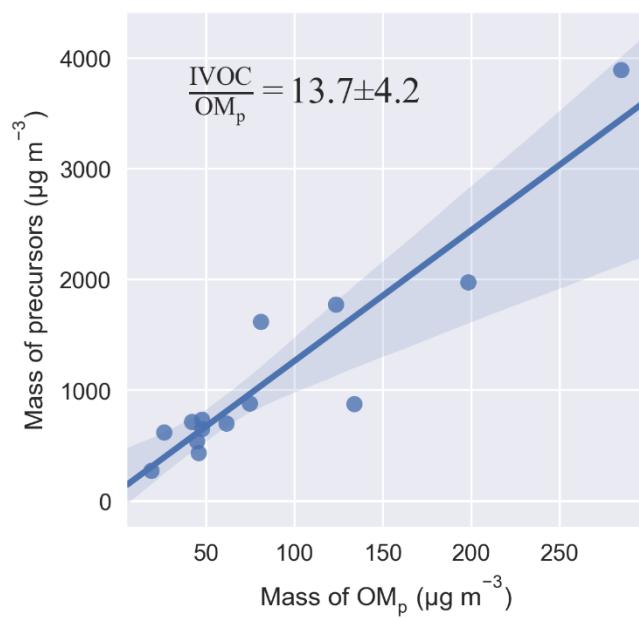
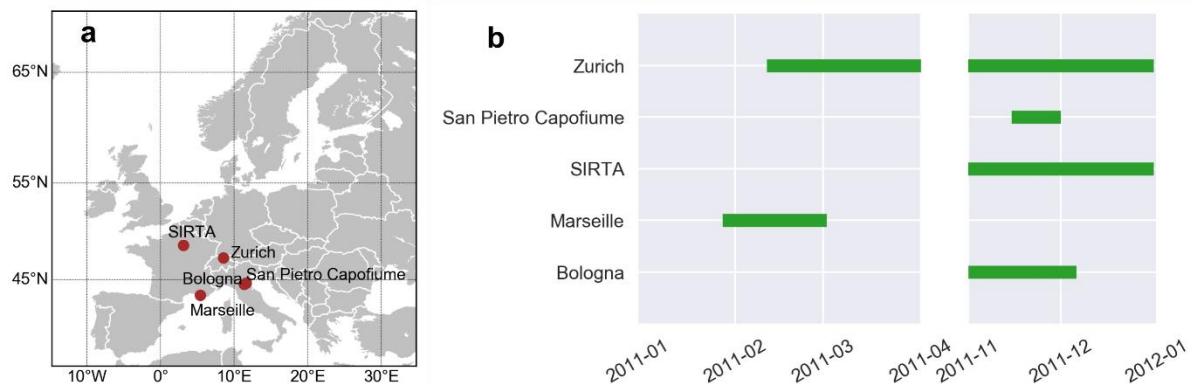


**Table S1:** Modeled and measured ratio of SOA to OA by different OA schemes.

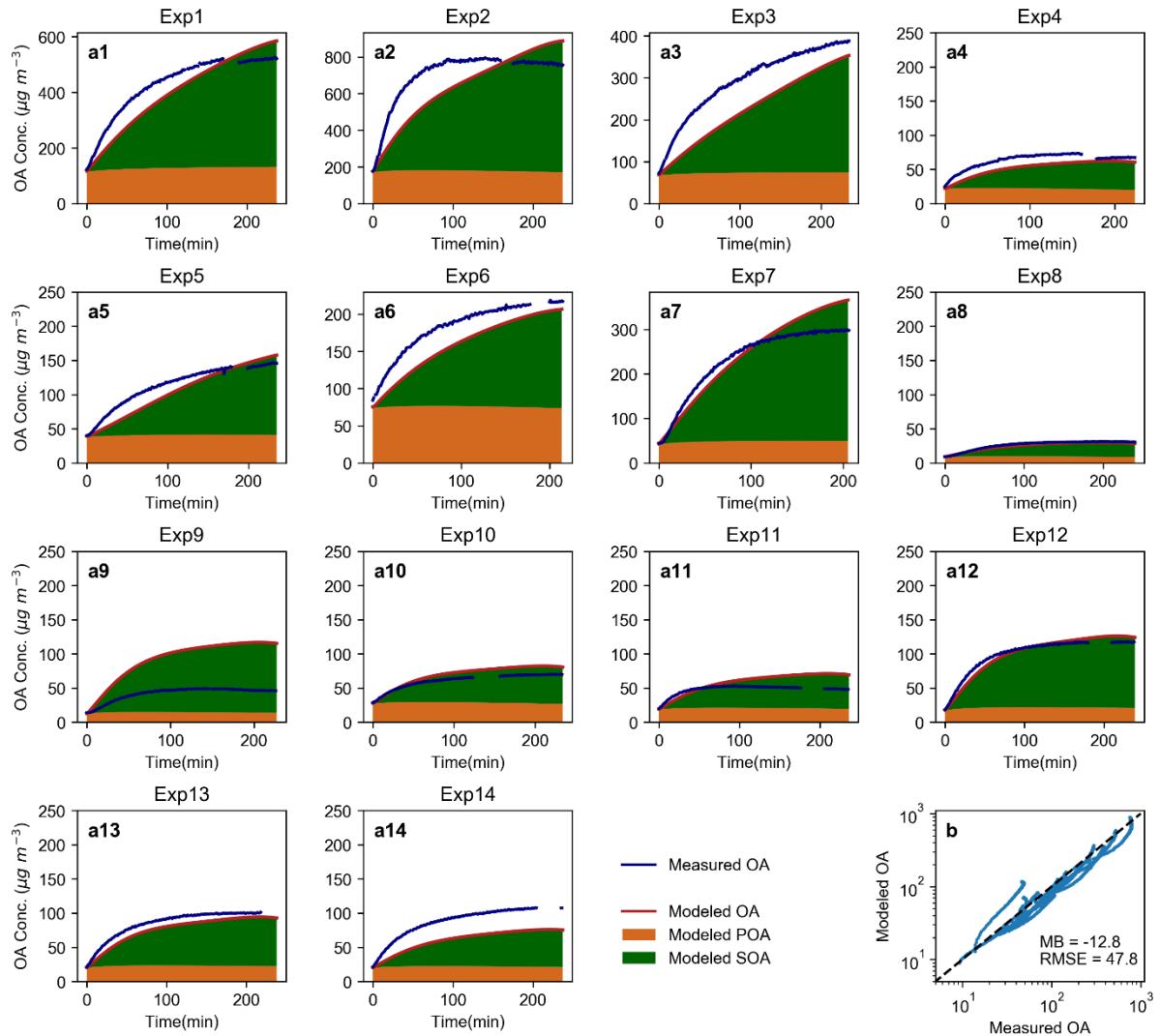
Site	Time	Source	Observed SOA/OA(%)	Modeled SOA/OA (%)				
				SOAP	VBS_ BASE	VBS_ 3POA	VBS_ noWLS	VBS_ WLS
Barcelona	Spring 2009	Crippa et al., 2014	49.0	54.1	79.6	57.2	58.2	62.8
Bologna	Winter 2011	Jiang et al., 2019	52.3	18.4	48.1	25.7	26.2	38.5
	Fall 2011		39.2	32.4	71.5	46.9	48.2	58.3
Cabauw	Spring 2008, 2009	Crippa et al., 2014	65.5	63.8	87.4	70.2	71.4	75.4
Chilbolton	Spring 2009	Crippa et al., 2014	59.0	63.8	84.2	64.5	66.0	69.9
Finokalia	Spring 2008	Crippa et al., 2014	78.5	61.5	89.3	74.2	75.4	80.3
