Tropical tropospheric aerosol sources and chemical composition observed at high-altitude in the Bolivian Andes

Supplementary material

Moreno et. al

--- Sampling site ---

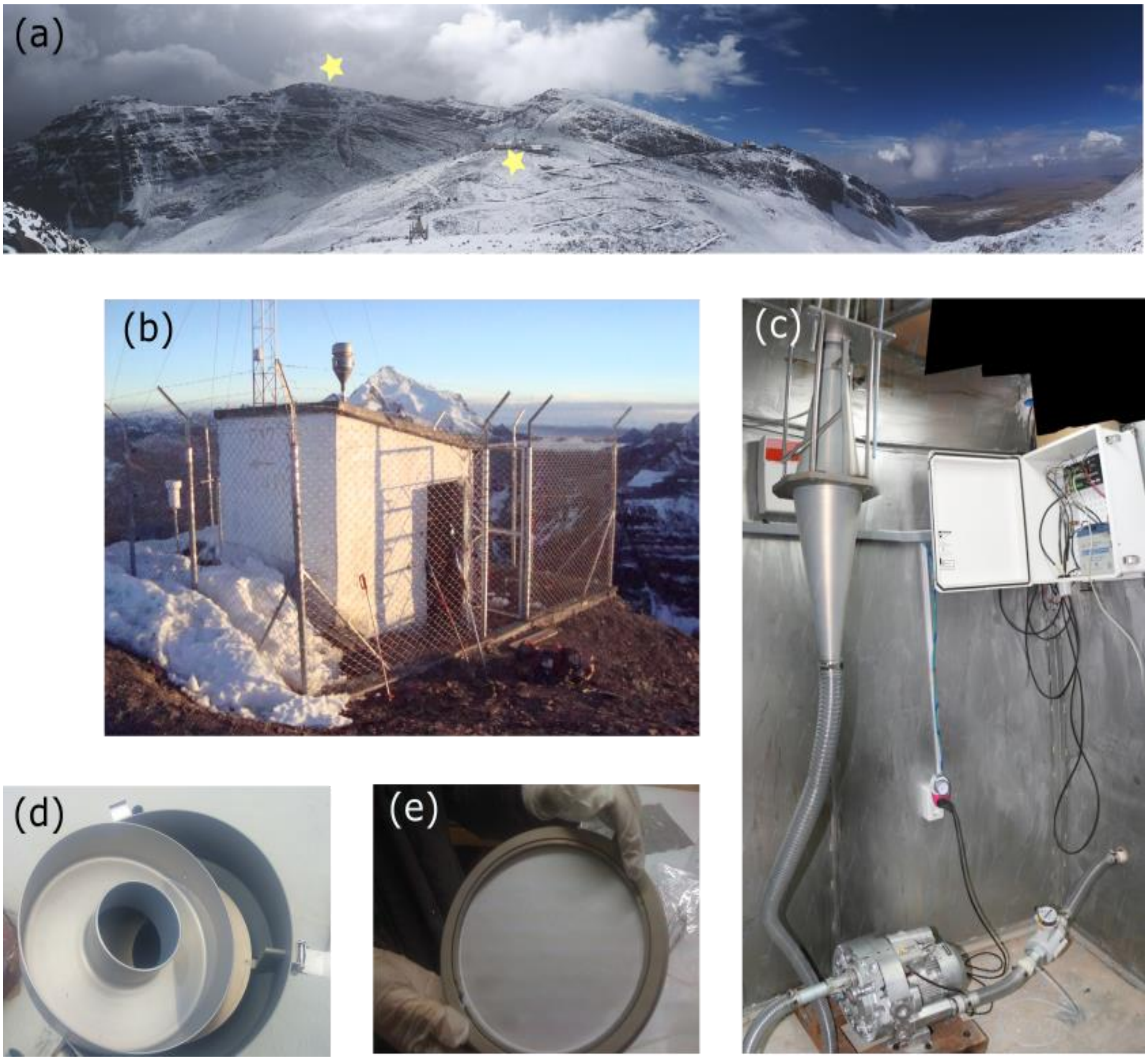

Figure S 1. (a) Measurement site: left star = high-volume sampling site, right star = main GAW-Cosmic Ray observatory (the sampling site for aerosol in the 1970-1980 was located just above the main observatory, not in the summit). (b) Sampling hut with the Digitel head visible. Also meteorological variables are collected at the site (the 10-meter tower partially visible has an anemometer on top) (c) High-Volume sampler system (d) Water-collection system inside the sampling head (missing until 2016) (e) Filter holder with clean filter as set for sampling. Pictures taken by I. Moreno

--- Standard temperature and pressure (STP) correction ---

The volume sampled in CHC was obtained in ambient conditions, 534 hPa, which is nearly half of the pressure at sea level. This implies that the volume sampled at CHC is equivalent to twice the volume sampled at sea-level. For comparison with other sites, the concentration was transformed to standard temperature and pressure (STP) conditions (P=1013.25 hPa, T=273ºK) taking into account the average meteorological conditions (pressure, P_{avg} and temperature, T_{avg}) for each sample (equations S1 and S2). When no meteorological parameters were available, the annual mean pressure and/or temperature were used to correct the volume to standard cubic meters.

$$C_{STP} = C_{CHC} \times F \text{ (equation S1)}$$

In which F mean value was 1.88 (σ = 0.01), ranging from 1.85 to 1.93 and was calculated for each sample following equation 2:

$$F = \frac{1013.25[hPa] \times (T_{avg}[°C] + 273)}{P_{avg}[hPa] \times 273[K]} \text{ (equation S2)}$$

Concentrations are then expressed in standard cubic meters for intercomparison with other sites (and other sites' ambient data were also converted to STP conditions). Note, however, that this extensively used simple correction does not take into account the effect of temperature and pressure changes in volatility of the measured species, and therefore the true reference values remain those measured at the site (Table S3)

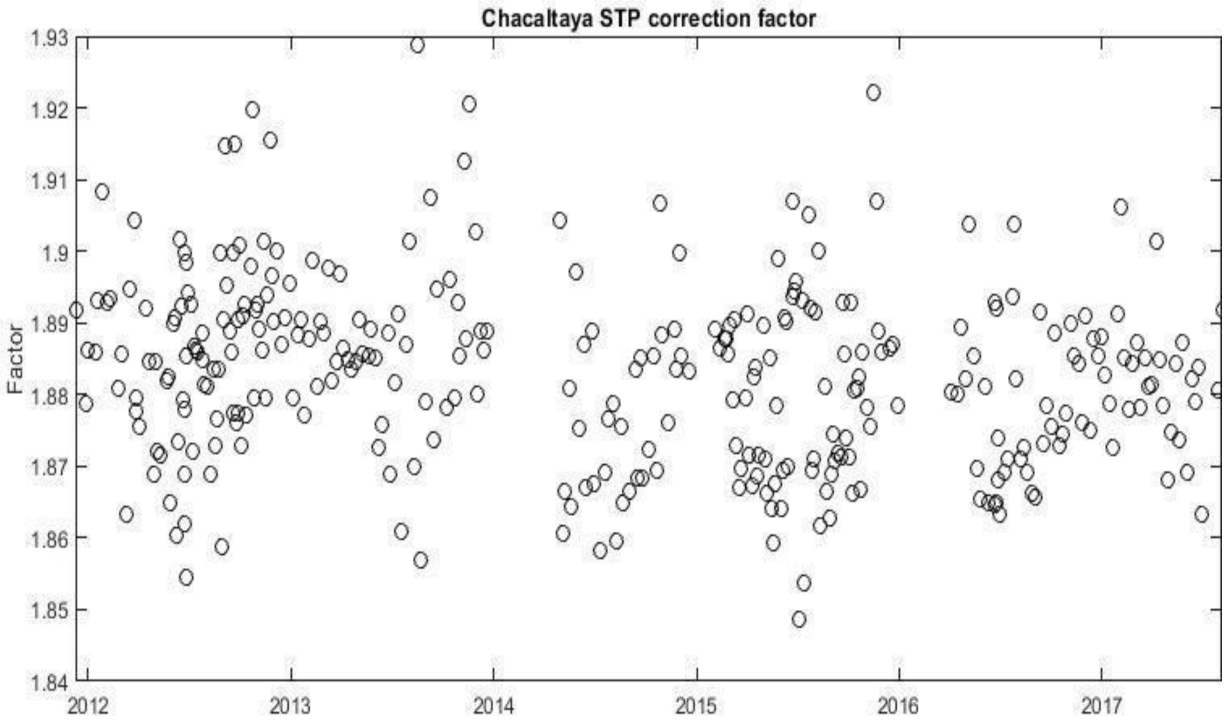

Figure S 2. Variability of STP correction factor for Chacaltaya samples

--- Detection limit and concentration data tables ---

Year (sample number)	2011-2014 (1-171)	2015 (173-248)	2016 (250-270)	2017 (273-289)	2016 (B42-B43)	2017 (B44-B48)	2016 (INHALE)	2019 (291-323)	2020 (324-330)	
Species	**QL**	**QL**	**QL**	**QL**	**QL**	**QL**	**QL**	**QL**	**QL**	**Units**
OC	0.03	0.09	0.08	0.03	0.20	0.20	0.05	0.08	0.02	$\mu g.m^{-3}$
EC	0.002	0.0001	0.003	0.001	0.02	0.02	0	$3x10^{-6}$	0	$\mu g.m^{-3}$
F^-	0.20	0.01	0.15	0.17	NA	NA	0.16	0.02	0.001	$ng.m^{-3}$
HCO_2^-	0.63	3.99	7.60	0.91	NA	NA	NA	0.99	0.03	$ng.m^{-3}$
$MeSO_3^-$	0.04	0.26	0	0.03	0.73	0.06	0.15	0.01	0.004	$ng.m^{-3}$
Cl^-	0.41	1.56	0.80	0.34	13.59	2.33	1.17	0.25	0.15	$ng.m^{-3}$
Br^-	0.10	0.12	0.08	0.09	NA	NA	0.09	0.02	0.03	$ng.m^{-3}$
NO_3^-	0.22	3.43	5.10	0.30	14.60	3.46	41.45	1.17	0.85	$ng.m^{-3}$
SO_4^{2-}	2.39	8.60	4.26	4.18	3.83	3.48	14.91	1.95	0.26	$ng.m^{-3}$
$C_2O_4^{-22-}$	0.21	0.44	0.15	0.16	1.67	1.05	0.84	0.39	0.06	$ng.m^{-3}$
Li^+		0.006			NA	NA	NA	0.03	0.001	$ng.m^{-3}$
Na^+	0.66	3.30	1.47	2.31	14.69	4.99	4.90	0.49	0.38	$ng.m^{-3}$
NH_4^+	0.96	2.88	3.95	1.61	9.81	5.40	6.63	1.06	0.60	$ng.m^{-3}$
K^+	0.39	0.81	0.60	0.34	7.96	1.63	17.76	0.47	0.39	$ng.m^{-3}$
Mg^{2+}	0.07	0.15	0.30	0.17	0.51	0.39	0.42	0.01	0.03	$ng.m^{-3}$
Ca^{2+}	1.49	1.83	1.40	2.21	10.17	2.53	2.29	0.12	0.07	$ng.m^{-3}$
arabitol	0.25	0.53	0.33	0.33	0.77	0.33	0.22	0.22	0.09	$ng.m^{-3}$
sorbitol	0.22	1.94	0.13	0.13	1.27	0.13	NA	1.08	0.04	$ng.m^{-3}$
mannitol	0.19	0.78	0.33	0.33	0.77	0.33	0.22	0.19	0.09	$ng.m^{-3}$
levoglucosan	0.27	0.26	1.65	1.65	0.62	1.65	0.17	0.95	0.46	$ng.m^{-3}$
mannosan	0.05	0.33	0.33	0.33	0.77	0.33	0.22	0.19	0.09	$ng.m^{-3}$
galactosan	0.29	0.13	0.13	0.13	0.31	0.13	0.09	0.08	0.04	$ng.m^{-3}$
glucose	0.51	0.33	0.33	0.33	0.77	0.33	0.22	0.19	0.09	$ng.m^{-3}$

Table S 1. Quantification limit for the analyzed species at Chacaltaya. Data from IGE, Grenoble.

