

1 **Supplementary Information**

2 3 **Direct measurement of N₂O₅ heterogeneous uptake** 4 **coefficients on atmospheric aerosols in southwestern** 5 **China and evaluation of current parameterizations**

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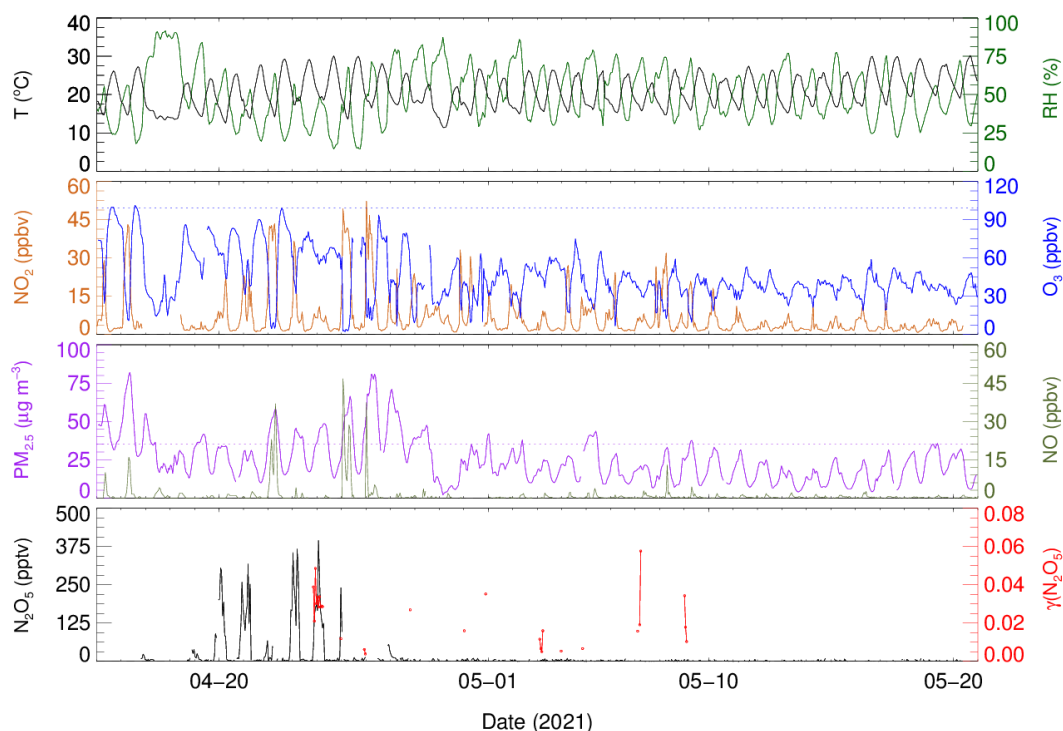
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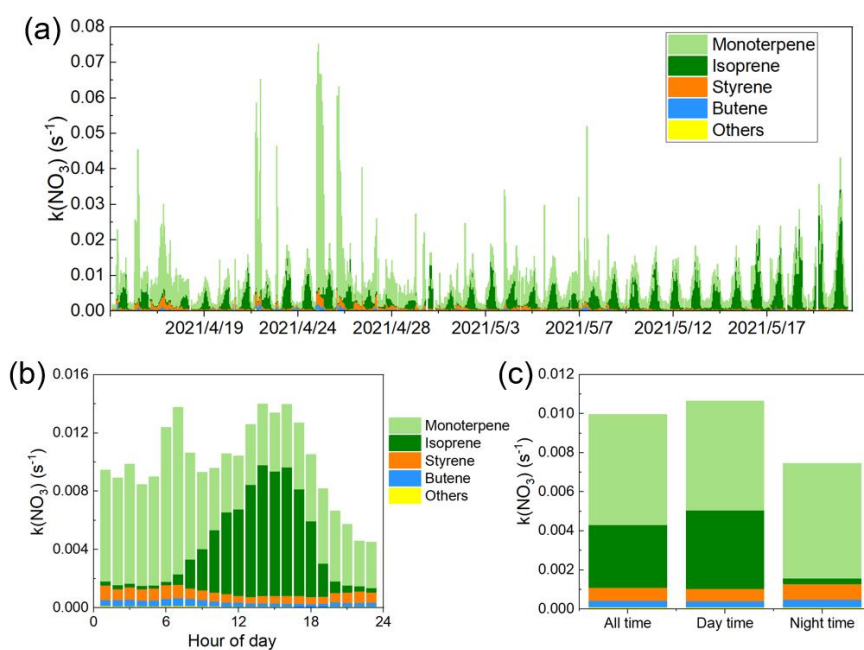
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41 **Figure S1.** Time series of measured concentrations of NO_2 , O_3 , NO , $\text{PM}_{2.5}$ and N_2O_5 ,
 42 the values of $\gamma(\text{N}_2\text{O}_5)$, and meteorological parameters of RH and T during the campaign.
 43 The blue line and the purple line represent Chinese national air quality standards for O_3
 44 and $\text{PM}_{2.5}$, respectively.

