Supplementary information

Fungal spores as a source of sodium salt particles in the Amazon basin

China et al.

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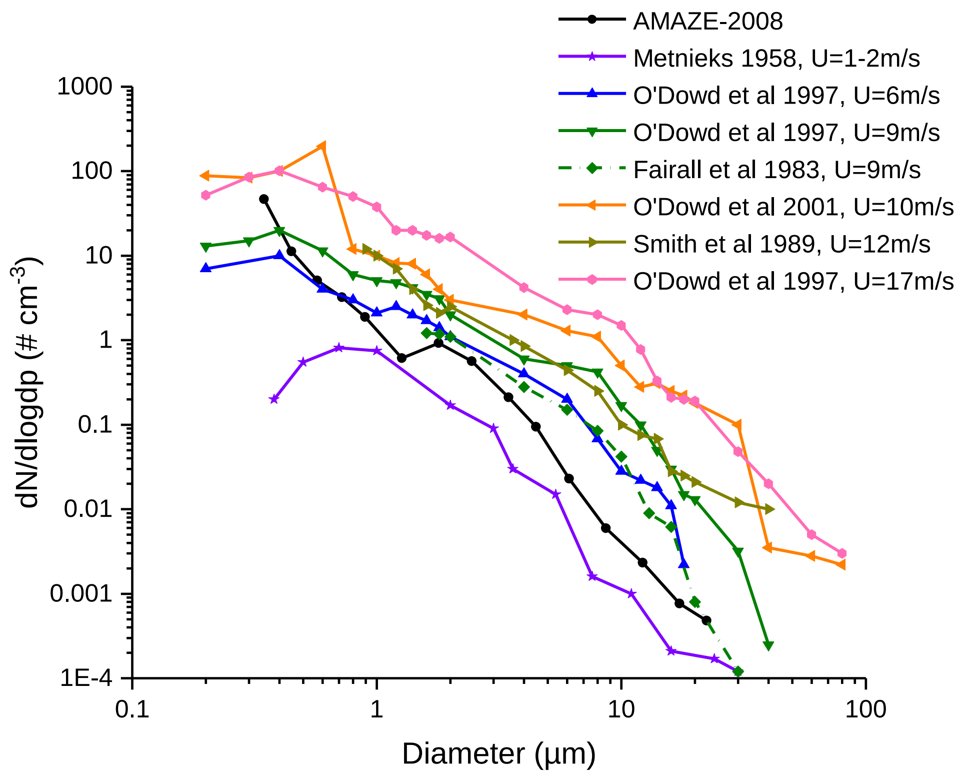
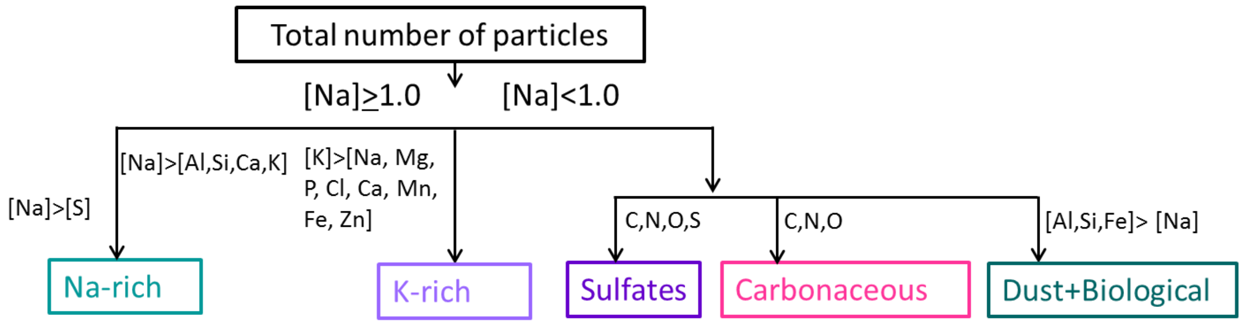


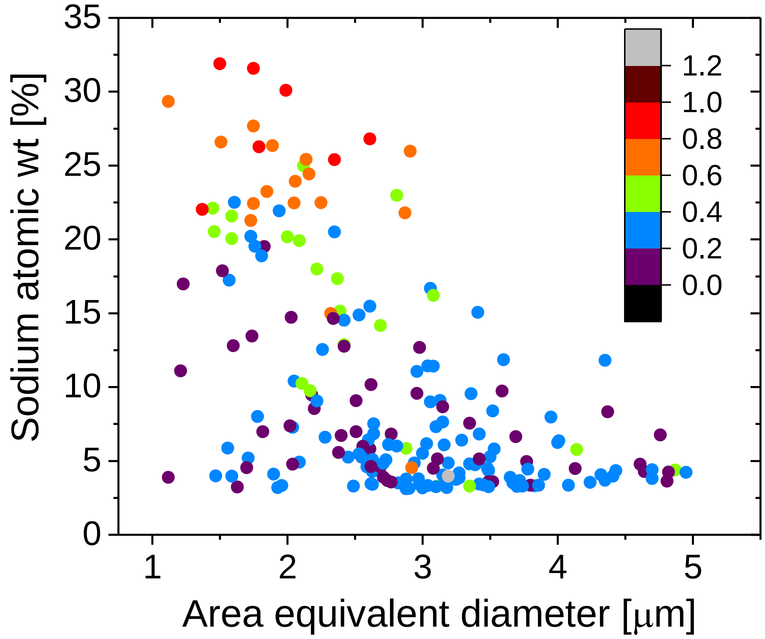
Supplementary Figure 1. Comparison of coarse particles in Amazonia size distribution with sea salt particles from different marine areas. Size distribution of sea salt particles for maritime air masses over different coastal sites (Metnieks[1], 1958 in Ireland; O'Dowd et al[2] 1997 in North East Atlantic coastal site; Fairall et al[3] 1952 in California coastal area; and Smith et al[4] 1989 in North East Atlantic coastal site) at several wind speeds, U. Size distribution data is from Lewis and Schwartz[5]. Average particle concentrations for the wet season from Amazonia (black line) were measured using an optical particle counter during the Amazonian Aerosol Characterization Experiment (AMAZE-08) field campaign during 2008.

