

# Natural Aerosols, sources and sinks

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# What is atmospheric aerosols??

- Primary aerosols – emitted directly into the atmosphere as particles or droplets
- Secondary aerosols – formed in the atmosphere through nucleation (new particle formation) and/or condensation on existing particles

What is the sources of natural atmospheric aerosols?

The oceans; 66 % of the Earth surface

The arid areas, semiarid to hyperarid 37% global land area

The ecosystems, e.g. the forests that cover about 30% of global land surface and the marine ecosystem in all the oceans



# Global aerosol sources (Tg/y)

Andreae et al., 2005

Global aerosol emissions			Total	Coarse	Fine	Natural fine	Anth. fine
Carbonaceous aerosols							
	Prim org	0-2 um	95		95		
		biomass burn					54
		fossile fuel					4
		Biogenic				35	
	Black carbon 0-2 um		10		10		
		open burning, biofuel					6
		Fossile fuel					4,5
	Secondary org		28		28		
		Biogenic				25	
		Anthropogenic					3,5
Sulfates			200		200		
	Biogenic sulfates					57	
	Volcanic sulfates					21	
	Anthropogenic sulfates						122
Nitrates			18		18	9	9
Industrial dust			100	70	30		30
Sea salt			10130				
	d<1				180	180	
	1-d-16			9940			
Mineral dust			1600				
	<1 um				165	165	
	1-2.5			496			
	2,5-10 um			992			
<b>Total</b>			<b>12181</b>	<b>11498</b>	<b>726</b>	<b>492</b>	<b>233</b>