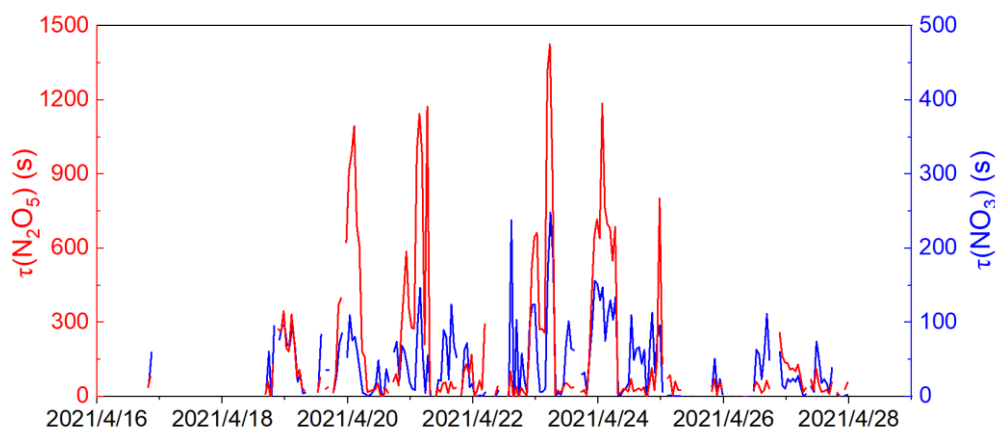
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47 **Figure S2.** NO_3 reactivity with VOCs. (a) Time series of VOCs contributions for
 48 $k(\text{NO}_3)$. (b) Mean diurnal profiles of $k(\text{NO}_3)$. (c) The contribution of VOCs categories

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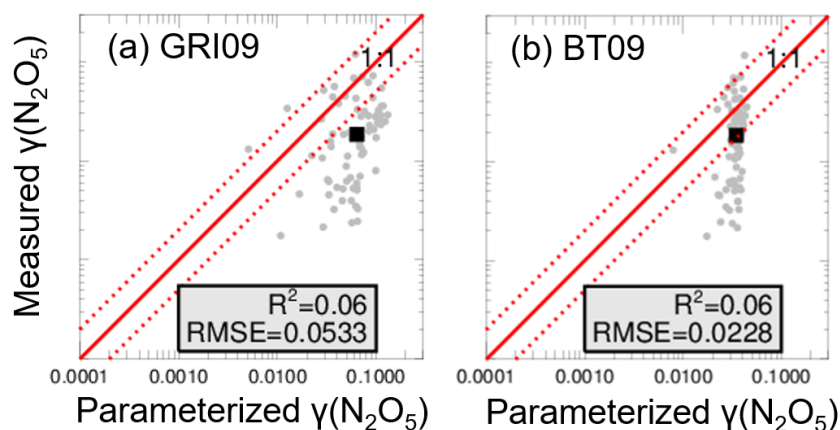


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51 **Figure S3.** Time series of N_2O_5 and NO_3 lifetime during the campaign.

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55 **Figure S4.** The distribution of parameterized $\gamma(\text{N}_2\text{O}_5)$ values of GRI09 (a) and BT09
56 (b).

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58 **Table S1.** Summary of field observation results of nocturnal NO_3 and N_2O_5
59 concentrations, $P(\text{NO}_3)$, and $\tau(\text{N}_2\text{O}_5)$ from various regions around the world in recent
60 years.