Supplementary Table 1. Aerosol sample identifications, local (AMT) sampling time, and duty cycle. Number of particles (N) analyzed and estimated margin of error (ME) at a given confidence interval (99%) applying binomial distribution[6].

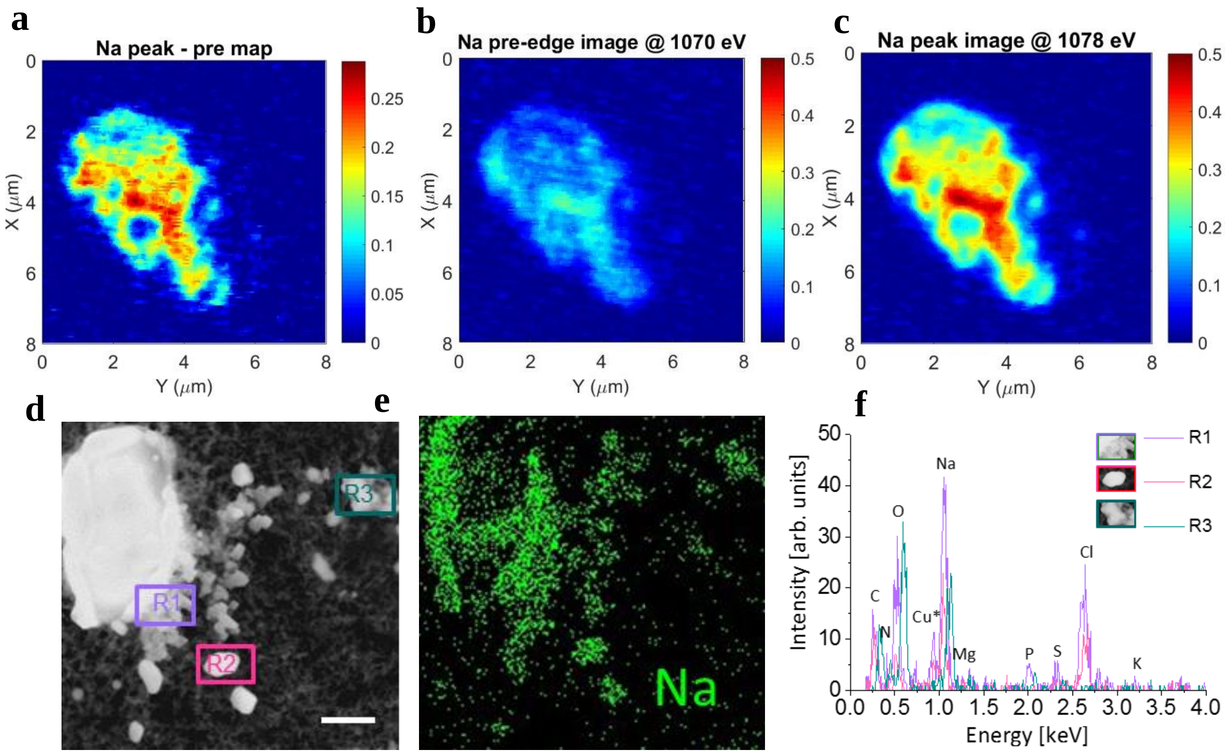
Sample #	Canopy	Sampling start	Sampling end	Duty cycle (%)	N	ME
1A	Above	01/28/2015 12:05 PM	01/28/2015 2:30 PM	100	580	4.8
1B	Below	01/28/2015 12:05 PM	01/28/2015 2:30 PM	100	472	5.4
2A	Above	01/28/2015 7:00 PM	01/29/2015 5:30 AM	75	305	6.7
2B	Below	01/28/2015 7:00 PM	01/29/2015 5:30 AM	75	366	6.1
3A	Above	01/30/2015 11:30 AM	01/30/2015 2:00 PM	100	180	8.7
3B	Below	01/30/2015 11:30 AM	01/30/2015 2:00 PM	100	220	7.9
4A	Above	01/30/2015 09:00 AM	02/02/2015 5:00 PM	75	240	7.5
4B	Below	01/30/2015 09:00 AM	02/02/2015 5:00 PM	75	320	6.5
5A	Above	02/02/2015 08:00 PM	02/04/2015 05:00 AM	75	524	5.1
5B	Below	02/02/2015 08:00 PM	02/04/2015 05:00 AM	75	614	4.7



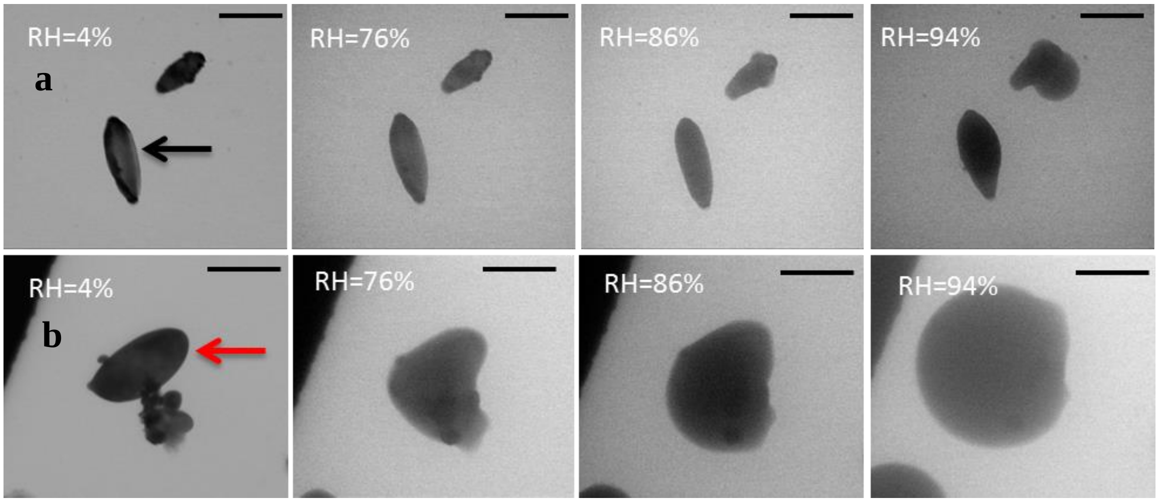
Supplementary Figure 2. Particle-type classification scheme. Total particles were classified into five groups based on their elemental compositions from CCSEM/EDX data.



