Implementation of ISORROPIA-lite thermodynamic module within EMAC, implications for aerosol composition, acidity, and radiative forcing.

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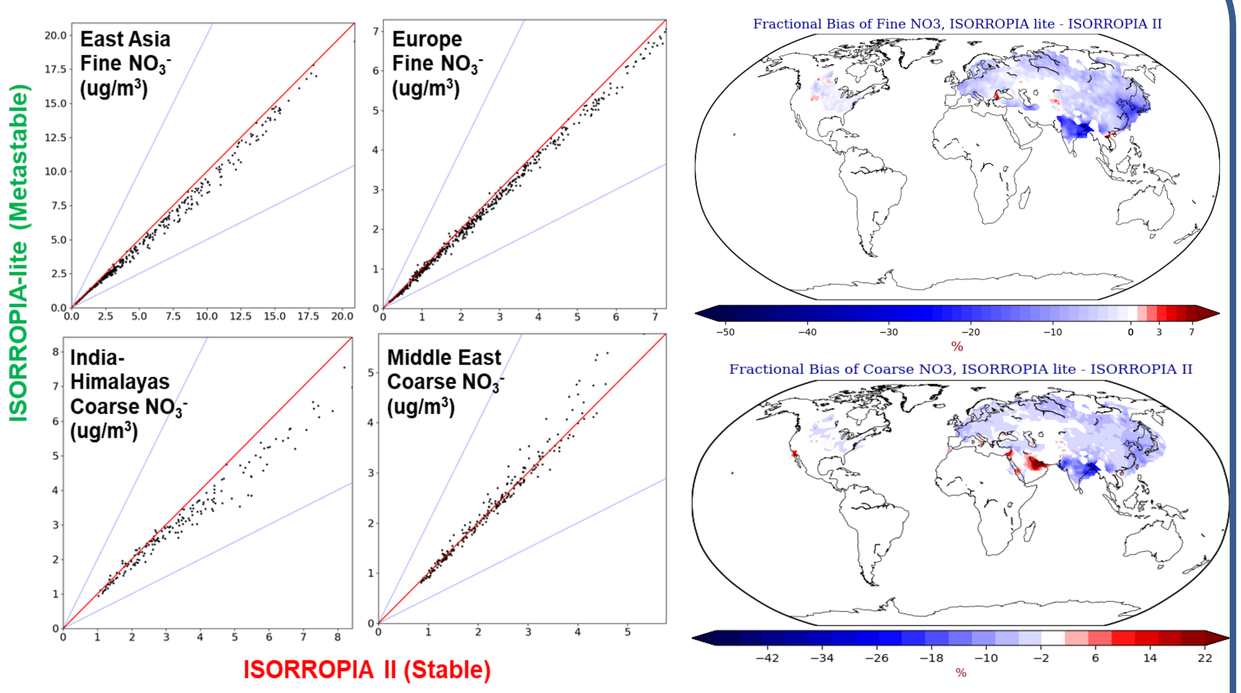
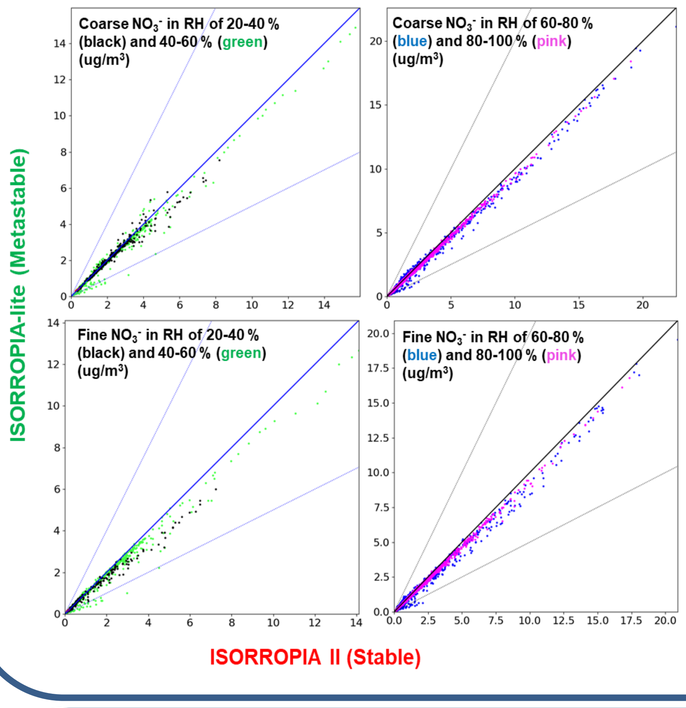
Abstract: This study focuses on the performance and results of a lighter and more computationally efficient version of the ISORROPIA II thermodynamic module, i.e., ISORROPIA-lite, in the global atmospheric and chemistry model EMAC. The main focus is to compare ISORROPIA-lite with ISORROPIA-II (in stable mode), and for that reason, simulations were completed in the ''forward'' problem configuration (gas + aerosol as input), for the years 2009 & 2010. An evaluation of the results of ISORROPIA-lite is also performed by comparing them with surface observations from three different networks in North America (IMPROVE), Europe (EMEP), and East Asia (EANET). The examined aerosol components are nitrate (NO_3^-) in the coarse and fine size modes, as well as sulphate (SO_4^{-2}), ammonium (NH_4^+), crustal ions (Na^+, Ca^+, K^+, Mg^+), water mass fraction (WMF) of aerosols and aerosol acidity (pH). Firstly, the model predictions compare quite well with the observations, apart from some overpredictions of $PM_{2.5}$ nitrate over Europe and East Asia (~2 & 5 μg m^{-3} respectively or 20%). The observed differences between ISORROPIA-lite and ISORROPIA-II are minimal except for some overpredictions by the latter in inorganic aerosol concentrations and underprediction of the WMF, producing in the majority Mean Error values <10%. Regarding acidity, ISORROPIA-lite produced somewhat more acidic particles (for about 2 pH units) with further sensitivity simulations showcasing that NH_3 plays a major role in the buffering of the accumulation mode pH. Regarding the computational efficiency of ISORROPIA-lite, it exhibited a speed up by 4% & 5% compared to ISORROPIA-II in metastable and stable mode respectively and was calculated based on the number of total time steps that EMAC performed during the same running period.

