



# Supplement of

# New methodology shows short atmospheric lifetimes of oxidized sulfur and nitrogen due to dry deposition

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### **17** The Supporting Information contains the following sections:

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25 Section 1. Supporting Tables S1 to S3.

Measurement	Instrument	Sampling time resolution (s)	Detection limit	Manufacturer
SO <sub>2</sub>	Thermo 43iTLE	1	0.7 ppbv	Thermo Fisher Scientific, Franklin, MA, USA
NOy	Thermo 42iTL	1	0.09 ppbv	Thermo Fisher Scientific, Franklin, MA, USA
pSO <sub>4</sub>	Aerosol Mass Spectrometer	10	0.048 ug m <sup>-3</sup>	Aerodyne Research Inc.

## 26 Table S1. Instrumentation and measurement details for SO<sub>2</sub>, NO<sub>y</sub> and pSO<sub>4</sub>.

**Table S2**. Measurement and model-derived estimates of cumulative deposition (%), transport

29 distance (km) and lifetimes (hrs) of TOS and TON for F7, F19 and F20. Geographic foot print

30 areas under the plumes for TOS and TON are also provided.

	Cumulative deposition (%)		e-folding transport distance (d <sub>1/e</sub> ) (km)			Lifetime ( $\tau = d_{1/e}/u$ ) (hrs)						
TOS	7	19	20		7	19	20		7	19	20	
Measurements	22±4	74±5	45±3		1230±290	71±1	210±	4	26	2.2	6.5	
Model	7	21	8		4300	500	2800		large	16	91	
Footprint (km <sup>2</sup> )	3500	5700	4200									
TON	7	19	20 (SP)	20 (NP)	7	19	20 (SP)	20 (NP)	7	19	20	
Measurements	31±11	49±11	62±14	34±6	360±14	190±7	62±1	290±30	7.6	5.6	1.9	9.0
Model	3	19	4	2	4300	650	2000	2400	91	23	63	78
Footprint (km <sup>2</sup> )	3500	5700	4200	3100								

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- **Table S3**. Equivalent dry deposition velocities  $V_d$  (cm s<sup>-1</sup>) determined from the aircraft
- 34 measurements (AC) and the model.  $SO_2$  and TON mixing ratios were taken from the average of
- the lowest ~40m (interpolated values) across the plume width for two sets of screens. SP=south
- 36 plume, NP=north plume for F20.

Flight	SO <sub>2</sub>		TON		
	AC	model	AC	model	
7	0.9±0.6	0.76	2.3±0.7	1.32	
	1.5±0.3	0.67	3.2±1.0	1.55	
mean	1.2±0.5	0.72	2.8±0.8	1.44	
19	2.3±0.5	0.52	1.9±0.6	1.10	
	2.8±0.5	0.67	1.3±0.4	1.35	
	2.3±0.4	0.70	1.5±0.5	1.40	
mean	2.4±0.4	0.63	1.6±0.5	1.28	
20	3.5±0.6	0.63	6.7±2.0	1.06	
			(SP)	(SP)	
			4.2±1.3	0.94	
			(NP)	(NP)	
	3.2±0.5	0.52	2.8±0.8	0.77	
			(SP)	(SP)	
			0.18±0.05	0.85	
			(NP)	(NP)	
mean	3.4±0.6	0.58	4.7±0.1.4	0.92	
			( <b>SP</b> )	( <b>SP</b> )	
			2.2±0.7	0.90	
			( <b>NP</b> )	( <b>NP</b> )	

- **Table S4**. Prescribed values used in the Monte-Carlo simulations with five different deposition
- 39 algorithms.