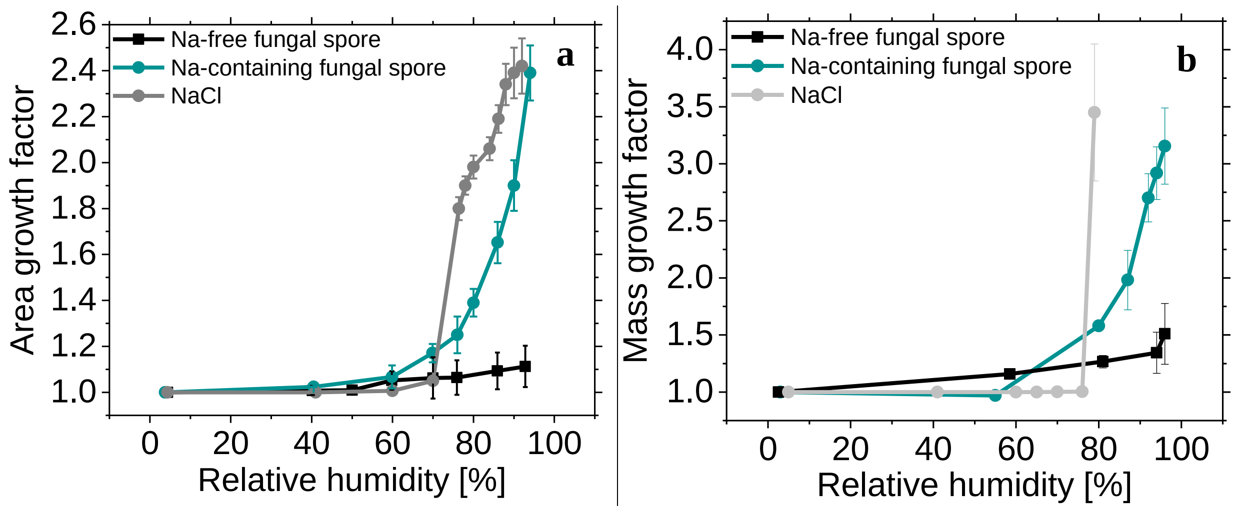
Supplementary Figure 3. Size dependence of sodium fraction in individual biological particles. Atomic weight percent of sodium in individual biological particles. Color bar indicates Cl/Na atomic ratio. For fresh particles, Cl/Na atomic ratio would be close to 1 for fresh sea salt particles.



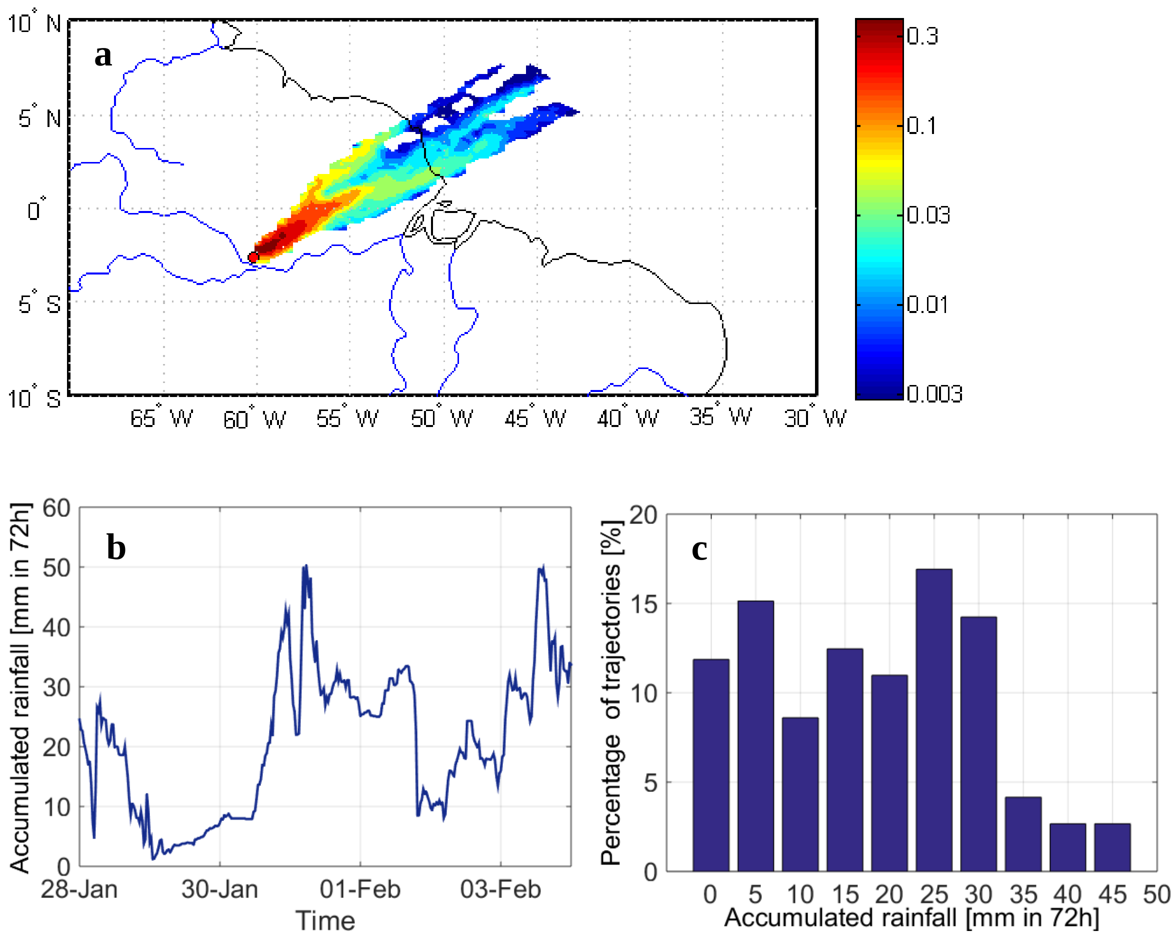
Supplementary Figure 4. Chemical composition of ruptured fungal spores. Scanning transmission X-ray microscopy images at a) Na map (optical density difference between Na K peak and pre-edge), b) Na K pre-edge (1070 eV) and c) peak (1078 eV) and. Color bars represent optical density. SEM image d) of a ruptured fungal spore and its fragments after prolonged water vapor exposure (8 hrs); e) elemental Na map; f) the EDX spectra acquired over three different regions of the particle (indicated by boxes in the SEM image) showing NaCl composition of the fragmented particles. Energies for three different regions are slightly offset for better visualization of the spectra.