**A breaking wave seen through a camera**

**The breaking wave entrain air into the water, forming a bubble cloud**

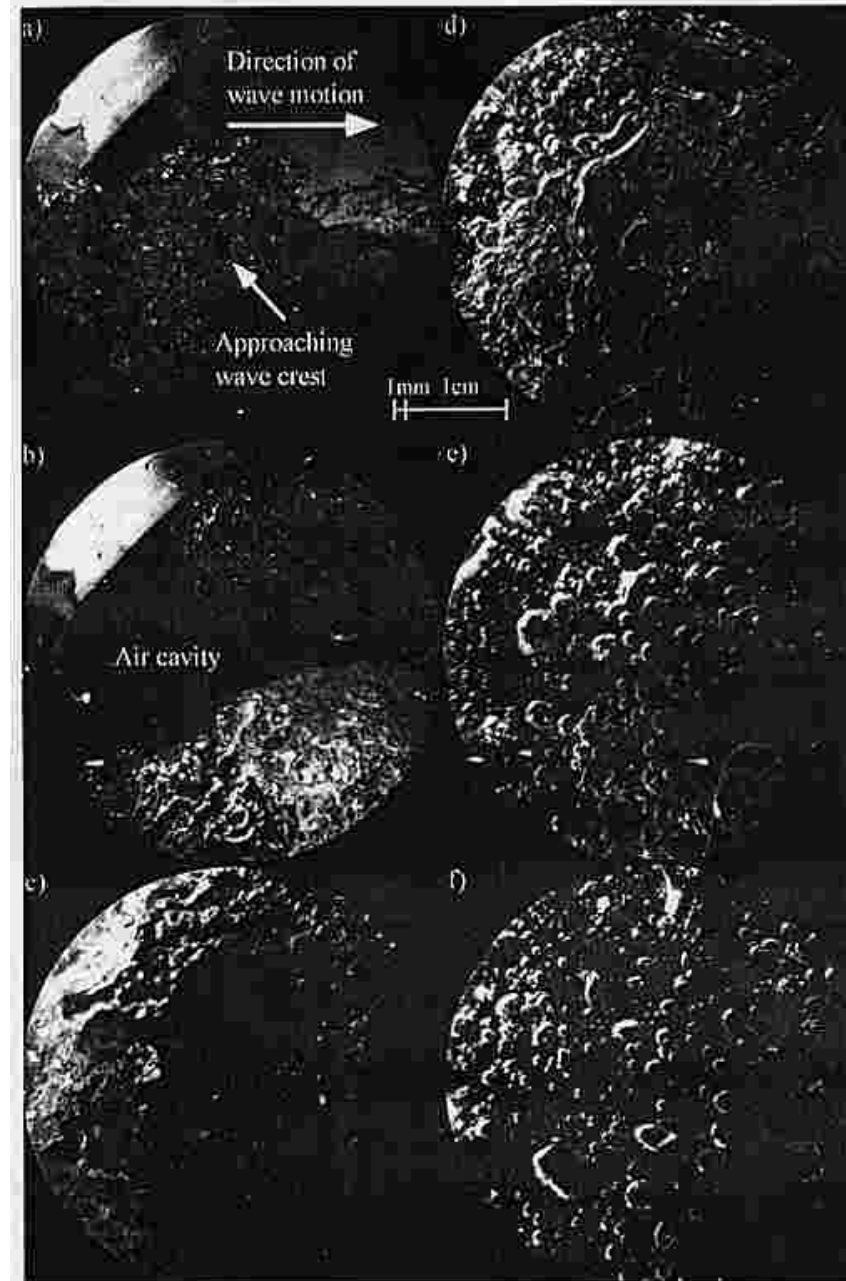
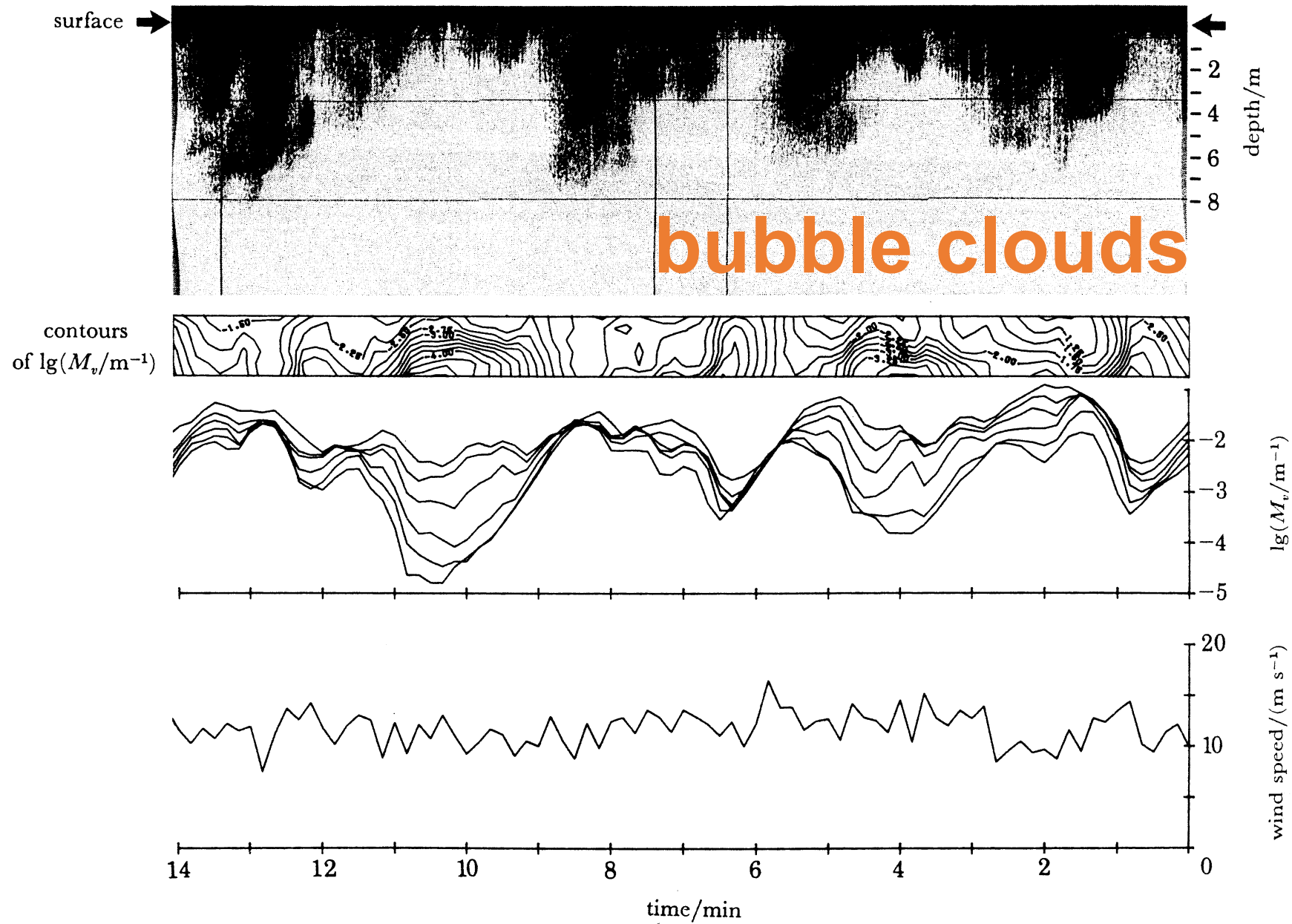