Ambient (534 hPa)		PM_{10}-A December 2011 to July 2013				$PM_{2.5}$ August 2013 to December 2015				PM_{10}-B April 2016 to August 2017				PM_{10}-C February 2019 to March 2020			
ng m^{-3}	Season	mean	**median**	**std**	**N>QL**	mean	**median**	**std**	N>QL	mean	**median**	**std**	N>QL	mean	**median**	**std**	N>QL
OC	annual	483	**371**	(343)	56	466	**354**	(385)	77	423	**320**	(320)	49	321	**280**	(237)	32
	DJFM	253	**208**	(155)	15	259	**214**	(194)	10	196	**185**	(127)	13	164	**153**	(86,8)	11
	A	328	**312**	(60,1)	5	304	**268**	(189)	6	172	**173**	(47,1)	5	271	**255**	(108)	4
	MJJA	530	**449**	(287)	22	552	**430**	(510)	33	483	**420**	(318)	21	447	**375**	(282)	15
	SON	710	**586**	(455)	14	472	**437**	(244)	28	719	**664**	(287)	10	335	**335**	(51,4)	2
EC	annual	49,4	**36,3**	(48,3)	36	42,9	**37,1**	(25,9)	74	41,4	**30,1**	(36,5)	49	35,0	**27,3**	(25,8)	32
	DJFM	22,3	**17,3**	(12,5)	12	27,5	**29,1**	(16,5)	8	19,5	**17,8**	(10,6)	13	20,1	**19,2**	(12,0)	11
	A	39,1	**39,1**	(6,34)	2	39,1	**36,9**	(30,6)	6	20,5	**18,5**	(5,42)	5	35,3	**32,7**	(19,3)	4
	MJJA	37,6	**23,9**	(31,9)	8	47,9	**43,5**	(24,7)	32	46,2	**35,8**	(31,9)	21	46,1	**37,3**	(31,5)	15
	SON	80,9	**60,7**	(61,3)	14	42,4	**33,8**	(27,6)	28	70,0	**51,7**	(52,9)	10	33,0	**33,0**	(1,61)	2
Li+	annual	0,028	**0,020**	(0,038)	48	0,017	**0,014**	(0,018)	52	0,006	**0,002**	(0,012)	19	0,008	**0,007**	(0,006)	27
	DJFM	0,011	**0,005**	(0,012)	10	0,014	**0,014**	(0,003)	2	0,003	**0,002**	(0,004)	4	0,008	**0,007**	(0,004)	9
	A	0,014	**0,014**	(0,009)	2	0,010	**0,010**	(0,001)	2	0,002	**0,002**	(0,001)	4	0,008	**0,006**	(0,006)	4
	MJJA	0,022	**0,021**	(0,012)	22	0,012	**0,009**	(0,008)	26	0,003	**0,002**	(0,003)	8	0,010	**0,007**	(0,008)	12
	SON	0,052	**0,035**	(0,062)	14	0,023	**0,020**	(0,025)	22	0,026	**0,015**	(0,024)	3	0,002	**0,002**	(0,000)	2
Na+	annual	13,5	**11,7**	(11,1)	54	13,5	**10,8**	(8,22)	71	16,0	**12,1**	(14,3)	41	6,44	**4,90**	(5,49)	31
	DJFM	8,44	**9,47**	(5,24)	15	8,81	**7,15**	(5,71)	7	11,4	**12,5**	(6,94)	6	4,37	**4,78**	(3,33)	11
	A	12,1	**14,0**	(6,53)	4	9,81	**8,94**	(5,10)	4	6,32	**5,85**	(2,96)	5	6,28	**6,84**	(3,49)	4
	MJJA	11,2	**8,6**	(8,68)	21	14,1	**13,3**	(8,25)	33	12,7	**11,4**	(6,43)	20	8,59	**5,88**	(6,91)	14
	SON	22,7	**22,0**	(14,8)	14	14,5	**11,4**	(8,84)	27	30,3	**25,4**	(21,8)	10	3,03	**3,03**	(1,11)	2
NH4+	annual	84,0	**76,9**	(56,3)	58	162	**157**	102	78	168	**163**	(104)	49	149	**132**	(85,3)	32
	DJFM	47,4	**42,9**	(41,0)	16	74,0	**30,1**	(98,3)	10	92,6	**58,0**	(77,8)	13	107	**122**	(62,9)	11
	A	83,4	**59,5**	(43,9)	5	186	**187**	(131)	6	128	**133**	(53,7)	5	215	**192**	(124)	4
	MJJA	71,0	**67,5**	(34,8)	23	188	**167**	(103)	34	169	**206**	(83,7)	21	169	**159**	(81,1)	15
	SON	147,3	**151,7**	(55,3)	14	158	**153**	(79,2)	28	283	**293**	(95,3)	10	99,2	**99,2**	(7,49)	2
K+	annual	13,0	**9,49**	(12,2)	54	14,2	**12,5**	(9,54)	77	18,3	**8,3**	(18,2)	47	13,8	**10,7**	(14,1)	31
	DJFM	4,61	**4,64**	(2,58)	13	4,36	**3,42**	(3,05)	9	4,41	**4,24**	(3,69)	11	4,31	**3,27**	(3,46)	10
	A	5,68	**4,58**	(3,73)	5	6,32	**5,84**	(4,84)	6	3,38	**2,67**	(2,33)	5	9,60	**8,51**	(5,10)	4
	MJJA	12,7	**11,0**	(9,03)	22	17,9	**16,1**	(10,4)	34	24,1	**22,0**	(21,2)	21	21,4	**15,1**	(16,7)	15
	SON	23,7	**18,6**	(15,9)	14	14,5	**12,7**	(7,31)	28	28,7	**29,6**	(11,1)	10	12,4	**12,4**	(1,81)	2
Mg2+	annual	3,85	**3,61**	(2,39)	56	4,56	**4,16**	(2,91)	75	3,63	**2,66**	(3,52)	49	3,52	**3,05**	(2,59)	32
	DJFM	2,49	**1,86**	(1,83)	15	1,98	**2,39**	(1,63)	9	1,67	**0,78**	(1,59)	13	2,38	**2,75**	(1,26)	11
	A	2,97	**1,72**	(2,72)	5	2,65	**1,75**	(2,21)	6	2,63	**2,22**	(2,16)	5	3,24	**2,91**	(1,83)	4
	MJJA	3,48	**3,12**	(1,30)	22	5,60	**4,50**	(3,24)	33	3,18	**2,61**	(1,78)	21	4,70	**3,57**	(3,15)	15
	SON	6,21	**6,62**	(2,61)	14	4,58	**4,56**	(2,24)	27	7,62	**5,01**	(5,42)	10	1,61	**1,61**	(0,20)	2
Ca2+	annual	38,9	**35,7**	(23,1)	55	48,8	**40,0**	(42,1)	79	30,9	**27,6**	(23,1)	48	27,6	**19,4**	(22,6)	32
	DJFM	21,0	**16,7**	(16,2)	14	15,3	**11,1**	(12,1)	11	12,8	**7,0**	(11,4)	12	12,2	**9,3**	(8,60)	11
	A	22,1	**18,9**	(15,3)	5	36,1	**31,2**	(27,4)	6	16,7	**20,1**	(11,7)	5	21,5	**22,3**	(9,74)	4
	MJJA	45,4	**43,1**	(17,7)	22	68,9	**59,6**	(52,3)	34	34,0	**31,6**	(16,4)	21	42,2	**42,0**	(24,7)	15
	SON	52,5	**50,4**	(25,6)	14	40,3	**39,2**	(22,7)	28	52,9	**44,1**	(29,2)	10	16,0	**16,0**	(6,32)	2
F-	annual	1,08	**0,78**	(1,22)	49	1,48	**0,94**	(1,31)	76	0,79	**0,60**	(0,62)	42				14
	DJFM	0,49	**0,46**	(0,22)	10	0,48	**0,32**	(0,30)	9	0,30	**0,25**	(0,22)	10		*Fluoride*		5
	A	0,49	**0,42**	(0,25)	5	1,00	**0,62**	(1,18)	6	0,29	**0,24**	(0,10)	3		*lost for*		0
	MJJA	0,84	**0,68**	(0,56)	20	1,92	**1,52**	(1,49)	33	1,01	**0,91**	(0,61)	20		*PM10C*		9
	SON	2,07	**1,47**	(1,86)	14	1,38	**0,94**	(1,11)	28	0,98	**0,90**	(0,68)	9				0
Cl-	annual	4,58	**3,24**	(3,72)	47	6,98	**5,14**	(5,79)	45	5,70	**4,06**	(6,25)	34				25
	DJFM	2,09	**1,69**	(1,37)	11	2,38	**2,38**	-	1	0,83	**0,83**	(0,67)	2		*Chloride*		7
	A	2,93	**2,85**	(0,93)	4	9,20	**9,20**	-	1	1,00	**1,32**	(0,56)	3		*lost for*		3
	MJJA	4,77	**3,58**	(4,10)	22	7,72	**5,62**	(6,62)	28	5,64	**4,73**	(4,37)	21		*PM10C*		14
	SON	7,58	**8,75**	(3,22)	10	5,74	**4,21**	(4,02)	15	8,83	**4,58**	(10,1)	8				1
Br-	annual	0,77	**0,68**	(0,49)	50	0,63	**0,49**	(0,42)	69	0,87	**0,58**	(0,82)	38	0,51	**0,41**	(0,43)	28
	DJFM	0,46	**0,28**	(0,58)	9	0,93	**0,83**	(0,83)	4	0,25	**0,22**	(0,09)	5	0,32	**0,13**	(0,33)	9
	A	0,36	**0,42**	(0,19)	5	0,25	**0,21**	(0,13)	5	0,28	**0,25**	(0,24)	4	0,33	**0,34**	(0,14)	4
	MJJA	0,70	**0,67**	(0,29)	22	0,61	**0,44**	(0,39)	32	0,89	**0,92**	(0,66)	20	0,69	**0,45**	(0,49)	14
	SON	1,23	**1,12**	(0,44)	14	0,67	**0,59**	(0,40)	28	1,43	**1,45**	(1,12)	9	0,38	**0,38**	-	1
NO3-	annual	78,8	**67,6**	(60,4)	58	44,9	**34,7**	(41,9)	76	58,1	**37,6**	(61,2)	45				29
	DJFM	26,1	**16,7**	(26,2)	17	20,3	**15,1**	(17,0)	9	15,6	**7,5**	(17,8)	11		*Nitrate*		8
	A	66,6	**79,8**	(27,2)	5	44,8	**22,6**	(69,2)	6	15,6	**9,7**	(14,2)	4		*lost for*		4
	MJJA	101	**99,7**	(44,7)	22	45,9	**35,4**	(32,8)	33	85,2	**59,8**	(72,4)	21		*$PM_{10}C$*		15
	SON	112	**122**	(77,1)	14	51,5	**41,8**	(48,8)	28	65,5	**48,4**	(40,7)	9				2
SO42-	annual	241	**214**	(175)	58	600	**560**	(425)	79	530	**492**	(407)	52	484	**476**	(267)	32
	DJFM	162	**130**	(147)	16	274	**150**	(370)	10	273	**179**	(243)	14	353	**353**	(190)	11
	A	221	**129**	(148)	5	573	**550**	(457)	6	468	**495**	(269)	5	635	**582**	(322)	4
	MJJA	179	**162**	(89)	23	728	**721**	(477)	35	485	**498**	(326)	23	567	**483**	(276)	15
	SON	441	**425**	(176)	14	562	**494**	(299)	28	1026	**1067**	(424)	10	279	**279**	(11,6)	2
$MeSO_3^-$	annual	3,49	**3,54**	(1,89)	54	4,17	**3,95**	(2,34)	76	3,57	**3,30**	(1,83)	48	2,40	**2,56**	(1,32)	32
	DJFM	1,80	**1,67**	(1,31)	12	1,91	**1,53**	(1,41)	8	2,00	**1,72**	(1,44)	12	2,00	**1,03**	(1,87)	11
	A	5,04	**4,58**	(1,85)	5	2,98	**3,53**	(1,53)	6	4,29	**3,29**	(2,54)	5	2,66	**2,95**	(0,96)	4
	MJJA	3,92	**3,60**	(1,70)	23	4,56	**4,13**	(2,03)	34	3,96	**3,65**	(1,58)	21	2,77	**2,76**	(0,81)	15

	SON	3,68	**4,46**	(1,78)	14	4,59	**4,02**	(2,66)	28	4,27	**4,12**	(1,43)	10	1,26	**1,26**	(0,29)	2
Formate	annual	6,52	**4,67**	(5,68)	48	7,33	**6,39**	(3,72)	50	14,66	**5,21**	(36,7)	16				27
	DJFM	4,41	**3,22**	(3,59)	13	4,87	**4,87**	-	1	4,62	**4,36**	(2,43)	4	*Formate lost for $PM_{10}C$*			8
	A	2,72	**2,85**	(0,77)	4	7,63	**7,63**	(0,67)	2	1,43	**1,43**	(0,31)	2				2
	MJJA	7,06	**6,63**	(4,62)	18	7,44	**6,39**	(3,58)	24	6,82	**5,21**	(5,98)	8				15
	SON	9,04	**7,02**	(8,14)	13	7,31	**5,91**	(4,13)	23	79,3	**79,3**	(101)	2				2
Oxalate	annual	17,4	**12,0**	(21,7)	57	23,4	**21,4**	(16,2)	76	20,2	**16,1**	(19,9)	45	28,2	**23,2**	(16,8)	32
	DJFM	14,6	**13,8**	(11,4)	17	14,5	**13,4**	(9,7)	11	9,8	**4,7**	(10,8)	11	20,7	**22,4**	(9,83)	11
	A	12,0	**12,0**	(11,1)	5	25,5	**21,3**	(17,8)	6	4,9	**4,4**	(5,0)	4	31,7	**23,6**	(20,2)	4
	MJJA	8,1	**1,9**	(10,9)	21	21,2	**21,3**	(12,7)	31	19,3	**16,6**	(15,5)	21	31,7	**23,2**	(19,8)	15
	SON	36,8	**31,4**	(32,8)	14	28,8	**24,6**	(19,7)	28	41,5	**40,4**	(25,2)	9	36,1	**36,1**	(9,83)	2
H (neq m^{-3})	annual	0,72	**0,36**	(1,60)	59	3,21	**2,73**	(3,23)	80	2,19	**1,41**	(2,75)	52	1,07	**0,42**	(1,34)	32
	DJFM	0,57	**0,06**	(1,23)	17	1,57	**1,55**	(1,87)	11	0,64	**0,48**	(0,56)	14	1,15	**1,26**	(1,25)	11
	A	0,37	**0,51**	(0,49)	5	1,74	**1,34**	(2,44)	6	1,97	**1,01**	(2,59)	5	0,82	**0,63**	(0,77)	4
	MJJA	-0,13	**0,02**	(0,81)	23	3,94	**3,23**	(3,66)	35	2,16	**1,51**	(1,97)	23	1,20	**0,43**	(1,61)	15
	SON	2,41	**1,78**	(1,99)	14	3,26	**2,62**	(3,00)	28	4,51	**3,25**	(4,48)	10	0,21	**0,21**	(0,28)	2
Glucose	annual	1,75	**1,40**	(1,11)	54	1,96	**1,61**	(1,40)	48	1,55	**1,31**	(0,93)	44	0,96	**0,84**	(0,60)	23
	DJFM	1,97	**1,76**	(1,52)	13	1,51	**1,49**	(0,72)	4	1,73	**1,19**	(1,11)	10	1,06	**0,84**	(0,82)	8
	A	1,25	**1,29**	(0,33)	5	2,07	**2,07**	(0,40)	2	0,76	**0,77**	(0,21)	5	0,83	**0,81**	(0,49)	3
	MJJA	1,33	**1,25**	(0,48)	22	2,12	**1,64**	(1,87)	21	1,30	**1,34**	(0,58)	19	1,01	**1,04**	(0,49)	10
	SON	2,37	**2,03**	(1,29)	14	1,87	**1,43**	(0,98)	21	2,23	**2,21**	(1,08)	10	0,58	**0,58**	(0,15)	2
Levoglucosan	annual	6,7	**2,8**	(12,1)	56	7,63	**4,53**	(10,1)	70	19,5	**14,1**	(21,9)	32	5,87	**3,62**	(5,67)	19
	DJFM	0,76	**0,59**	(0,74)	15	1,50	**1,30**	(1,54)	5	3,37	**3,11**	(1,41)	4	1,48	**1,31**	(1,04)	5
	A	1,40	**1,2**	(0,61)	5	1,07	**1,01**	(0,72)	4	2,20	**2,20**	-	1	-	-	-	0
	MJJA	9,8	**5,1**	(16,4)	22	10,6	**4,85**	(13,6)	33	25,9	**17,5**	(26,9)	17	8,12	**7,94**	(6,06)	12
	SON	10,3	**5,8**	(10,3)	14	6,12	**5,37**	(3,81)	28	16,9	**17,0**	(11,6)	10	3,36	**3,36**	(0,96)	2
Mannosan	annual	1,55	**0,52**	(2,9)	17	1,09	**0,42**	(1,9)	50	3,67	**2,16**	(5,04)	25	0,73	**0,58**	(0,70)	14
	DJFM	-	-	-	0	0,34	**0,34**	-	1	0,44	**0,41**	(0,11)	3	0,09	**0,09**	(0,04)	2
	A	-	-	-	0	0,04	**0,04**	-	1	-	-	-	0	-	-	-	0
	MJJA	1,44	**0,43**	(3,09)	14	1,5	**0,47**	(2,25)	24	5,56	**4,07**	(6,40)	13	0,87	**0,68**	(0,73)	11
	SON	2,06	**1,38**	(2,17)	3	0,76	**0,42**	(1,62)	24	2,02	**2,13**	(1,41)	9	0,47	**0,47**	-	1
Galactosan	annual	1,06	**0,41**	(2,61)	23	0,73	**0,30**	(1,20)	48	2,66	**1,24**	(4,29)	26	0,58	**0,55**	(0,50)	14
	DJFM	-	-	-	0	0,17	**0,17**	-	1	0,25	**0,25**	(0,06)	2	0,05	**0,05**	(0,03)	2
	A	0,07	**0,07**	(0,03)	2	-	-	-	0	0,35	**0,35**	-	1	-	-	-	0
	MJJA	1,83	**0,48**	(3,68)	11	1,14	**0,51**	(1,63)	23	4,38	**2,55**	(5,59)	13	0,68	**0,56**	(0,51)	11
	SON	0,40	**0,35**	(0,40)	10	0,36	**0,27**	(0,26)	24	1,14	**1,16**	(0,88)	10	0,59	**0,59**	-	1
Arabitol	annual	2,03	**1,41**	(2,32)	58	0,90	**0,78**	(0,47)	44	0,56	**0,52**	(0,17)	31	0,30	**0,27**	(0,20)	22
	DJFM	1,47	**1,16**	(0,95)	17	0,36	**0,41**	(0,24)	3	0,77	**0,70**	(0,29)	4	0,36	**0,28**	(0,34)	6
	A	1,25	**1,01**	(0,62)	5	0,79	**0,79**	(0,11)	2	0,36	**0,36**	(0,01)	2	0,37	**0,37**	(0,05)	2
	MJJA	1,41	**0,89**	(1,16)	22	0,97	**0,91**	(0,48)	18	0,56	**0,52**	(0,13)	17	0,27	**0,23**	(0,14)	12
	SON	3,95	**2,97**	(3,87)	14	0,93	**0,78**	(0,47)	21	0,52	**0,52**	(0,14)	8	0,21	**0,21**	(0,12)	2
Mannitol	annual	0,72	**0,57**	(0,58)	49	0,89	**0,72**	(0,65)	43	0,97	**0,86**	(0,55)	40	0,62	**0,49**	(0,46)	13
	DJFM	0,75	**0,58**	(0,71)	12	0,98	**1,23**	(0,52)	3	1,35	**1,12**	(0,79)	9	0,89	**0,95**	(0,64)	5
	A	0,41	**0,42**	(0,12)	5	1,34	**1,34**	(0,53)	2	0,42	**0,38**	(0,08)	3	0,46	**0,46**	(0,04)	2
	MJJA	0,58	**0,55**	(0,24)	19	0,85	**0,75**	(0,48)	20	0,84	**0,85**	(0,35)	19	0,41	**0,26**	(0,26)	5
	SON	1,04	**0,93**	(0,77)	13	0,87	**0,61**	(0,85)	18	1,06	**1,03**	(0,50)	9	0,60	**0,60**	-	1