Component	Input Range	Units	Algorithm	Reference
Friction velocity (U*)	0.2 to 0.6	unitless	All 5 algorithms	Oski-ôtin ground site observations
Obukhov Length (L)	-200 to -350	m	All 5 algorithms	Oski-ôtin ground site observations
Reference Height (Z)	40 to 45	m	All 5 algorithms	Estimate of AOSR
Roughness Length (z0)	0.6 to 1	unitless	All 5 algorithms	Grassi et al., 2013
Schmidt Number (S <sub>c</sub> )	0.8 to 2	unitless	All 5 algorithms	Oski-ôtin ground site observations
Leaf Area Index (LAI)	2 to 5	unitless	All 5 algorithms	Makar et al., 2018; Brook et al., 1999
Minimum Leaf stomatal resistance for H <sub>2</sub> O (rs <sub>min</sub> )	100 to 250	s/m	ZHANG, C5DRY, WESLEY, GEM_MACH	Zhang et al., 2003
Canopy Height (h <sub>c</sub> )	6 to 18	m	C5DRY	Estimate of AOSR
Ground resistance (R <sub>g</sub> )	100 to 250	s/m	All 5 algorithms	Wesley et al., 1989
Solar Radiation (SolarRG)	450	W/m <sup>2</sup>	ZHANG, C5DRY, WESLEY, GEM- MACH	Oski Otin ground site observations
Mesophyll resistance (R <sub>m</sub> )	0.03 to 0.05	s/m	ZHANG, C5DRY, NOAH-GEM	Makar et al., 2018
In canopy aerodynamic	20 to 60	s/m	ZHANG, WESLEY, GEM-	Zhang et al., 2003

resistance			MACH, NOAH-	
reference (R <sub>ac0</sub> )			GEM	
Cuticle resistance (R <sub>cut</sub> )	500 to 1000	s/m	C5DRY	Based on calculations of R <sub>cut</sub> from the other deposition algorithms
Dry cuticle reference (R <sub>cut,d0</sub> )	2000	s/m	ZHANG, WESLEY, NOAH- GEM	Zhang et al., 2002
Dry cuticle reference (R <sub>cuti</sub> )	1000	s/m	GEM-MACH	Makar et al., 2018
Surface Temperature (T <sub>s</sub> )	20 to 25	°C	ZHANG, C5DRY, WESLEY, GEM- MACH	Aircraft observations
Relative Humidity (RH)	55 to 70	%	ZHANG, C5DRY, GEM-MACH, NOAH-GEM	Aircraft observations
Solar Zenith Angle (Theta)	65 to 75	unitless	ZHANG, C5DRY	https://www.esrl.noaa.gov/gmd/ grad/antuv/SolarCalc.jsp
Slope gas exchange data (m)	9 to 10	unitless	NOAH-GEM	Zhang et al., 2002
Intercept gas exchange data (b)	0.01 to 0.04	unitless	NOAH-GEM	Zhang et al., 2002
Net CO <sub>2</sub> assimilation rate (A <sub>n</sub> )	1e <sup>-6</sup> to 4e <sup>-6</sup>	mol C/m²/s	NOAH-GEM	Baldocchi et al., 1997
RH fraction at the leaf surface (h <sub>s</sub> )	0.5 to 1	unitless	NOAH-GEM	Estimated range

Atmospheric	101300	Pa	NOAH-GEM	Aircraft observations
pressure (P)				
CO <sub>2</sub> partial	23 to 37	Pa	NOAH-GEM	Niyogi et al., 2009
pressure at the				
leaf surface ( $C_s$ )				
Ambient T at	20 to 25	°C	ZHANG, C5DRY,	Aircraft observations
height Z (T <sub>a</sub> )			GEM-MACH	
T <sub>min</sub>	-5 to 0	°C	ZHANG, C5DRY,	Makar et al., 2018
			GEM-MACH	
T <sub>max</sub>	40 to 45	°C	ZHANG, C5DRY,	Makar et al., 2018
			GEM-MACH	
T <sub>opt</sub>	15 to 30	°C	ZHANG, C5DRY,	Makar et al., 2018
			GEM-MACH	
Molecular	0.1085	cm <sup>2</sup> /s	All 5 algorithms	Massman et al., 1998
diffusivity of				
$SO_2(D_c)$				
Molecular	0.2178	cm <sup>2</sup> /s	All 5 algorithms	Massman et al., 1998
diffusivity of				
water (D <sub>H2O</sub> )				
	1	1		



