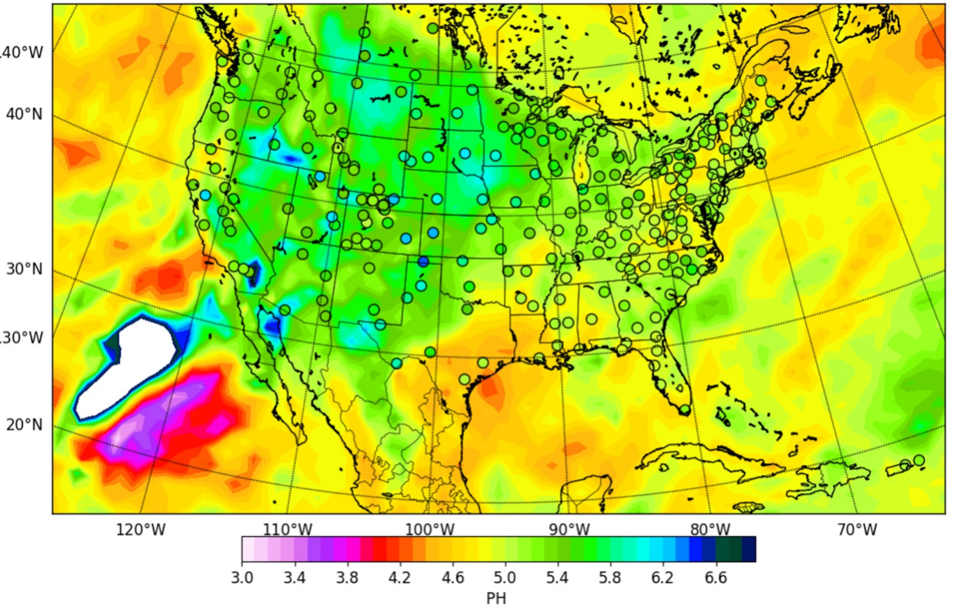
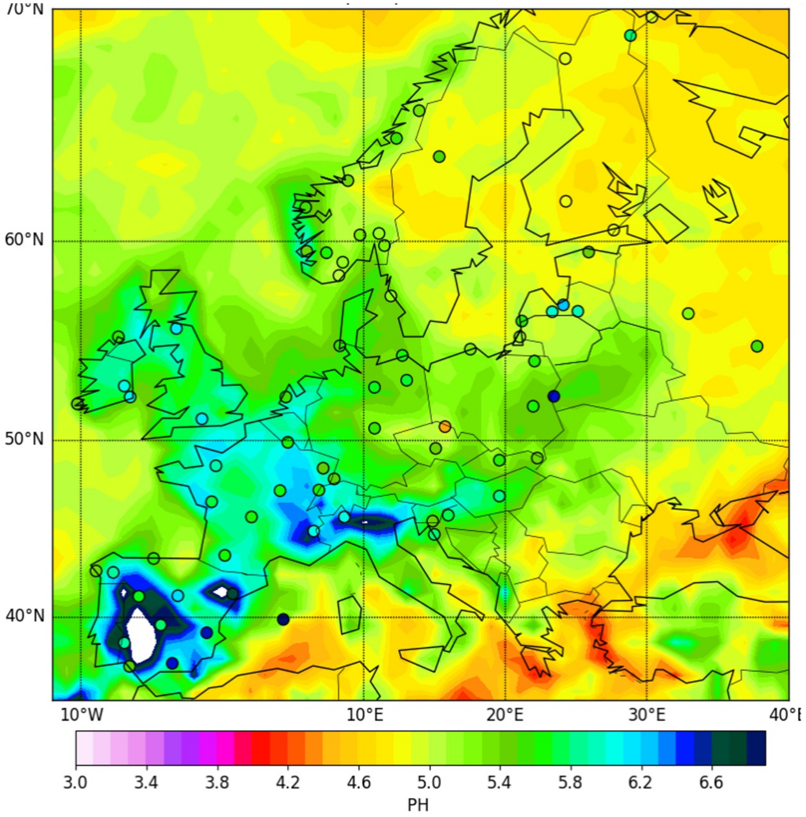
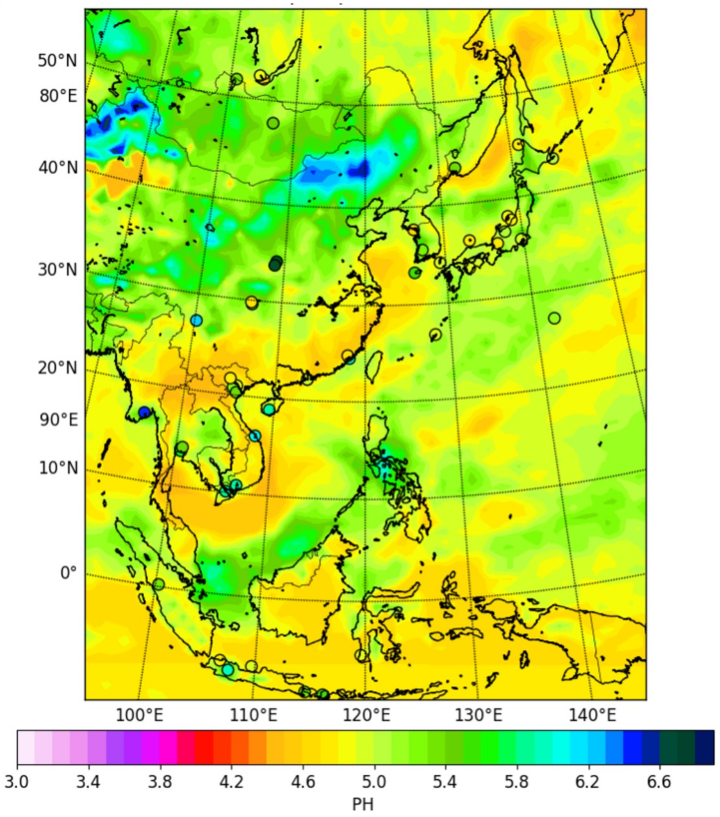
Evaluation of rain pH versus routine observations over US (NTN/NADP), Europe (EMEP) and Asia (EANET) in 2019







