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Aerosol composition trends during 2000–2020: in-depth insights from model predictions and multiple worldwide near-surface observation datasets

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Aerosol Chemistry Climate Model Description

Model configuration

In the model configuration used, EMAC calculates fields of gas phase species online through the Module Efficiently Calculating the Chemistry of the Atmosphere (MECCA) submodel (Sander et al., 2019). MECCA calculates the concentration of a range of gases, including aerosol precursor species such as SO_2, NH_3, NO_x, DMS, H_2SO_4 and DMSO. The concentrations of the major oxidant species (OH, H_2O_2, NO_3, and O_3) are also calculated online. The loss of gas phase species to the aerosol through heterogeneous reactions (e.g., N_2O_5 to form HNO_3) is treated using the MECCA_KHET submodel (Jöckel et al., 2010). The aqueous phase oxidation of SO_2 and the uptake of HNO_3 and NH_3 in cloud droplets are treated by the SCAV submodel (Tost et al., 2006; Tost et al., 2007).

Aerosol microphysics and gas/aerosol partitioning are calculated by the Global Modal-aerosol eXtension (GMXe) module (Pringle et al., 2010). The aerosol size distribution is described by 7 interacting lognormal modes (4 hydrophilic and 3 hydrophobic modes). The modes cover the aerosol size spectrum (nucleation, Aitken, accumulation and coarse). The aerosol composition within each mode is uniform with size (internally mixed), though can vary between modes (externally mixed). The removal of gas and aerosol species through dry deposition is calculated within the DRYDEP submodel (Kerkweg et al., 2006) based on the big leaf approach. The sedimentation of aerosols is calculated within the SEDI submodel (Kerkweg et al., 2006) using a first order trapezoid scheme. Cloud properties and microphysics are calculated by the CLOUD submodel utilizing the detailed two-moment microphysical scheme of Lohmann and Ferrachat (2010) and considering a physically based treatment of the processes of liquid (Karydis et al., 2017) and ice crystal (Bacer et al., 2018) activation.

State of the art modules for the inorganic thermodynamics

The inorganic aerosol composition is computed with the ISORROPIA-lite thermodynamic equilibrium model (Kakavas et al., 2022) as implemented in EMAC by Milousis et al. (2024). ISORROPIA-lite is an accelerated and simplified version of the widely used ISORROPIA-II aerosol thermodynamics model which calculates the gas/liquid/solid equilibrium partitioning of the K^+-Ca^{2+}-Mg^{2+}-$NH4^+$-Na^+-$SO4^{2-}$-NO^{3-}-Cl^--H_2O aerosol system. ISORROPIA-lite assumes that the aerosol is always in a metastable state (i.e., it is composed only of a supersaturated aqueous phase) and uses binary activity coefficients from precalculated look-up tables to minimize the computational cost. ISORROPIA-lite provides almost identical results with ISORROPIA-II in a metastable mode and reduces its computational cost by 35% (Kakavas et al., 2022). The application of ISORROPIA-lite in EMAC improved the computational speed of the model by 4% (Milousis et al., 2024). The assumption of thermodynamic equilibrium is a good approximation for fine mode aerosols which can reach equilibrium within the time frame of one model timestep. However, the equilibrium timescale for large particles is typically larger than the timestep of the model (Meng and Seinfeld, 1996). To account for kinetic limitations, the process of gas/aerosol partitioning is calculated in two stages (Pringle et al., 2010). In the first stage the amount of the gas phase species that are able to kinetically condense onto the aerosol phase within the model timestep is calculated assuming diffusion limited condensation (Vignati et al., 2004). In the second stage ISORROPIA-lite re-distributes the mass between the gas and the aerosol phase assuming instant equilibrium between the two phases.

State of the art module for organic aerosol

The organic aerosol composition and evolution in the atmosphere is calculated by a computationally efficient version of the ORACLE module (Tsimpidi et al., 2014) which simulates a wide variety of semi-volatile organic products separating them into bins of logarithmically spaced effective saturation concentrations. In this application ORACLE uses three surrogate species with effective saturation concentration at 298 K of $C^* = 10^{-2}$, 10^{1}, and 10^{4} μg m^{-3} to cover the volatility range of LVOCs, SVOCs and IVOCs emissions from biomass burning and other combustion sources (biofuel and fossil fuel combustion, and other urban sources). These organic compounds are allowed to partition between the gas and aerosol phases resulting in the formation of POA. The least volatile fraction, at 10^{-2} μg m^{-3}, describes the low volatility organics in the atmosphere that are mostly in the particulate phase even in remote locations. The 10 μg m^{-3} volatility bin describes the semivolatile organics in the atmosphere which partition between the particle and gas phase at atmospheric conditions. Finally, even under highly polluted conditions the majority of the material in the 10^{4} μg m^{-3} volatility bin will exist almost exclusively in the vapor phase. Photochemical reactions that modify the volatility of the emitted organic compounds that remain in the gas phase are taken into account and the oxidation products are simulated separately in the module to keep track of the SOA formation from SVOC and IVOC emissions. LVOCs are not allowed to participate in photochemical reactions since they are already in the lowest volatility bin. A similar approach is followed for SOA formed from VOCs. In the this version of ORACLE, it is assumed that the oxidation of the anthropogenic (alkanes, aromatics, and olefins) and biogenic (isoprene and monoterpenes) VOC species results in two products for each precursor distributed in two volatility bins with effective saturation concentrations at 298 K equal to 1 and 10^3 μg m^{-3} at 298 K. Overall, we have assumed that functionalization and fragmentation processes after any subsequent photochemical aging as a result of the reaction with OH results in a net average decrease of volatility by a factor of 10^3 for SOA produced by SVOC/IVOC and anthropogenic VOC, without a net average change of volatility for SOA produced by biogenic VOC (Tsimpidi et al., 2014). In total 18 organic compounds are simulated explicitly, i.e., 9 in each of the gas and aerosol phases. Based on the saturation concentration of each organic compound, ORACLE calculates the partitioning between the gas and particle phases by assuming bulk equilibrium and that all organic compounds form a pseudo-ideal solution. A schematic overview of the ORACLE module and the different aerosol types and chemical processes considered here is provided in Figure S1.