	Fall 2011	Jiang et al., 2019	100	69.5	91.4	78.6	79.3	83.4
Helsinki	Fall 2009	Crippa et al., 2014	71.0	76.1	77.1	53.8	54.8	58.4
Hyytiälä	Spring 2009	Crippa et al., 2014	93.0	88.1	88.2	71.6	73.8	77.4
	Fall 2008		90.0	94.7	93.3	82.6	83.7	85.1
K-Puszta	Fall 2008	Crippa et al., 2014	77.0	51.3	79.5	57.7	59.6	68.9
Mace Head	Spring 2008, 2009	Crippa et al., 2014	63.0	92.5	95.4	87.6	88.1	89.3
Marseille	Winter 2011	Jiang et al., 2019	50.7	38.4	64.4	38.9	40.1	48.6
	Summer 2011		89.5	63.9	89.4	74.2	74.4	76.0
Melpitz	Spring 2008, 2009	Crippa et al., 2014	87.5	82.7	92.4	80.5	81.6	83.7
	Fall 2008		78.0	81.3	91.2	77.8	78.8	81.0
Montsec	Fall 2011	Jiang et al., 2019	91.4	85.0	92.8	81.5	82.1	84.6
Paris	Winter 2011	Jiang et al., 2019	37.2	16.4	33.7	15.5	16.1	22.7
