Supplemental Information for "A framework for expanding aqueous chemistry in the Community Multiscale Air Quality (CMAQ) model version 5.1"

Table S1. Gas phase species[•] that participate in phase transfer and associated gas phase diffusion and accommodation coefficients. Unless otherwise indicated, Henry's Law coefficients are the base model's original values (from CMAQ subroutine hlconst.F).

Reaction	**H_{law} (M/atm)**	**$-\Delta H/R$ (K)**	**α[a]**	**D_g (m^2/s) x 10^5[b]**
SO_2 (gas) ↔ SO_2 (aq)	1.4	2.9E+03	1.10 x 10^{-1}	1.28
HNO_3 (gas) ↔ HNO_3 (aq)	2.1E+05	8.7E+03	8.68 x 10^{-2}	1.32
CO_2 (gas) ↔ H_2CO_3 (aq)	3.6E-02	2.2E+03	1.50 x 10^{-4}	1.55
NH_3 (gas) ↔ NH_4OH (aq)	6.1E+01	4.2E+03	9.10 x 10^{-2}	2.30
H_2O_2 (gas) ↔ H_2O_2 (aq)	8.3E+04	7.4E+03	1.53 x 10^{-1}	1.46
O_3 (gas) ↔ O_3 (aq)	1.14E-02	2.3E+03	1.00 x 10^{-1}	1.48
HCOOH (gas) ↔ HCOOH (aq)	8.9E+03	6.1E+03	2.29 x 10^{-2}	1.53
MHP (gas) ↔ MHP (aq)	3.1E+02	5.2E+03	6.76 x 10^{-3}	1.31
PAA (gas) ↔ PAA (aq)	8.4E+02	5.3E+03	1.90 x 10^{-2}	1.02
HCl (gas) ↔ HCl (aq)	1.9E+01	6.0E+02	1.16 x 10^{-1}	1.89
GLY (gas) ↔ GLY (aq)	3.6E+05		2.30 x 10^{-2}[c]	1.15[a]
MGLY (gas) ↔ MGLY (aq)	3.2E+04[e]		2.30 x 10^{-2}[c]	1.15[d]
IEPOX (gas) ↔ IEPOX (aq)	2.7E+06[f]		2.00 x 10^{-2}[f]	1.00[f]
MAE (gas) ↔ MAE (aq)	1.2E+05[f]		2.00 x 10^{-2}[f]	1.00[f]
HMML (gas) ↔ HMML (aq)	1.2E+05[f]		2.00 x 10^{-2}[f]	1.00[f]

[•] H_2SO_4 is instantaneously transferred to aerosol SO_4 and N_2O_5 to HNO_3 (gas) at the start of cloud processing. [a]CAPRAM 2.4 tables (http://projects.tropos.de/capram/capram_24.html)*. [b]Hermann et al. (2000)*. [c]Lim et al. (2005)*. [d]Assumed equal to D_g for glyoxal. [e]The Henry's Law coefficient for methylglyoxal is replaced with that of glyoxal when the CB05 gas phase mechanism is used (Carlton et al., 2010). [f]Pye et al. (2013). *and the original sources therein

Table S2. Dissociation reactions and equilibrium constants. Ionic species are considered explicitly and dissociation equilibrium reactions are described as a set of forward and backward reactions. Activity coefficients are rolled into the forward and backward reaction rates.

Reaction	**$K_{eq,298}$[a] [M]**	**$-\Delta E_a/R$ [K][#]**	**k_b[a,d] [M^{-1} s^{-1}]**
$SO_2 \leftrightarrow HSO_3^- + H^+$	1.39x10^{-2}	1.87x10^{3}	2.0x10^{8}
$HSO_3^- \leftrightarrow SO_3^{2-} + H^+$	6.72x10^{-8}	3.55x10^{2}	5.0x10^{10}
$HNO_3 \leftrightarrow NO_3^- + H^+$	1.7x10^{1}		5.0x10^{10}
$NH_4OH \leftrightarrow NH_4^+ + OH^-$	1.77x10^{-5}	-7.10x10^{2}	3.4x10^{10}
$H_2CO_3 \leftrightarrow HCO_3^- + H^+$	4.3x10^{-7}	-9.95x10^{2}	6.4x10^{4}
$HCO_3^- \leftrightarrow CO_3^{2-} + H^+$	4.68x10^{-11}	-1.79x10^{3}	5.0x10^{10}
$HCOOH \leftrightarrow HCOO^- + H^+$	1.8x10^{-4} [b]	-2.00x10^{1}	5.0x10^{10}
$HCl \leftrightarrow Cl^- + H^+$	1.74x10^{6} [b]	6.90x10^{3}	5.0x10^{10}
$H_2O \leftrightarrow OH^- + H^+$	1.8x10^{-16} [c]	-6.95x10^{3}	1.4x10^{11}
$H_2SO_4 \leftrightarrow HSO_4^- + H^+$	1.0x10^{3} [b]		5.0x10^{10}
$HSO_4^- \leftrightarrow SO_4^{2-} + H^+$	1.02x10^{-2}	2.45x10^{3}	1.0x10^{11}