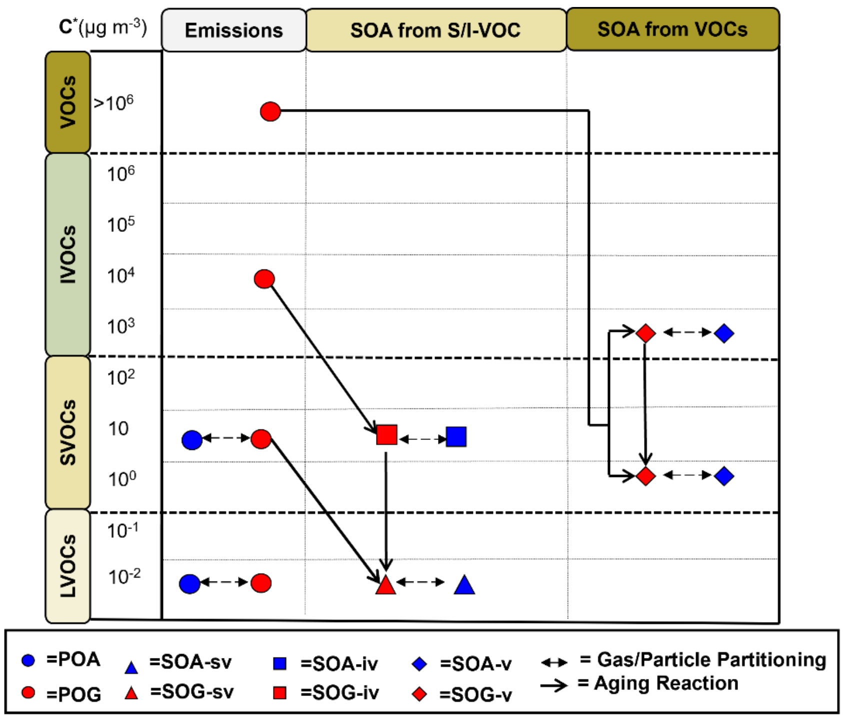

Figure S1: Schematic of the VBS resolution and the formation procedure of POA and SOA from LVOCs, SVOCs, IVOCs and VOCs emissions in ORACLE-lite. Red indicates that the compound is in the vapor phase and blue in the particulate phase. The circles correspond to primary organic material that can be emitted either in the gas or in the aerosol phase. The triangles indicate the formation of SOA from SVOCs by fuel combustion and biomass burning sources, while the squares show SOA from IVOCs by fuel combustion and biomass burning sources, and the diamonds the formation of SOA from anthropogenic and biogenic VOC sources. The partitioning processes, the aging reactions and the names of the species used to track all compounds are also shown.

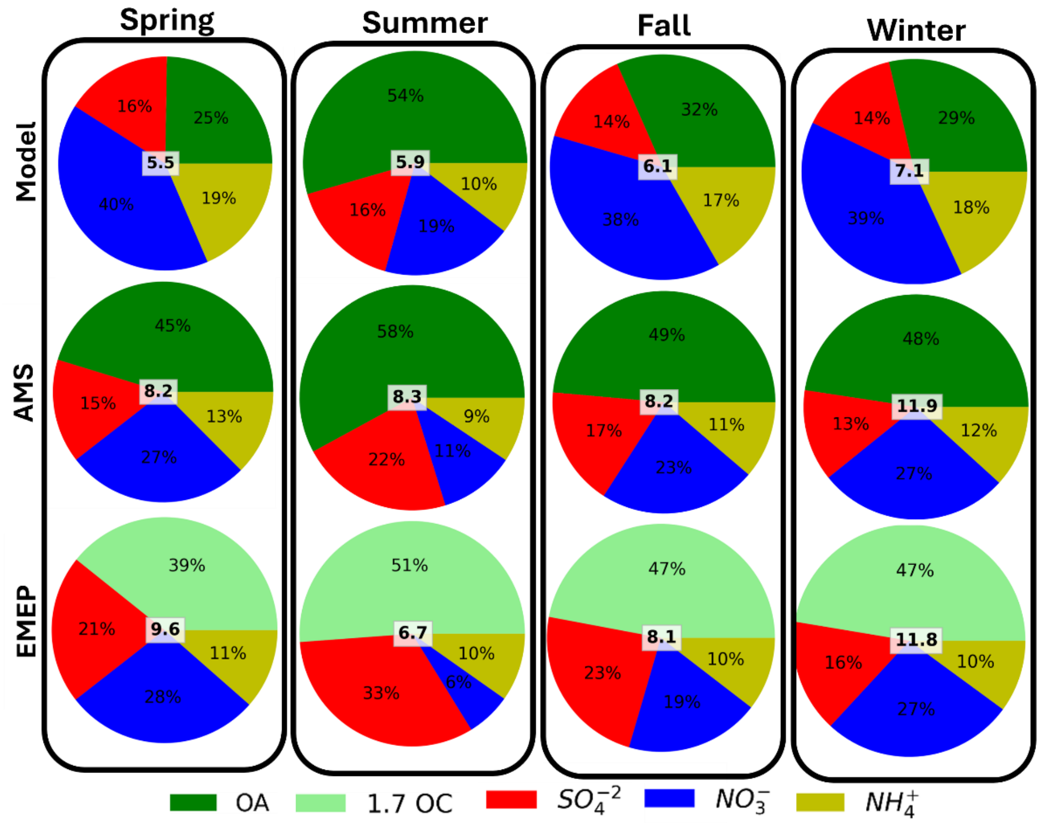

Figure S2. Seasonal aerosol composition in Europe from EMAC simulation, AMS observations and EMEP filter measurements during 2000-2020. The white boxes indicate the seasonal mean of the total PM concentration (in µg m^{-3}), the numbers in the respective pie slices indicate the relative contribution of each element (in %).

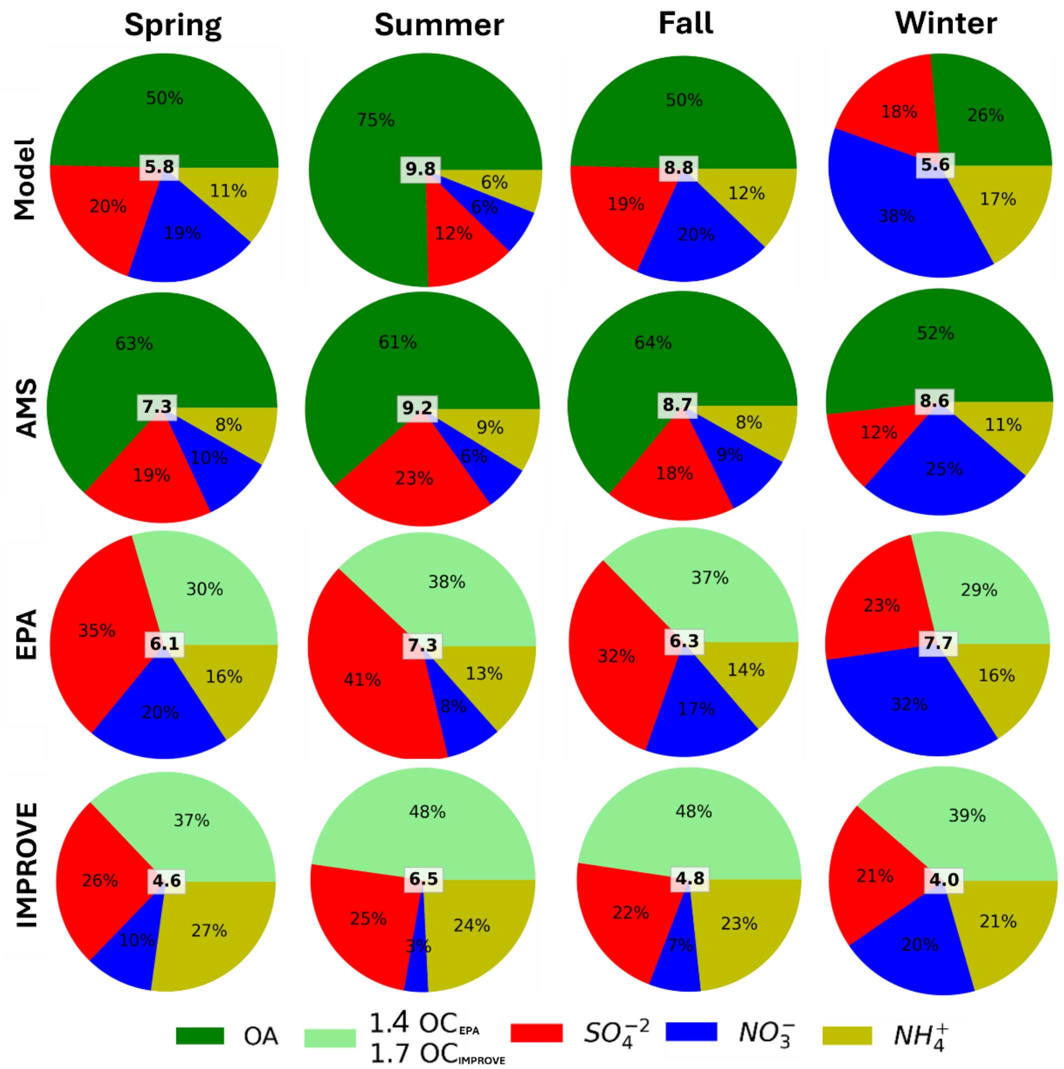

Figure S3. Seasonal aerosol composition in North America from EMAC simulation, AMS observations and IMPROVE and EPA filter measurements during 2000-2020. The white boxes indicate the seasonal mean of the total PM concentration (in µg m^{-3}), the numbers in the respective pie slices indicate the relative contribution of each element (in %).