Table S 2. Ambient data for Chacaltaya

STP (1013 hPa, 0°C)		**$PM_{10}A$** December 2011 to July 2013				**$PM_{2.5}$** August 2013 to December 2015				**$PM_{10}B$** April 2016 to August 2017				**$PM_{10}C$** February 2019 to March 2020			
ng/m3	Season	mean	**median**	**std**	N	mean	**median**	**std**	N	mean	**median**	**std**	N	mean	**median**	**std**	N
OC	bulk	913	**703**	(648)	56	881	**669**	(729)	77	801	**605**	(604)	49	607	**529**	(449)	32
	DJFM	479	**393**	(294)	15	491	**405**	(367)	10	371	**349**	(240)	13	311	**290**	(164,3)	11
	A	621	**591**	(113,7)	5	575	**508**	(357)	6	325	**328**	(89,1)	5	512	**482**	(205)	4
	MJJA	1003	**849**	(543)	22	1044	**814**	(965)	33	914	**795**	(601)	21	846	**709**	(533)	15
	SON	1343	**1108**	(861)	14	894	**827**	(462)	28	1360	**1256**	(543)	10	634	**634**	(97,3)	2
EC	bulk	92,8	**68,2**	(90,8)	36	80,7	**69,7**	(48,6)	74	77,7	**56,5**	(68,7)	49	65,8	**51,3**	(48,5)	32
	DJFM	41,9	**32,6**	(23,4)	12	51,6	**54,6**	(30,9)	8	36,6	**33,5**	(19,9)	13	37,8	**36,1**	(22,5)	11
	A	73,4	**73,4**	(11,91)	2	73,5	**69,4**	(57,4)	6	38,5	**34,8**	(10,19)	5	66,3	**61,5**	(36,2)	4
	MJJA	70,6	**44,8**	(59,9)	8	90,1	**81,7**	(46,4)	32	86,8	**67,2**	(59,9)	21	86,6	**70,2**	(59,1)	15
	SON	151,9	**114,0**	(115,1)	14	79,7	**63,5**	(51,8)	28	131,6	**97,2**	(99,4)	10	62,1	**62,1**	(3,02)	2
Li^+	bulk	0,053	**0,038**	(0,070)	48	0,032	**0,025**	(0,033)	52	0,012	**0,005**	(0,023)	19	0,015	**0,013**	(0,012)	27
	DJFM	0,020	**0,010**	(0,022)	10	0,026	**0,026**	(0,005)	2	0,006	**0,004**	(0,007)	4	0,014	**0,013**	(0,007)	9
	A	0,027	**0,027**	(0,017)	2	0,019	**0,019**	(0,002)	2	0,004	**0,003**	(0,002)	4	0,015	**0,012**	(0,011)	4
	MJJA	0,042	**0,040**	(0,022)	22	0,023	**0,017**	(0,016)	26	0,006	**0,004**	(0,006)	8	0,018	**0,013**	(0,015)	12
	SON	0,098	**0,066**	(0,116)	14	0,044	**0,038**	(0,046)	22	0,048	**0,027**	(0,045)	3	0,004	**0,004**	(0,001)	2
Na^+	bulk	25,2	**21,9**	(20,8)	54	25,2	**20,1**	(15,38)	71	29,9	**22,7**	(26,8)	41	12,05	**9,17**	(10,28)	31
	DJFM	15,80	**17,72**	(9,81)	15	16,50	**13,39**	(10,68)	7	21,3	**23,3**	(12,98)	6	8,18	**8,95**	(6,23)	11
	A	22,7	**26,2**	(12,22)	4	18,37	**16,73**	(9,54)	4	11,83	**10,94**	(5,55)	5	11,75	**12,80**	(6,53)	4
	MJJA	21,0	**16,2**	(16,25)	21	26,3	**24,9**	(15,44)	33	23,7	**21,4**	(12,04)	20	16,08	**11,00**	(12,94)	14
	SON	42,5	**41,2**	(27,7)	14	27,1	**21,4**	(16,54)	27	56,6	**47,5**	(40,9)	10	5,67	**5,67**	(2,08)	2
NH_4^+	bulk	158,1	**144,8**	(106,0)	58	305	**295**	192	78	316	**306**	(196)	49	281	**248**	(160,5)	32
	DJFM	89,1	**80,8**	(77,1)	16	139,3	**56,8**	(185,0)	10	174,3	**109,2**	(146,4)	13	202	**229**	(118,5)	11
	A	157,0	**112,0**	(82,6)	5	349	**351**	(247)	6	241	**251**	(101,0)	5	406	**361**	(233)	4
	MJJA	133,6	**127,1**	(65,6)	23	353	**315**	(194)	34	319	**388**	(157,5)	21	318	**300**	(152,6)	15