FIG. 7. Images taken in a breaking wave crest showing the crushing of an air cavity. Successive images were taken every 90 milliseconds. (a) The approaching wave crest. (b) The air cavity trapped by the overturning wave crest. (c)-(f) The evolving air-water mixture behind the wave crest.

FIGURE 11. Sonograph,  $M_v$ , and wind records from Oban in southwesterly winds:

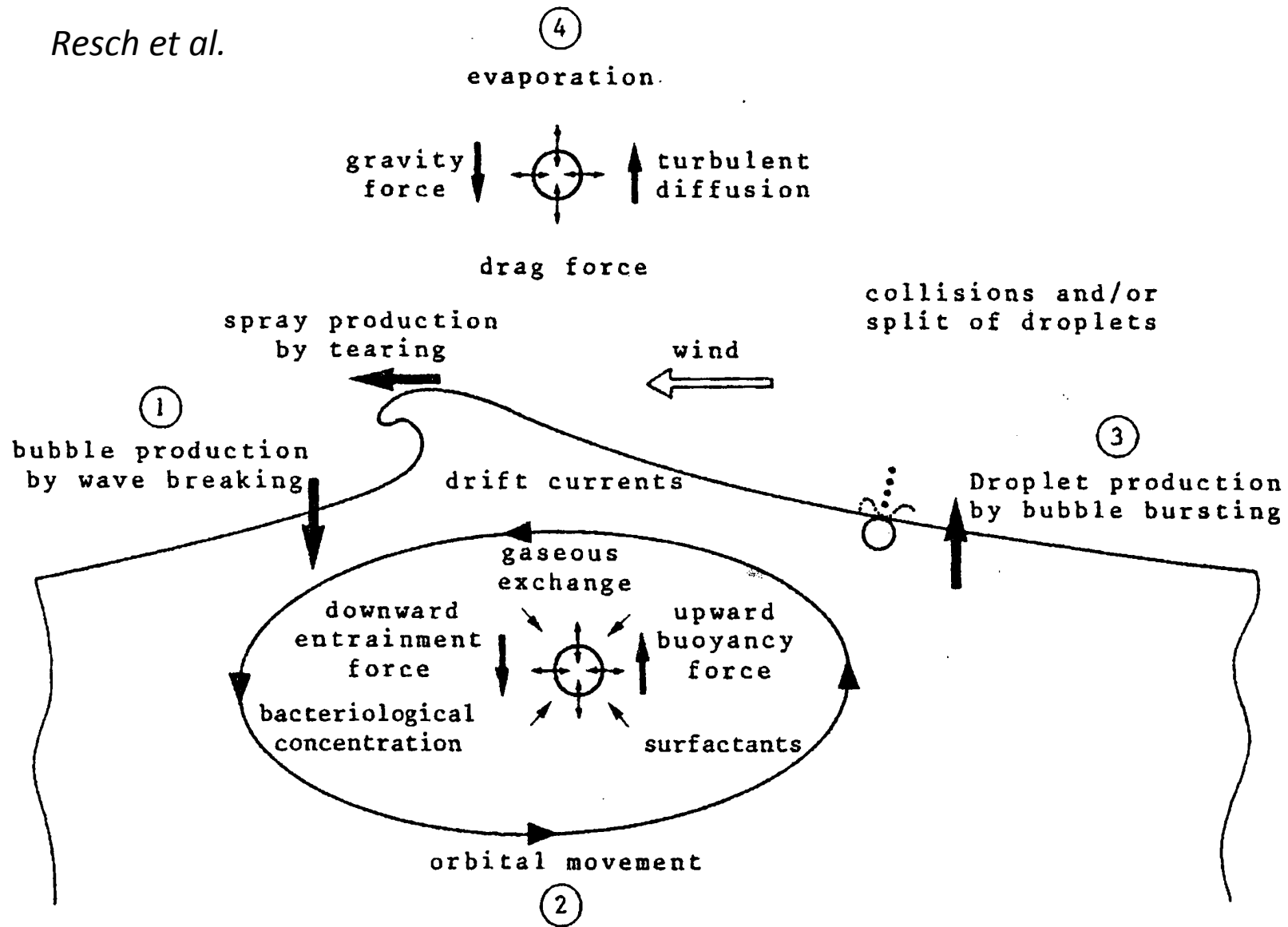


Fig. 2. Physical processes leading to the production of marine liquid aerosols responsible for air-sea particulate exchanges. Circled numbers refer to chronological steps indicated in Figure 1. We are mainly interested in this study by step 3. The ultimate goal is to propose a model for the transfer function between air bubbles in seawater and liquid aerosols in the atmosphere.

**The established conceptual model:  
There are Jet and Film drops and that form large and  
small particles respectively**