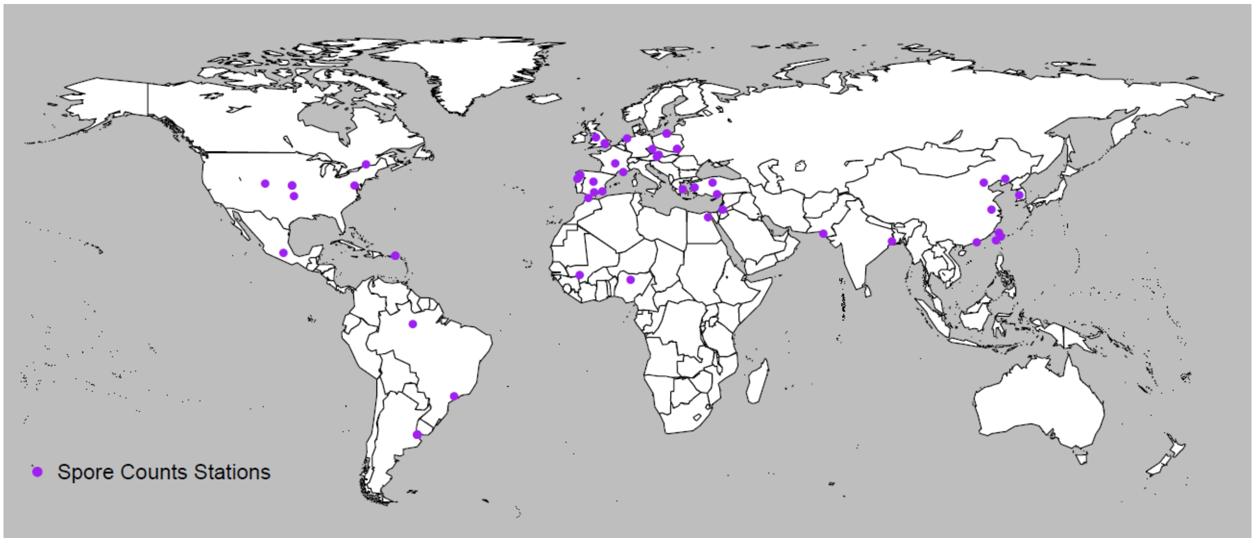
Supplementary Figure 5. Dynamic ESEM images of biological particles during hydration experiments. An environmental scanning electron microscope (ESEM, Quanta 3D model, FEI, Inc.) with a Peltier cooling stage was used for hydration experiments. a) Sodium-free biological particle is indicated by the black arrow and b) a sodium-containing biological particle is indicated by a red arrow. First, the sample on TEM grids was loaded onto the cooling stage at 278 K. Next, by controlling the water vapor in the SEM chamber, the relative humidity (RH) was increased. Scale bars are 5 µm.



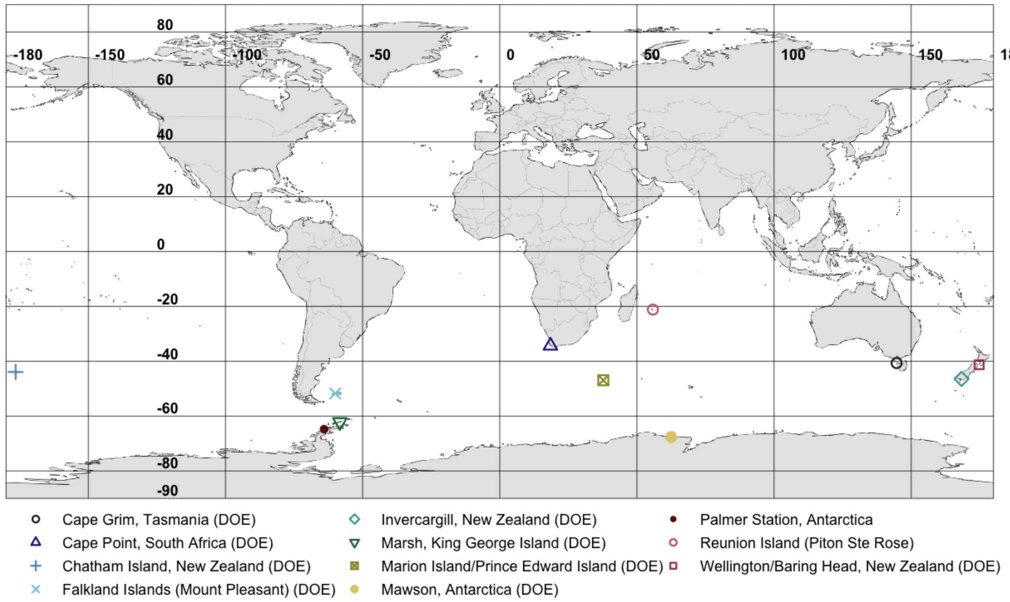
Supplementary Figure 6. Hygroscopic growth factors of fungal spore. a) Area equivalent diameter growth factors (D_{wet}/D_{dry}) of sodium-free fungal spore (black line), sodium-containing fungal spore (red line) from ESEM experiments. Gray line indicates the growth factor of laboratory generated NaCl particles. b) Mass growth factor of sodium-free fungal spore and sodium-free fungal spore particles using in situ scanning transmission X-ray microscopy (STXM) humidification experiments[7].



