## **Supplementary Materials**

## An evaluation of global organic aerosol schemes using airborne observations

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Campaign	Organic Aerosol	NO <sub>x</sub>	Isoprene	со	
ARCPAC	<b>C-ToF-AMS</b> (A.M. Middlebrook)	<b>NOAA NO<sub>y</sub>O</b> ₃ (T.B. Ryerson)	<b>PTR-MS</b> (J.A. de Gouw, C. Warneke)	VUV Resonance Fluorescence (J.S. Holloway)	
ARCTAS	<b>HR-ToF-AMS</b> (J.L. Jimenez)	NCAR 4 channel Chemiluminescence (A.J. Weinheimer, F.M. Flocke, D.J. Knapp, D.D. Montzka, I.B. Pollack)	<b>TOGA</b> (E. Apel, R. Hornbrook)	<b>DACOM</b> (G.S. Diskin, G. Sachse)	
EUCAARI	<b>C-ToF-AMS</b> (H. Coe)				
OP3	<b>C-ToF-AMS</b> (H. Coe)				
CalNex	<b>C-ToF-AMS</b> (A.M. Middlebrook)	NOAA NO <sub>y</sub> O₃ (T.B. Ryerson, I.B. Pollack)	<b>PTR-MS</b> (J.A. de Gouw, C. Warneke)	VUV Resonance Fluorescence (J.S. Holloway)	
DC3	HR-ToF-AMS (J.L. Jimenez)	NOAA NO <sub>y</sub> O₃ (T.B. Ryerson, I.B. Pollack)	<b>PTR-MS</b> (T. Mikoviny, A. Wisthaler)	<b>DACOM</b> (G.S. Diskin, G Sachse)	
SENEX	<b>C-ToF-AMS</b> (A.M. Middlebrook)	<b>NOAA NO<sub>y</sub>O</b> 3 (T.B. Ryerson, I.B. Pollack)	<b>PTR-MS</b> (M. Graus)	VUV Resonance Fluorescence (J.S. Holloway)	
SEAC4RS	HR-ToF-AMS (J.L. Jimenez)	<b>NOAA NO<sub>y</sub>O</b> ₃ (T.B. Ryerson, I.B. Pollack, J. Peischl)	WAS (D.R. Blake)	<b>DACOM</b> (G.S. Diskin, G. Sachse)	
GoAmazon	HR-ToF-AMS (J.E. Shilling)		<b>PTR-MS</b> (J.E. Shilling)	Los Gatos ICOS Analyzer (S.R. Springston)	
FRAPPE	<b>C-ToF-mAMS</b> (R. Bahreini)	NCAR 2-channel Chemiluminescence (A.J. Weinheimer, D.D. Montzka)	<b>TOGA</b> (E. Apel, R. Hornbrook)	Aero-Laser VUV Fluorescence (T.L. Campos and F.M. Flocke)	
KORUS-AQ	<b>HR-AMS</b> (J.L. Jimenez, P. Campuzano-Jost)	NCAR 4-channel Chemiluminescence (A.J. Weinheimer, D.D. Montzka)	<b>PTR-MS</b> (P. Eichler, L. Kaser, T. Mikoviny, M. Müller, A. Wisthaler)	DACOM (G.S. Diskin, S.E. Pusede)	
ATom	HR-ToF-AMS (J.L. Jimenez)	<b>NOAA NO<sub>y</sub>O₃</b> (T.B. Ryerson, J. Peischl, C. Thompson)	<b>TOGA</b> (E. Apel, R. Hornbrook)	<b>QCLS</b> (B.C. Daube, S.C. Wofsy, R. Commane, E. Kort)	

**Table S1**. An overview of the instrumentation and associated primary investigators for the organic aerosol and trace gas observations used in this analysis.

Nitrogen oxides were measured using photolysis rates and NO/O<sub>3</sub> chemiluminescence techniques (Ryerson et al., 2000), carbon monoxide levels were measured using a Differential Absorption Carbon monOxide Measurement (DACOM) instrument (Sachse et al., 1987) or a VUV resonance fluorescence approach (Gerbig et al., 1999), isoprene concentrations were observed using a Proton Transfer Reaction Mass Spectrometer (de Gouw and Warneke, 2007), a Trace Organic Gas Analyzer (Apel et al., 2010) or a whole air sampling approach (Colman et al., 2001) and sulfate aerosol loadings were measured using an AMS.

Regime	Description	Percentage of Dataset	Mean OA	Median OA	Std. Dev. OA	Mean Isoprene	Mean NO <sub>x</sub>	Mean CO
А	Dominant anthropogenic Influence	38.8%	2.0	0.6	3.2	0.05	0.97	145
F	Dominant pyrogenic Influence	7.3%	4.5	2.7	5.3	0.14	0.17	151
В	Dominant biogenic Influence	4.0%	3.2	2.8	2.8	1.48	0.17	123
AF	Anthropogenic and Pyrogenic Influence	6.8%	3.8	1.6	5.0	0.05	0.81	160
AB	Anthropogenic and Biogenic Influence	14.1%	4.2	2.8	4.1	0.61	0.36	115
AFB	No dominant influence from any one source category.	10.2%	3.2	2.5	2.9	0.10	0.38	115
R	Remote / Clean (concentrations under $0.2 \ \mu g \ / \ sm^3$ )	18.9%	0.1	0.1	0.3	0.05	0.08	71
Aggregate			2.4	0.7	3.7	0.26	0.56	126

**Table S2.** An overview of the different regimes. Statistics (mean, median, standard deviation) are listed for the observational data categorized into the individual regimes. OA data is in units of  $\mu g \text{ sm}^{-3}$ . Mean observations for isoprene, nitrogen oxides and carbon monoxide are in units of parts per billion (ppb).



**Figure S1.** Superimposed distributions from the simple (blue) and complex (red) schemes with the observations in black for the different campaigns. Vertical lines represent median values for the different distributions.



**Figure S2**. Mean vertical profiles (in km) for the observed and simulated OA and sulfate across the different regimes. The profiles are binned at 200m intervals. Observations are in black. For the OA, the complex scheme is in red while the simple scheme is in blue. Model sulfate is in green.



**Figure S3**. A comparison of the simulated OA loadings averaged by grid-box over the vertical dimension. Panel (d) provides an overview of the column-averaged 'best fit' scheme based on the ability to minimize the mean bias.



**Figure S4.** Distribution in the ratio and bias between the observed and modelled organic aerosol concentrations for each model scheme across the 17 campaigns. The boxes denote the  $25^{\text{th}}$  and  $75^{\text{th}}$  percentile of the distribution, while the whiskers denote the  $5^{\text{th}}$  and  $95^{\text{th}}$  percentile. The ratio plots have been overlaid with violin plots describing the entire distribution.



Figure S5. Comparison of complex (red), simple (blue) and observed (grey) organic aerosol to carbon monoxide.



**Figure S6**. A comparison of model-observation OA bias and binned observations for a) relative humidity, b) Temperature, c) Sulfate, d) Isoprene, e) CO and f)  $NO_x$  for the complex (red) and simple (blue) schemes across the aggregate dataset. The best fit line is shown in black.



**Figure S7.** A statistical evaluation of the OA model skill for the complex and simple schemes against a modified treatment that interchanges the POA and SOA from both schemes.

## References

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