#### 42 Section S2. Supporting Figures S1 to S5.



Figure S1. AMS total mass ( $\Sigma$ (p-Organics, pSO<sub>4</sub>, pNO<sub>3</sub>, pNH<sub>4</sub>)) (gray points) compared with mass 44 45 estimated from the UHSAS (black points) and the AMS CE-corrected mass (red points). The 46 particle collection efficiency (CE) of the AMS was investigated by comparing the total AMS-derived 47 mass with the mass estimated from the size distribution measurements of the UHSAS. Number 48 concentrations measured by the UHSAS over a size range of 60 nm to  $1\mu$ m (matching that of the AMS) 49 were converted to volume concentrations using mid-point bin diameters and assuming spherical shapes. Volume concentrations were then converted to mass concentrations using densities weighted by the AMS 50 51 components. A CE of 0.5 was determined for both F7 and F20, and for F19 it was 1.0. Detailed 52 investigations and discussions on the CE of the AMS can be found in the literature (e.g. Middlebrook et al., 2012; Dunlea et al., 2009; Kleinman et al., 2008; Quinn et al, 2006). 53 54



Figure S2. Emissions-normalized deposition fluxes of (a) TOS and (b) TON derived from the aircraft-based measurements (solid symbols and lines) and the GEM-MACH model (open





61 Figure S3. Probability distributions of a) Ra, b) Rb, and c) Rc (s/m) for SO2 derived from Monte Carlo simulations using 5 different deposition algorithms. 62





Figure S4  $R_{cut}$ ,  $R_c$ , (s/m) and  $V_d$  (cm/s) for SO<sub>2</sub> as a function of pH as derived from Monte Carlo simulations with the GEM-MACH deposition algorithm. 65



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**Figure S5**. A strong diurnal cycle was seen in the  $V_d$  for SO<sub>2</sub> as determined from the vertical

- 68 gradient methodology at the Oski-ôtin site in the AOSR, with a full stability correction (S24).
- 70 the diurnal cycle of eddy diffusivity observed at this site.

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- 73 Section S3. Ground-based SO<sub>2</sub> fluxes. SO<sub>2</sub> fluxes were estimated using an eddy
- recovariance/vertical gradient method with data collected on a 32m tower at the Oski-ôtin air
- 75 quality station (57.1837 ° N, 111.6395 ° W) in Fort McKay, centrally located in the AOSR. The
- observation method was similar to that reported previously (Wu et al., 2018); ultrasonic
- anemometers (model CSAT-3, Campbell Scientific, USA) were collocated with inlets of <sup>1</sup>/<sub>2</sub>"
- 78 Teflon tubing at 32m, 18m and 8m above ground, drawing sampled air to gas analyzers at the
- <sup>79</sup> base of the tower (Thermo Environmental 43i TCL). Data presented here were collected
- 80 between June 6-8, 2018. Eddy diffusivities were calculated from the difference in wind speed at
- 81 32m and 8m combined with the momentum flux determined through eddy covariance at 18m,
- 82 and stability-corrected following Högström et al. 1996. The determined dry deposition velocities
- for  $SO_2$  are shown in Figure S4. Only daytime data (between 19 and 1 UTC) unaffected by
- 84 structural disturbances (e.g. flow through the tower) were included in the comparison with the
- aircraft results aligning with the typical flight times. Resulting deposition velocities for  $SO_2$  had
- 86 a median of 4.1 cm s<sup>-1</sup> and a trimmed mean of 4.9 cm s<sup>-1</sup> (standard error 1.2 cm s<sup>-1</sup>).

#### 87 Section S4. SO<sub>2</sub> chemical losses

- 88 The most significant oxidant that reacts in the gas phase with  $SO_2$  is the hydroxyl radical, OH, to
- Previous aircraft studies have shown that, in the absence of clouds, SO<sub>2</sub>
- 90 oxidation by OH is the main pathway for SO<sub>2</sub> loss in industrial plumes in summertime (Brock et
- al., 2002; Miyakawa et al., 2007). The transformation flights were all conducted during midday
- 92 under clear sky conditions, hence the contribution of cloud aqueous chemistry towards  $pSO_4$
- 93 production during the study flights is minimal. The potential loss of  $SO_2$  to reactions with
- alkenes to form organosulfates (Shang et al., 2016) and with criegee biradicals to form  $H_2SO_4$
- 95 (Boy et al., 2013; Mauldin et al, 2012; Huang et al., 2015) would not be accounted for in the 96 mass balance of the S mass in SO<sub>2</sub> presented above but would be <1% of the SO<sub>2</sub> conversion.
- Regardless, since sulfates are detected as pSO<sub>4</sub> by the AMS (Farmer et al., 2010), any SO<sub>2</sub>
- 98 chemical loss other than by the reaction with OH would still be captured in the mass balance of
- 99 TOS.
- 100