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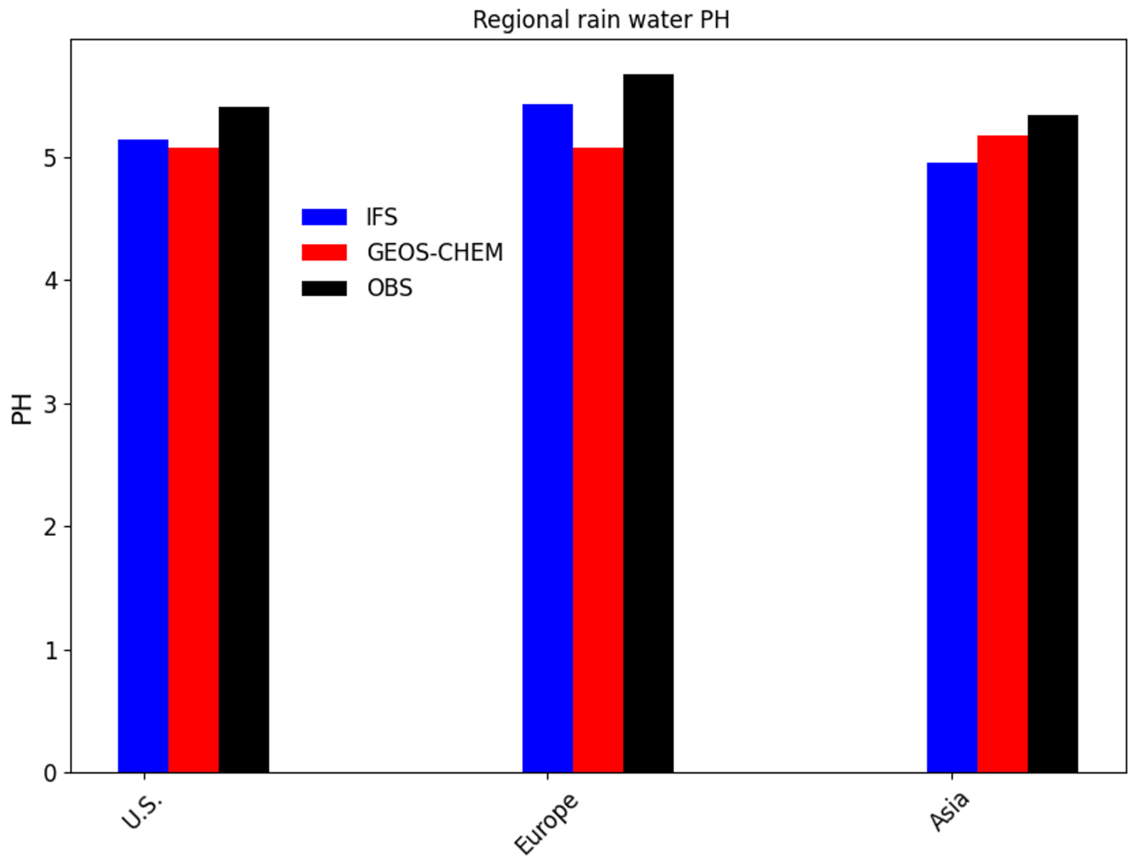
Evaluation of regional rainwater pH