Location	Site type	Period	NO_3 (pptv) (night)	N_2O_5 (pptv) (night)	$P(\text{NO}_3)$ (ppbv/h)	$\tau(\text{N}_2\text{O}_5)$ (s)	Reference
Kunming, China	Suburban	2021.04	5.7 ± 3.2	33.4 ± 75.2 (395.1)	0.6 ± 0.1	185 ± 294	This work
Beijing, China	Rural	2016.05	27	73 ± 90 (937)	1.2 ± 0.9	270 ± 240	(Wang et al., 2018)
Beijing, China	Urban	2016.09	-	36.7	1.4 ± 1.7	-	(Wang et al., 2017a)

Beijing, China	Suburban	2016.02	-	~1400	-	-	(Wang et al., 2020b)
Wangdu, China	Rural	2014.07	-	~200 (430)	1.7±0.6	76.9	(Tham et al., 2016)
Mountain Tai, China	Suburban	2014.07	-	6.8±7.7	0.5±0.4	74	(Wang et al., 2017c)
Jinan, China	Urban	2014.08	-	22±12 (278)	-	-	(Wang et al., 2017b)
Shanghai, China	Urban	2011.08	16±9	310±380	1.1±1.1	-	(Wang et al., 2013)
Taizhou, China	Rural	2018.05	4.4±2.2	26.0±35.7 (492)	1.0×0.5	43 ± 52	(Wang et al., 2020a)
Taizhou, China	Suburban	2018.05	4.4±2.2	26.0 ± 35.7	1.2±0.4	55±68	(Li et al., 2020)
Changzhou, China	Suburban	2019.05	-	61.0 ± 63.1 (477.2)	2.8 ± 1.6	-	(Zhai et al., 2023)
Hongkong, China	Island	2012.08	7 ±12	17±33 (336)	-	76±61	(Yan et al., 2019)
Hongkong, China	Coastal	2013.11	-	~11800	0.26	~13 h	(Brown et al., 2016)
Shenzhen, China	Coastal	2019.09	-	56±89 (1420)	2.9±0.5	-	(Niu et al., 2022)
Heshan, China	Urban	2019.09	~90	64±145	2.5±2.1	-	(Wang et al., 2022)
South China Sea	Island	2021.11	10 ±13	120±129	1.4±0.7	30±42	(Wang et al., 2024)
Seoul, Korea	Urban, tower	2015.05	-	4100±1200, 2600±1600 (5000)	1.3	1800	(Brown et al., 2017)
Southern Spain	Coastal	2008.11	-	~500	-	-	(Crowley et al., 2011)
Northwestern, Europe	Coastal, airborne	2010.07	-	670	-	15~120 min	(Morgan et al., 2015)
Taunus Observatory, Germany	Rural	2011.08	-	~800	~1.8	-	(Phillips et al., 2016)
East coast USA	Coastal	2002.06	17	84	-	-	(Brown et al., 2004)
California, USA	Coastal	2004.01	-	~200	-	~30 min	(Wood et

USA							al., 2005)
Salt Lake Valley, USA	Airborne	2017.01	-	0~2	0~2	-	(McDuffie et al., 2019)
Lower Fraser Valley, Canada	Suburban	2012.07	-	1.4 (23)	-	-	(Osthoff et al., 2018)

61 The values in brackets are the maximum of N₂O₅ concentration.

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64 **Table S2.** Summary of global field observation results of $\gamma(\text{N}_2\text{O}_5)$.

Location	Period	Site type	$\gamma(\text{N}_2\text{O}_5)$	Method	Reference
Kunming, China	2021.04	Suburban	0.0018~0.12 (0.23±0.21)	AFTS	This work
Beijing, China	2016.02	Suburban	0.001-0.02 (0.0046)	Box model	(Wang et al., 2020b)
Beijing, China	2016.05	Rural	0.012-0.055 (0.034)	Products	(Wang et al., 2018)
Beijing, China	2016.09	Urban	0.025-0.072 (0.048)	Steady-state	(Wang et al., 2017a)
Beijing, China	2018.1	Urban	0.0075-0.0149	Steady-state	(Xia et al., 2021)
Beijing, China	2019.01	Tower	0.0005-0.2 (0.05)	Box model	(Chen et al., 2020)
Beijing, China	2020.12	Urban	0.0045-0.12 (0.042±0.026)	AFTS	(Chen et al., 2022)
Wangdu, China	2014.06	Rural	0.005-0.039	Products	(Tham et al., 2018)
Wangdu, China	2014.06	Rural	0.0012-0.072	Steady-state	(Lu et al., 2022)
Wangdu, China	2017.12	Rural	0.006-0.015	Steady-state	(Xia et al., 2021)
Mountain Tai, China	2014.07	Suburban	0.021-0.102 (0.061)	Steady-state	(Wang et al., 2017c)
Mountain Tai, China	2018.03	Suburban	0.001-0.019 (0.01)	AFTS	(Yu et al., 2020)
Jinan, China	2014.08	Urban	0.042-0.092 (0.069)	Steady-state	(Wang et al., 2017b)
Taizhou, China	2018.06	Suburban	0.027-0.107 (0.08)	Steady-state	(Li et al., 2020)