101 OH concentrations were estimated using ratios of selected volatile organic compounds (VOCs) that react almost exclusively with OH (during the daytime) and a methodology as described 102 103 previously (Kleinman et al., 2003). Lagrangian transport times were determined from the aircraft-based wind speed measurements and the transit time of air between successive screens 104 (Liggio et al., 2016). It is possible that there will be cross plume gradients in  $SO_2$  and VOC 105 concentrations given their different sources from each facility. However, the VOC canisters 106 were not instantaneous, but were  $\sim 30$  s long, representing a spatial grab of  $\sim 2-3$  km at the speed 107 of the aircraft. These VOC's represent the average VOC concentration from numerous sources 108 109 on site, and their spatial footprint overlaps significantly with the SO<sub>2</sub> source footprint in these facilities. The uncertainties ranged from 17 to 58%, which attempts to account for uncertainties 110 associated with the selection of the reference hydrocarbon concentrations, the slope 111 determination, transport times, and reaction rate constants. OH concentrations derived using the 112 ratio of toluene to benzene and plume box modeling for F19 (Liggio et al., 2016) were consistent 113 within the uncertainties. 114

#### 115 Section S5. Modelled dry deposition fluxes and dry deposition velocities

Dry deposition fluxes estimated are compared with those predicted from an air quality model, 116 117 Global Environmental Multiscale - Modelling Air-quality and CHemistry (GEM-MACH). GEM-MACH is a comprehensive on-line chemical reaction transport model (Moran et al., 2010) 118 that has recently been used to estimate acidic deposition downwind of the AOSR (Makar et al., 119 2018) using a 2.5 km grid cell resolution. A detailed description of GEM-MACH appears 120 elsewhere (Makar et al., 2018; Akingunola et al., 2018; Gordon et al., 2018). The model 121 includes parameterizations for gas-phase chemistry, aqueous chemistry and cloud processing of 122 gases and aerosols, inorganic heterogeneous chemistry, secondary organic aerosol formation, and 123 aerosol microphysics. The model version used here employs a 12-bin sectional approach to 124 resolve particle size distribution, and eight aerosol species (sulfate, nitrate, ammonium, 125 secondary organic aerosol, primary organic aerosol, black carbon, sea-salt, and crustal material), 126 and incorporates aerosol direct and indirect feedbacks with the meteorological code's radiative 127 transfer (Makar et al., 2015a; Makar et al., 2015b). Gas phase deposition of N and S compounds 128 is determined through a commonly used resistance methodology with deposition velocities 129 130 calculated using inferential methods (Makar et al., 2018). The deposition fluxes are incorporated into the vertical diffusion operator as a flux boundary condition. Further details on the 131

132 formulation of GEM-MACH are provided elsewhere (Makar et al., 2018 and references therein).

133 The model plume boundaries were determined separately for  $NO_x$  and  $SO_2$  plumes, using the

assumption that the plume edge corresponds to background concentrations, as was the case for

the observed plumes. Model and observed screens did not necessarily spatially coincide due to

differences between the modelled and observed wind fields (Tables 1, 2). However, the same

strategy was used to set up downwind model screen locations as in the observations (specifically,determining the plume center at one hour's advection time downwind from the sources, placing

determining the plume center at one hour's advection time downwind from the sources, placingthe first screen perpendicular to this direction and centred on the plume centreline, calculating a

one-hour forward trajectory for the second screen and repeating the process for the second and

subsequent screens). The intersection of the screen lines with the 0.1 maximum concentration

142 contours for  $SO_2$  and  $NO_x$  respectively, determined the boundaries of the screens for the  $SO_2$  and

143 NO<sub>x</sub> plumes. Boundaries were also adjusted to correspond with the 0.2 and 0.3 maximum

144 concentration contours which resulted in small differences (<5%) in the derived deposition

145 fluxes. In F7, the modelled and actual plume locations were very similar; however, in F19 and

146 F20, the modelled plumes were not exactly in the same geographical location as the observations

147 because of differences in advection direction (Tables 1, 2).

148 The spatially averaged dry deposition velocities for SO<sub>2</sub>, pSO<sub>4</sub> and TON are compared with 149 those obtained using inferential methods from GEM-MACH. The measurement and model

- results for all three flights are listed in Table S2.
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