[a]Warneck and Williams (2012)*. [b]Seinfeld and Pandis(2006)*. [c]Removed [H_2O] concentration of 55.5 M from the original equilibrium constant. [d]Graedel and Weschler (1981)*. *and the original sources therein

$k_f = Keq_{i,T} \times k_b$ [#]Note that while there are some minor differences between some of the equilibrium constants used here compared to AQCHEM, box model tests indicate that these small differences only have a minimal impact on predicted concentrations

Table S3. Chemical kinetic reactions.

Reaction	Rate coefficient in appropriate units of M,s (298K); $-E_a/R$ (K) [$]		Notes
$HSO_3^- + H_2O_2 + H^+ \rightarrow SO_4^{2-} + 2 H^+$	7.45E+7 / (1 + 13 [H+]); -4756.08		
$SO_2 + O_3 \rightarrow SO_4^{2-} + 2 H^+$	2.4E+4; 0.0		Q
$HSO_3^- + O_3 \rightarrow SO_4^{2-} + H^+$	3.7E+5; -5530.88		Q
$SO_3^{2-} + O_3 \rightarrow SO_4^{2-}$	1.5E+9; -5280.56		Q
$HSO_3^- + MHP + H^+ \rightarrow SO_4^{2-} + 2 H^+$	1.9E+7; -3799.5		
*$HSO_3^- + PAA + H^+ \rightarrow SO_4^{2-} + 2 H^+$	3.6E+7; -3999.2		*
*$HSO_3^- + PAA \rightarrow SO_4^{2-} + H^+$	7E+2; 0.0		
SO_2 (+ Mn(II)) $\rightarrow SO_4^{2-} + 2 H^+$	7.5E+2; 0.0		#
HSO_3^- (+ Mn(II)) $\rightarrow SO_4^{2-} + H^+$	7.5E+2; 0.0		#
SO_3^{2-} (+ Mn(II)) $\rightarrow SO_4^{2-}$	7.5E+2; 0.0		#
SO_2 (+ Fe(III)) $\rightarrow SO_4^{2-} + 2 H^+$	2.6E+3; 0.0		#
HSO_3^- (+ Fe(III)) $\rightarrow SO_4^{2-} + H^+$	2.6E+3; 0.0		#
SO_3^{2-} (+ Fe(III)) $\rightarrow SO_4^{2-}$	2.6E+3; 0.0		#
SO_2 (+Mn(II) + Fe(III)) $\rightarrow SO_4^{2-} + 2 H^+$	1E+10; 0.0		#
HSO_3^- (+ Mn(II) + Fe(III)) $\rightarrow SO_4^{2-} + H^+$	1E+10; 0.0		#
SO_3^{2-} (+ Mn(II) + Fe(III)) $\rightarrow SO_4^{2-}$	1E+10; 0.0		#
GLY + OH• → 0.04 ORGC	3E+10; 0.0		Fixed OH•
MGLY + OH• → 0.04 ORGC	3E+10; 0.0		Fixed OH•
	k_1 ($M^{-2}s^{-1}$)	k_2 ($M^{-2}s^{-1}$)	
IEPOX + H_2O → 2-methyltetrol	9E-04	1.3E-05	Rate = KIEPOX * Parent Hydrocarbon * Nucleophile * Q where KIEPOX = k_1*[H^+] + k_2*[$HSO4^-$] H_2O concentration remains fixed (55.5 M)
IEPOX + SO_4^{2-} → IEPOX_OS	2E-04	2.9E-06	
IEPOX + NO_3^- → IEPOX_ON[§]	2E-04	2.9E-06	
IEPOX + 2-methyltetrol → dimers	2E-04	2.9E-06	
IEPOX + IEPOX_OS → dimers	2E-04	2.9E-06	
IEPOX + IEPOX_ON → dimers[§]	2E-04	2.9E-06	
MAE + H_2O → 2-MG	9E-04	1.3E-05	
MAE + SO_4^{2-} → MPAN_OS	2E-04	2.9E-06	
MAE + NO_3^- → MPAN_ON[§]	2E-04	2.9E-06	
MAE + 2-MG → dimers[§]	2E-04	2.9E-06	
MAE + MPAN_OS→ dimers[§]	2E-04	2.9E-06	
MAE + MPAN_ON → dimers[§]	2E-04	2.9E-06	
HMML + H_2O → 2-MG	9E-04	1.3E-05	
HMML + SO_4^{2-} → MPAN_OS	2E-04	2.9E-06	
HMML + NO_3^- → MPAN_ON[§]	2E-04	2.9E-06	
HMML + 2-MG → dimers[§]	2E-04	2.9E-06	
HMML + MPAN_OS → dimers[§]	2E-04	2.9E-06	
HMML + MPAN_ON → dimers[§]	2E-04	2.9E-06	