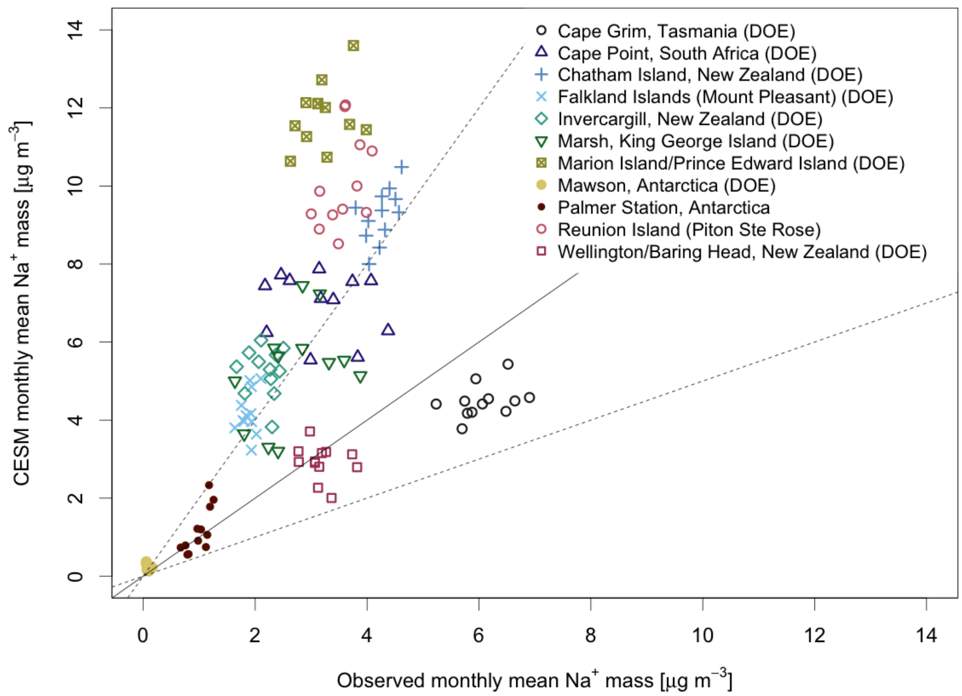
Supplementary Figure 7. Airmass transport patterns during the sampling period. Backward trajectory analysis using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model. Backtrajectories ending at the at the ZF2 site were calculated every 30 minutes within the sampling period (28 January-04 February, 2015) using HYSPLIT model, comprehending 337 trajectories in total. Backtrajectory duration was 72 h, final height was 100 m, and 1 degree GDAS (Global Data Assimilation System) meteorological fields were used in the calculations. **a,** Density of 72h backtrajectories ending at the ZF2 site (symbolized by a red circle) in central Amazon basin sampling site. The color bar represents the fraction of the 337 trajectories passing over a region. **b,** Time series of rainfall integrated for each backtrajectory. **c,** Histogram of rainfall integrated for each backtrajectory. Approximate travel time of marine air masses to the sampling site was ~2.5-3 days.



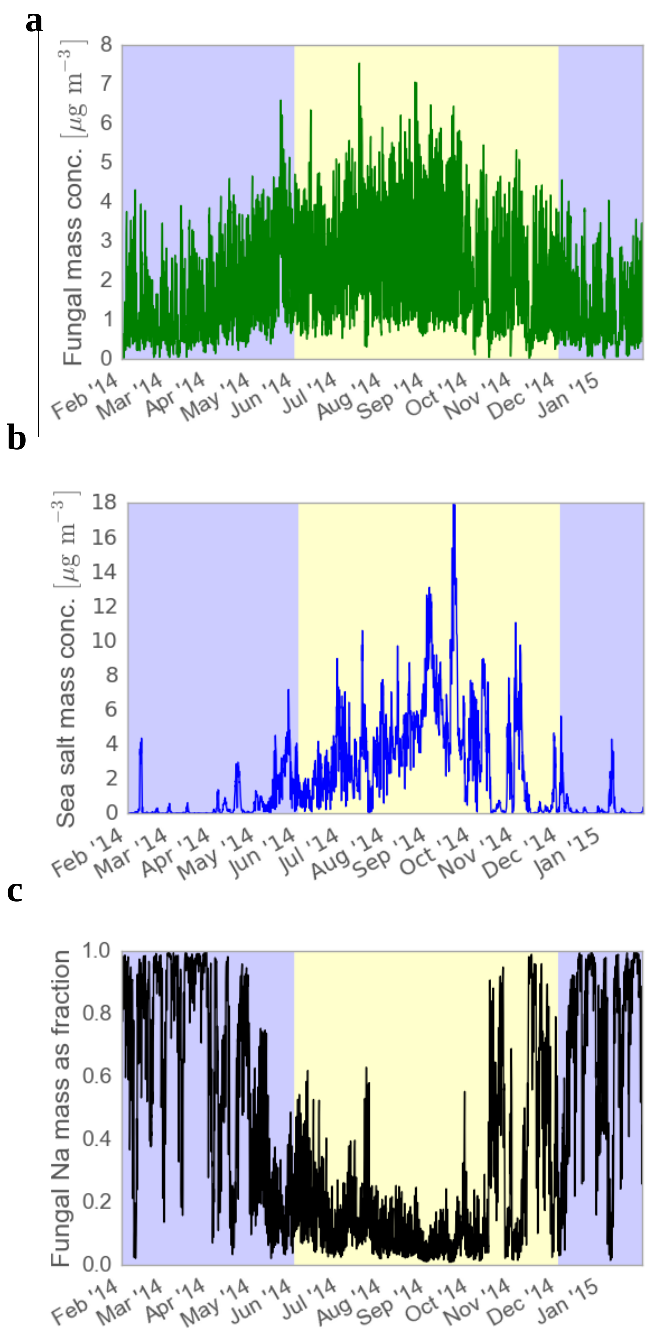
Supplementary Figure 8. Global map of spore counts and measurement site. Purple dots represent the measurement where spore counts are available and used in Fig. 3 and Supplementary Table 2.