	SON	277,3	**285,6**	(104,1)	14	297	**288**	(149,0)	28	533	**552**	(179,3)	10	186,7	**186,7**	(14,10)	2
K+	bulk	24,2	**17,70**	(22,8)	54	26,4	**23,3**	(17,82)	77	34,1	**15,5**	(34,0)	47	25,7	**19,9**	(26,3)	31
	DJFM	8,61	**8,66**	(4,81)	13	8,14	**6,39**	(5,70)	9	8,22	**7,91**	(6,88)	11	8,04	**6,10**	(6,46)	10
	A	10,61	**8,56**	(6,97)	5	11,80	**10,90**	(9,04)	6	6,32	**4,99**	(4,35)	5	17,93	**15,89**	(9,51)	4
	MJJA	23,8	**20,4**	(16,86)	22	33,4	**30,0**	(19,4)	34	45,0	**41,1**	(39,6)	21	39,9	**28,2**	(31,3)	15
	SON	44,3	**34,7**	(29,6)	14	27,0	**23,7**	(13,64)	28	53,6	**55,2**	(20,6)	10	23,1	**23,1**	(3,38)	2
Mg2+	bulk	7,28	**6,83**	(4,51)	56	8,62	**7,87**	(5,51)	75	6,86	**5,04**	(6,66)	49	6,66	**5,76**	(4,89)	32
	DJFM	4,71	**3,52**	(3,45)	15	3,75	**4,52**	(3,08)	9	3,16	**1,48**	(3,01)	13	4,49	**5,20**	(2,38)	11
	A	5,62	**3,25**	(5,13)	5	5,01	**3,31**	(4,18)	6	4,96	**4,20**	(4,08)	5	6,12	**5,51**	(3,47)	4
	MJJA	6,58	**5,91**	(2,47)	22	10,58	**8,50**	(6,12)	33	6,01	**4,92**	(3,37)	21	8,88	**6,75**	(5,96)	15
	SON	11,74	**12,51**	(4,94)	14	8,65	**8,62**	(4,23)	27	14,41	**9,48**	(10,24)	10	3,05	**3,05**	(0,38)	2
Ca2+	bulk	72,4	**66,4**	(43,0)	55	90,8	**74,5**	(78,4)	79	57,4	**51,3**	(43,0)	48	51,4	**36,2**	(42,0)	32
	DJFM	39,0	**31,1**	(30,1)	14	28,5	**20,6**	(22,5)	11	23,8	**13,1**	(21,2)	12	22,7	**17,3**	(16,01)	11
	A	41,1	**35,1**	(28,4)	5	67,2	**58,0**	(51,0)	6	31,0	**37,4**	(21,9)	5	40,0	**41,5**	(18,12)	4
	MJJA	84,6	**80,2**	(32,9)	22	128,2	**110,9**	(97,3)	34	63,4	**58,8**	(30,5)	21	78,5	**78,1**	(45,9)	15
	SON	97,7	**93,9**	(47,6)	14	75,1	**73,0**	(42,3)	28	98,5	**82,0**	(54,4)	10	29,7	**29,7**	(11,76)	2
F-	bulk	2,05	**1,48**	(2,31)	49	2,80	**1,79**	(2,49)	76	1,49	**1,13**	(1,17)	42				14
	DJFM	0,92	**0,87**	(0,41)	10	0,91	**0,60**	(0,56)	9	0,57	**0,47**	(0,41)	10		*Fluoride*		5
	A	0,93	**0,80**	(0,47)	5	1,88	**1,18**	(2,24)	6	0,55	**0,45**	(0,18)	3		*lost for*		0
	MJJA	1,58	**1,28**	(1,06)	20	3,64	**2,88**	(2,82)	33	1,92	**1,72**	(1,15)	20		*PM10C*		9
	SON	3,91	**2,78**	(3,51)	14	2,61	**1,77**	(2,11)	28	1,86	**1,70**	(1,30)	9				0
Cl-	bulk	8,72	**6,17**	(7,08)	47	13,28	**9,79**	(11,01)	45	10,85	**7,73**	(11,91)	34				25
	DJFM	3,97	**3,22**	(2,60)	11	4,54	**4,54**	(0,00)	1	1,57	**1,57**	(1,27)	2		*Chloride*		7
	A	5,58	**5,43**	(1,77)	4	17,51	**17,51**	(0,00)	1	1,91	**2,52**	(1,07)	3		*lost for*		3
	MJJA	9,08	**6,82**	(7,80)	22	14,70	**10,70**	(12,60)	28	10,73	**9,01**	(8,32)	21		*PM10C*		14
	SON	14,42	**16,66**	(6,12)	10	10,93	**8,02**	(7,66)	15	16,82	**8,71**	(19,3)	8				1
Br-	bulk	1,45	**1,27**	(0,92)	50	1,18	**0,91**	(0,79)	69	1,63	**1,09**	(1,54)	38	0,96	**0,78**	(0,80)	28
	DJFM	0,86	**0,52**	(1,09)	9	1,74	**1,56**	(1,55)	4	0,46	**0,42**	(0,16)	5	0,61	**0,24**	(0,62)	9
	A	0,68	**0,78**	(0,35)	5	0,46	**0,39**	(0,24)	5	0,52	**0,47**	(0,45)	4	0,61	**0,64**	(0,27)	4
	MJJA	1,31	**1,26**	(0,54)	22	1,15	**0,82**	(0,72)	32	1,66	**1,73**	(1,24)	20	1,30	**0,83**	(0,92)	14
	SON	2,31	**2,09**	(0,83)	14	1,26	**1,10**	(0,75)	28	2,69	**2,72**	(2,11)	9	0,71	**0,71**	(0,00)	1
NO3-	bulk	149,1	**127,9**	(114,2)	58	84,9	**65,6**	(79,2)	76	109,9	**71,1**	(115,8)	45				29
	DJFM	49,5	**31,6**	(49,6)	17	38,5	**28,5**	(32,2)	9	29,6	**14,2**	(33,6)	11		*Nitrate*		8
	A	126,1	**151,0**	(51,5)	5	84,7	**42,8**	(130,9)	6	29,4	**18,3**	(26,9)	4		*lost for*		4
	MJJA	191	**188,6**	(84,6)	22	86,9	**67,1**	(62,0)	33	161,3	**113,2**	(136,9)	21		*PM10C*		15
	SON	213	**230**	(145,9)	14	97,5	**79,1**	(92,3)	28	124,0	**91,6**	(77,0)	9				2
SO42-	bulk	451	**399**	(327)	58	1121	**1047**	(795)	79	991	**920**	(761)	52	905	**889**	(499)	32
	DJFM	302	**242**	(274)	16	513	**280**	(691)	10	511	**335**	(455)	14	659	**659**	(355)	11
	A	413	**242**	(277)	5	1071	**1028**	(855)	6	874	**925**	(502)	5	1186	**1089**	(602)	4
	MJJA	335	**303**	(167)	23	1360	**1347**	(892)	35	907	**931**	(609)	23	1061	**903**	(517)	15
	SON	825	**795**	(329)	14	1051	**923**	(559)	28	1917	**1995**	(792)	10	521	**521**	(21,7)	2
$MeSO_3$	bulk	6,58	**6,67**	(3,56)	54	7,86	**7,45**	(4,41)	76	6,73	**6,22**	(3,45)	48	4,52	**4,83**	(2,48)	32
	DJFM	3,38	**3,15**	(2,47)	12	3,61	**2,89**	(2,66)	8	3,76	**3,25**	(2,71)	12	3,78	**1,93**	(3,53)	11
	A	9,51	**8,64**	(3,49)	5	5,63	**6,66**	(2,88)	6	8,10	**6,20**	(4,78)	5	5,01	**5,56**	(1,82)	4
	MJJA	7,39	**6,79**	(3,21)	23	8,60	**7,78**	(3,82)	34	7,46	**6,88**	(2,99)	21	5,22	**5,20**	(1,53)	15
	SON	6,94	**8,41**	(3,37)	14	8,65	**7,58**	(5,01)	28	8,06	**7,76**	(2,70)	10	2,37	**2,37**	(0,54)	2
Formate	bulk	12,25	**8,78**	(10,69)	48	13,80	**12,02**	(6,99)	50	27,57	**9,80**	(68,9)	16				27
	DJFM	8,29	**6,05**	(6,74)	13	9,16	**9,16**	-	1	8,69	**8,19**	(4,57)	4		*Formate*		8
	A	5,12	**5,36**	(1,45)	4	14,34	**14,34**	(1,27)	2	2,68	**2,68**	(0,59)	2		*lost for*		2
	MJJA	13,28	**12,46**	(8,68)	18	13,99	**12,02**	(6,74)	24	12,83	**9,80**	(11,25)	8		*PM10C*		15
	SON	16,99	**13,21**	(15,31)	13	13,75	**11,11**	(7,77)	23	149,2	**149,2**	(191)	2				2
Oxalate	bulk	32,8	**22,6**	(40,8)	57	44,0	**40,3**	(30,5)	76	38,0	**30,3**	(37,4)	45	53,2	**43,6**	(31,7)	32
	DJFM	27,6	**26,1**	(21,4)	17	27,3	**25,2**	(18,2)	11	18,5	**8,8**	(20,4)	11	39,0	**42,2**	(18,52)	11
	A	22,5	**22,6**	(20,9)	5	48,0	**40,1**	(33,6)	6	9,3	**8,3**	(9,4)	4	59,8	**44,4**	(38,0)	4
	MJJA	15,2	**3,7**	(20,5)	21	39,9	**40,1**	(23,9)	31	36,4	**31,2**	(29,2)	21	59,7	**43,7**	(37,4)	15
	SON	69,4	**59,2**	(61,7)	14	54,3	**46,3**	(37,1)	28	78,3	**76,2**	(47,5)	9	68,1	**68,1**	(18,52)	2
H (neq/m3)	bulk	1,35	**0,65**	(2,99)	59	5,97	**5,16**	(6,02)	80	4,02	**2,62**	(5,12)	52	1,92	**0,71**	(2,50)	32
	DJFM	1,06	**0,13**	(2,28)	17	2,92	**2,89**	(3,44)	11	1,14	**0,86**	(1,03)	14	2,09	**2,28**	(2,33)	11
	A	0,69	**0,93**	(0,91)	5	3,17	**2,41**	(4,52)	6	3,60	**1,80**	(4,81)	5	1,39	**1,04**	(1,48)	4
	MJJA	-0,22	**0,07**	(1,52)	23	7,33	**6,09**	(6,82)	35	4,00	**2,78**	(3,65)	23	2,16	**0,69**	(2,99)	15
	SON	4,53	**3,30**	(3,73)	14	6,06	**4,87**	(5,59)	28	8,31	**6,04**	(8,40)	10	0,34	**0,34**	(0,54)	2
Glucose	bulk	3,29	**2,64**	(2,10)	54	3,69	**3,04**	(2,64)	48	2,91	**2,46**	(1,75)	44	1,81	**1,59**	(1,12)	23
	DJFM	3,70	**3,32**	(2,86)	13	2,84	**2,81**	(1,35)	4	3,26	**2,24**	(2,09)	10	1,99	**1,59**	(1,54)	8
	A	2,36	**2,44**	(0,62)	5	3,90	**3,90**	(0,75)	2	1,44	**1,46**	(0,40)	5	1,56	**1,52**	(0,93)	3
	MJJA	2,51	**2,35**	(0,90)	22	4,00	**3,09**	(3,52)	21	2,44	**2,53**	(1,10)	19	1,89	**1,95**	(0,93)	10
	SON	4,46	**3,82**	(2,43)	14	3,53	**2,69**	(1,84)	21	4,19	**4,15**	(2,03)	10	1,08	**1,08**	(0,29)	2
Levoglucosan	bulk	12,6	**5,3**	(22,7)	56	14,29	**8,47**	(18,9)	70	36,6	**26,5**	(41,1)	32	11,00	**6,77**	(10,62)	19
	DJFM	1,42	**1,10**	(1,38)	15	2,80	**2,44**	(2,88)	5	6,31	**5,82**	(2,63)	4	2,76	**2,45**	(1,95)	5
	A	2,62	**2,2**	(1,14)	5	2,01	**1,89**	(1,34)	4	4,12	**4,12**	-	1	-	-	-	0
	MJJA	18,4	**9,6**	(30,6)	22	19,9	**9,08**	(25,4)	33	48,5	**32,7**	(50,4)	17	15,21	**14,87**	(11,35)	12
	SON	19,2	**10,9**	(19,2)	14	11,46	**10,06**	(7,13)	28	31,7	**31,8**	(21,7)	10	6,29	**6,29**	(1,80)	2
Mannosan	bulk	2,92	**0,99**	(5,5)	17	2,06	**0,80**	(3,7)	50	6,93	**4,07**	(9,51)	25	1,38	**1,10**	(1,32)	14
	DJFM	-	-	-	0	0,65	**0,65**	-	1	0,83	**0,77**	(0,21)	3	0,17	**0,17**	(0,07)	2
	A	-	-	-	0	0,07	**0,07**	-	1	-	-	-	0	-	-	-	0
	MJJA	2,71	**0,81**	(5,83)	14	2,8	**0,89**	(4,24)	24	10,48	**7,68**	(12,08)	13	1,64	**1,29**	(1,37)	11
	SON	3,89	**2,61**	(4,09)	3	1,43	**0,80**	(3,06)	24	3,82	**4,01**	(2,67)	9	0,89	**0,89**	-	1
Galactosan	bulk	1,99	**0,76**	(4,92)	23	1,38	**0,57**	(2,26)	48	5,01	**2,35**	(8,09)	26	1,09	**1,04**	(0,94)	14
	DJFM	-	-	-	0	0,32	**0,32**	-	1	0,47	**0,47**	(0,12)	2	0,09	**0,09**	(0,05)	2
	A	0,14	**0,14**	(0,06)	2	-	-	-	0	0,65	**0,65**	(0,00)	1	-	-	-	0
	MJJA	3,46	**0,91**	(6,93)	11	2,16	**0,97**	(3,07)	23	8,25	**4,81**	(10,54)	13	1,27	**1,06**	(0,96)	11
	SON	0,75	**0,66**	(0,75)	10	0,68	**0,52**	(0,50)	24	2,15	**2,19**	(1,66)	10	1,11	**1,11**	-	1
Arabitol	bulk	3,76	**2,63**	(4,31)	58	1,67	**1,45**	(0,88)	44	1,04	**0,96**	(0,32)	31	0,56	**0,51**	(0,38)	22
	DJFM	2,72	**2,16**	(1,76)	17	0,67	**0,76**	(0,45)	3	1,42	**1,30**	(0,53)	4	0,68	**0,52**	(0,63)	6
	A	2,32	**1,88**	(1,16)	5	1,46	**1,46**	(0,21)	2	0,66	**0,66**	(0,02)	2	0,69	**0,69**	(0,10)	2
	MJJA	2,62	**1,65**	(2,15)	22	1,79	**1,68**	(0,90)	18	1,03	**0,96**	(0,24)	17	0,50	**0,43**	(0,26)	12
	SON	7,33	**5,51**	(7,18)	14	1,72	**1,46**	(0,88)	21	0,96	**0,96**	(0,26)	8	0,38	**0,38**	(0,21)	2
Mannitol	bulk	1,38	**1,09**	(1,10)	49	1,69	**1,37**	(1,24)	43	1,85	**1,63**	(1,04)	40	1,17	**0,92**	(0,87)	13
	DJFM	1,42	**1,09**	(1,36)	12	1,86	**2,33**	(1,00)	3	2,57	**2,13**	(1,49)	9	1,69	**1,80**	(1,21)	5
	A	0,78	**0,80**	(0,23)	5	2,55	**2,55**	(1,01)	2	0,79	**0,72**	(0,16)	3	0,87	**0,87**	(0,07)	2
	MJJA	1,10	**1,04**	(0,46)	19	1,61	**1,42**	(0,90)	20	1,60	**1,61**	(0,67)	19	0,77	**0,50**	(0,50)	5

	SON	1,97	**1,76**	(1,47)	13	1,64	**1,15**	(1,62)	18	2,01	**1,95**	(0,95)	9	1,13	**1,13**	-	1

Table S 3. Data for Chacaltaya taken to standard conditions

		$PM_{10}A$ vs $PM_{2.5}$			**$PM_{10}B$ vs $PM_{2.5}$**			**$PM_{10}C$ vs $PM_{2.5}$**			**$PM_{10}A$ vs $PM_{10}B$**			**$PM_{10}B$ vs $PM_{10}C$**			**$PM_{10}A$ vs $PM_{10}C$**		
	Season	p	**H**	**PM10A/ PM2,5**	p	**H**	**PM10B/ PM2,5**	p	**H**	**PM10C/ PM2,5**	p	**H**	**PM10A/ PM10B**	**p**	**H**	**PM10B/ PM10C**	p	**H**	**PM10A/ PM10C**
OC	bulk	0,74	0	0	0,24	0	0	<0,01	**1**	**0,69**	0,21	0	0	0,17	0	0	0,01	**1**	**1,50**
	DJFM	0,72	0	0	0,48	0	0	0,25	0	0	0,38	0	0	0,56	0	0	0,03	**1**	**1,54**
	A	0,43	0	0	0,18	0	0	0,91	0	0	0,01	**1**	**1,91**	0,29	0	0	0,56	0	0
	MJJA	0,74	0	0	0,63	0	0	0,46	0	0	0,43	0	0	0,85	0	0	0,26	0	0
	SON	0,06	0	0	0,02	**1**	**1,52**	0,59	0	0	0,46	0	0	0,03	**1**	**2,15**	0,33	0	0
EC	bulk	0,73	0	0	0,17	0	0	0,07	0	0	0,55	0	0	0,74	0	0	0,42	0	0
	DJFM	0,73	0	0	0,40	0	0	0,49	0	0	0,53	0	0	0,91	0	0	0,88	0	0
	A	0,86	0	0	0,43	0	0	0,91	0	0	0,10	0	0	0,41	0	0	0,80	0	0
	MJJA	0,19	0	0	0,51	0	0	0,55	0	0	0,51	0	0	0,95	0	0	0,26	0	0
	SON	0,02	**1**	**1,90**	0,07	0	0	1	0	0	0,58	0	0	0,36	0	0	0,20	0	0
Li+	**bulk**	0,02	**1**	**1,66**	0	**1**	**0,38**	<0,01	**1**	**0,48**	0	**1**	**4,38**	0,01	**1**	**0,79**	0	**1**	**3,47**
	DJFM	0,27	0	0	0,13	0	0	0,07	0	0	0,08	0	0	0,15	0	0	0,78	0	0
	A	1	0	0	0,13	0	0	0,53	0	0	0,13	0	0	0,11	0	0	0,53	0	0
	MJJA	<0,01	**1**	**1,80**	<0,01	**1**	**0,24**	0,18	0	0	0	**1**	**7,60**	0,02	**1**	**0,31**	<0,01	**1**	**2,34**
	SON	0,04	**1**	**2,23**	1	0	0	0,02	**1**	**0,10**	0,59	0	0	0,20	0	0	0,02	**1**	**21,9**
Na+	bulk	0,52	0	0	0,60	0	0	<0,01	**1**	**0,48**	0,34	0	0	0	**1**	**2,48**	<0,01	**1**	**2,09**
	DJFM	0,94	0	0	0,63	0	0	0,10	0	0	0,37	0	0	0,05	**1**	**2,60**	0,03	**1**	**1,93**
	A	0,69	0	0	0,41	0	0	0,49	0	0	0,19	0	0	0,90	0	0	0,20	0	0
	MJJA	0,06	0	0	0,68	0	0	0,01	**1**	**0,61**	0,23	0	0	0,03	**1**	**1,47**	0,21	0	0
	SON	0,03	**1**	**1,56**	0,01	**1**	**2,09**	0,02	**1**	**0,21**	0,46	0	0	0,03	**1**	**10,0**	0,03	**1**	**7,5**
NH4+	bulk	0	**1**	**0,52**	0,68	0	0	0,68	0	0	0	**1**	**0,50**	0,38	0	0	0	**1**	**0,56**
	DJFM	0,73	0	0	0,31	0	0	0,11	0	0	0,10	0	0	0,49	0	0	0,01	**1**	**0,44**
	A	0,33	0	0	0,79	0	0	0,76	0	0	0,31	0	0	0,19	0	0	0,06	0	0
	MJJA	<0,01	**1**	**0,38**	0,86	0	0	0,69	0	0	0,00	**1**	**0,42**	0,75	0	0	0,00	**1**	**0,42**
	SON	0,97	0	0	<0,01	**1**	**1,80**	0,38	0	0	0,00	**1**	**0,5**	0,03	**1**	**2,86**	0,20	0	0
K+	bulk	0,12	0	0	0,73	0	0	0,23	0	0	0,72	0	0	0,71	0	0	0,92	0	0
	DJFM	0,74	0	0	0,65	0	0	1	0	0	0,64	0	0	1	0	0	0,60	0	0
	A	0,93	0	0	0,43	0	0	0,35	0	0	0,31	0	0	0,03	**1**	**0,35**	0,29	0	0
	MJJA	0,02	**1**	**0,71**	0,87	0	0	0,87	0	0	0,35	0	0	0,80	0	0	0,08	0	0
	SON	0,03	**1**	**1,64**	<0,01	**1**	**1,99**	1	0	0	0,17	0	0	0,12	0	0	0,33	0	0
Mg2+	bulk	0,23	0	0	0,01	**1**	**0,80**	0,03	**1**	**0,77**	0,16	0	0	0,55	0	0	0,40	0	0
	DJFM	0,44	0	0	1	0	0	0,65	0	0	0,15	0	0	0,18	0	0	0,96	0	0
	A	1	0	0	1	0	0	0,48	0	0	1	0	0	0,56	0	0	0,73	0	0
	MJJA	0,01	**1**	**0,62**	<0,01	**1**	**0,57**	0,27	0	0	0,44	0	0	0,10	0	0	0,26	0	0
	SON	0,05	**1**	**1,36**	0,33	0	0	0,06	0	0	0,98	0	0	0,03	**1**	**4,72**	0,07	0	0
Ca2+	bulk	0,42	0	0	0,01	**1**	**0,63**	<0,01	**1**	**0,57**	0,05	0	0	0,44	0	0	0,02	**1**	**1,41**
	DJFM	0,29	0	0	0,64	0	0	0,74	0	0	0,08	0	0	0,83	0	0	0,12	0	0
	A	0,54	0	0	0,13	0	0	0,35	0	0	0,69	0	0	0,73	0	0	0,90	0	0
	MJJA	0,12	0	0	<0,01	**1**	**0,49**	0,09	0	0	0,06	0	0	0,30	0	0	0,84	0	0
	SON	0,10	0	0	0,31	0	0	0,10	0	0	0,84	0	0	0,06	0	0	0,10	0	0
F-	bulk	0,04	**1**	**0,73**	0,00	**1**	**0,53**	*F- lost PM10C*			0,22	0	0	*F- lost PM10C*			*F- lost PM10C*		
	DJFM	0,84	0	0	0,11	0	0				0,05	**1**	**1,62**						
	A	0,66	0	0	0,26	0	0				0	0	0						
	MJJA	0	0	0	0	0	0				0	0	0						
	SON	0	0	0	0	0	0				0	0	0						
Cl-	bulk	0,01	**1**	**0,66**	0,10	0	0	*Cl- lost for PM10C*			0,81	0	0	*Cl- lost for PM10C*			*Cl- lost for PM10C*		
	DJFM	0,67	0	0	0,67	0	0				0,15	0	0						
	A	0,40	0	0	0,50	0	0				0,06	0	0						
	MJJA	0,09	0	0	0,30	0	0				0,52	0	0						
	SON	0,17	0	0	0,77	0	0				0,51	0	0						
Br-	bulk	0,09	0	0	0,67	0	0	0,11	0	0	0,71	0	0	0,19	0	0	0,01	**1**	**1,51**
	DJFM	0,50	0	0	0,29	0	0	0,15	0	0	0,52	0	0	0,61	0	0	0,34	0	0
	A	0,42	0	0	0,90	0	0	0,41	0	0	0,41	0	0	0,69	0	0	1,00	0	0
	MJJA	0,12	0	0	0,32	0	0	0,51	0	0	0,55	0	0	0,59	0	0	0,41	0	0
	SON	<0,01	**1**	**1,84**	0,03	**1**	**2,14**	0,37	0	0	0,73	0	0	0,60	0	0	0,13	0	0
NO_3-	bulk	<0,01	**1**	**1,76**	0,58	0	0	*NO_3- lost for $PM_{10}C$*			0,04	**1**	**1,36**	*NO_3- lost for $PM_{10}C$*			*NO_3- lost for $PM_{10}C$*		
	DJFM	1	0	0	0,17	0	0				0,26	0	0						
	A	0,18	0	0	0,91	0	0				0,03	**1**	**4,28**						
	MJJA	<0,01	**1**	**2,20**	0,04	**1**	**1,86**				0,11	0	0						
	SON	0,01	**1**	**2,18**	0,20	0	0				0,24	0	0						
SO_4^{-2}	bulk	0,00	**1**	**0,40**	0,32	0	0	0,28	0	0	0,00	**1**	**0,45**	0,94	0	0	0,00	**1**	**0,50**
	DJFM	0,62	0	0	0,70	0	0	0,17	0	0	0,22	0	0	0,22	0	0	0,01	**1**	**0,46**
	A	0,33	0	0	0,93	0	0	0,76	0	0	0,15	0	0	0,41	0	0	0,06	0	0
	MJJA	0,00	**1**	**0,25**	0,05	**1**	**0,67**	0,27	0	0	0,00	**1**	**0,37**	0,49	0	0	0,00	**1**	**0,32**
	SON	0,34	0	0	0,00	**1**	**1,82**	0,12	0	0	0,00	**1**	**0,43**	0,12	0	0	0,15	0	0
$MeSO_3^-$	bulk	0,18	0	0	0,19	0	0	<0,01	**1**	**0,58**	0,98	0	0	<0,01	**1**	**1,49**	<0,01	**1**	**1,46**
	DJFM	0,85	0	0	0,91	0	0	0,78	0	0	0,84	0	0	0,93	0	0	1,00	0	0
	A	0,08	0	0	0,66	0	0	0,35	0	0	0,69	0	0	0,56	0	0	0,06	0	0
	MJJA	0,40	0	0	0,33	0	0	<0,01	**1**	**0,61**	0,83	0	0	0,01	**1**	**1,43**	0,01	**1**	**1,42**
	SON	0,45	0	0	0,96	0	0	0,04	**1**	**0,27**	0,62	0	0	0,03	**1**	**3,40**	0,10	0	0
Fornate	bulk	0,02	**1**	**0,89**	0,07	0	0	*Formate lost for PM10C*			0,92	0	0	*Formate lost for PM10C*			*Formate lost for PM10C*		
	DJFM	0,57	0	0	1	0	0				0,62	0	0						
	A	0,13	0	0	0,33	0	0				0,13	0	0						
	MJJA	0,73	0	0	0,14	0	0				0,60	0	0						
	SON	0,92	0	0	0,15	0	0				0,23	0	0						