RESCH ET AL.: MARINE LIQUID AEROSOL PRODUCTION FROM BURSTING OF AIR BUBBLES

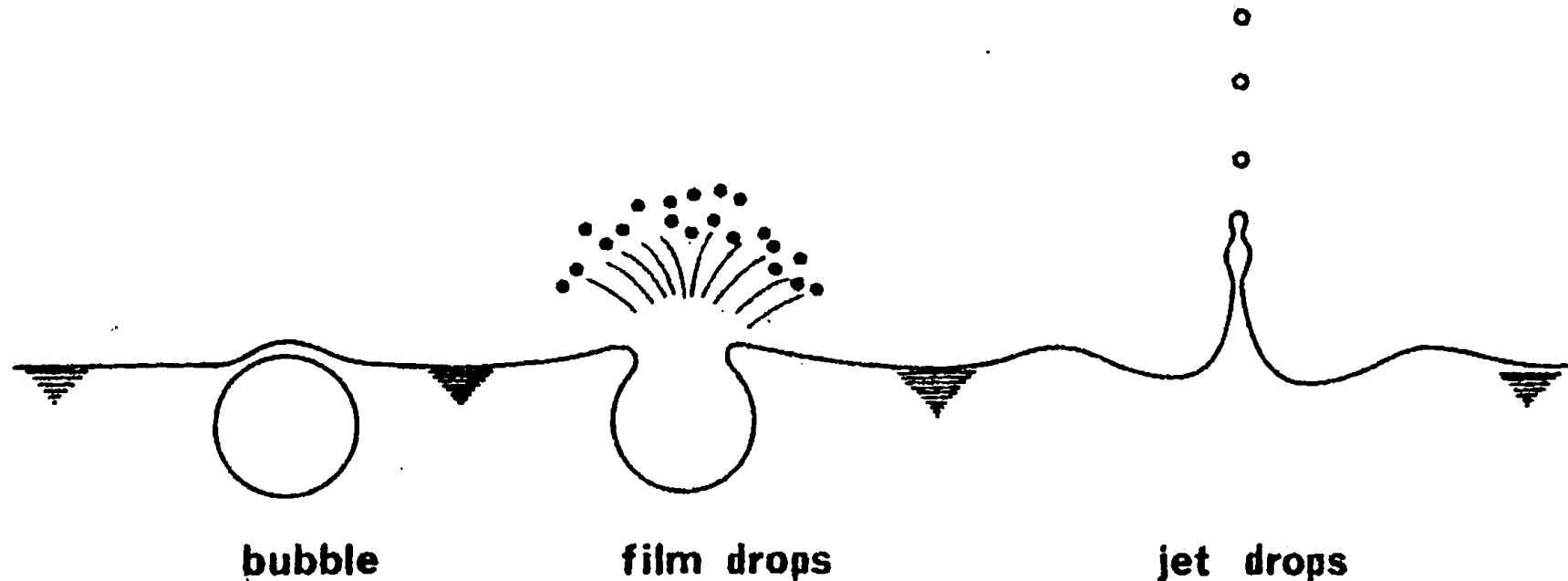


Fig. 3. Schematic representation of the production of film drops and jet drops by bubble bursting. In a time sequence the onset of film cap breaking is detected after  $150 \mu\text{s}$ , film drops are detected after  $300 \mu\text{s}$ , and jet drops appear between 1 and 30 ms after the rupture of the bubble cap.