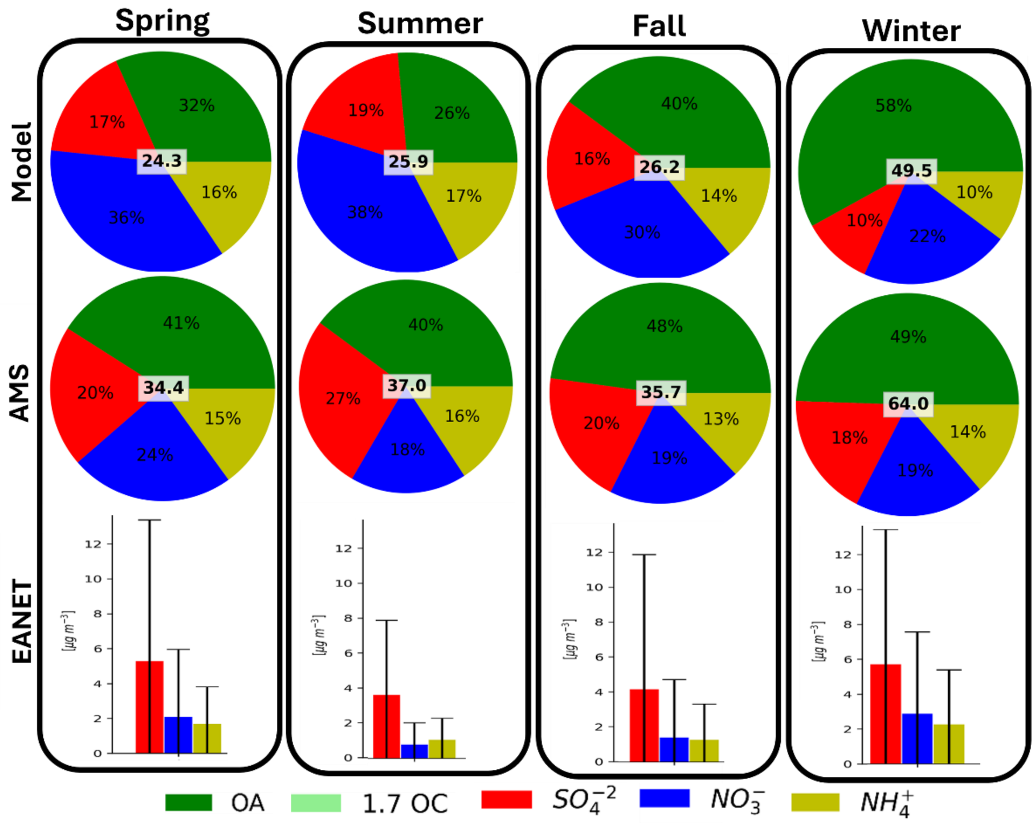

Figure S4. Seasonal aerosol composition in Eastern Asia from EMAC simulation, AMS observations and EANET filter measurements during 2000-2020. The white boxes indicate the seasonal mean of the total PM concentration (in µg m^{-3}), the numbers in the respective pie slices indicate the relative contribution of each element (in %).

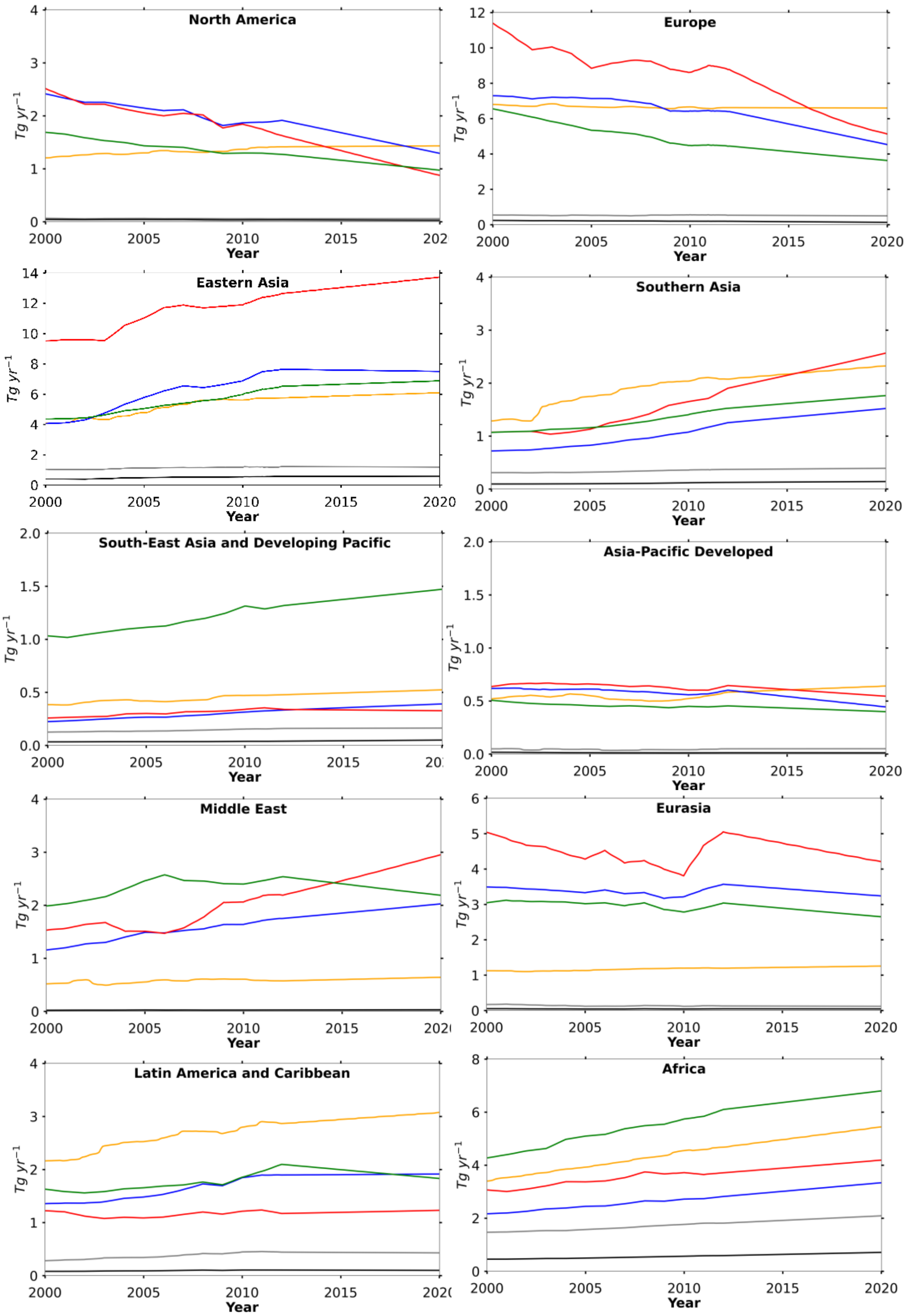

Figure S5. Temporal evolution of pollutant emissions in Tg yr^{-1} of OC (grey), BC (black), NH_3 (orange), NO_x (blue), SO_2 (red), and VOC (green) during the period 2000-2020 in the 10 regions considered according to WGIII AR6.