Evaluation of regionally averaged 2019 rain pH (surface, precipitation weighted) versus observation and output from GEOS-CHEM (2013, from Shah et al. 2020, ACP).



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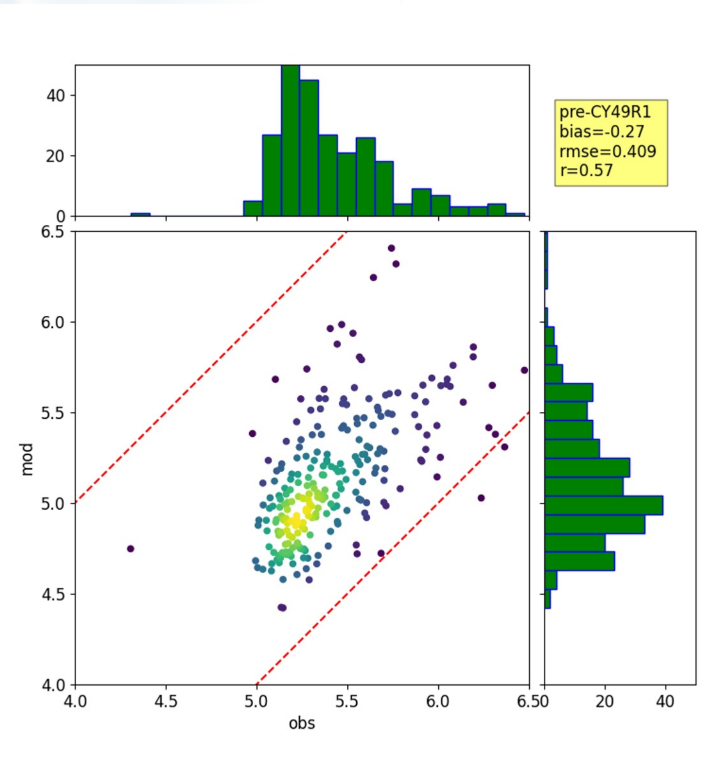
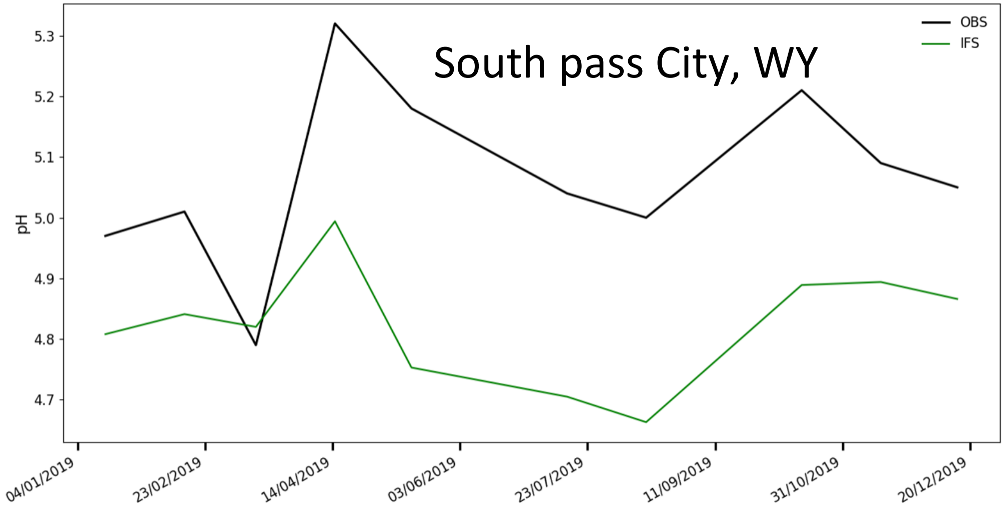
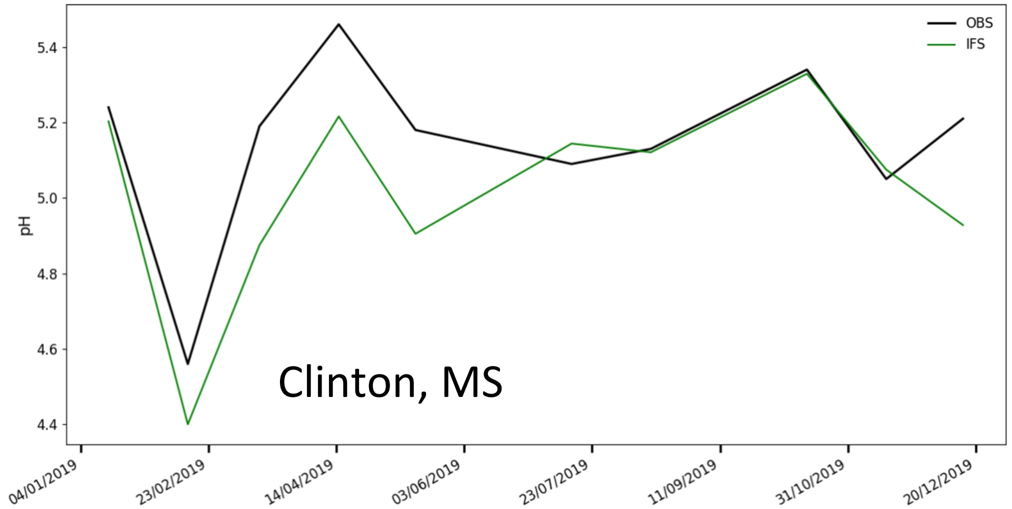
Evaluation of US rainwater pH

Evaluation of monthly simulated rain pH (at surface, precipitation weighted) versus NTN obs in 2019







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Conclusion/summary

- The use of EQSAM4Clim in cycle 49R1 improves substantially the nitrogen life cycle by reducing that large positive biases in both NO3- and NH4+.
- It also allows for the computation and use of aerosol, cloud, rain acidity, which can act as a future ECMWF product
- EQSAM4Clim in the IFS requires very little extra computing cost
- New diagnostic output of these quantities are experimental products! More evaluation is needed.

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