[$] Rate coefficients are equal to those used in the standard version of AQCHEM. [Q] Aqueous diffusion correction factor applied; [*] included here as two reactions to accommodate the original single rate expression which included pH dependent and pH independent elements. [#] A S(VI) inhibition factor of the following form is applied to these reaction rates: $(1+75[S(VI)]^{0.67})^{-1}$. (Species in parentheses on the left hand side of the rate expression indicate species that affect the reaction rate calculation but whose concentrations are not consumed in the reaction.) [§] Included here but not included in the version of AQCHEM-KMTI released with CMAQv5.1

Table S4. CMAQ species and local AQCHEM-KMT(I) species. The “G”, “L”, “A”, and “WD” prefixes in column 1 refer to the gas-phase, aqueous-phase, Aitken aerosol, and wet deposition portion of the species respectively. Shaded cells include species specific to AQCHEM-KMTI. CMAQ aerosol concentration and wet deposition indices are of the form: [species, mode].

AQCHEM-KMT(I) local dynamic species	CMAQ species	Name and Notes
G_SO2, L_SO2, WD_SO2	GAS(LSO2), GASWDEP(LSO2)	Sulfur dioxide
G_HNO3, L_HNO3, WD_HNO3	GAS(LHNO3)/AEROSOL(LNO3, 2/3), GASWDEP(LHNO3)	Nitric acid, nitrate
G_CO2, L_H2CO3, WD_CO2	GAS(LCO2), GASWDEP(LCO2)	Carbon dioxide
G_NH3, L_NH4OH, WD_NH4OH	GAS(LNH3), AEROSOL(LNH4, 2/3), GASWDEP(LNH3)	Ammonia, ammonium
G_H2O2, L_H2O2, WD_H2O2	GAS(LH2O2), GASWDEP(LH2O2)	Hydrogen peroxide
G_O3, L_O3, WD_O3	GAS(LO3), GASWDEP(LO3)	Ozone
G_HCOOH, L_HCOOH, WD_HCOOH	GAS(LFOA), GASWDEP(LFOA)	Formic acid
G_MHP, L_MHP, WD_MHP	GAS(LMHP), GASWDEP(LMHP)	Methylhydroxyperoxide (CH3OOH)
G_PAA, L_PAA, WD_PAA	GAS(LPAA), GASWDEP(LPAA)	peroxyacetic acid (CH3C(O)OOH)
G_HCL, L_HCL, WD_HCL	GAS(HCL), GASWDEP(HCL)	Hydrochloric acid
G_GLY, L_GLY, WD_GLY	GAS(LGLY), GASWDEP(LGLY)	Glyoxal
G_MGLY, L_MGLY, WD_MGLY	GAS(LMGLY), GASWDEP(LMGLY)	Methylglyoxal
A_NO3AKN	AEROSOL(LNO3,1)	Aitken nitrate
A_NH4AKN	AEROSOL(LNH4,1)	Aitken ammonium
A_CLAKN	AEROSOL(LCL,1)	Aitken chloride
A_NAAKN	AEROSOL(LNA,1)	Aitken sodium
A_SO4AKN	AEROSOL(LSO4,1)	Aitken sulfate
A_PECAKN	AEROSOL(LEC,1)	Aitken elemental carbon
A_POAAKN	AEROSOL(LPOA,1)	Aitken primary OA
A_PRIAKN	AEROSOL(LPRI,1)	Aitken other primary
L_SO4MIN2, WD_H2SO4	AEROSOL(LSO4, 2/3), AERWDEP(LSO4, 2/3)	Sulfate
L_NO3MIN, WD_NO3MIN	GAS(LHNO3), AEROSOL(LNO3, 2/3), AERWDEP(LNO3, 2/3)	Nitrate
L_NH4PLUS, WD_NH4PLUS	GAS(LNH3), AEROSOL(LNH4, 2 and 3), AERWDEP(LNH4, 2/3)	Ammonium
L_CLMIN, WD_CLMIN	AEROSOL(LCL, 2/3), AERWDEP(LCL, 2/3)	Chloride
L_PRIACC, WD_PRIACC	AEROSOL(LPRI, 2), AERWDEP(LPRI, 2)	Other primary
L_PECACC, WD_PECACC	AEROSOL(LEC, 2), AERWDEP(LEC, 2)	Elemental carbon
L_ORGC, WD_ORGC	AEROSOL(LORGC, 2), AERWDEP(LORGC, 2)	Cloud SOA
L_POAACC, WD_POAACC	AEROSOL(LPOA, 2), AERWDEP(LPOA, 2)	Primary OA
L_HPLUS, WD_HPLUS	HPWDEP	H^+
L_OHMIN	--	OH^-