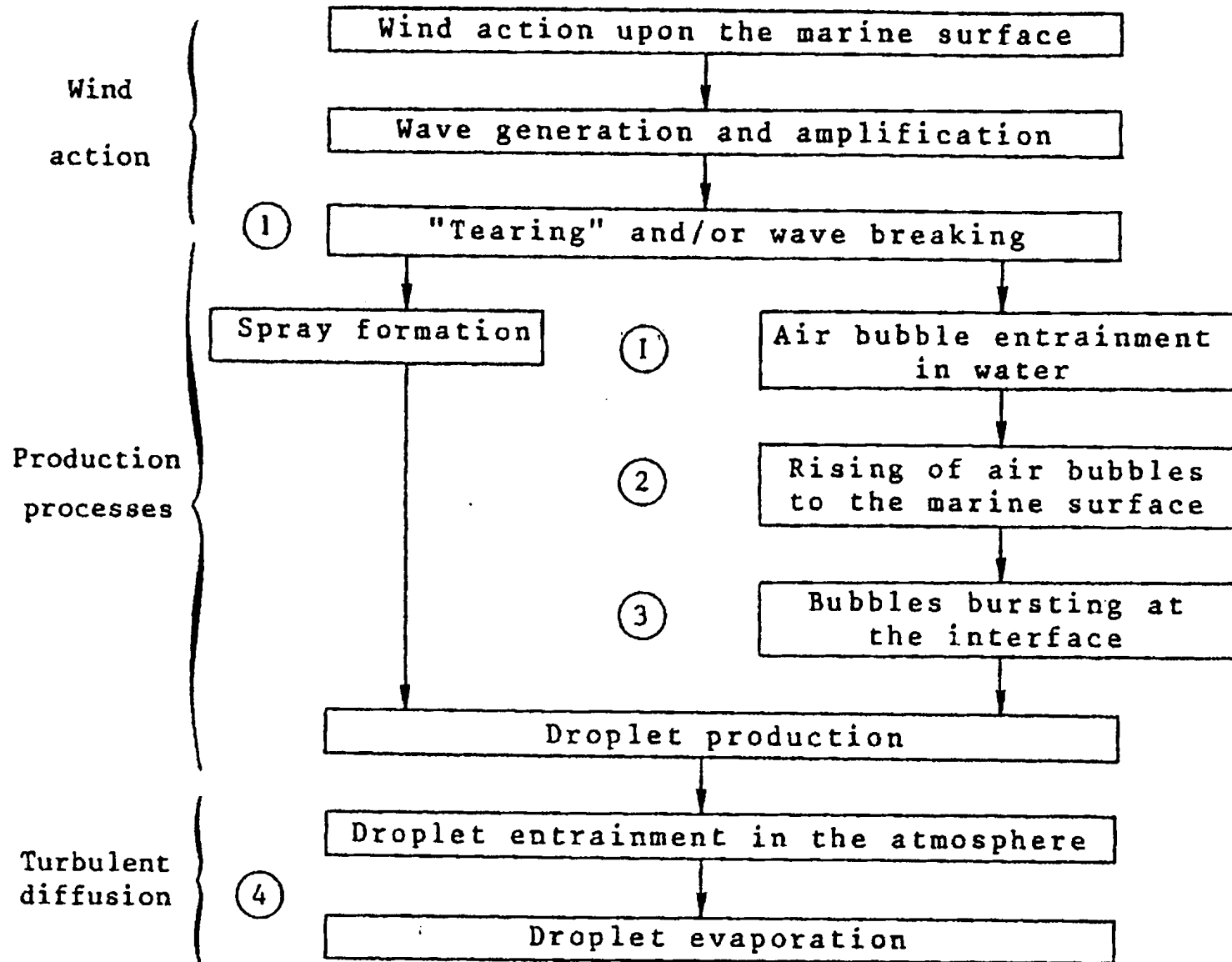


Fig. 1. Various steps involved in the process of air-sea particulate exchange.