	Fall 2011		44.5	23.8	57.6	31.9	32.9	41.0
Payerne	Spring 2009	Crippa et al., 2014	84.0	72.4	86.5	69.0	70.5	75.4
	Fall 2008		81.0	76.9	88.2	72.0	73.0	76.6
Puy de Dome	Spring 2009	Crippa et al., 2014	76.0	52.5	84.2	65.4	67.8	75.6
	Fall 2008		90.0	59.8	85.1	66.5	68.1	74.6
San Pietro Capofiume	Fall 2008	Jiang et al., 2019	36.3	38.3	74.7	50.9	52.5	62.1
Vavihill	Spring 2009		69	73.0	86.7	69.0	70.7	74.8
Zurich	Winter 2011	Jiang et al., 2019	68.2	34.0	59.2	34.2	35.7	44.4
	Spring 2011		66.8	72.1	86.7	68.9	70.3	74.3
	Summer 2011		69.5	86.6	93.3	82.5	82.7	83.6
	Fall 2011		62.6	78.2	89.2	73.8	74.7	77.3



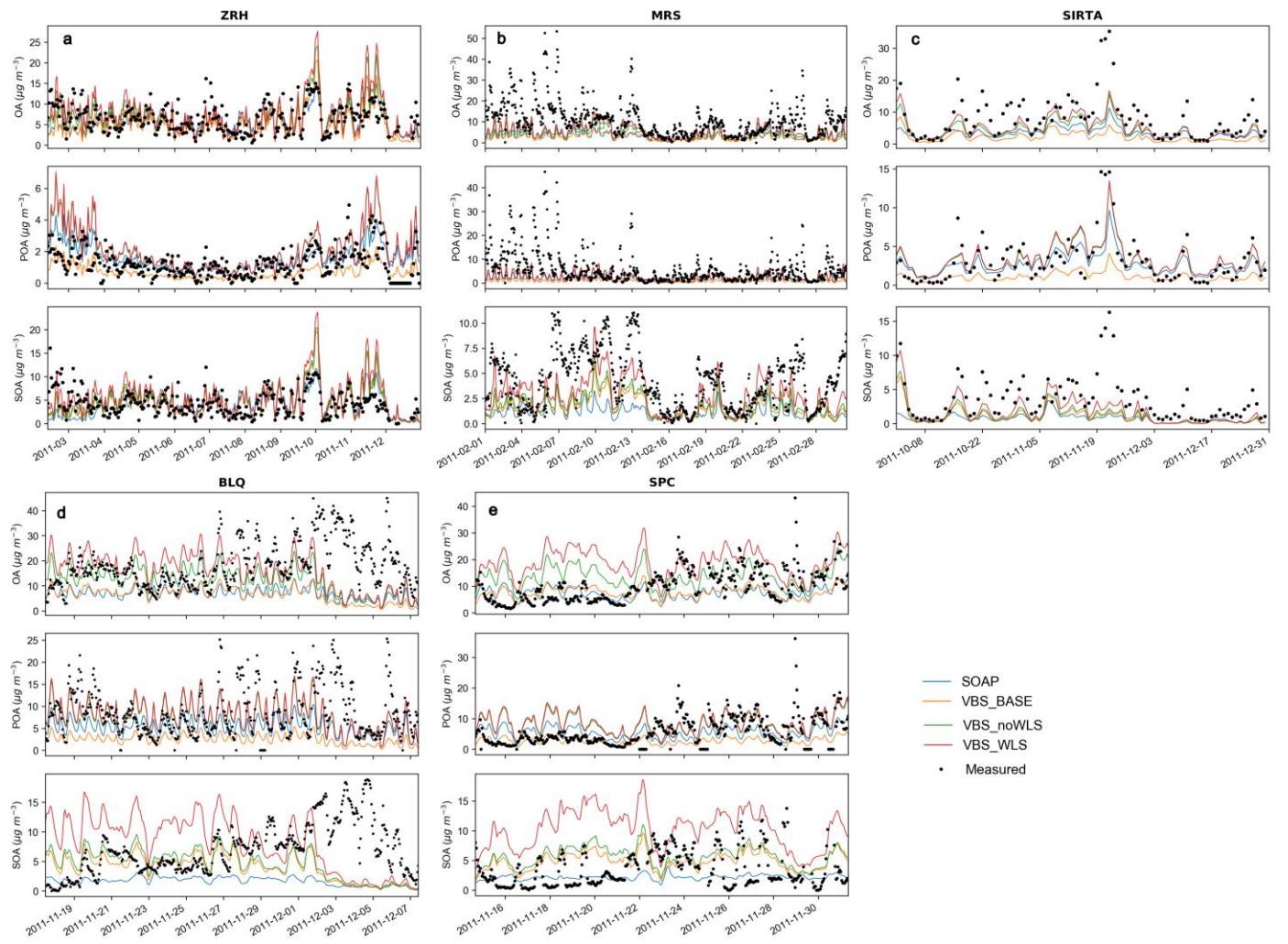
**Figure S1:** The relationship between mass of IVOC precursors and the mass of primary OM ( $\text{OM}_p$ ).