Oxalate	bulk	0,00	1	0,75	0,09	0	0	0,13	0	0	0,41	0	0	0,01	**1**	**0,72**	0,00	1	0,62
	DJFM	0,89	0	0	0,17	0	0	0,13	0	0	0,22	0	0	0,03	**1**	**0,48**	0,07	0	0
	A	0,18	0	0	0,04	**1**	**0,19**	0,61	0	0	0,41	0	0	0,03	**1**	**0,16**	0,19	0	0
	MJJA	0,00	1	0,38	0,42	0	0	0,16	0	0	0,02	**1**	**0,42**	0,04	**1**	**0,61**	0,00	1	0,25
	SON	0,51	0	0	0,11	0	0	0,34	0	0	0,40	0	0	0,73	0	0	0,70	0	0
Glucose	bulk	0,57	0	0	0,08	0	0	<0,01	**1**	**0,49**	0,22	0	0	0,01	**1**	**1,60**	0,00	**1**	**1,81**
	DJFM	0,78	0	0	0,84	0	0	0,37	0	0	0,64	0	0	0,12	0	0	0,06	0	0
	A	0,10	0	0	0,10	0	0	0,20	0	0	0,06	0	0	1	0	0	0,25	0	0
	MJJA	0,24	0	0	0,11	0	0	0,03	**1**	**0,47**	0,84	0	0	0,21	0	0	0,16	0	0
	SON	0,17	0	0	0,41	0	0	0,03	**1**	**0,31**	0,98	0	0	0,03	**1**	**3,86**	0,02	**1**	**4,11**
Levoglucosan	bulk	0,07	0	0	<0,01	**1**	**2,56**	0,70	0	0	<0,01	**1**	**0,35**	0,01	**1**	**3,33**	0,33	0	0
	DJFM	0,54	0	0	0,11	0	0	0,69	0	0	<0,01	**1**	**0,23**	0,06	0	0	0,14	0	0
	A	0,73	0	0	0,4	0	0	-	-	-	-	-	-	-	-	-	-	-	-
	MJJA	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
	SON	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Mannosan	bulk	0,75	0	0	<0,01	**1**	**3,37**	0,63	0	0	<0,01	**1**	**0,42**	<0,01	**1**	**5,03**	0,86	0	0
	DJFM	.-	-	-	0	0	0	0	0	0	0	0	0	0	0	0	0,00	0	0
	A	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	MJJA	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0,00	0	0
	SON	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0,00	0	0
Galactosan	bulk	0,64	0	0	<0,01	**1**	**3,63**	0,77	0	0	<0,01	**1**	**0,40**	0,01	**1**	**4,59**	0,54	0	0
	DJFM	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	A	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	MJJA	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	SON	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Arabitol	bulk	<0,01	**1**	**2,26**	<0,01	**1**	**0,63**	<0,01	**1**	**0,33**	<0,01	**1**	**3,61**	<0,01	**1**	**1,87**	<0,01	**1**	**6,75**
	DJFM	<0,01	**1**	**4,03**	0,23	0	0	0,90	0	0	0,10	0	0	0,07	0	0	<0,01	**1**	**4,02**
	A	0,38	0	0	0,33	0	0	0,33	0	0	0,10	0	0	1	0	0	0,10	0	0
	MJJA	0,33	0	0	<0,01	**1**	**0,58**	<0,01	**1**	**0,28**	<0,01	**1**	**2,54**	<0,01	**1**	**2,05**	<0,01	**1**	**5,20**
	SON	<0,01	**1**	**4,26**	0,01	**1**	**0,56**	0,03	**1**	**0,22**	<0,01	**1**	**7,61**	0,04	**1**	**2,52**	0,02	**1**	**19,2**
Mannitol	bulk	0,08	0	0	0,20	0	0	0,06	0	0	0,00	**1**	**0,74**	0,01	**1**	**1,58**	0,32	0	0
	DJFM	0,54	0	0	0,73	0	0	0,79	0	0	0,01	**1**	**0,55**	0,24	0	0	0,51	0	0
	A	0,10	0	0	0,20	0	0	0,33	0	0	0,79	0	0	0,80	0	0	0,57	0	0
	MJJA	0,06	0	0	0,64	0	0	0,03	**1**	**0,48**	0,02	**1**	**0,69**	0,01	**1**	**2,07**	0,20	0	0
	SON	0,17	0	0	0,07	0	0	0,95	0	0	0,50	0	0	0,40	0	0	0,57	0	0

Table S 4. Comparison of median concentration between different periods. p= p score of Wilcoxon rank sum test (significance level set to 0.05). H = Null hypothesis: H=1, medians are different; H=0, medians are the same. When medians are different (bold), mass ratios between two different periods were calculated.

--- Complete dataseries and saccharide analysis ---

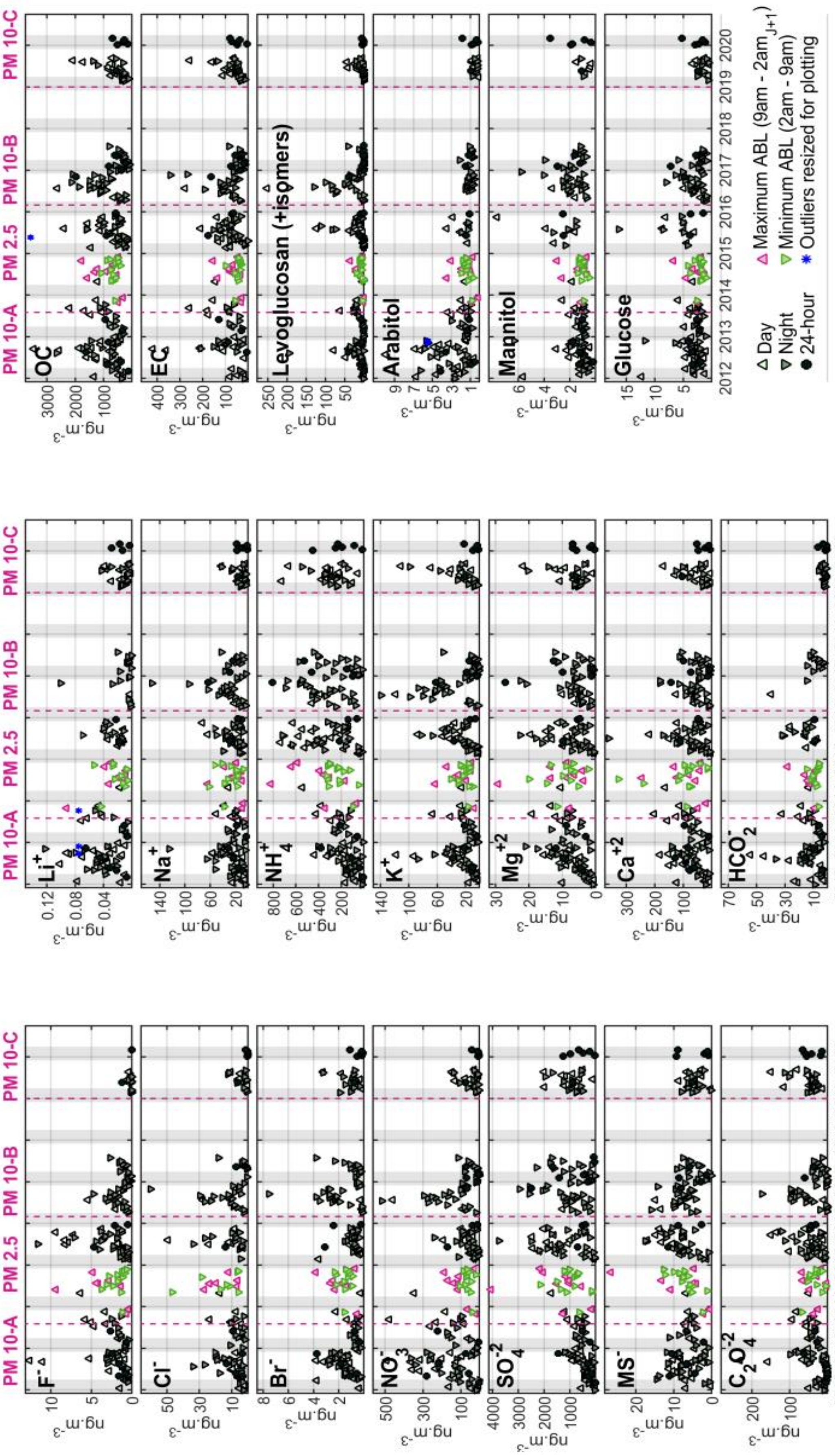

Figure S 3 Concentration of ions, OC, EC and anhydrosugars measured in Chacaltaya at STP conditions. The dashed lines show the difference between PM_{10} and PM 2.5 sampling periods. Upward pointing triangles are for daytime sampling, downward pointing triangles for nighttime and circles for 24-hour sampling. The color markers identify sampling conditions more likely to represent the difference between maximum (pink) and minimum (green) atmospheric boundary layer influence at Chacaltaya. Blue asterisks are extreme outliers whose values are found in the supplementary material. Gray shaded area highlights the wet season (DJFM). Note that F^-, Cl^-, NO_3^- and formiate concentrations for PM_{10}-C samples are suspected of presenting losses, and were excluded from the analysis but presented here to show the concentrations' drop.