Whitecap  
cover  
increase  
with the  
wind  
speed

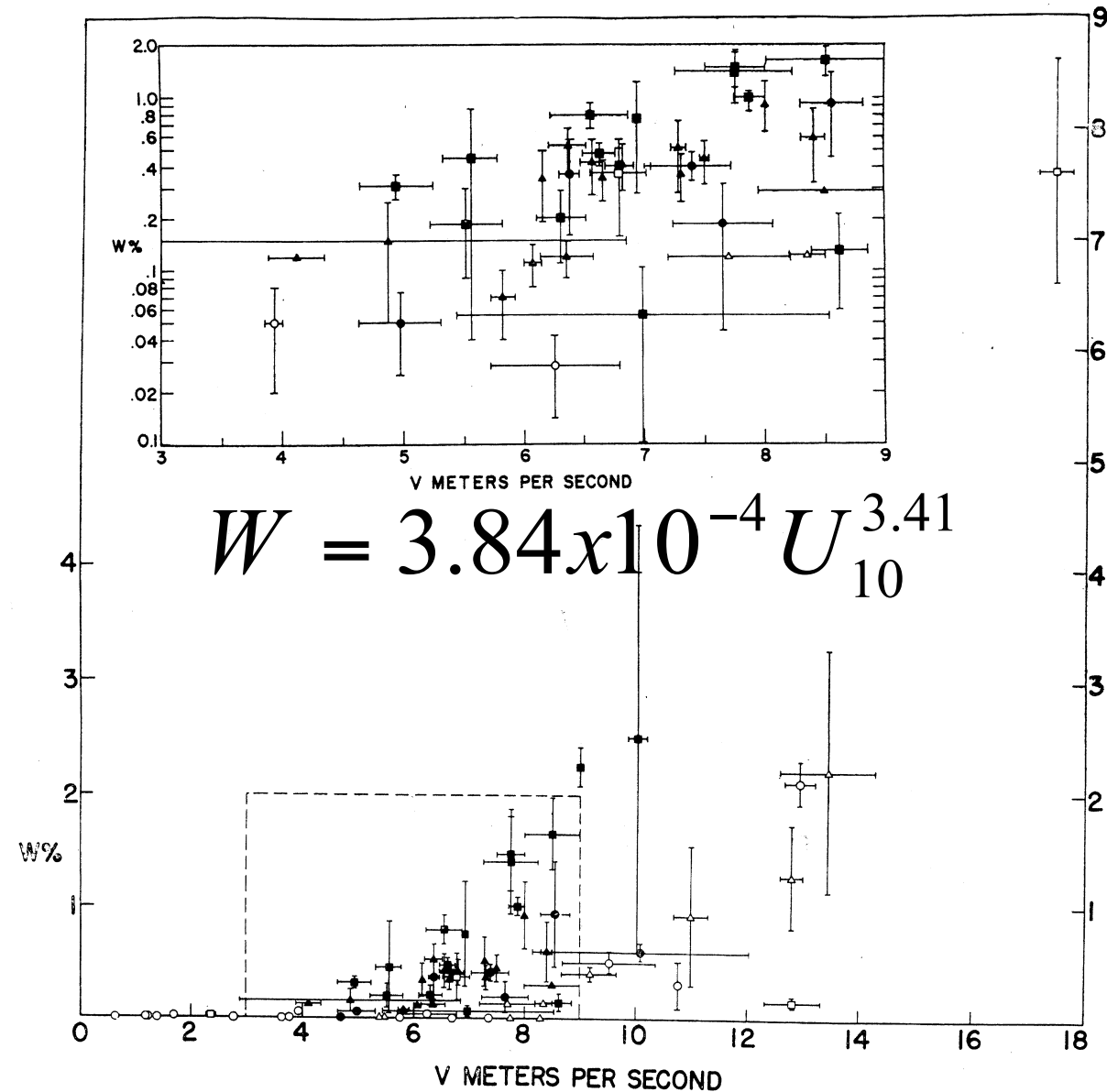


FIG. 1. Percent oceanic whitecap coverage ( $W$ ) vs 10 m elevation wind speed ( $V$ ): squares, observations when atmosphere was thermally stable; triangles, stability near neutral ( $-0.4\text{C} < \Delta T < 0.6\text{C}$ ); circles, thermally unstable. Filled symbols represent BOMEX results, open symbols all other results (see Table 1). Vertical bars indicate standard deviations from mean values. Horizontal bars span range of wind speeds measured during observation periods. Insert shows semi-log representation of data from within box formed by dashed line.