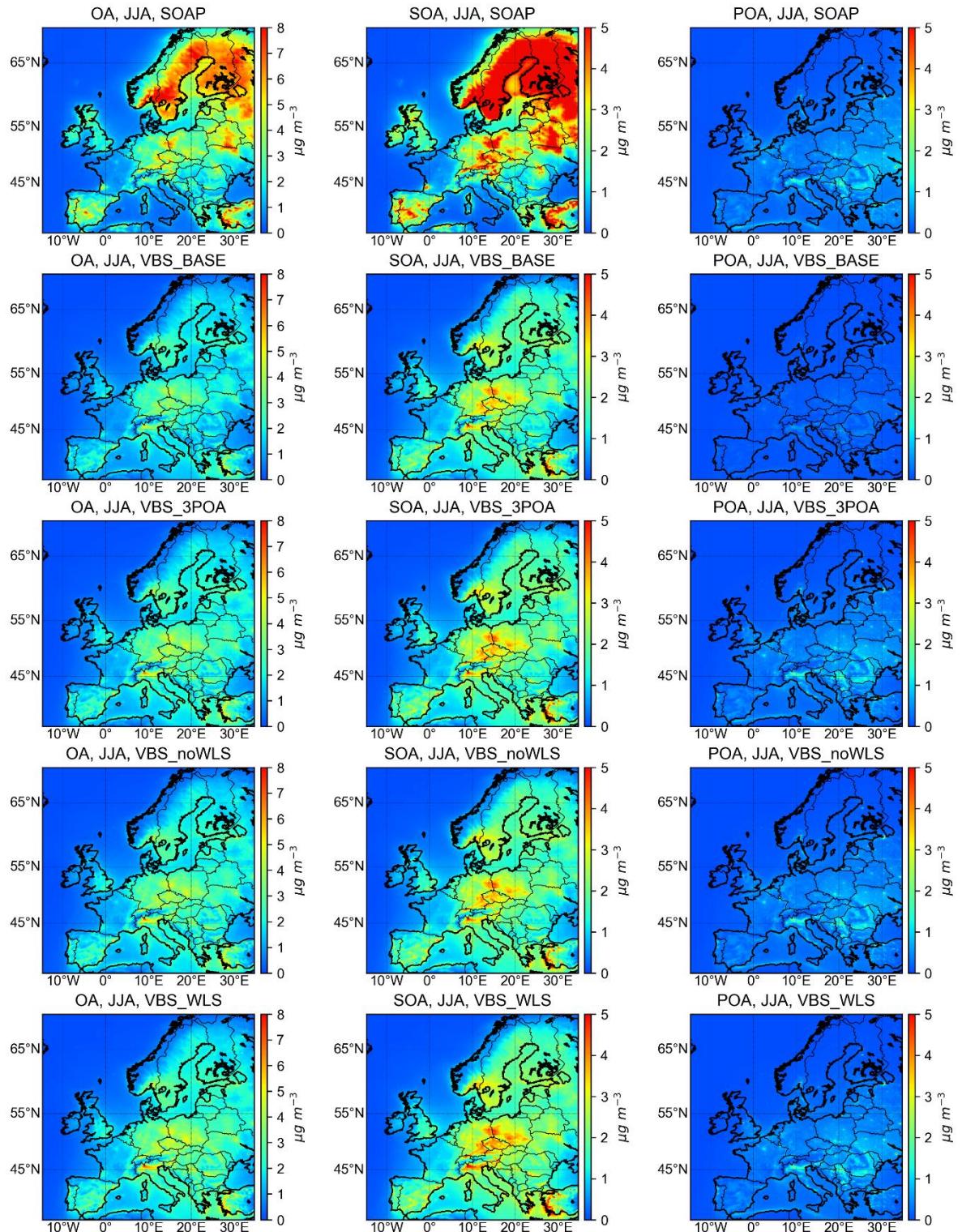
**Figure S2:** Spatial distribution (a) and observation periods (b) of observations.



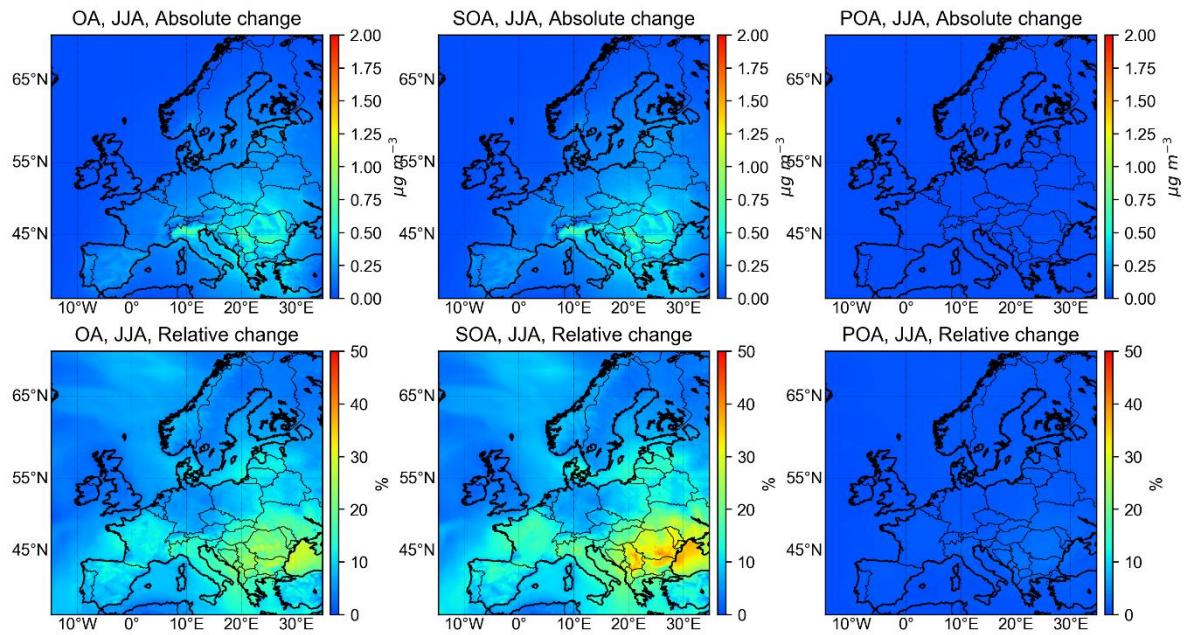
**Figure S3:** Modeled OA with optimized parameterization assuming no vapor wall loss (a) and comparison with measurements (b).



**Figure S4:** Temporal variations of measured and observed OA, POA and SOA by different OA schemes.



**Figure S5:** Modeled OA, SOA and POA in summer (JJA, June–July–August) by different OA schemes.



**Figure S6:** Differences in modeled OA, SOA and POA in summer (JJA, June–July–August) by VBS schemes with (VBS\_WLS) and without (VBS\_noWLS) vapor wall corrections.