In figure S3, the higher nitrate concentration of PM_{10}-A (149 ng m^{-3}) compared to PM_{10}-B (110 ng m^{-3}) remains unexplained, with the hypothesis being that a shift of NO_3^- towards coarser particles was due to the presence of crustal particles (such as observed by Wang et al., 2013), more day-time sampling in that period (which may have included more NO_3^- from the urban area), and/or shifts in the gas/phase equilibrium (Trebs, 2005) under lower sulfate contributions compared to the rest of the time series

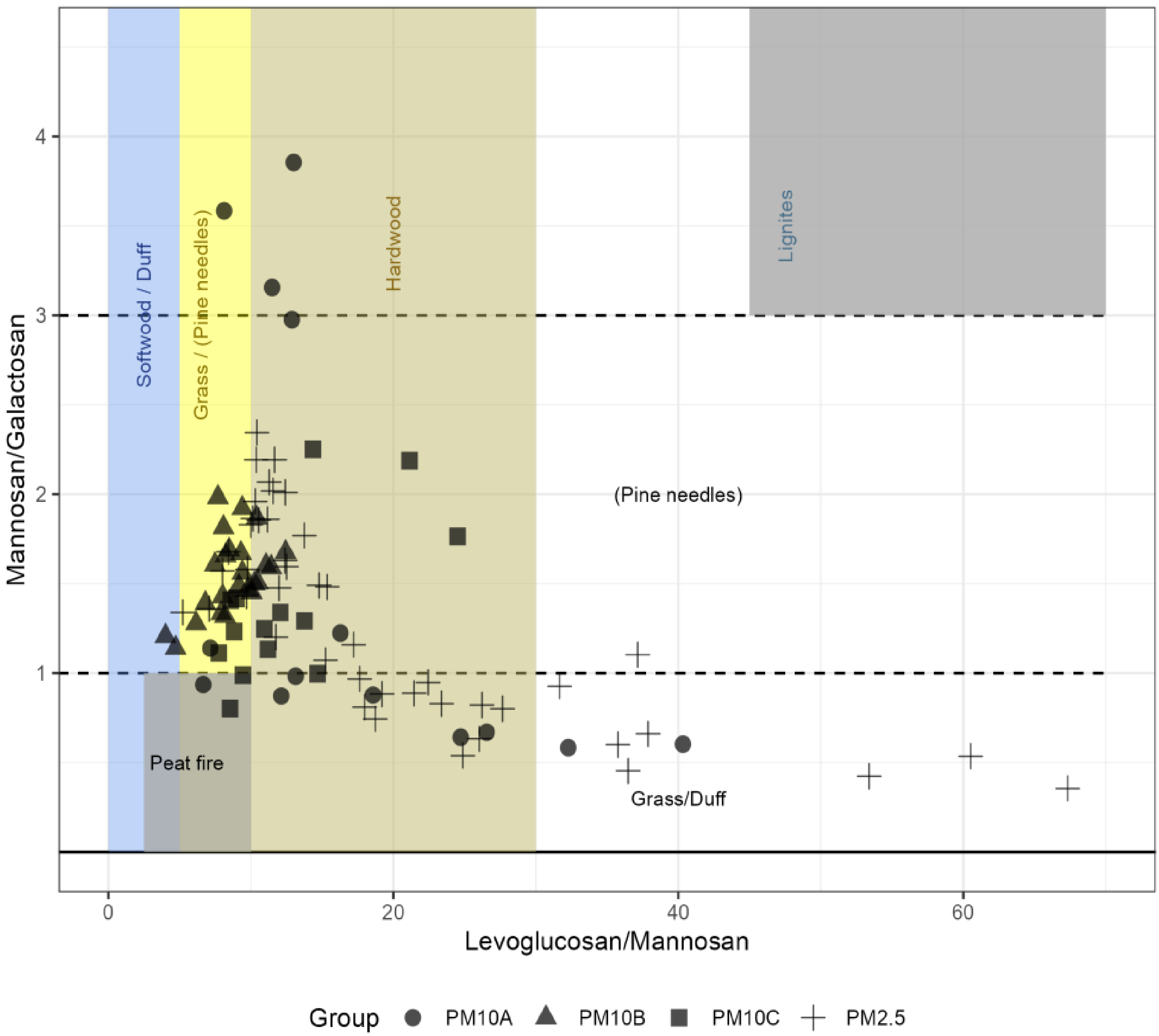

Figure S 4 Diagnostic diagram based on mass ratios of levoglucosan and its stereoisomers. Simplified plot adapted from (Marynowski and Simoneit, 2022) and (Xu et al., 2019) to cover the range of Chacaltaya data. Note that conifer forests are absent in the region and therefore the substrate "pine needles" is negligible for our case.

The highest levoglucosan+mannosan+galactosan concentration is 180 ng m^{-3} and it was found in 2016 (25 - 29/Jul/2016), early in the biomass burning season, as part of a regional event that spanned until mid-November. In our record, 2016 and 2017 were the years with the highest levoglucosan concentrations (Figure S3).

--- Backtrajectory complementary information ---

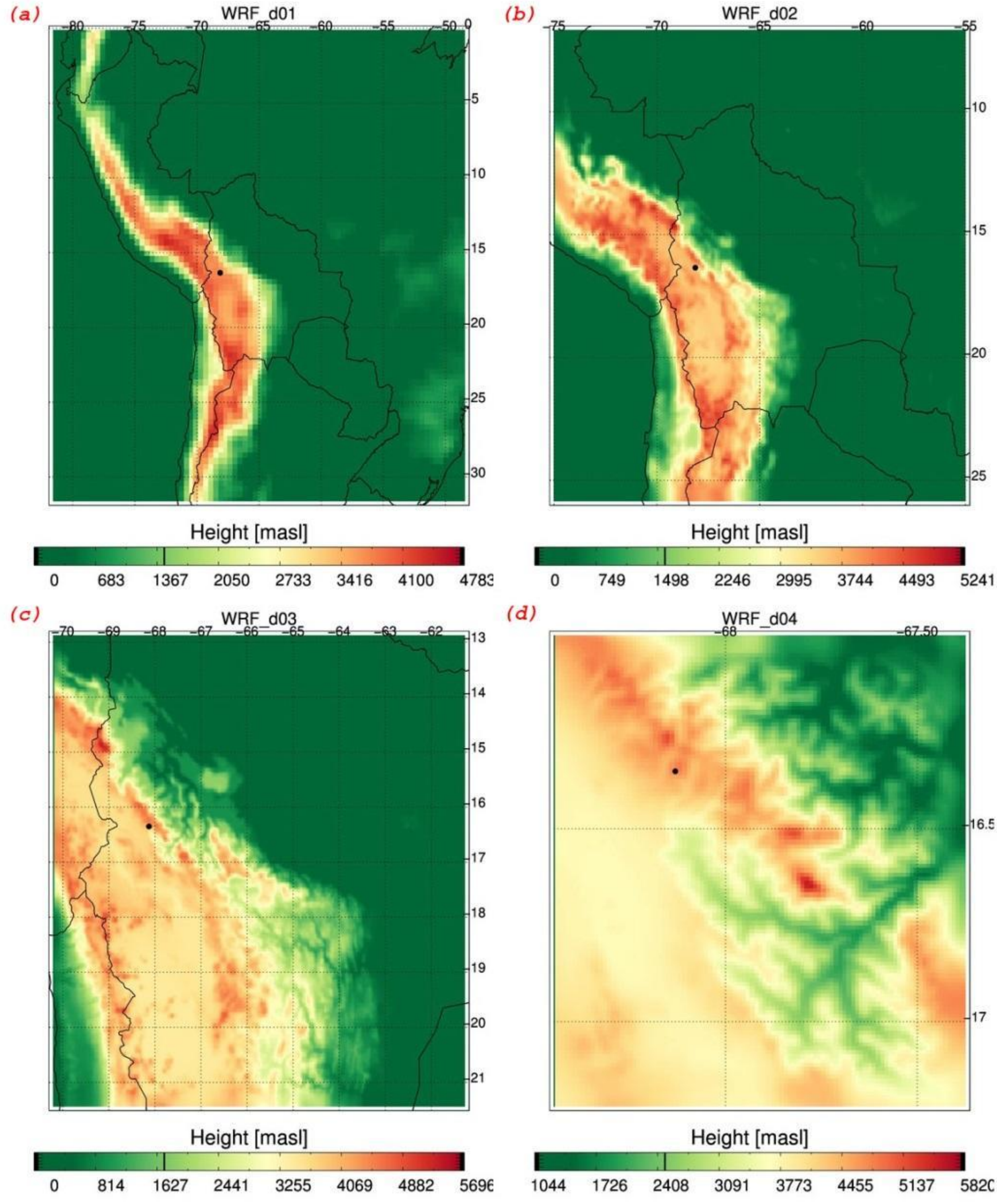

Figure S 5. Spatial resolution of four domains simulated by WRF model: a) Domain 1 (d01), b) Domain 2 (d02), c) Domain 3 (d03), and d) Domain 4 (d04). The black dot indicates Chacaltaya location. Prepared by F. Velarde

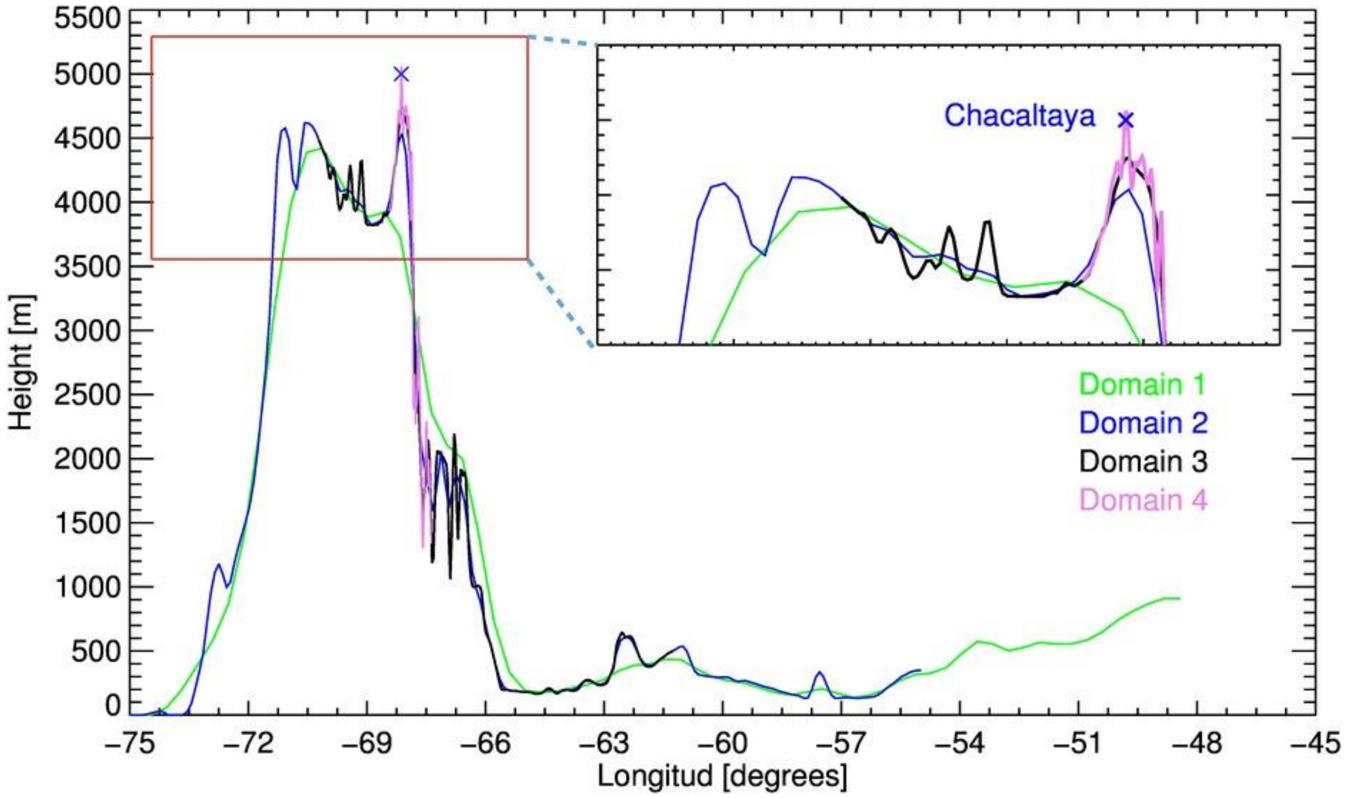

Figure S 6. Topography for the WRF domains used to obtain Hysplit backtrajectories. Latitudinal section at 16.351ºS for all domains. Note that domain 4 represents better the real altitude of Chacaltaya and the complex terrain around. Prepared by F. Velarde

Domain	Spatial resolution [km]	Height of Chacaltaya [masl]
WRFd01	38.00x38.00	3728.14
WRFd02	9.50x9.50	4535.99
WRFd03	3.17x3.17	4732.34
WRFd04	1.06x1.06	5058.44

Table S 5. WRF nested domains for generating meteorological data to be used in Hysplit Desktop v. 4.8. In all domains 28 pressure levels were used. Prepared by F. Velarde

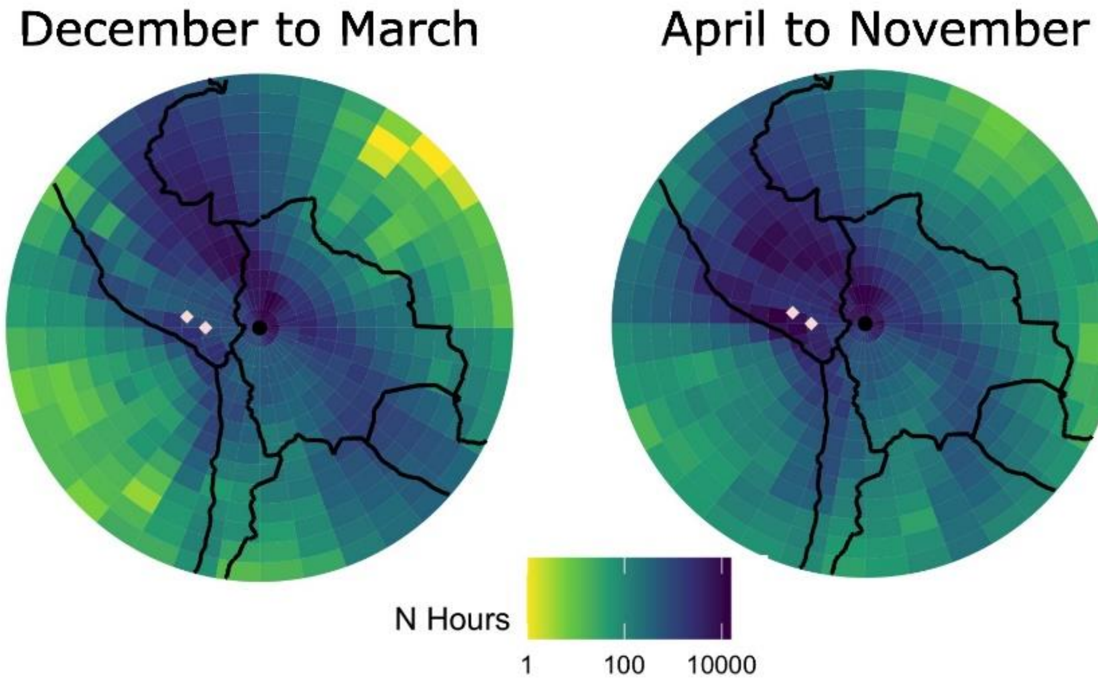

Figure S 7. Seasonality. WRF-based backtrajectories for all the samples taken for the wet season (December to March) and the rest of the year. The pink rhombuses show Sabancaya and Ubinas locations.