Table S1: Number of observation sites and datasets per subcontinent and region type for PM_1 and OA factor analysis.

	#Sites of PM_1 data	#PM_1 datasets	#Sites of OA factor analysis	#OA datasets
Europe	58	431	46	130
Rural	29	240	25	84
Downwind	17	155	13	24
Urban	12	36	8	22
North-America	35	88	27	68
Rural	17	46	12	35
Downwind	5	7	4	6
Urban	13	35	11	27
Eastern Asia	48	153	45	148
Rural	17	44	17	37
Downwind	11	16	10	16
Urban	20	93	18	95
Latin America & Caribbean	10	18	8	14
Rural	5	7	2	2
Downwind	2	4	3	5
Urban	3	7	3	7
Africa	3	14	3	5
Rural	1	12	1	3
Downwind	1	1	1	1
Urban	1	1	1	1
Southern Asia	5	19	5	19
Rural	1	5	1	5
Downwind	0	0	0	0
Urban	4	14	4	14
South-East Asia & Developing Pacific	3	4	1	2
Rural	2	2	0	0
Downwind	0	0	0	0
Urban	1	2	1	2
Asia-Pacific Developed	7	17	5	12
Rural	5	13	4	10
Downwind	1	1	0	0
Urban	1	3	1	2
Total	169	744	140	398

Table S2: Percentage change (%) in pollutant emissions between the 2000s and 2010s.

	OC	BC	NH_3	NO_x	SO_2	ARO	ALK	OLE	C5H8	TERP	VOC
Southern Asia	27	49	81	112	140	58	93	45	27	129	65
Europe	-9	-45	-3	-38	-55	-47	-44	-41	-52	4	-45
Africa	42	56	60	54	37	61	40	86	76	16	59
South-East Asia & Developing Pacific	30	51	36	75	27	39	43	53	44	99	43
Latin America & Caribbean	53	23	42	41	1	13	12	10	24	127	12
Eurasia	-30	-11	12	-7	-17	-22	-1	-19	10	179	-13
Asia-Pacific Developed	2	-30	23	-28	-14	-11	-29	-30	-28	-13	-12
Middle East	26	50	24	75	93	14	9	7	-3	119	10
North America	-12	-38	19	-47	-65	-36	-45	-59	-73	-4	-42
Eastern Asia	14	48	50	85	44	99	40	10	112	244	58

Table S3: Statistical evaluation of EMAC PM1 components against AMS and ACSM datasets over North America, Europe, Eastern Asia, Southeast Asia and developing Pacific, and Asia Pacific developed, Latin America and Caribbean, and Africa, during 2000–2020.

		Number of datasets	**Mean observed** [μg m^{-3}]	**Mean predicted** [μg m^{-3}]	**MAGE** [μg m^{-3}]	**MB** [μg m^{-3}]	**NME** [%]	**NMB** [%]	**RMSE** [μg m^{-3}]	**R**	R^2
North America	OA	86	4.77	4.56	3.05	-0.2	64.1	-4.24	4.29	-1.09	0.21
	SO_4^{2-}	88	1.63	1.18	0.85	-0.45	52.37	-27.72	1.27	0.14	0.55
	NO_3^-	88	1.07	1.1	0.94	0.04	87.79	3.35	1.45	0.07	0.36
	NH_4^+	87	0.81	0.67	0.51	-0.14	62.97	-17.29	0.71	-0.09	0.28
Europe	OA	442	4.59	2.18	2.73	-2.41	59.54	-52.56	3.95	-0.22	0.49
	SO_4^{2-}	431	1.54	0.91	0.79	-0.63	51.08	-41.17	1.22	-0.03	0.5
	NO_3^-	431	2.07	1.98	1.09	-0.09	52.54	-4.17	1.74	0.38	0.62
	NH_4^+	431	1.03	0.93	0.44	-0.1	42.33	-9.38	0.68	0.36	0.61
Eastern Asia	OA	159	19.3	13.64	9.41	-5.65	48.74	-29.3	13.19	0.16	0.6
	SO_4^{2-}	153	8.54	4.52	4.44	-4.02	52.05	-47.12	6.47	-0.39	0.41
	NO_3^-	152	8.47	9.12	3.58	0.65	42.3	7.67	4.81	0.3	0.59
	NH_4^+	152	5.99	4.21	2.63	-1.78	43.96	-29.76	3.79	0.08	0.53
Latin America & Caribbean	OA	21	8.39	5.51	5.46	-2.89	65.04	-34.41	8.12	-0.05	0.32
	SO_4^{2-}	19	1.21	0.84	0.44	-0.37	36.29	-30.40	0.59	0.70	0.90
	NO_3^-	19	1.43	0.70	0.91	-0.73	63.78	-50.91	1.93	0.12	0.50
	NH_4^+	19	0.95	0.37	0.58	-0.57	61.46	-60.68	1.11	0.11	0.60
Africa	OA	14	3.42	5.51	2.41	2.09	70.34	61.04	3.30	-1.37	0.54
	SO_4^{2-}	14	1.93	1.23	1.06	-0.71	55.12	-36.55	1.36	-0.31	0.31
	NO_3^-	14	0.42	0.80	0.43	0.38	103.78	91.22	0.67	-4.45	0.82
	NH_4^+	14	0.80	0.48	0.47	-0.32	58.86	-39.59	0.54	-1.28	0.30
South Asia & Developing Pacific	OA	23	33.26	18.59	19.24	-14.67	57.85	-44.10	28.99	0.31	0.79
	SO_4^{2-}	23	7.01	4.02	4.28	-2.99	61.07	-42.66	5.48	-0.12	0.46
	NO_3^-	22	5.39	5.21	3.10	-0.17	57.56	-3.23	4.53	0.57	0.75
	NH_4^+	22	5.24	2.76	3.42	-2.48	65.36	-47.39	5.25	0.13	0.61
Asia - Pacific Developed	OA	17	5.80	2.54	3.40	-3.26	58.64	-56.14	4.64	-0.28	0.61
	SO_4^{2-}	17	4.84	2.30	2.66	-2.53	54.96	-52.35	3.46	-0.45	0.60
	NO_3^-	17	0.79	1.53	1.16	0.74	146.03	92.84	1.40	-1.04	0.22
	NH_4^+	17	1.58	0.79	0.89	-0.80	56.35	-50.29	1.05	-1.48	0.37