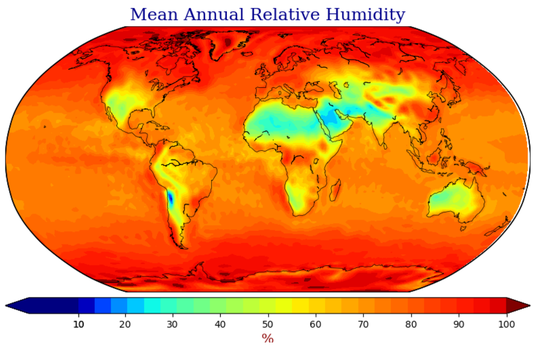
Model performance in NO_3^- over specific regions



- Disagreements in coarse & fine NO_3^- (up to 4 μg m^{-3} or 50 %) over Himalayan Region with characteristic low RH values < 40% that do not favor nitrate aerosol formation in metastable.
- Reverse behavior in coarse mode NO_3^- over Middle East due to low water content available for HNO_3 condensation in stable.
- Better agreement over North America, East Asia and Europe where annual RH values are higher

Relative Humidity dependence of NO_3^-

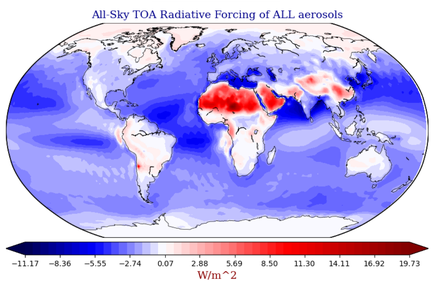




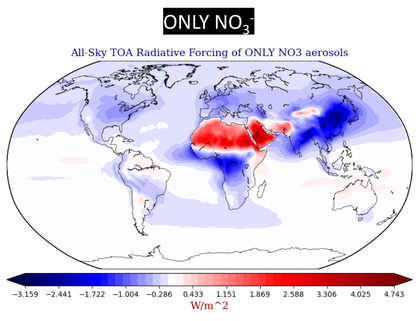
- Bigger biases in the lower RH ranges (20 – 60 %).
- Along with mid-range temperatures, NO_3^- aerosol formation is not favored by the supersaturated state.
- Regions governed by such atmospheric conditions have most disagreements.

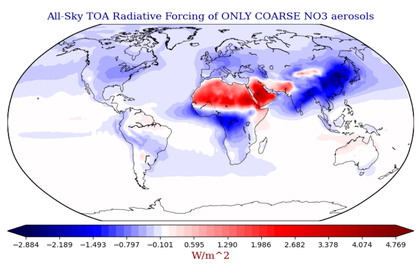
Direct TOA Radiative Forcing (10 year Period)

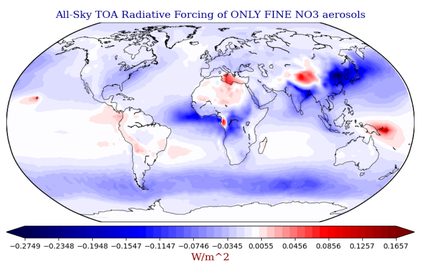
ALL AEROSOLS



ONLY NO_3^-







TOA RF (W m^{-2})	Net	LW	SW
ALL aerosols	- 1.48	+ 0.44	- 1.93
ALL NO_3^-	- 0.14	+ 0.23	- 0.37
Coarse NO_3^-	- 0.12	+ 0.23	- 0.35
Fine NO_3^-	- 0.02	~ 0	- 0.02

ONLY COARSE NO_3^- ONLY FINE NO_3^-

- The aerosol Direct Radiative Forcing is controlled more by the effect of the SW radiation flux.
- The NO_3^- Direct Radiative Forcing is controlled more by the Coarse Mode particle phase.