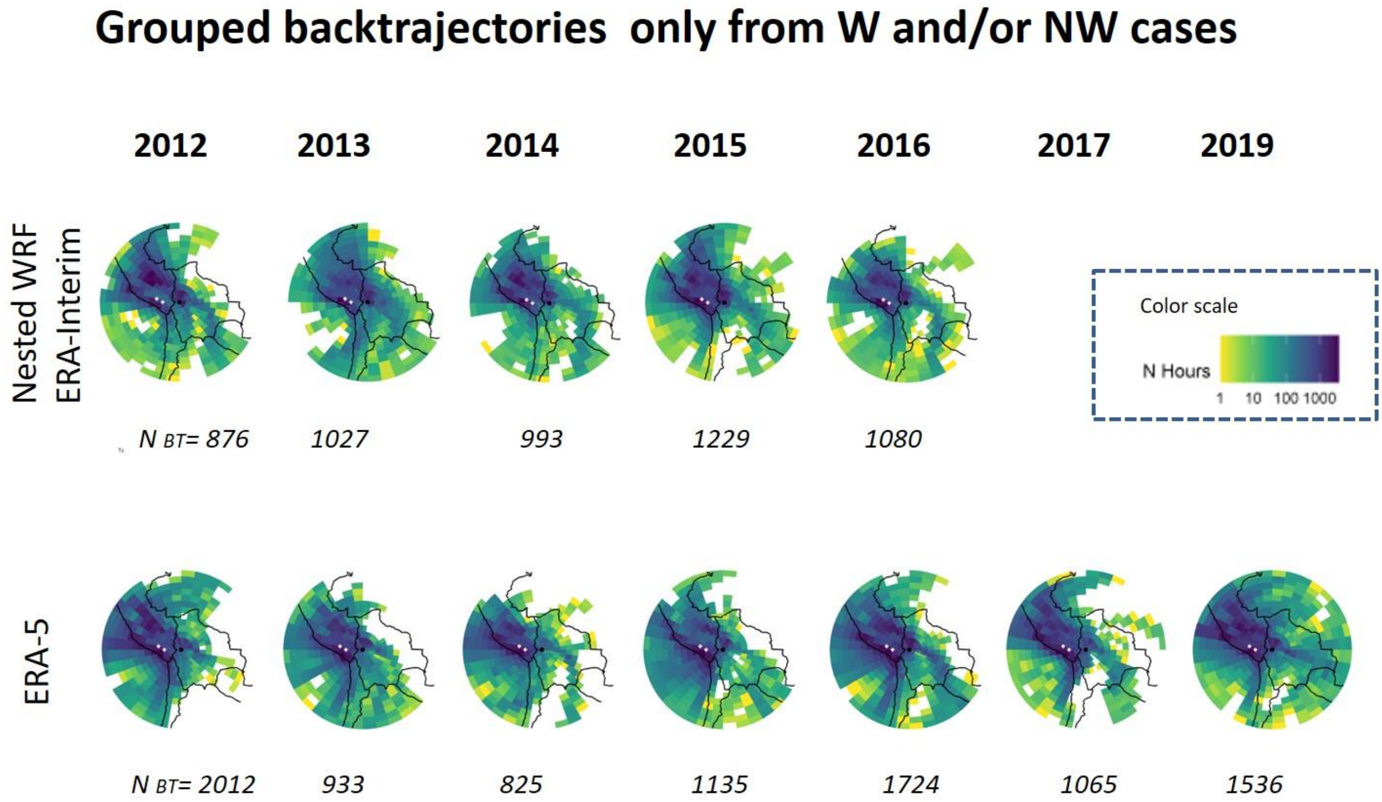

Figure S 8. Comparison of nested WRF (only central point) and ERA-5 backtrajectories (the only one point available) based on samples that present North (N) and/or Northwest (NW) pathways. The classification was made for each dataset (as there are not necessarily coincident) and grouped for the entire year. The number of 96h-bactrajectories used for each plot is in italics. The color scale corresponds to the number of hours of accumulated air passages over a pixel.

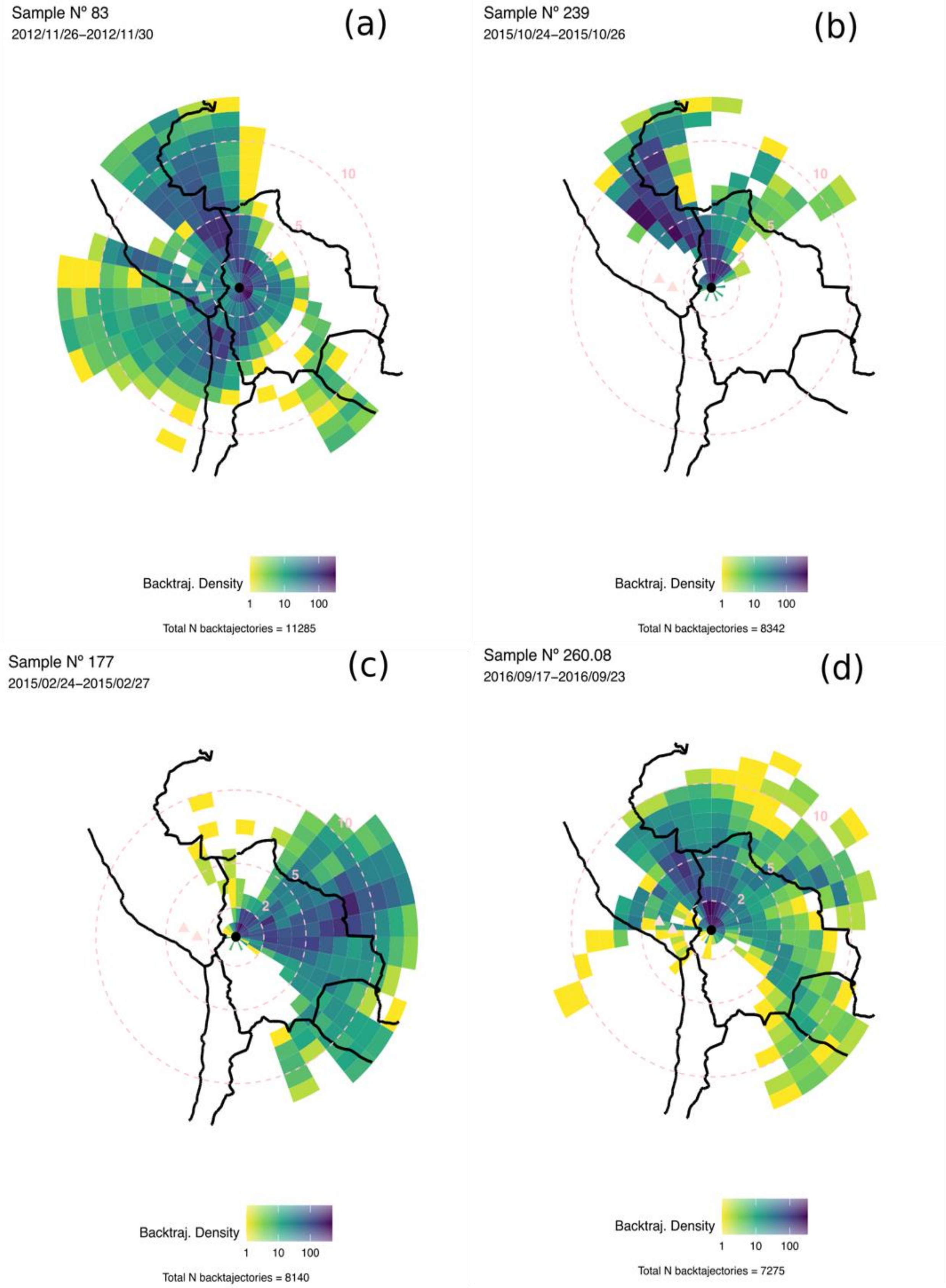

Figure S 9. Examples of backtrajectories obtained by nested WRF data with ERA-Interim boundary conditions. (a) Sample 83 from November. Mixed origins with Amazonian, Altiplano and Pacific influences with high chloride content. (b) Sample 239 from October with a clear north pathway (Amazonia and montane forests). It presents high oxalate content (c) Sample 177 from February with Amazonian and some Chaco influences, extremely low concentration for most species including $MeSO_3^-$. (d) Sample 260.08 from October. Mixed origins with high concentrations of biomass burning emissions.

In Figure S 9 we present a set of examples of WRF-based back-trajectories from mixed directions (a, d) and a dominant direction (b, c). It must be noted that a marked direction does not always bring the same chemical fingerprint. For instance, sample 239 (d) presents an oxalate concentration >p90, and it is similar to the backtrajectory pattern for samples 16 and 74 (not shown), for which oxalate is <p10 and for which the latter arabitol presents maximum values. In another study that used dispersion for assessing the origin of the air masses (Aliaga et. al., 2021), it was found that the further the trajectory goes, the higher it tends to be. Therefore, in this regard, trajectories such as (a) and (d) would bring air from higher layers than (b), which may have a higher near-ground influence.

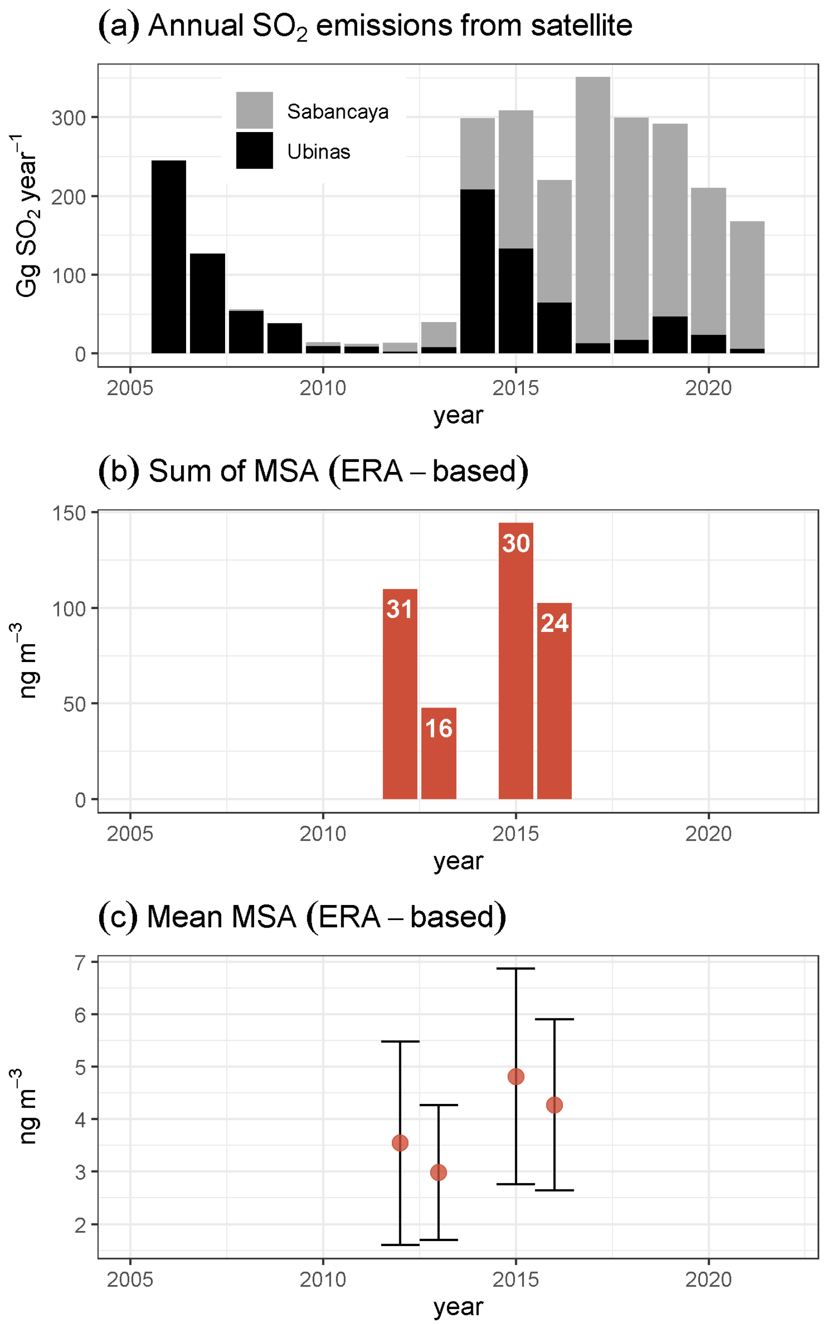

Figure S 10. Control case for marine transport using ERA-5 backtrajectories in complement to volcanic transport. Only years with at least 9 months of filter data were used. (a) Bulk SO_2 emissions for Sabancaya (grey) and Ubinas (black) from https://so2.gsfc.nasa.gov/measures.html (b) Accumulated SO_4^{-2} measured in the filters taken under W and/or SW influence grouped per year based on ERA-5 backtrajectories. The number of samples used is in white letters inside the red (c) Mean concentration and standard deviation for the selected samples. Note that there is no statistically significant difference between 2015-2016 and 2012.