Table S4: North America: Rural and remote regions

Location	Coordinates	Dates	# datasets	References
Yorkville, GA	33.9 N 85.05 W	Jul '12, Dec '12 – Jan '13	2	Xu et al. (2015)
Centreville, AL	32.94 N 87.18 W	Jun '13 -Jul' 13	1	Xu et al. (2015)
Blodgett-Forest, CA	38.895 N 120.634 W	Aug '07 – Sep '07	2	Q. Zhang and Jimenez, Worton et al. (2011), Farmer et al. (2011)
Duke-Forest, NC	35.98 N 79.09 W	Sep '04	1	Q. Zhang and Jimenez, Jimenez et al. (2009), Stroud et al. (2007)
Manitou-Forest,CO	39.08 N 105.07 W	Jul '11- Aug '11	2	Q. Zhang and Jimenez, Palm et al. (2016)
Nova-Scotia, CAN	43.76 N 66.1 W	Jul '04 - Aug '04	2	Q. Zhang and Jimenez, Jimenez et al. (2009), Holzinger et al. (2007)
Pinnacle-State Park, NY	42.09 N 77.21 W	Jul '04	1	Q. Zhang and Jimenez, Jimenez et al. (2009), Bae et al. (2007)
Point Reyes, CA	38.07 N 122.81 W	Jul '05	1	Q. Zhang and Jimenez, Ervens et al. (2010)
Southern-Great - Plains (SGP), OK	36.605 N 97.485 W	Nov '10 - Jun '12	20	Q. Zhang and Jimenez, Parworth et al. (2015)
Storm-Peak, CO	40.5 N 106.7 W	Apr '04	1	Q. Zhang and Jimenez, Richardson et al. (2007), Tsimpidi et al. (2016)
Trinidad Head, CA	41.054 N 124.151 W	Apr '02 - May '02	2	Q. Zhang and Jimenez, Millet et al. (2004)
Mt. Whistler, CAN, BC	50.01 N 122.95 W	Apr '06 - May '06	2	Q. Zhang and Jimenez, Sun et al. (2009)
Whiteface Mt., NY	44.35 N 73.85 W	Jul '02 - Aug '02	2	Q. Zhang and Jimenez, Hogrefe et al. (2004)
Rocky Mountain Nat. Park, CO	40.278 N 105.545 W	Jul '10	1	Schurman et al. (2015a)
Look Rock, TN	35.633 N 83.942 W	Feb '13 – Dec '13	4	Budisulistiorini et al. (2016)
Durham, NH	43.11 N 70.95 W	Jul '04 - Aug '04	1	Cottrell et al. (2008)
Peaked Mt., WY	43.793 N 110.959 W	Aug '11	1	Schurman et al. (2015 b)

Table S5: North America: Urban regions

Location	Coordinates	Dates	# datasets	References
Atlanta, Georgia Tech, GA	33.779 N 84.396 W	Jul '12 – Aug '12, Feb '13	2	Xu et al. (2015)
Atlanta, Jefferson Street (JST), GA	33.778 N 84.417 W	Aug '11 – Dec '12, Mar '14 – Feb '15	14	Budisulistiorini et al. (2013, 2014, 2016), Rattanavaraha et al. (2017), Xu et al. (2015)
Las Vegas, NV	36.177 N 115.196 W	Jan '08	1	Brown et al. (2012)
Boulder, CO	40.0 N 105.253 W	Jun '03	1	Q. Zhang and Jimenez, Jimenez et al. (2009), Nemitz et al. (2008)
Fresno, CA	36.81 N 119.778 W	Jan '10, Dec '14 - Jan '15	3	Q. Zhang and Jimenez, Ge et al. (2012 a, b), Collier et al. (2018).
Houston, TX	29.8 N 95.4 W	Aug '00 - Sep '00	2	Q. Zhang and Jimenez, Jimenez et al. (2009)
New York City, NY	40.736 N 73.822 W	Jul '01, Jan '04, Jul '09 – Sep '09	4	Q. Zhang and Jimenez, Jimenez et al. (2009), Drewnick et al. (2004), Weimer et al. (2006), Sun et al. (2011), Ng et al. (2011)
Pasadena, CA	34.141 N 118.122 W	May '10 - Jun '10	2	Q. Zhang and Jimenez, Hayes et al. (2013), Thompson et al. (2012)
Pittsburgh, PA	40.45 N 79.95 W	Sep '02	1	Q. Zhang and Jimenez, Jimenez et al. (2009), Zhang et al. (2005)
Riverside, CA	33.972 N 117.323 W	Jul '05 - Aug '05	2	Q. Zhang and Jimenez, Docherty et al. (2011)
Slocan Park, Vancouver, CAN	49.244 N 123.048 W	Aug '01	1	Boudries et al. (2004), Jimenez et al. (2009)
Langley Vancouver, CAN	49.05 N 122.606 W	Aug '01	1	Boudries et al. (2004)
Yakima, WA	46.589 N 120.528 W	Jan '13	1	Bottenus et al. (2018)

Table S6: North America: Urban and industrial downwind regions

Location	Coordinates	Dates	# datasets	References
Cool, CA	38.883 N 121.0 W	Jun ’10	1	Q. Zhang and Jimenez, Setyan et al. (2012)
Upton, NY	40.871 N 72.89 W	Jul ’11 - Aug ’11	2	Q. Zhang and Jimenez, Zhou et al. (2016), Sedlacek III et al. (2012)
Davis, CA	38.54 N 121.78 W	Jan ’11	1	Q. Zhang and Jimenez
Great Smoky Mountains site, TN	35.674 N 83.942 W	Jul ’13	1	Budisulistiorini et al. (2015)
East Coast Cruise, US	37.95 N 74.3 W	Jul ’02 - Aug ’02	2	Q. Zhang and Jimenez, Tsimpidi et al. (2016), De Gouw et al. (2005)