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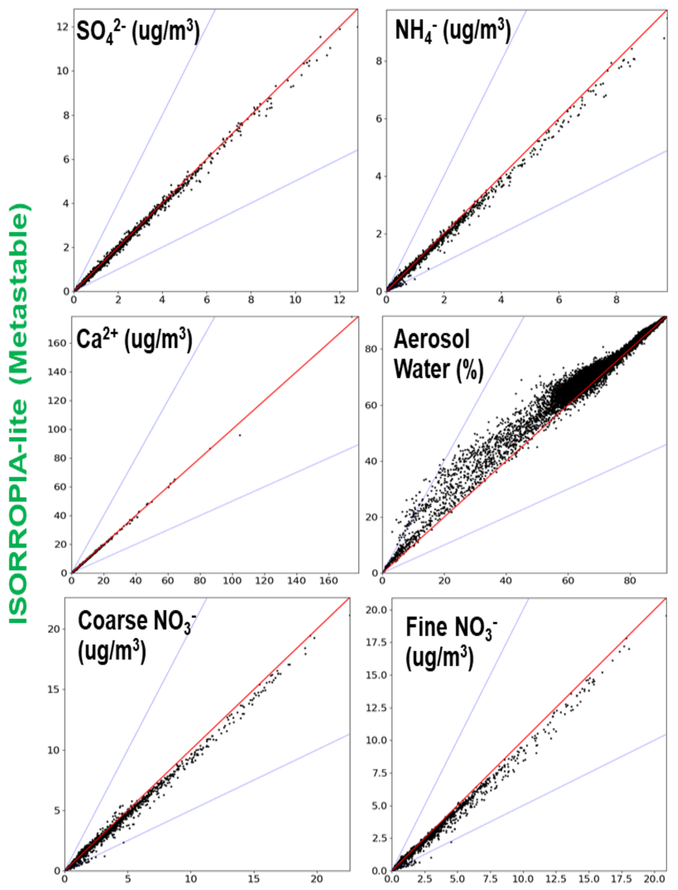
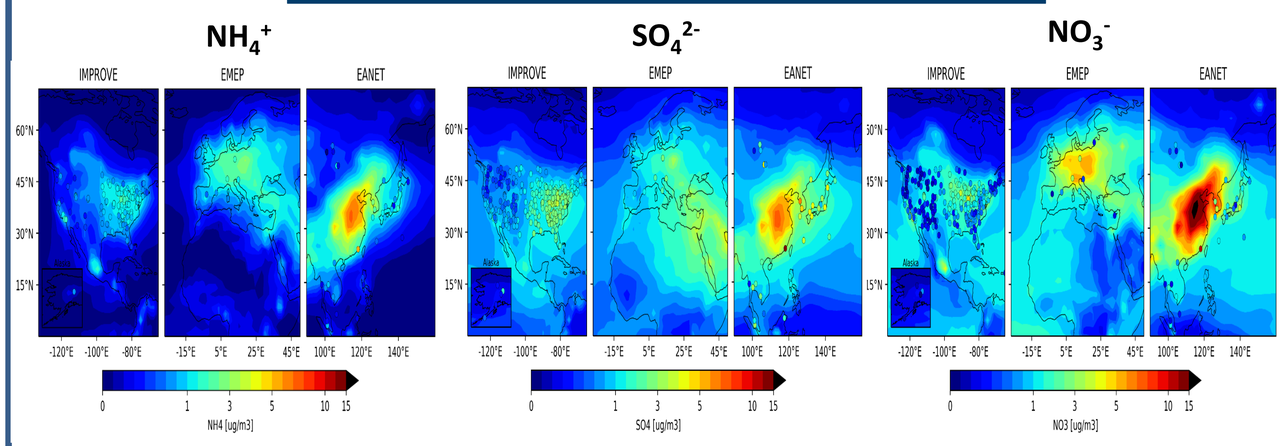
Comparison Against ISORROPIA II (Stable)



	Mean Bias (μg m^{-3})	Normalized Mean Error (%)
Coarse NO_3^-	-0.033	6.2
Fine NO_3^-	-0.025	7.6
HNO_3	-0.012	12.6
NH_4^+	-0.004	6.3
SO_4^{-2}	-0.012	3.0
Na^+	-0.460	8.1
Ca^{+2}	-0.021	4.0
K^+	-0.018	5.2