Location	Period	Site type	$\gamma(\text{N}_2\text{O}_5)$	Method	Reference
Changzhou, China	2019.06	Suburban	0.057-0.123 0.001-0.024	Steady-state Parameterization	(Zhai et al., 2023)
Hongkong, China	2013.11	Suburban	0.004-0.029 (0.014)	Steady-state	(Brown et al., 2016)
Hongkong, China	2013.11	Suburban	0.0005-0.016 (0.004)	Box model	(Yun et al., 2018)
Heshan, China	2017.03	Suburban	0.002-0.067 (0.02)	AFTS	(Yu et al., 2020)
Heshan, China	2019.10	Urban	0.0019-0.077 (0.0317)	Products	(Wang et al., 2022)
Shenzhen, China	2019.10	Coastal	0.002-0.068 (0.027 \pm 0.02) 0.005-0.08 (0.031 \pm 0.02)	Products Box model	(Niu et al., 2022)
New England, USA	2002.08	Ship	0.03-0.04	Steady-state	(Aldener et al., 2006)
New England, USA	2004.02	Airborne	0.0016-0.02	Steady-state	(Brown et al., 2006)
Texas, USA	2006.10	Airborne	0.0005-0.006 (0.0039)	Steady-state	(Brown et al., 2009)
Boulder, USA	2008.07	Tower	0.0009-0.012 (0.003)	AFTS	(Bertram et al., 2009)
Seattle, USA	2008.08	Coastal	0.005-0.04	AFTS	(Bertram et al., 2009)
California, USA	2009.09	Coastal	0.00003-0.029 (0.0054)	AFTS	(Riedel et al., 2012)
Los Angeles, USA	2010.05	Airborne	0.001-0.01	Steady-state	(Chang et al., 2016)
Colorado, USA	2011.02	Tower	0.002-0.1 (0.04)	Box model	(Wagner et al., 2013)
Eastern, USA	2015.02	Airborne	0.00002-0.175 (0.014)	Box model	(McDuffie et al., 2018)
Salt Lake Valley, USA	2017.01	Airborne	0.001-0.1 (0.076)	Box model	(McDuffie et al., 2019)
NW Europe/UK	2010.06	Airborne	0.0076-0.03	Steady-state	(Morgan et al.,

Location	Period	Site type	$\gamma(\text{N}_2\text{O}_5)$	Method	Reference
SW Germany	2011.08	Suburban	0.004-0.11 (0.028)	Products, Steady-state	2015) (Phillips et al., 2016)

65 The values in brackets are mean values of $\gamma(\text{N}_2\text{O}_5)$.

66 AFTS: aerosol flow tube system;

67 Steady-state: steady state approximation;

68 Products: products formation rate analysis;

69 Box model: inverse iterative box model simulation.

Table S3. Summary of the details and performances of the parameterizations discussed in this study.