Table S7: Europe: Rural and remote regions

Location	Coordinates	Dates	# data	References
Mt. Åreskutan, SE	63.43 N, 13.08 E	Jul ’03	1	Drewnick et al. (2007)
Birkenes, NO	58.389 N, 8.252 E	Aug ’12 - Dec ’12, Oct ’19 - Dec ’19	8	EBAS/NILU
Hyltemossa, SE	56.098 N, 13.419 E	Oct ’17 - Dec ’17, Jan ’20 - May ’20	8	EBAS/NILU
Vavihill, SE	56.017 N, 13.15 E	Oct ’08 - Nov ’08, Mar ’09	3	EBAS/NILU, Tsimpidi et al. (2016)
Mace Head, IE	53.326 N, 9.887 W	Aug ’02, Jun ’06, May ’08, Jun ’08, Mar ’09	5	EBAS/NILU, Q. Zhang and Jimenez, Dall’Osto et al. (2010), Coe et al. (2006), Crippa et al. (2014), Tsimpidi et al. (2016)
Chilbolton, UK	51.145 N, 1.44 W	Dec ’08 - Mar ’09	4	Q. Zhang and Jimenez, Crippa et al. (2014)
K-Puszta, HU	46.96 N, 19.58 E	Sep ’08 - Oct ’08	2	Q. Zhang and Jimenez, Crippa et al. (2014)
Aukštaitija, LT	55.46 N, 26.0 E	Aug ’13, Apr ’16 - Sep ’16	3	Pauraite et al. (2019 a, b)
Finokalia, Crete, EL	35.33 N, 25.66 E	Aug ’01, May ’08, Mar ’09, Aug ’12 – Sep ’12	4	Schneider et al. (2004),Q. Zhang and Jimenez, Crippa et al. (2014), Pikridas et al. (2010), Hildebrandt et al. (2010), Bougiatioti et al. (2014)
Island of Lampedusa, IT	35.518 N, 12.631 E	Jun ’13	1	Mallet et al. (2019)
Cabauw, NL	51.97 N, 4.926 E	May ’08, Mar ’09, Jul ’12 – May ‘13	13	Crippa et al. (2014), Mensah et al. (2011), Schlag et al. (2016)
Melpitz,DE	51.53 N, 12.934 E	Jan ’07 - Feb ’07, May ’08 - Oct ’08, Mar ’09, Jun ’14 – Nov ‘17	45	EBAS/NILU, Q. Zhang and Jimenez, Crippa et al. (2014), Poulain et al. (2011)
Puy-de-Dôme, FR	45.767 N, 2.95 E	Sep ’08 - Oct ’08, Mar ’09, Jun ’10, Apr ’15 - Feb ’16, Jul ’16 - Oct ’16, Jan ’18 - Dec ’19	37	Q. Zhang and Jimenez, Tsimpidi et al. (2016), Freney et al. (2011), Farah et al. (2021) EBAS/NILU
Po Valley (Ispra), IT	45.8 N, 8.633 E	Mar ’13 – Feb ’14	4	Bressi et al. (2016)
Jungfraujoch, CH	46.548 N, 7.984 E	May ’08, Aug ’12 – Sep ’13	6	Lanz et al. (2010)
Magadino, CH	46.15 N, 8.92 E	Sep ’13 – Oct ‘14	5	Chen et al. (2021)
Montsec, ES	42.05 N, 0.717 E	Jul ’11 – Sep ’11, Jan ’12 – Mar ’12	2	Ripoll et al. (2015)
Cape Corse Corsica, FR	42.969 N, 9.38 E	Jun ’13, Jul ’13	2	Arndt et al. (2017), Michoud et al. (2017)
Montseny national park, ES	41.779 N, 2.36 E	Feb ’09 - Mar ’09, Jun ’12 – Jul ’13	15	Q. Zhang and Jimenez, Crippa et al. (2014), Minguillón et al. (2011, 2015)
Capo Granitola, Sicily, IT	37.58 N, 12.66 E	Jul ’13	1	Rinaldi et al. (2017)
Preila, LT	55.917 N, 21.067 E	Mar ’14	1	Ulevicius et al. (2016)
Cyprus, CY	35.03 N, 33.05 E	Mar ’15	1	Debevec et al. (2017)
Mt. Cimone, IT	44.183 N, 10.7 E	Jun ’12 – Jul ‘12	1	Rinaldi et al. (2015)
Hyytiälä, FI	61.85 N, 24.283 E	Mar ’03 –Apr ’03, Apr ’05, May ’08, Sep ’08 – Oct ’08, Mar ’09 –Sep ’08, Jul ’10 – Jan ’11, Apr ’12 –Aug ’13, Feb ’14 – Oct ’14, Aug ’16 –Dec ’16, May ’17 – Jul ’18, Feb ’19 – Apr ‘20	68	Q. Zhang and Jimenez, EBAS/NILU, Allan et al. (2006), Raatikainen et al. (2010), Raatikainen et al. (2010), Äijälä et al. (2019), Corrigan et al. (2013), Heikkinen et al. (2020, 2021)
Payerne, CH	46.813 N, 6.945 E	Jun ’06, Jan ‘07 – Feb ’07, Sep ’08 – Oct ’08, Mar ‘09	6	EBAS/NILU, Lanz et al. (2010), Crippa et al. (2014)
Harwell, UK	51.567 N, 1.317 W	Sep ’08 – Oct ‘08	1	Crippa et al. (2014)
Massongex, CH	46.243 N, 6.135 E	Nov ’06 – Dec ‘06	1	Perron et al. (2010), Lanz et al. (2010)
HÄrkingen, CH	40.74 N, 7.82 E	May ’05	1	Lanz et al. (2010)
Reiden, CH	47.25 N, 7.95 E	Feb ’06	1	Lanz et al. (2010)

Table S8: Europe: Urban regions

Location	Coordinates	Dates	# datasets	References
Edinburgh, UK	55.955 N 3.184 W	Nov ’00	1	Q. Zhang and Jimenez, Jimenez et al. (2009), Allan et al. (2003)
Manchester, UK	53.476 N 2.234 W	Jun ’01, Jan ’02	2	Q. Zhang and Jimenez,Tsimpidi et al. (2016), Allan et al. (2003)
Zürich, CH	47.378 N 8.531 E	Jul ’05, Jan ’06, Jun ’16 – Jul ‘16	3	Lanz et al. (2007, 2008, 2010), Tsimpidi et al. (2016), Stefenelli et al. (2019)
Grenoble, FR	45.19 N 5.71 E	Jan ’09	1	Favez et al. (2010), Lanz et al. (2010)
Mainz, DE	50.0 N 8.2 E	Sep ’04	1	Q. Zhang and Jimenez, Vester et al. (2007)
Athens, EL	37.97 N 23.72 E	Dec ’13 – Feb‘14	3	Fourtziou et al. (2017)
Creil, FR	49.26 N 2.47 E	Mar ’15	1	Petit et al. (2017)
Metz, FR	49.11 N 6.22 E	Mar ’15	1	Petit et al. (2017)
Lyon, FR	45.76 N 4.85 E	Mar ’15	1	Petit et al. (2017)
Paris, FR	48.829 N 2.359 E	Jul ’09, Jan ’10 – Feb ‘10	2	Freutel et al. (2013), Fountoukis et al. (2015)
Marseille, FR	43.306 N 5.394 E	Jul ’08, Jan ’11 – Feb ’11, Feb ’17 - Apr ’18	16	Bozzetti et al. (2017), Haddad et al. (2013), Chazeau et al. (2021)
Barcelona, ES	41.387 N 2.116 E	Mar ’09, Aug ’13, Jun ’14 – May ’15, Nov ’17 – Oct ’18	10	Mohr et al. (2012), Minguillón et al. (2016), Via et al. (2021)

Table S9: Europe: Urban and industrial downwind regions

Location	Coordinates	Dates	# datasets	References
Helsinki, FI	60.2 N 24.95 E	Feb ’09 - May ’09	4	Q. Zhang and Jimenez, [m], Crippa et al. (2014), Timonen et al. (2013)
Po Valley, IT	44.65 N 11.617 E	Apr ’08	1	Q. Zhang and Jimenez, Saarikoski et al. (2012)
Taunus, DE	50.224 N 8.449 E	Jul ’04	1	Q. Zhang and Jimenez, Jimenez et al. (2009), Dusek et al. (2006), Hings et al. (2007)
Bush, UK	55.859 N 3.205 W	Jun ’06	1	EBAS/NILU, [n]
Chelmsford, UK	51.737 N 0.429 E	Aug ’03	1	Q. Zhang and Jimenez, Jimenez et al. (2009), Cubison et al. (2006)
Weybourne, UK	52.951 N 1.122 E	May ’04	1	Q. Zhang and Jimenez, Tsimpidi et al. (2016), Gysel et al. (2007)
Roveredo, CH	46.239 N 9.12 E	Mar ’05, Dec ’05	2	Alfarra et al. (2007), Lanz et al. (2010)
Livry-Gargan, GOLF, Paris, FR	48.934 N 2.547 E	Jul ’09, Jan ’10 – Feb ‘10	2	Freutel et al. (2013), Fountoukis et al. (2015)
Palaiseau, SIRTA Paris, FR	48.709 N 2.159 E	Jul ’09, Jan ’10 – Feb ’10, Feb’12 – Dec ’17, Jan ’19 - Apr ’20	37	Freutel et al. (2013), Crippa et al. (2013), Fountoukis et al. (2015), Petit et al. (2014), Zhang et al. (2019b), EBAS/NILU, [o]
Villeneuve, FR	50.611 N 3.14 E	Oct ’16 – Nov ’17, Jul ’18 – Apr ’20	35	EBAS/NILU, [p]
Dunkirk Harbour, FR	51.052 N 2.354 E	Jul ’13 – Sep ‘14	5	Zhang et al. (2021)
Prague-Suchdol, CZ	50.127 N 14.385 E	Jun ’12 – Jul ’12, Jan’13 – Feb ‘13	2	Kubelová et al. (2015)
Bucharest, RO	44.35 N 26.03 E	Jun ’12 – Jul ’12, Jan ’13, Aug ’16 - Feb ’18	21	Marmureanu et al. (2020), EBAS/NILU, [q]
Patras, EL	38.298 N 21.809 E	Jun ’12	1	Kostenidou et al. (2015)
Athens, Agia Paraskevi, EL	37.995 N 23.816 E	Jul ’12	1	Kostenidou et al. (2015)
Hohenpeißenberg, DE	47.8 N 11.03 E	May ’02, Mar ’15, May ’15 – Oct ’15, Jan ’17 - Oct ’18, Feb‘ 19 - Dec ’19	40	Q. Zhang and Jimenez, Lanz et al. (2010) Hock et al. (2008), EBAS/NILU, [r], [s]
Puijo, Kuopio, FI	62.902 N 27.65 E	Oct ’08	1	Crippa et al. (2014), Leskinen et al. (2012)