Mg^+	-0.057	7.0
Cl^-	-0.819	8.3
H_2O	1.320	3.6

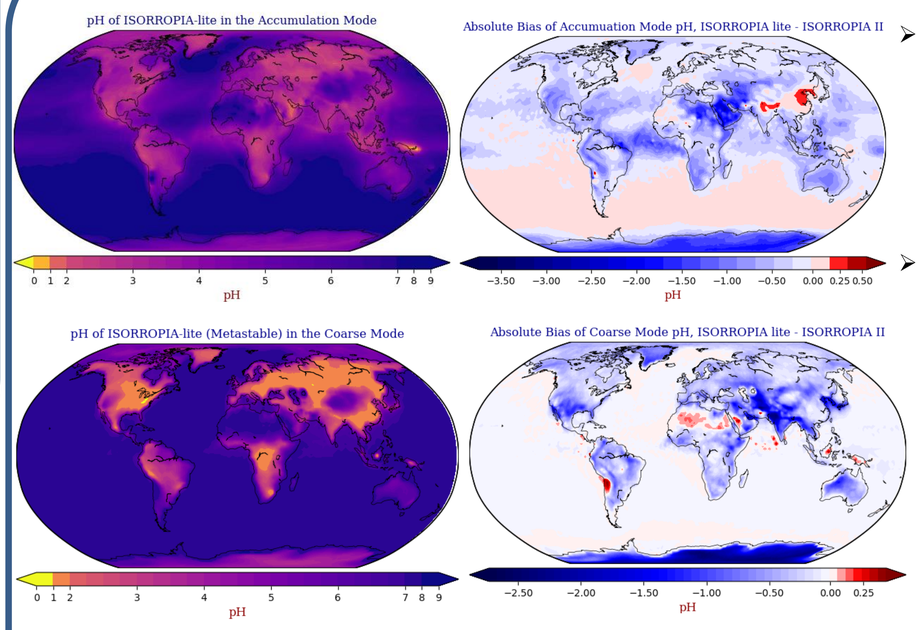
- A few underpredictions of up to 1 μg m^{-3} (10-20 %), for NH_4^+ and SO_4^{2-}.
- Excellent agreement for crustal ions.
- Slightly larger biases-in low concentrations- up to 3 μg m^{-3} (~40 %) for coarse & fine NO_3^-.
- Overprediction of aerosol water due to the different aerosol state assumption.