--- Clustering to identify seasonality patterns ---

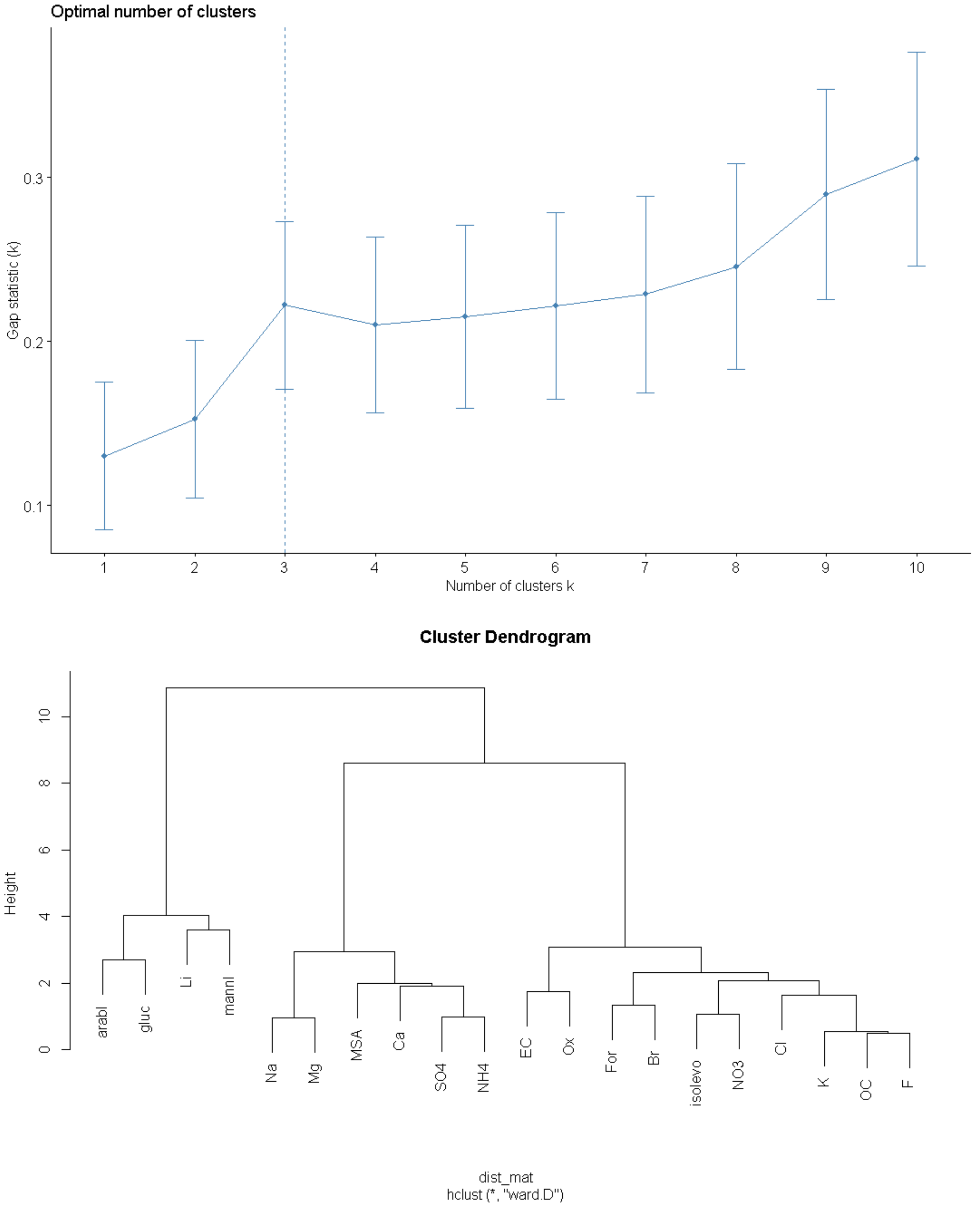

Figure S 11. Clustering by k-means of median concentration for ions, EC, OC, saccharides. Upper panel: k-means method optimal number of clustering. Lower panel: hierarchical clustering based on the Euclidean distance of concentrations using Ward method, prepared with R.

--- Meteorological information ---

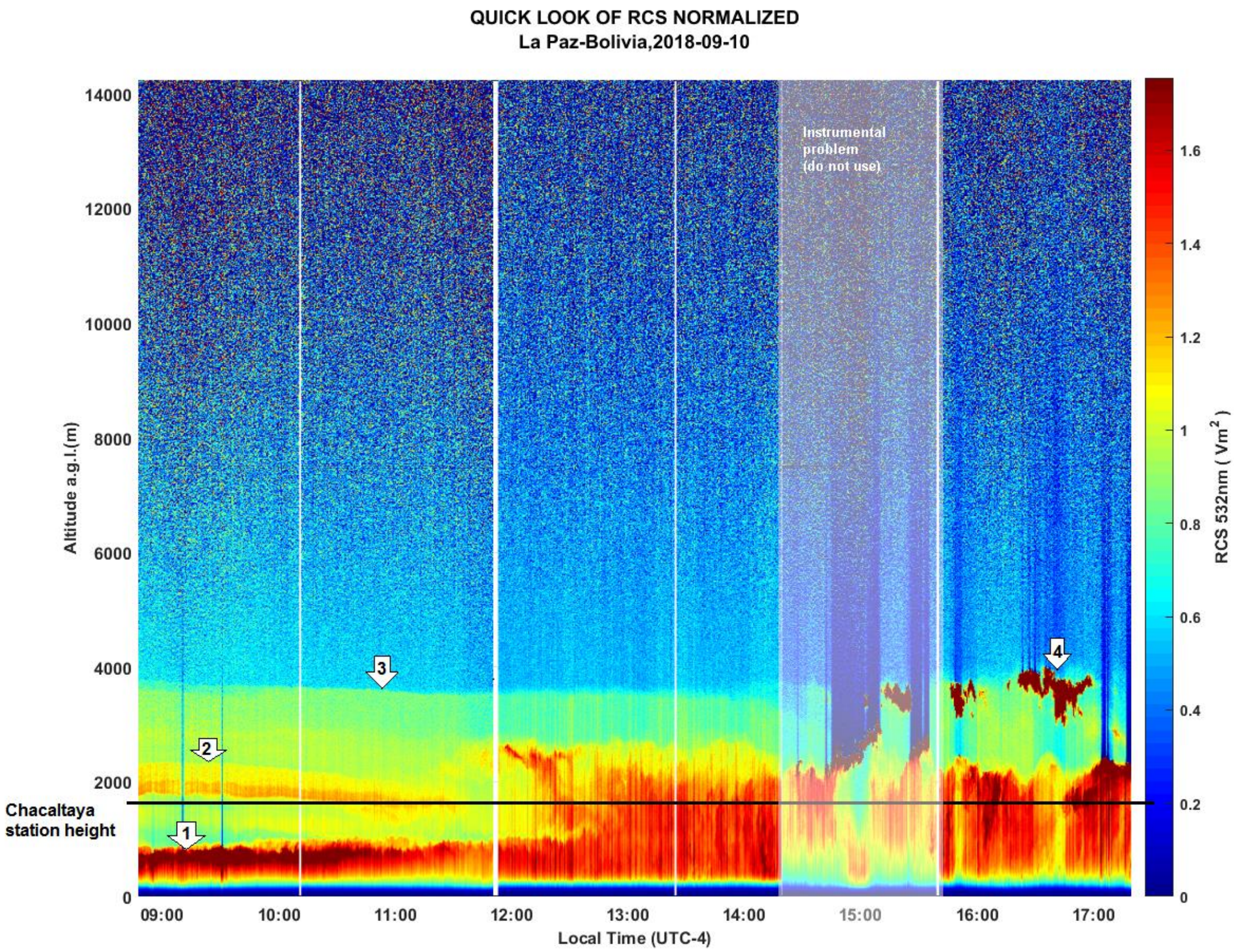

Figure S 12. Range-corrected signal (RCS) of lidar measurements at the Cota Cota location (LP-EA in Figure 1) showing an example of a multi-layered boundary/mixing layer. Ground level is already 3640 m a.s.l. The horizontal line shows the Chacaltaya station height. The cleared area between 14:30 and 15:30 corresponds to an instrumental problem for which the absolute RCS is not correct. The numbered arrows stand for:
1= Top of the valley mixing layer
2= Top of the nocturnal residual layer
3= Top of the regional residual layer
4 = Clouds/ condensation level.
The image was taken during the core of the biomass burning season, when smoke and other by-products are emitted in large quantities in the Amazonian basin (with a comparatively small influence from the Altiplano). Prepared by R Forno, MF Sánchez, the use is authorized by the authors.

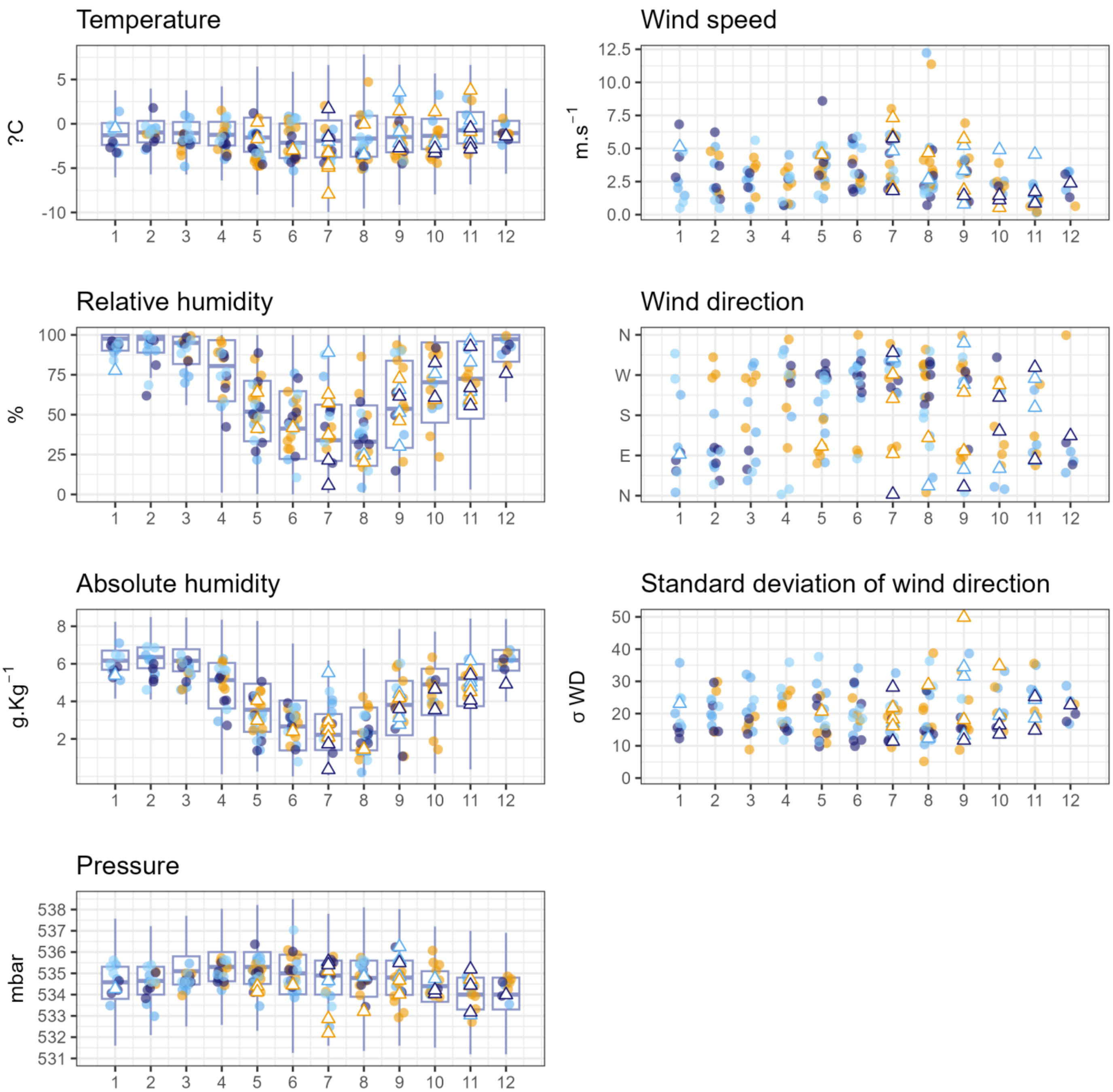

Figure S 13. Average meteorological conditions for each sample (for wind, vector average was used). Blue boxplots represent the climatology (data from 2011 to 2020), not presented for winds for clarity. Brown markers = PM_{10}-A, yellow markers = PM2.5, dark green markers = PM_{10}-B, light green markers = PM_{10}-C. Open triangles represent the outliers that were removed from Figure 4 and Figure 3S.

---- Outlier values not presented in the figure 4 and figure S3 ----

Ys	Ms	Ds	SN	ST	HeadN	ng/m3 amb	Species defining the outlier
2012	1	16	4	2	10	6,59	glucose
						2,97	mannitol
2012	7	27	47	2	10	5,38	arabitol
2012	8	9	49	-2	10	104	isolevo
2012	9	4	57	2	10	192	EC
2012	9	4	57	2	10	5,86	F
2012	9	14	60	2	10	6,67	F
						223	EC
						123	oxalate
2012	9	17	61	-2	10	0,114	Li
2012	10	26	74	-2	10	14,4	arabitol
2012	11	5	77	-2	10	66,3	Na
2012	11	16	80	-2	10	11,1	arabitol
						0,244	Li
2012	11	26	83	-2	10	3,33	mannitol
2013	9	9	124	4	2.5	0,123	Li
2014	5	29	145	5	2.5	2177	SO4
						15,5	Mg
						13,7	H
						240	Ca
2014	7	11	151	-5	2.5	176	Ca
2014	10	27	166	5	2.5	14,1	MSA
2015	5	12	197	6	2.5	3010	OC
2015	7	3	212	-6	2.5	6,14	F
						195	Ca
2015	7	31	218	-6	2.5	9,34	glucose
						17,3	H
						2007	SO4
2015	8	7	220	6	2.5	25,9	Cl
2015	9	4	226	6	2.5	13,1	H
2015	11	16	243	6	2.5	4,03	mannitol
2015	11	23	245	-6	2.5	10,9	H
2016	7	25	260	6	10	134	isolevo
						151	formato
2016	11	18	262	-6	10	182	EC
2016	12	2	263	99	10	189	EC
2016	12	16	265	-6	10	3,03	mannitol
2016	7	29	260.01	-6	10	104	isolevo
2016	9	16	260.08	-6	10	4,02	Br
						12,7	H
2016	10	21	260.13	-6	10	49,8	Na
2016	10	28	260.14	-6	10	31,5	Cl
						10,8	H
						81,3	Na
						17	Mg
2016	11	4	260.16	99	10	14,3	Mg

Table S 6. Outliers of the dataseries

---- References ----

Marynowski, L. and Simoneit, B. R. T.: Saccharides in atmospheric particulate and sedimentary organic matter: Status overview and future perspectives, Chemosphere, 288, 132376, https://doi.org/10.1016/j.chemosphere.2021.132376, 2022.

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Xu, J., Jia, C., He, J., Xu, H., Tang, Y.-T., Ji, D., Yu, H., Xiao, H., and Wang, C.: Biomass burning and fungal spores as sources of fine aerosols in Yangtze River Delta, China – Using multiple organic tracers to understand variability, correlations and origins, Environmental Pollution, 251, 155–165, https://doi.org/10.1016/j.envpol.2019.04.090, 2019.