L_FEPLUS3, WD_FEPLUS3[%]	AEROSOL(LFEACC, 2), AERWDEP(LFEACC, 2), AEROSOL(LSOILC, 3), AERWDEP(LSOILC, 3), AEROSOL(LANTHC, 3), AERWDEP(LANTHC, 3)	Soluble iron($^{3+}$); [%]AQCHEM species include contributions from coarse aerosol
L_MNPLUS2, WD_MNPLUS2[%]	AEROSOL(LMNACC, 2), AERWDEP(LMNACC, 2) , AEROSOL(LSOILC, 3), AERWDEP(LSOILC, 3), AEROSOL(LANTHC, 3), AERWDEP(LANTHC, 3)	Soluble manganese($^{2+}$); [%]AQCHEM species include contributions from coarse aerosol
L_HSO3MIN, (WD_SO2)	GAS(LSO2)	Bisulfite
L_SO3MIN2, (WD_SO2)	GAS(LSO2)	Sulfite
L_HCO3MIN, (WD_CO2)	GAS(LCO2)	Bicarbonate
L_CO3MIN2, (WD_CO2)	GAS(LCO2)	Carbonate
L_HCOOMIN, (WD_HCOOH)	GAS(LFOA)	Formate
L_HSO4MIN, (WD_H2SO4)	AEROSOL(LSO4, 2/3)	Bisulfate
L_CAPLUS2, WD_CAPLUS2[%]	AEROSOL(LCAACC, 2), AERWDEP(LCAACC, 2), AEROSOL(LSOILC, 3), AERWDEP(LSOILC, 3), AEROSOL(LANTHC, 3), AERWDEP(LANTHC, 3), AEROSOL(LSEASC, 3), AERWDEP(LSEASC, 3)	Calcium($^{2+}$); [%]AQCHEM species include contributions from coarse aerosol
L_MGPLUS2, WD_MGPLUS2[%]	AEROSOL(LMGACC, 2), AERWDEP(LMGACC, 2), AEROSOL(LSOILC, 3), AERWDEP(LSOILC, 3), AEROSOL(LANTHC, 3), AERWDEP(LANTHC, 3), AEROSOL(LSEASC, 3), AERWDEP(LSEASC, 3)	Magnesium($^{2+}$); [%]AQCHEM species include contributions from coarse aerosol
L_KPLUS, WD_KPLUS[%]	AEROSOL(LKACC, 2), AERWDEP(LKACC, 2), AEROSOL(LSOILC, 3), AERWDEP(LSOILC, 3), AEROSOL(LANTHC, 3), AERWDEP(LANTHC, 3), AEROSOL(LSEASC, 3), AERWDEP(LSEASC, 3)	Potassium($^{+}$); [%]AQCHEM species include contributions from coarse aerosol
L_NAPLUS, WD_NAPLUS[%]	AEROSOL(LNA, 2), AERWDEP(LNA, 2), AEROSOL(LSOILC, 3), AERWDEP(LSOILC, 3), AEROSOL(LANTHC, 3), AERWDEP(LANTHC, 3), AEROSOL(LSEASC, 3), AERWDEP(LSEASC, 3)	Sodium($^{+}$); [%]AQCHEM species include contributions from coarse aerosol
G_IEPOX, L_IEPOX, WD_IEPOX	GAS(LIEPOX), GASWDEP(LIEPOX)	Isoprene epoxydiols

