

# **“Aerosol water parameterization: a single parameter framework” by S. Metzger et al.**

## **Reply to Anonymous Referee 2 by S. Metzger et al.**

We thank the anonymous referee for her/his time to review this work and the constructive comments. Complementing our reply to referee 1 with below answers, we hope to have satisfactorily addressed all points.

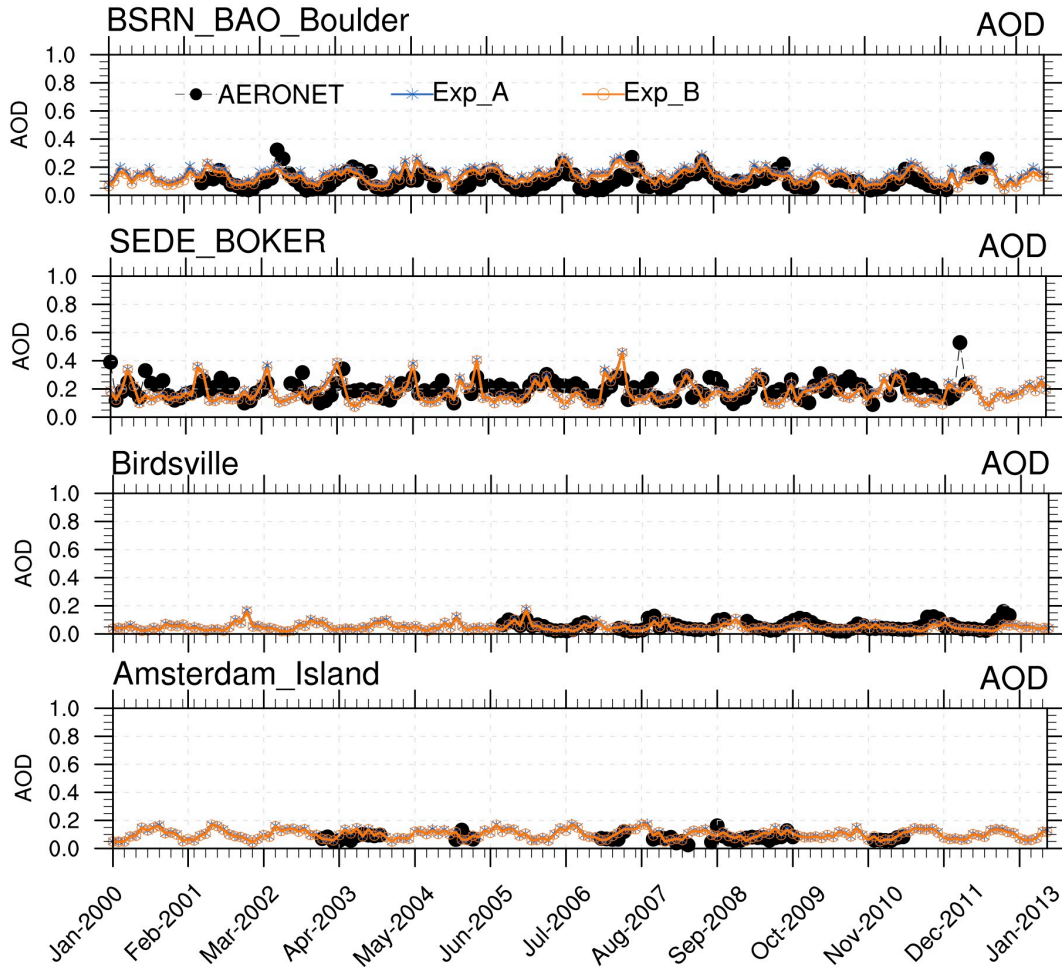
As stated in section 2.6 (Mixed solution RHD, p33507, line 12-15), we follow ISORROPIA II (Fountoukis and Nenes, 2007) to calculate the liquid-solid partitioning of multi-component aerosols using a mutual deliquescence RH range. To allow for an analytical solution, our calculation approach is different. The RH range depends on minimum and maximum thresholds (RHDMIN and RHDMAX, respectively) that can differ for certain aerosol compositions from ISORROPIA II. Both, the differences in the calculation and applications have been detailed in the manuscript (and complemented by the supplement).

We do not describe in this manuscript the case of efflorescence, since the manuscript only considers the so-called deliquescence branch of the aerosol hysteresis loop, where the relative humidity (RH) increases. Once the RH increases above certain thresholds, individual aerosol compounds deliquesce until the aerosol is fully deliquesced at a given RH. Depending on the multi-component aerosol composition, the thresholds lie within RHDMIN and RHDMAX, whereas for pure compounds the threshold is simply given by the RHD (with RHDMIN = RHDMAX = RHD); see Eqs. (13-21, pp33507 ff). The relative humidity of deliquescence (RHD) is prescribed (using values of ISORROPIA II).