Parameterization name	Factors considered	Parameterization	Reference	R ²	RMSE	Median
RIE03	Mass concentration of aerosol sulfate and nitrate (μg/m ³)	$\gamma = f \times r_1 + (1-f) \times r_2$ where, $r_1 = 0.02$ $r_2 = 0.002$ $f = \text{mass SO}_4^{2-} / (\text{mass SO}_4^{2-} + \text{mass NO}_3^-)$	(Riemer et al., 2003)	0	0.0223	0.017
DAV08	[SO ₄ ²⁻], [NO ₃ ⁻], [NH ₄ ⁺], RH, T	$\gamma = x_1 \times \gamma_1^* + x_2 \times \gamma_2^* + x_3 \times \gamma_3^*$ where, $\lambda_1 = -4.10612 + 0.02386 \times \text{RH} - 0.23771 \times \max((T-291), 0)$ $\gamma_1 = 1 / (1 + e^{-\lambda_1})$ $\gamma_1^* = \min(\gamma_1, 0.08585)$ $\lambda_2 = (-4.10612 - 0.80570) + 0.02386 \times \text{RH} + (-0.23771 + 0.10225) \times \max((T-291), 0)$ $\gamma_2 = 1 / (1 + e^{-\lambda_2})$ $\gamma_2^* = \min(\gamma_2, 0.053)$ $\lambda_3 = -8.10774 + 0.04902 \times \text{RH}$ $\gamma_3 = 1 / (1 + e^{-\lambda_3})$ $\gamma_3^* = \min(\gamma_3, 0.0154)$ $x_3 = [\text{NO}_3^-] / ([\text{NO}_3^-] + [\text{SO}_4^{2-}])$ $x_2 = \max(0, \min(1 - x_3, [\text{NH}_4^+] / ([\text{NO}_3^-] + [\text{SO}_4^{2-}]) - 1))$ $x_1 = 1 - x_2 - x_3$	(Davis et al., 2008)	0.02	00317	0.034

Parameterization name	Factors considered	Parameterization	Reference	R ²	RMSE	Median
		(1=NH ₄ HSO ₄ , 2=(NH ₄) ₂ SO ₄ , 3=NH ₄ NO ₃)				
BT09	ALWC, [NO ₃ ⁻], [Cl ⁻], V _a , S _a	$\gamma = \frac{4 V_a}{c S_a} K_H k'_{2f} \left(1 - \frac{1}{\left(\frac{k_3 [\text{H}_2\text{O}]}{k_{2b} [\text{NO}_3^-]} \right) + 1 + \left(\frac{k_4 [\text{Cl}^-]}{k_{2b} [\text{NO}_3^-]} \right)} \right)$ <p>where, $K_H=51$, Henry's Law Coefficient (Fried et al., 1994) $k'_{2f}=\beta - \beta_e^{-\delta[\text{H}_2\text{O}]}$ $\beta=1.15 \times 10^6$ (s⁻¹) $\delta=0.13$ (M⁻¹) $\frac{k_3}{k_{2b}}=0.06$ $\frac{k_4}{k_{2b}}=29$</p>	(Bertram and Thornton, 2009)	0.06	0.0228	0.034
BT09 w/o Cl	ALWC, [NO ₃ ⁻], V _a , S _a	$\gamma = \frac{4 V_a}{c S_a} K_H k'_{2f} \left(1 - \frac{1}{\left(\frac{k_3 [\text{H}_2\text{O}]}{k_{2b} [\text{NO}_3^-]} \right) + 1} \right)$ <p>parameters are same as BT09.</p>	(Bertram and Thornton, 2009)	0.07	0.0202	0.020
GRI09	ALWC, [NO ₃ ⁻], V _a , S _a	$\gamma = \frac{4 V_a}{c S_a} K_H k'_{2f} \left(1 - \frac{1}{\left(\frac{k_3 [\text{H}_2\text{O}]}{k_{2b} [\text{NO}_3^-]} \right) + 1} \right)$	(Griffiths et al., 2009)	0.06	0.0533	0.063

Parameterization name	Factors considered	Parameterization	Reference	R ²	RMSE	Median
		Where, $K_H=51$ $\frac{k_3}{k_{2b}}=1/30$ $k'_{2f}=5 \times 10^6$				
YU20	ALWC, $[\text{NO}_3^-]$, $[\text{Cl}^-]$, V_a , S_a	$\gamma = \frac{4 V_a}{c S_a} K_H k'_{2f} \left(1 - \frac{1}{\left(\frac{k_3 [\text{H}_2\text{O}]}{k_{2b} [\text{NO}_3^-]} \right) + 1 + \left(\frac{k_4 [\text{Cl}^-]}{k_{2b} [\text{NO}_3^-]} \right)} \right)$ <p>where, $K_H=51$ $k'_{2f}=[\text{H}_2\text{O}] \times 3 \times 10^4$ $\frac{k_3}{k_{2b}}=0.033$ $\frac{k_4}{k_{2b}}=3.4$</p>	(Yu et al., 2020)	0.09	0.02	0.019
EJ05	Mass fraction of aerosol sulfate and organic, RH,	$\gamma = \text{mass SO}_4^{2-} / \text{dry mass} \times \gamma_1 + \text{mass organic} / \text{dry mass} \times \gamma_2$ <p>where, $\gamma_1 = \alpha \times 10^\beta$ $\alpha = 2.79 \times 10^{-4} + 1.3 \times 10^{-4} \times \text{RH} - 3.43 \times 10^{-6} \times \text{RH}^2 + 7.52 \times 10^{-8} \times \text{RH}^3$</p>	(Evans and Jacob, 2005)	0	0.0228	0.019