G_IMAE, L_IMAE, WD_IMAE	GAS(LIMAE), GASWDEP(LIMAE)	Methacrylic acid epoxide
G_IHMML, L_IHMML, WD_IHMML	GAS(LIHMML), GASWDEP(LIHMML)	Hydroxymethylmethyl-□-lactone
L_IETET, WD_IETET	AEROSOL(LIETET, 2), AERWDEP(LIETET, 2)	2-methyltetrols
L_IEOS, WD_IEOS	AEROSOL(LIEOS, 2), AERWDEP(LIEOS, 2)	IEPOX-derived organosulfate
L_IEON, WD_IEON**	AEROSOL(LIEON, 2), AERWDEP(LIEON, 2)	IEPOX-derived organonitrate. **not included in CMAQv5.1 but used here
L_DIMER, WD_DIMER	AEROSOL(LDIMER, 2), AERWDEP(LDIMER, 2)	dimers
L_IMGA, WD_IMGA	AEROSOL(LIMGA, 2), AERWDEP(LIMGA, 2)	2-methylglyceric acid
L_IMOS, WD_IMOS	AEROSOL(LIMOS, 2), AERWDEP(LIMOS, 2)	MPAN-derived organosulfate
L_IMON, WD_IMON**	AEROSOL(LIMON, 2), AERWDEP(LIMON, 2)	MPAN-derived organonitrate. **not included in CMAQv5.1 but used here
G_OH, L_OH*	GAS(LHO)	Hydroxyl radical. *Mapped in from CMAQ but kept constant during/after cloud processing

Table S5. Box model test scenarios. Initial aerosol species concentrations are held constant for the variable scenarios and are associated with an initial droplet pH of ~5. Default gas concentrations were set at 1 ppb and modal aerosol species concentrations were set to 0.1 $\mu g/m^3$, with the following exceptions: O_3 = 50 ppb, CO_2 = 340 ppm, H_2SO_4 = 0.1 ppb, N_2O_5 = 0.1 ppb, SO_4^{2-} = 4.2 $\mu g/m^3$, Primary inorganic (inert) = 2.1 $\mu g/m^3$, Primary OA = 1.1 $\mu g/m^3$, SOA = 2.1 $\mu g/m^3$, EC = 1.1 $\mu g/m^3$, Cl^- = 2.3 $\mu g/m^3$, NH_4^+ = 2.2 $\mu g/m^3$, NO_3^- = 4.1 $\mu g/m^3$, Na, accumulation mode Fe, Mn, Ca = 0.3 $\mu g/m^3$, coarse soil and anthropogenic coarse = 2 $\mu g/m^3$, coarse sea salt cations = 1 $\mu g/m^3$, Pressure = 1 atm.

Variable input	Values
Liquid Water content (g/m^3)	0.05, 0.1, 0.2, 0.3
Precipitation rate (mm/hr)	0, 2
Temperature (K)	270, 283
Daylight (binary)	0, 1
Integration time (s)	60, 300, 720
SO_2 (ppb)	1, 5, 10
H_2O_2 (ppb)	0.1, 1, 5
(M)GLY (ppb)	0.5, 2
OH (ppb)	10^{-3}, 10^{-5}, 10^{-7}
NH_3 (ppb)	1, 5
HNO_3 (ppb)	1, 5