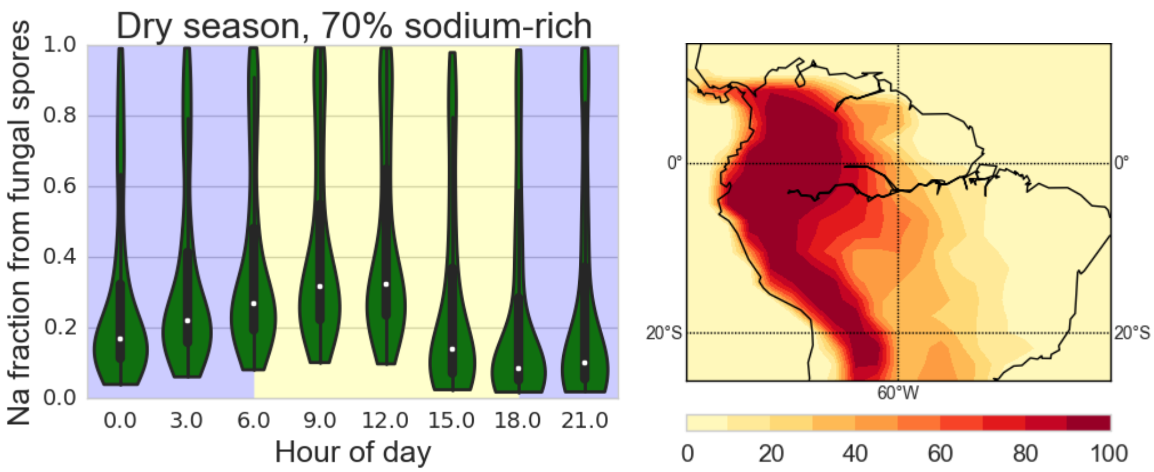
Supplementary Figure 9. Global map of sodium measurement sites, from the AEROCE/SEAREX. Symbols represent locations where measurements of the atmospheric aerosol sodium mass concentration are available and are compared with model-estimated sodium mass concentration. Note that many sites in this network sampled aerosol only for certain wind directions (sector sampling); sector-sampled sites were excluded from this comparison to remove impacts of sampling bias.



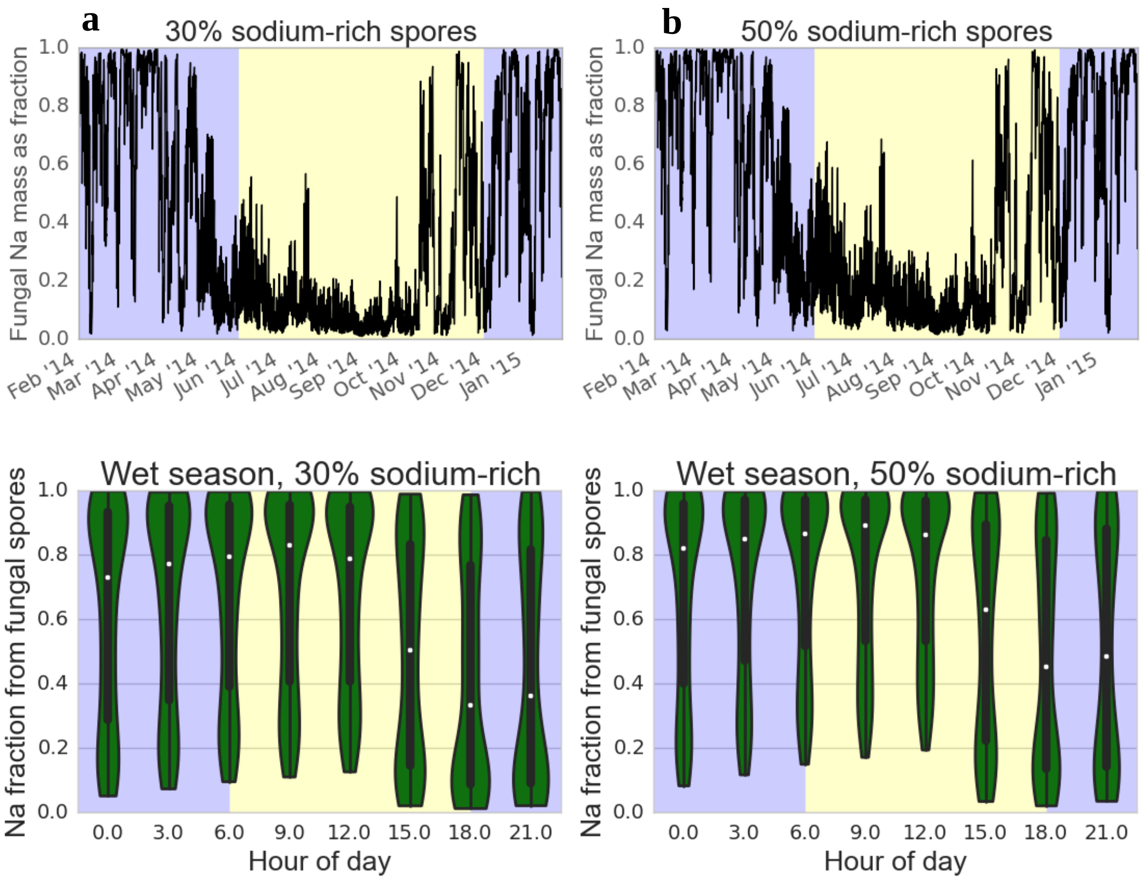
Supplementary Figure 10. Comparison of climatological monthly mean model-simulated and observed sodium mass from sea-salt. Observations are multi-year climatological monthly means of all available years from the AEROCE/SEAREX observational dataset (http://aerocom.met.no/databenchmarks.html)[8-10], collected at sampling sites shown in Supplementary Figure 8. Note that to ensure a representative comparison, this analysis has excluded sites in the measurement network that sampled aerosol only for certain wind directions (sectored sampling). The solid line represents the 1:1 line, and the dashed lines represent an over- or under-estimate of a factor of two (2:1 and 1:2).