Table S10: Eastern Asia: Rural and remote regions

Location	Coordinates	Dates	# datasets	References
China				
Gucheng, NCP, CN	39.15 N 115.73 E	Nov ’18	2	Sun et al. (2020 b)
Daban Mt., Menyuan, Tibet	37.608 N 101.257 E	Sep ’13	2	Du et al. (2015)
Nam Co, Tibet	30.767 N 90.95 E	Jun ’15	1	Xu et al. (2018a)
Mt. Everest, Tibet	28.36 N 86.95 E	Apr ’16 - May ’16	1	Zhang et al. (2018a)
Mt. Waliguan, Tibet	36.28 N 100.9 E	Jul ’17	1	Zhang et al. (2019)
Mt. Yulong, Tibet	27.2 N 100.2 E	Mar ’15 - Apr ’15	1	Zheng et al. (2017)
Lake Hongze, YRD	33.23 N 118.33 E	Mar ’11 - Apr ’11	1	Zhu et al. (2016)
Mt. Wuzhi, Hainan	18.84 N 109.49 E	Mar ’15 - Apr ’15	1	Zhu et al. (2016)
Mt. Tai, NCP	36.251 N 117.101 E	Jun ’10 - Oct ’10, Apr ’11, Jun ’11, Oct ’11 - Jan ’12	6	Zhang et al. (2014b)
Lin’an, YRD	44.285 N 119.73 E	Nov ’11 - Dec ’11, Mar ’13	2	Zhang et al. (2015b) Zhang et al. (2015e)
Back Garden, PRD	23.49 N 113.03 E	Jul ’06	1	Xiao et al. (2011)
Kaiping, PRD	22.32 N 112.53 E	Oct ’08 - Nov ’08	2	Huang et al. (2011)
Liulihe, NCP	39.6 N 116.03 E	Oct ’14 - Nov ’14 Apr ’15 Aug ’15 - Jan ’16	4	Hua et al. (2018)
Dianshan Lake, YRD	31.097 N 120.989 E	Jul ’15 - Jun ’16	12	Zhao et al. (2020)
South Korea				
Boseong	34.76 N 127.21 E	Nov ’12	1	Lee et al. (2017)
Baengnyeong Island	37.967 N 124.63 E	Oct ’01, May ’11 - Nov ’11	7	Lee et al. (2015)
Jeju-Island	33.51 N 126.5 E	Apr ‘01	1	Topping et al. (2004)

Table S11: Eastern Asia: Urban regions

Location	Coordinates	Dates	# data	References
China				
Shanghai, Puding EMS	31.23 N 121.53 E	May '10 - Jun '10	1	Huang et al. (2012)
Shanghai, Xuhui district	31.10 N 121.25 E	Dec '16 - Jan '17	1	Zhu et al. (2018)
Lanzhou (CAREERI), NW	36.05 N 103.862 E	Jul '12 - Aug '12	1	Xu et al. (2014)
Lanzhou (University), NW	36.05 N 103.85 E	Jan '14	1	Xu et al. (2016)
Lanzhou (Gansu MA), NW	36.04 N 103.879 E	Nov '14	1	Zhang et al. (2017)
Nanjing downtown, YRD	32.05 N 118.767 E	Jun '13, Oct '13, Dec '13	3	Zhang et al. (2015c), Zhang et al. (2015d)
Nanjing, Jiangxin Island, YRD	32.03 N 118.7 E	Aug '14 - Sep '14	1	Ge et al. (2017)
Nanjing Olympic center, Jiangxin Island, YRD	32.009 N 118.736 E	Apr '15	1	Wang et al. (2016 b)
Nanjing NUIST, YRD	32.206 N 118.707 E	Feb '15 - Mar '15	1	Ge et al. (2017), Zhou et al. (2020), Wang et al. (2016 a)
Beijing, CAMS, NCP	39.981 N 116.325 E	Jul '06	1	Sun et al. (2010), Tsimpidi et al. (2016)
Beijing, CMA, NCP	39.949 N 116.323 E	Jan '08, Apr '08, Jun '08, Oct '08	4	Zhang et al. (2013)
Peking University Campus, NCP	39.99 N 116.31 E	Jul '08 - Sep '08, Nov '10 - Dec '10, Aug '11 - Sep '11, Apr '12 - Feb '14, Jul '15 - Nov '15	11	Huang et al. (2010), Hu et al. (2016a) Hu et al. (2017), Li et al. (2018a) Duan et al. (2019)
Beijing, Institute of Atmospheric Physics (IAP), NCP	39.974 N 116.371 E	Jul '11 - May '13, Dec '13 - Jan '14, Jun '14 - Mar '15, Sep '15, Nov '16 - Dec '16, Jun '17, Jan '18 - Jun '18, Nov '18 - Mar '19, Jan '20 - Mar '20	47	Sun et al. (2015), Sun et al. (2018), Jiang et al. (2013), Jiang et al. (2015), Xu et al. (2018b), Zhang et al. (2014a), Sun et al. (2020 a), Sun et al. (2016 b), Zhang et al. (2015a; 2016), Zhou et al. (2020), Zhang et al. (2016), Xu et al. (2017;2019a, b), Zhao et al. (2017), Li et al. (2020 b)
Hangzhou, YRD	30.208 N 120.212 E	Aug '16 - Sep '16	2	Li et al. (2018b)
Shenzhen, University Campus, PRD	22.591 N 113.975 E	Jan '09 - Feb '09, Oct '09 - Nov '09, Aug '11 - Sep '11, Jan '15	4	Huang et al. (2010), He et al. (2011) Gong et al. (2013), Cao et al. (2018)
Donguan, PRD	23.0 N 113.7 E	Dec '13 - Jan '14	1	Lan et al. (2018)
Hong Kong, Monkok, PRD	22.322 N 114.169 E	Sep '13 - Dec '13, Apr '17	4	Sun et al. (2016), Niu et al. (2021)
Xi'an, NW	34.229 N 108.887 E	Dec '12	1	Wang et al. (2014)
Baoji, NW	34.355 N 107.143 E	Mar '14	1	Wang et al. (2017)
Tianjin, NCP	39.2 N 117.1 E	Sep '10	1	Zhang et al. (2012)
Xinxian, NCP	35.3 N 113.9 E	Jun '17	1	Li et al. (2018a)
Handan, NCP	36.57 N 114.5 E	Dec '15 - Jan '16	1	Li et al. (2017)
Shijiazhuang, NCP	38.034 N 114.541 E	Jan '14 - Feb '14	1	Huang et al. (2019)
Ziyang, Sichuan Basin	30.15 N 104.64 E	Dec '12	1	Hu et al. (2016b)
South Korea				
Seoul (KIST)	37.6 N 127.05 E	Dec '15 - Jun '16, Feb '19 - Mar '19	3	Kim et al. (2017), Kim et al. (2018) Kim et al. (2020)
Gwangju GIST campus	35.217 N 126.833 E	Sep '11, Dec '11, May '12, Jul '12, Nov' 13, May '15 - Jun '15	6	Park et al. (2013), Lee et al. (2017), Cho et al. (2018)