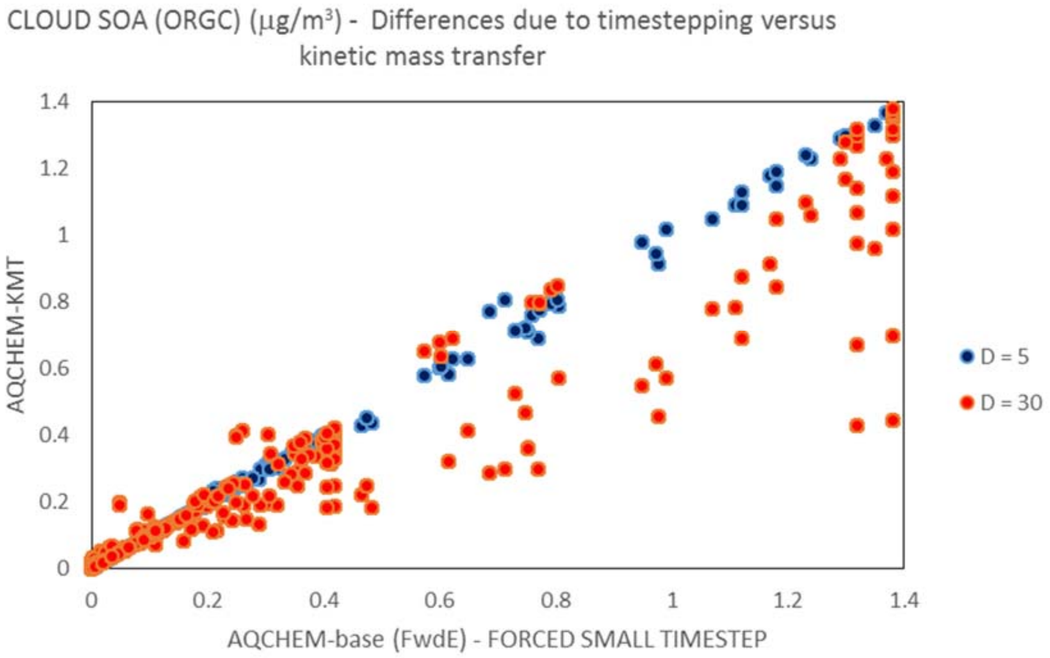

Figure S1: Cloud SOA (ORGC) predicted by AQCHEM-KMT at 5 (blue) and 30 μm (red) droplet diameters versus standard AQCHEM with a forced small time step. Compared to Figure 2 where AQCHEM is run with its standard time-stepping procedure, it appears that the differences between the models for ORGC are driven by both mass transfer limitations as well as the time-stepping procedure (which in standard AQCHEM is based on sulfate production only).

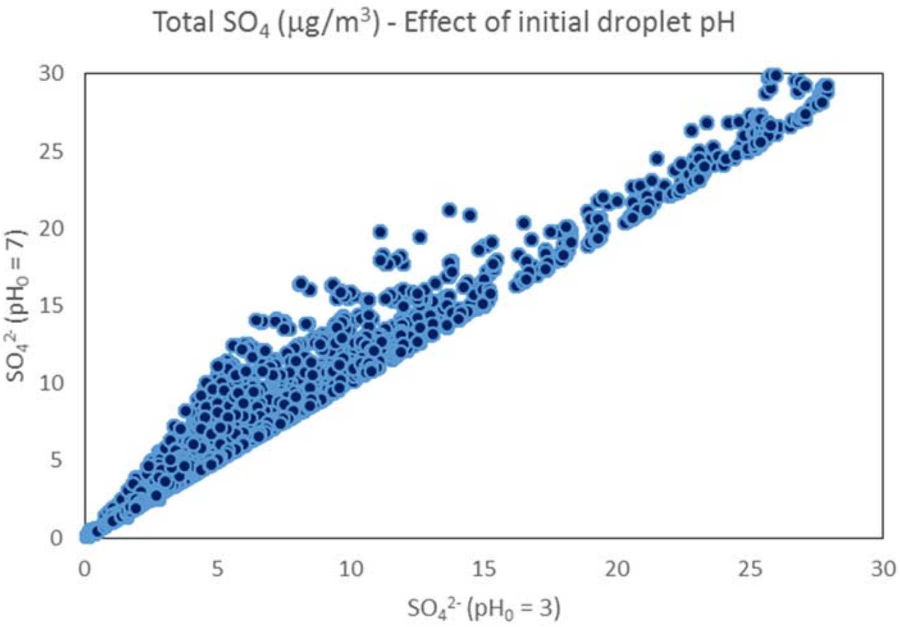

Figure S2: Predicted SO_4^{2-} concentrations for AQCHEM-KMT for initial pH of 7 versus 3.