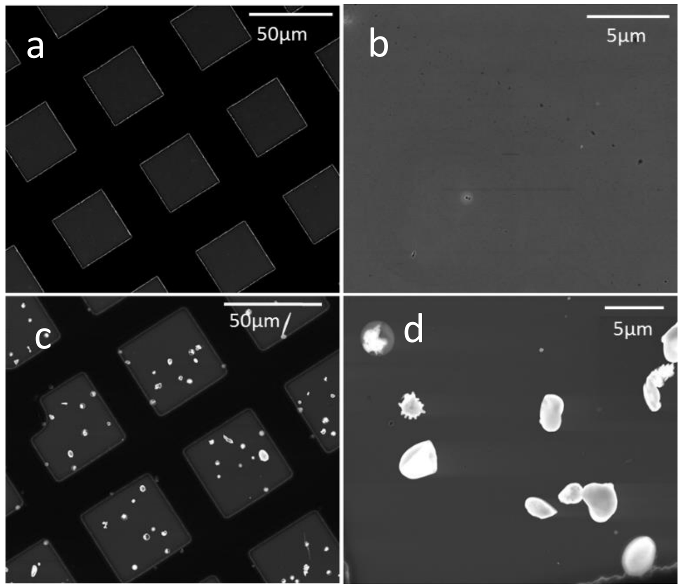
Supplementary Figure 11. Modeled time series of Na-containing particles from fungal spores and transported marine aerosol for March 2014 – Feb 2015. (a) Simulated fungal spore mass concentration. (b) Simulated sea salt mass concentration. (c) Mass fraction of Na, estimated from modeling results in airborne particles attributed to fungal spores. Background shading indicates season (blue for wet, yellow for dry). Results are from the nearest modeled grid point to the ZF2 tower, located at 2.84° S 60.0° W, in the model's lowest (near-surface) layer.



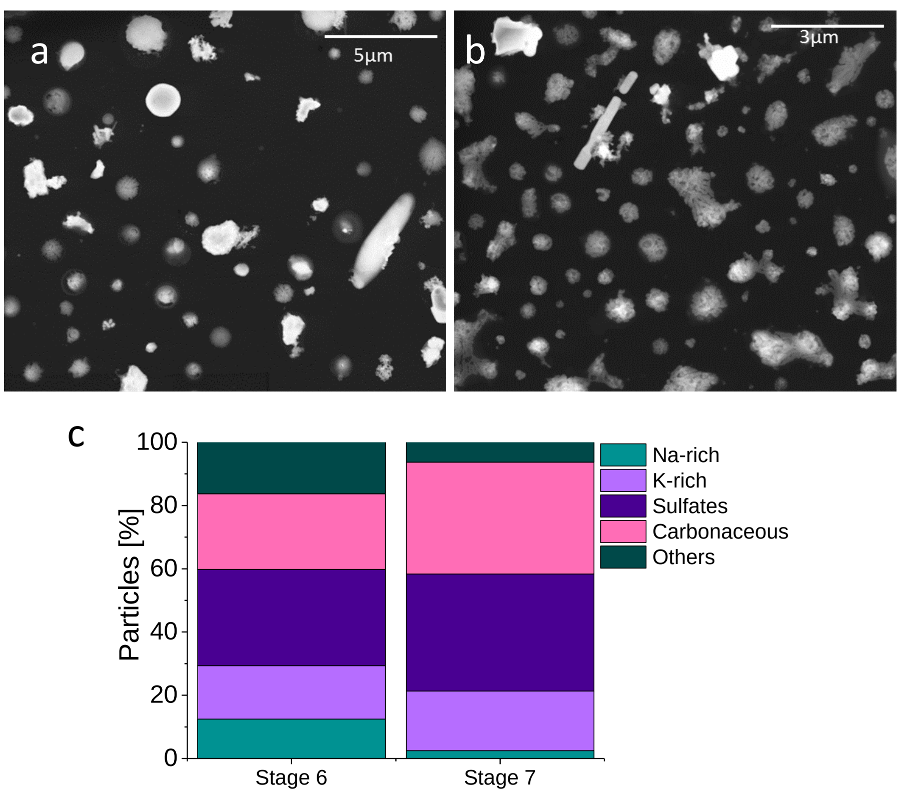
Supplementary Figure 12. Simulation of sodium contribution of biological particles to total sodium budget during dry season. Sodium fraction from fungal spores during dry season at the model's nearest grid point to the sampling site. Figure 3 shows similar data for the wet season. White dots represent the median, thick lines represent 25-75 percentile, and thin vertical lines represent the range of minimum-to-maximum values; violin plot represents a kernel density distribution of the sodium fraction values. Background shading indicates time of day, blue represents night [1800-0600] and yellow represents day [0600-1800].



Supplementary Figure 13. Modeled time series of sodium mass contribution from fungal spores assuming different number fractions of sodium-rich spores. Modeled time series (March 2014 – Feb 2015) of sodium mass percentage contributed by fungal spores assuming a) 30% and b) 50% of spores are sodium-rich.



Supplementary Figure 14. Investigation of particle contamination. SEM images show a) low magnification and b) high magnification image of blank sample; c) low magnification and d) high magnification image of collected sample.



Supplementary Figure 15. Fraction of Na-rich particles in smaller size (stages 6 and 7) particles. Representative SEM images of particles collected on a) stage 6 (0.56-1.0 µm), b) stage 7 (0.32-0.56 µm), c) number fraction of different particle classes for particles collected on stages 6 and 7. Fraction of Na-rich particles is <15%.