Table S12: Eastern Asia: Urban and industrial downwind regions

Location	Coordinates	Dates	# datasets	References
China				
Xinglong, NCP	40.4 N 117.667 E	Nov ’18 - Jan ’19	1	Li et al. (2020 b)
Changping campus of Peking University, NCP	40.14 N 116.11 E	Jun ’16	1	Li et al. (2019)
Jianxing, YRD	30.8 N 120.8 E	Jul ’10, Dec ’10	2	Huang et al. (2013)
Heshan, PRD	22.711 N 112.927 E	Nov ’10	1	Gong et al. (2012)
Panyu, PRD Dazhengang Mt.	23.0 N 113.35 E	Nov ’14 - Dec ’14	1	Qin et al. (2017)
Hong Kong, Clear Water Bay, PRD	22.338 N 114.268 E	May ’11, Sep ’11 - Feb ’12 Oct ’16	5	Li et al. (2015), Li et al. (2019)
Xianghe, NCP	39.798 N 116.958 E	Jun ’13	1	Sun et al. (2016 b)
Xinzhou, NCP	38.07 N 112.12 E	Jul ’14 - Aug ’14	1	Wang et al. (2016)
Xingtai, Xinfuqu NMO, NCP	37.18 N 114.37 E	May ’16 - Jun ’16	1	Zhang et al. (2018b)
Yufa, Huang Pu University, NCP	39.515 N 116.305 E	Aug ’06 - Sep ’06	1	Gunthe et al. (2011)
Changdao Island, NCP	37.99 N 120.7 E	Mar ’11 - Apr ’11	1	Hu et al. (2013)

Table S13: South Asia

Location	Coordinates	Dates	# datasets	References
Rural and remote areas of India				
Mount Ghats	17.92 N 73.65 E	Jun ’15, Mar ’16 - Feb ’17	5	Singla et al. (2017), Mukherjee et al. (2018)
Urban areas of India				
Kanpur, campus IIT	26.46 N 80.33 E	Nov ’12 - Apr ’13 Dec ’14 - Aug ’15, Sep ’16 - Oct ’16	6	Chakraborty et al. (2015), Kumar et al. (2016), Chakraborty et al. (2016a), Zhou et al. (2020), Chakraborty et al. (2016b; 2018)
Ahmedabad	23.0 N 72.6 E	Sep ’17 - Oct ’17	1	Singh et al. (2019)
Delhi, IITD campus South	28.545 N 77.193 E	Dec ’16 - Sep ’17	4	Gani et al. (2019), Zhou et al. (2020)
Old Delhi, campus IGDTUW	28.664 N 77.232 E	Jun ’18 - Nov’18	3	Cash et al. (2021)

Table S14: South-East Asia and Developing Pacific

Location	Coordinates	Dates	# datasets	References
Rural and remote areas of Borneo Malaysia				
Sabah Oil Palm plantage, Borneo, Malaysia	5.25 N 118.454 E	May ’08 - Jun ’08	1	Hewitt et al. (2010)
Sabah rainforest, Borneo, Malaysia	4.98 N 117.844 E	Apr ’08 - Jul ’08	1	Hewitt et al. (2010)
Urban				
Singapore, Nanyang Technological University	1.348 N 103.683 E	Oct ’15	1	Budisulistiorini et al. (2018)
Singapore, National University campus	1.3 N 103.77 E	May ’17 - Jun ’17	1	Rivellini et al. (2020)

Table S15: Asia Pacific Developed

Location	Coordinates	Dates	# datasets	References
Rural and remote areas				
Gunn Point, Northern Australia	12.249 S 131.045 E	Jun ’14	1	Mallet et al. (2017), Milic et al. (2017)
Brisbane, Queens-land, Australia	27.5 S 153.0 E	Sep ’11, Nov ’12	2	He et al. (2016)
Fukue-Island, Japan	32.78 N 128.66 E	Mar ’03 - May ’03	3	Zhang et al. (2007), Q. Zhang and Jimenez Takami et al. (2005), Zhang et al. (2007)
Cape Hedo, Okinawa, Japan	26.87 N 128.25 E	Mar ’03, Oct ’03 - Dec ’03, Apr ’04, Mar ’05	6	Q. Zhang and Jimenez, [t], Zhang et al. (2007), Takami et al. (2007)
Wakayama, Japan	34.07 N 135.52 E	Aug ’10	1	Han et al. (2014)
Urban				
Tokyo, University of Tokyo, Komaba	35.662 N 139.678 E	Jul ’03 - Aug ’03, Jan ’04 - Feb ’04, Aug ’04	3	Q. Zhang and Jimenez,Takegawa et al. (2006) Tsimpidi et al. (2016) Miyakawa et al. (2008)
Urban downwind				
Saitama, Japan	36.083 N 139.55 E	Aug ’04	1	Miyakawa et al. (2008)

Table S16: Latin America & Caribbean

Location	Coordinates	Dates	# datasets	References
Rural and remote regions				
Manaus W, Brazil, Amazonia	3.196 S 60.671 W	Aug ’14 - Oct ’14	1	de Sá et al. (2019)
Manaus NE, Brazil, Amazonia	2.146 S 59.005 W	Aug ’14 - Oct ’14	1	de Sá et al. (2019)
Manaus NW, Brazil, Amazonia	2.595 S 60.209 W	Feb ’08 - Mar ’08, Feb ’13	3	Chen et al. (2009), Q. Zhang and Jimenez
Porto Velho, Brazil Amazonia	8.69 S 63.87 W	Sep ’12	1	Artaxo et al. (2013)
Bird Island, Sub Antarctic	54.0 S 38.05 W	Nov ’10	1	Schmale et al. (2013)
Urban				
Mexico City, Mexico	19.358 N 99.1 W	Apr ’03 - Mar ’06	2	Q. Zhang and Jimenez, Salcedo et al. (2006), Tsimpidi et al. (2016), Aiken et al. (2009)
Santiago de Chile, Chile	33.45 S 70.69 W	Aug ’11 - Nov ’11	4	Q. Zhang and Jimenez, Carbone et al. (2013)
Tijuana, Maxico	32.52 N 117.037 W	May ’10	2	Takahama et al. (2013)
Urban and industrial downwind regions				
Manaus downwind west, Brazil	3.146 S 60.128 W	Aug ’14 - Oct ’14	1	de Sá et al. (2019)
Chacaltaya, Bolivia	16.35 S 68.131 W	Apr ’18 -Mar ’19	4	Mota (2020)
Mexico city, Universidad Tecnologica de Tecamac, Mexico	19.7 N 98.98 W	Mar ’06	1	Tsimpidi et al. (2011)
Pico de Tres Padres, Mexico	19.6 N 99.12 W	Mar ’06	1	Tsimpidi et al. (2011)

Table S17: Africa

Location	Coordinates	Dates	# datasets	References
Rural and remote areas				
Welegund, South Africa	26.569 S 26.939 E	Jun '14	1	Tiitta et al. (2014), Q. Zhang and Jimenez
Urban				
Kigali City, Rwanda	1.959 S 30.065 E	May '18	1	Bimenyimana (2018)
Urban and industrial downwind				
Mbour, Senegal	14.394 N 16.959 W	Mar '15 - Jun '15	1	Rivellini et al. (2017)

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