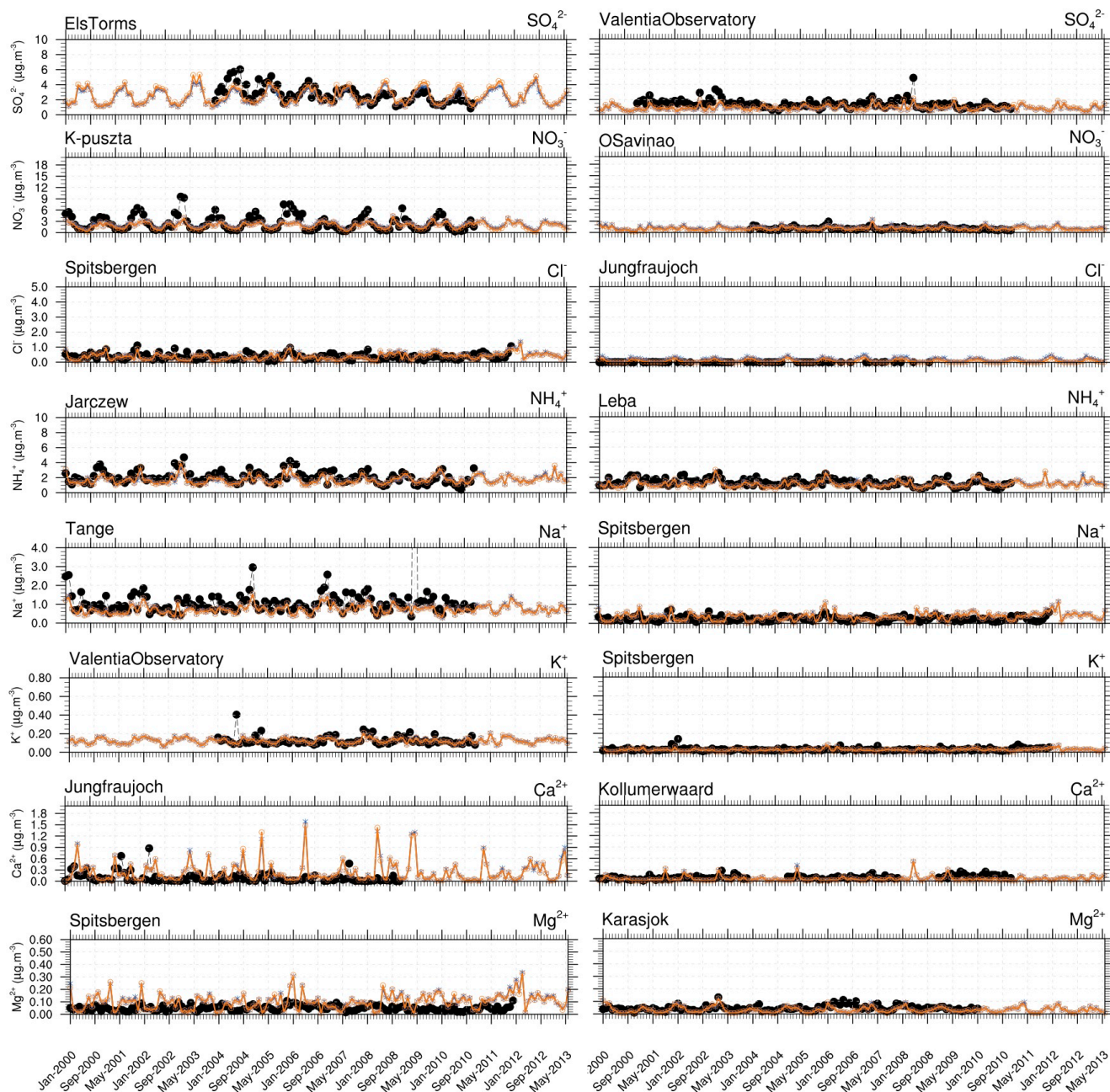
As also stated by referee 1, for the majority of chemical transport models, ISORROPIA II is executed in metastable mode in case of decreasing RH (efflorescence path). Then, a mutual deliquescence RH range (or RHD) is not required. Since this case is “trivial” compared to the deliquescence case, it is not considered here (“trivial” since mostly constant RH-thresholds such as 40% are used in 3-D applications due to the lack of aerosol composition dependent efflorescence measurements - especially in case of mixed solutions). But the vi-framework is valid for the entire range of water activity ( $a_w$ ), i.e., independent of deliquescence or efflorescence assumption (see Appendix A1 and Figs. A1-A2). It can be equally used for so-called metastable aerosols, for which the formation of solid salts is generally not considered (see our reply to referee 1 w.r.t. the addition to the MS).

Figure 4 and 6 (below) show a 3-D modeling example, which considers the aerosol hysteresis loop based on the full gas-liquid-solid partitioning (deliquescence path) and a 40% RH-threshold (efflorescence path, i.e., solids are formed instantaneously in case RH drops below). Both gas-

aerosol partitioning models (EQSAM4clim and ISORROPIA II) are embedded exactly in the same way in our EMAC chemistry-climate model and yield similar global applications results. The results represent a typical long-term (13 years) application (monthly means) and will be part of a follow-up study to the current manuscript. The results are shown here to demonstrate the applicability of EQSAM4clim for global applications if based on the aerosol hysteresis loop (deliquescence and efflorescence path). The differences compared to ISORROPIA II are negligible for these EMAC results and both simulations are close to the long-term observations.



**Figure 4.** AOD (monthly mean) at four AERONET stations that have long-term data available and which represent different aerosol types and climatic regions. AERONET observations (black line), EMAC - Exp A (ISORROPIA II, blue line), Exp B (EQSAM4clim, orange line); [http://aeronet.gsfc.nasa.gov/cgi-bin/type/piece\\_of\\_map\\_opera\\_v2\\_new](http://aeronet.gsfc.nasa.gov/cgi-bin/type/piece_of_map_opera_v2_new).



**Figure 6.** Lumped aerosol concentrations (liquids+solids) of the EMAC simulations (Fig. 4) and EMEP observations: sulfate ( $\text{SO}_4^{2-}$ ), nitrate ( $\text{NO}_3^-$ ), chloride ( $\text{Cl}^-$ ), ammonium ( $\text{NH}_4^+$ ), sodium ( $\text{Na}^+$ ), potassium ( $\text{K}^+$ ), calcium ( $\text{Ca}^{2+}$ ) and magnesium ( $\text{Mg}^{2+}$ ); each species is compared with two different station observations (left and right columns, black lines); EMAC simulations based on two gas-liquid-solid partitioning schemes: ISORROPIA II (blue line), EQSAM4clim (orange line).