Supplementary Table 2. Observed mean fungal spore concentrations at various ecosystem and geographical locations. Observed concentration (Obs. Conc.), minimum concentration (Min. Conc.), maximum concentration (Min. Conc.) are reported in number of spore per m^{-3}.

No#	Longitude	Latitude	Altitude	Start date	End date	Obs Conc.	Min. Conc.	Max. Conc.	Ecosystem	Ref. no
1	2,95	45,76	1465	04.03.2003	06.03.2003	222	53	390	Mixed forest	11
2	15,76	47,71	1644	01.03.2000	30.03.2000	773	104	2031	Mixed forest	12
3	6,6	53,16	5	01.01.1981	31.12.1983	258	42	1225	Urban	13
4	-73,75	45,46	0	16.10.2002	16.10.2002	615	492	738	Urban	14
5	-0,11	51,5	64	01.06.1961	31.10.1961	7500	2000	13000	Urban	15
6	-2,98	53,4	92	01.06.1961	31.10.1961	9000	3000	15000	Urban	15
7	5,36	43,28	0	01.01.1962	31.12.1962	69	3	411	Urban	16
8	-59,4	-1,91	174	21.07.2001	21.07.2001	12476	4764	20188	Evergreen broadleaf forest	17
9	14,41	50,08	0	01.10.1988	15.12.1988	17	NA	NA	Urban	18
10	7,63	11,18	690	01.07.1959	30.07.1959	128	20	237	Croplands	19
11	-46,63	-23,55	0	01.03.2007	31.03.2007	3768	NA	NA	Urban	20
12	-5,36	35,56	250	01.01.2009	31.12.2012	1434	177	12082	Urban	21
13	22	50,03	0	01.01.2001	31.12.2002	2163	NA	NA	Urban	22
14	22,08	50,03	0	01.04.2001	31.12.2002	2119	NA	NA	Croplands	22
15	18,8	54,6	0	01.01.1998	31.03.1998	105	0	1000	Water	23
16	18,8	54,6	0	01.07.1998	30.09.1998	223	0	600	Water	23
17	18,85	54,52	0	02.08.1995	17.09.1995	19	0	122	Water	23
18	-8,38	41,51	5	01.01.2005	31.12.2007	934	NA	NA	Mixed forest	24
19	-8,65	41,18	20	01.01.2005	31.12.2007	531	NA	NA	Urban	24
20	-7,85	42,33	0	01.01.2002	31.12.2002	964	NA	NA	Urban	25
21	-64,73	18,33	0	01.07.2000	30.07.2000	45	30	60	Evergreen broadleaf forest	26
22	-64,79	18,33	0	18.07.2000	08.08.2001	21,24	18,25	61	Water	27
23	34,25	36,6	0	18.03.2002	22.10.2002	1702	100	8510	Croplands	28
24	114,26	22,33	0	01.09.2002	30.04.2003	269,5	28,5	1963	Forest	29

25	-104,86	39,75	0	01.01.1987	31.12.1995	409	NA	NA	Urban	30
26	-96,56	39,18	0	01.04.1953	30.10.1953	24499	837	48162	Urban	31
27	-96,56	39,18	0	01.12.1953	30.03.1954	715	170	1261	Urban	31
28	-95,93	36,11	0	01.01.2002	31.12.2002	24121	53	48188	Urban	32
29	-57,95	-34,91	15	01.07.2000	15.09.2000	284	NA	2000	Urban	33
30	-57,95	-34,91	15	15.09.2000	15.12.2000	395	NA	2000	Urban	33
31	-57,95	-34,91	15	15.12.2000	15.03.2001	814	NA	3478	Urban	33
32	-57,95	-34,91	15	15.03.2001	30.06.2001	715	NA	4763	Urban	33
33	-3,58	37,18	0	01.01.1994	31.12.1994	832	NA	NA	Urban	34
34	32,86	39,93	15	01.01.1990	31.12.1990	2917	17	5817	Urban	35
35	-0,35	51,81	0	04.05.1954	30.09.1954	11500	6400	10000	Croplands	36
36	121,1	25,05	0	01.03.2003	31.12.2004	2255	NA	NA	Water	37
37	120,2	23	0	01.12.2000	30.04.2001	28683	NA	NA	Urban	38
38	-8	12,65	0	01.02.2001	31.03.2001	225	80	370	Urban	39
39	127,38	36,35	0	01.01.1995	31.12.1995	3014	100	5929	Urban	40
40	121,6	23,96	0	01.04.1993	31.03.1996	4839	NA	NA	Urban	41
41	36,1	32,01	0	01.12.2008	30.09.2009	7541	3066	12017	Urban	42
42	23,71	37,98	30	01.01.1998	31.12.2001	1055	60	6328	Urban	43
43	-99,18	19,31	0	01.08.1988	28.02.1989	351	45	3195	Urban/shrub	44
44	-0,98	37,6	0	01.01.1994	31.12.1999	655	10	1301	Croplands	45
45	67	24,85	0	01.01.2010	31.12.2010	310	157	469	Urban	46
46	-3,75	40,45	0	01.01.2003	31.12.2003	609	166	1614	Urban	47
47	16,36	48,2	0	01.01.2002	31.12.2002	49	NA	NA	Forest	48
48	31,55	29,86	0	01.01.2006	29.02.2007	216	101	331	Urban	49
49	116,38	39,91	0	01.06.2003	30.05.2004	1164	23	13959	Urban	50
50	118,76	32,05	0	21.03.1998	14.07.1998	655	NA	NA	Urban	51
51	123,06	41,13	0	01.12.1990	30.11.1991	1797	NA	NA	Urban	52
52	-77,33	39,23	0	01.09.1978	01.12.1980	695	4	6885	Suburban	53
53	27,41	38,61	74	01.01.2004	31.12.2005	541	415	780	Urban	54
54	88,16	22,66	10	01.10.1996	30.09.1998	879	119	1639	Croplands	55

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