Parameterization name	Factors considered	Parameterization	Reference	R ²	RMSE	Median
	T	$\beta=4 \times 10^{-2} \times (T-294)$, (T \geq 282K) $\beta=-0.48$, (T<282K) $\gamma_2=RH \times 5.2 \times 10^{-4}$, (RH<57%) $\gamma_2=0.03$, (RH \geq 57%)				
BT09+Rie09	ALWC, [NO ₃ ⁻], [Cl ⁻], V _a , S _a , organic coating	$\frac{1}{\gamma} = \frac{1}{\gamma_{core}} + \frac{1}{\gamma_{org.coat}}$ <p>where, γ_{core}=BT09 $\gamma_{org.coat} = \frac{4RTD_{org}H_{org}R_c}{clR_p}$ R, gas constant (atm·m³/mol·K) T, temperature (K) D_{org}H_{org}=εD_{aq}H_{aq} ε=0.03 H_{aq}=5000, Henry's Law Coefficient in aqueous core (mol m⁻³ atm⁻¹) D_{aq}=1×10⁻⁹, N₂O₅ Liquid Diffusion Coefficient (m² s⁻¹) R_p, median particle total radius, (m) R_c=R_p-l, particle core radius (m) l=R_p×(1-β^{1/3}), organic coating thickness (m) β=V_{inorganic}/(V_{organic}+V_{inorganic})</p>	(Bertram and Thornton, 2009; Anttila et al., 2006; Riemer et al., 2009)	0.03	0.0278	0.012
BT09+Rie09(wG14)	ALWC, [NO ₃ ⁻], [Cl ⁻],	$\frac{1}{\gamma} = \frac{1}{\gamma_{core}} + \frac{1}{\gamma_{org.coat}}$	(Gaston et al., 2014)	0.07 (O/C=0.8);	0.0201 (O/C=0.8);	0.019 (O/C=0.8);

Parameterization name	Factors considered	Parameterization	Reference	R ²	RMSE	Median
	V _a , S _a , organic coating, O/C, RH	same as BT09+Rie09 except, $\varepsilon=0.06$, (RH \leq 30% and O/C \geq 0.7) $\varepsilon=0.008$, (RH \leq 30% and O/C \leq 0.7) $\varepsilon=0.3$, (30% \leq RH \leq 70% and O/C \geq 0.7) $\varepsilon=0.05$, (30% \leq RH \leq 70% and O/C \leq 0.7) $\varepsilon=1.0$, (70% \leq RH and O/C \geq 0.7) $\varepsilon=0.8$, (70% \leq RH and O/C \leq 0.7)		0.07 (O/C=0.5)	0.0248 (O/C=0.5)	0.006 (O/C=0.5)
MD18	ALWC, [NO ₃ ⁻], [Cl ⁻], V _a , S _a , organic coating, O/C, RH	$\frac{1}{\gamma} = \frac{1}{\gamma_{core}} + \frac{1}{\gamma_{org.coat}}$ same as BT09+Rie09 except, γ_{core} =BT09 w/o Cl k'_{2f} =[H ₂ O] \times 2.14 \times 10 ⁵ $\frac{k_3}{k_{2b}}$ =0.04 $\varepsilon=0.15\times$ O/C+0.0016 \times RH	(McDuffie et al., 2018)	0.05 (O/C=0.8); 0.04 (O/C=0.5)	0.0205 (O/C=0.8); 0.0208 (O/C=0.5)	0.022 (O/C=0.8); 0.018 (O/C=0.5)

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