Model Evaluation vs Observations

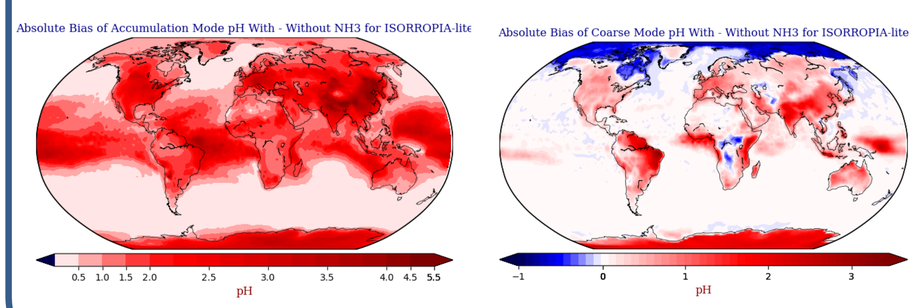


- Exceptional agreement for **NH_4^+** over North America and Europe. Model **overprediction** of ~2 μg m^{-3} (~10 %) over East Asia.
- Exceptional agreement for **SO_4^{2-}** over North America and Europe. Slightly stronger **overprediction** of East Asia concentrations.
- Best agreement for **NO_3^-** over North America. Model **overprediction** over Europe (~2 μg m^{-3} or 20 %) and East Asia (~ 5 μg m^{-3} or 20%).

Estimated Aerosol Acidity



- **Metastable** state produces **more acidic** accumulation mode aerosols particularly over arid regions, due to no or extremely low water available for pH calculation by stable.
- The more basic pH values of East Asia are due to higher aerosol water present in metastable state.
- **Metastable** state produces **more acidic** coarse mode aerosols over polluted regions, although with smaller bias (~1 pH).
- More efficient dust removal along with non-existent salt precipitation in **metastable**, lead to more acidic particles.



- The presence of NH_3 drives the pH of accumulation mode to be significantly more basic.
- Effect on coarse mode pH is similar, but not so evident.
- Should NH_3 be present, NH_4NO_3 would be formed in arid regions which is less acidic (than KNO_3 or $CaNO_3$).

Conclusions:

- ❑ Successful implementation of ISORROPIA-lite in EMAC showing very good agreement with ISORROPIA II, while reproducing observations competently.
- ❑ The most disagreements occur in regions with low-to-mid RH values (20 – 60 % range) due to particle state differentiation.
- ❑ Metastable case produces in general more acidic particles than stable state (1-2 pH), with strong NH_3 buffering in Accumulation Mode.
- ❑ Magnified estimate of Direct TOA Radiative Forcing for aerosols (- 1.48 Wm^{-2}) but much more realistic for NO_3^- particles (- 0.14 Wm^{-2}).
- ❑ Computational speed-up in comparison to ISORROPIA II measured up to 5%.