Monahan 1971

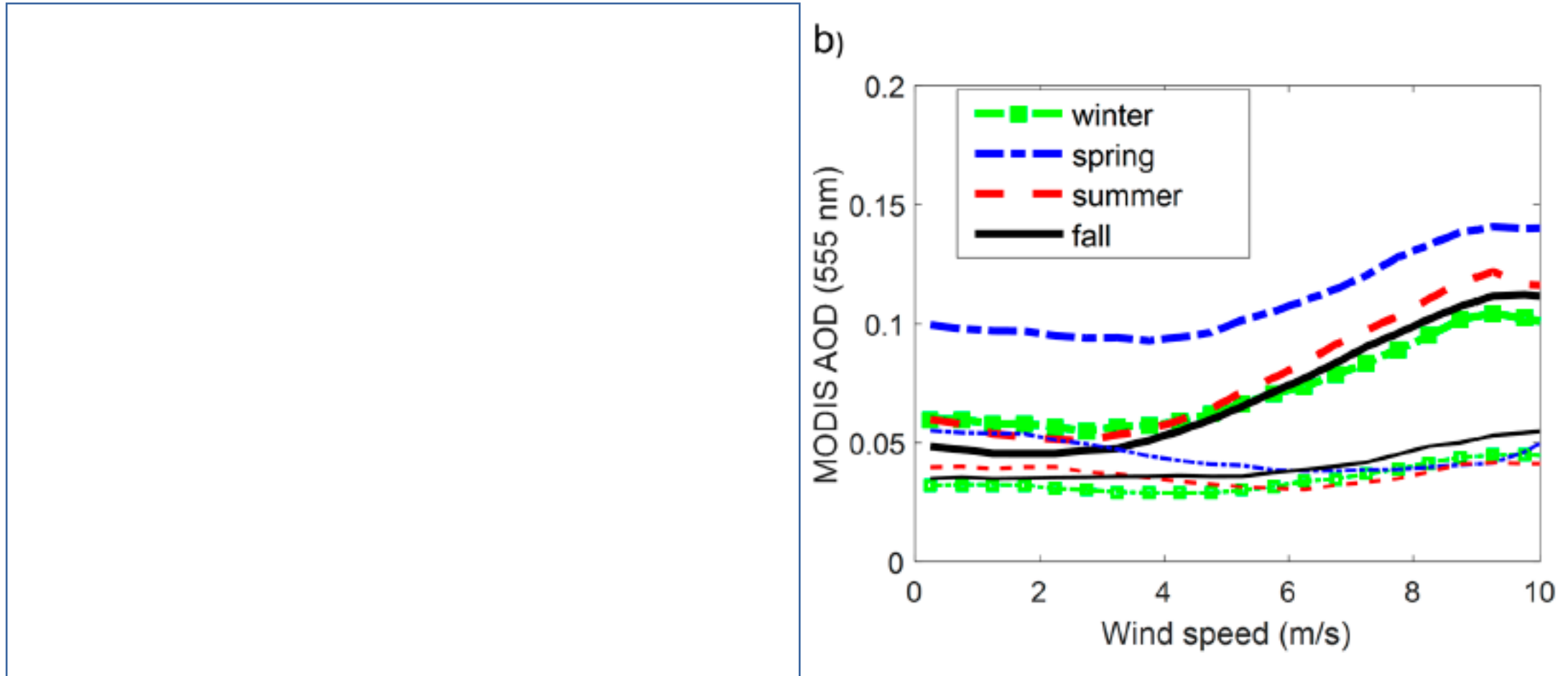
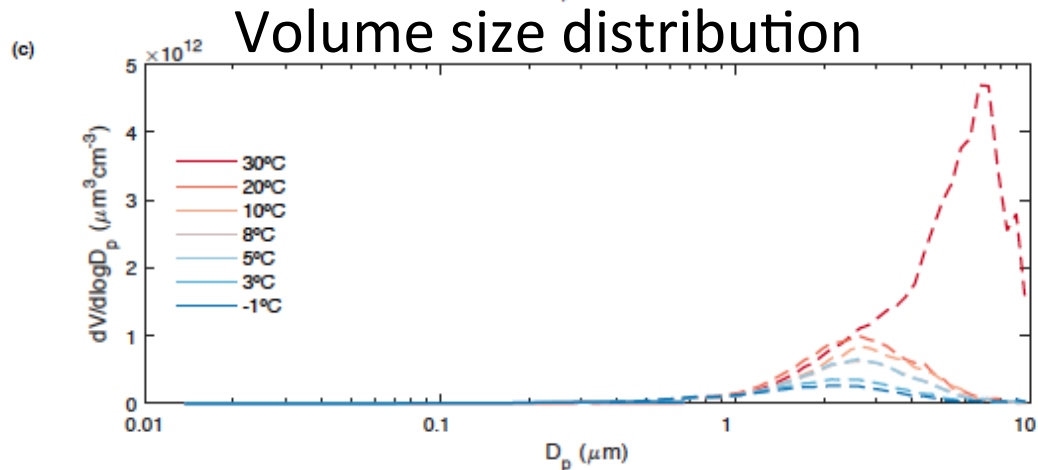
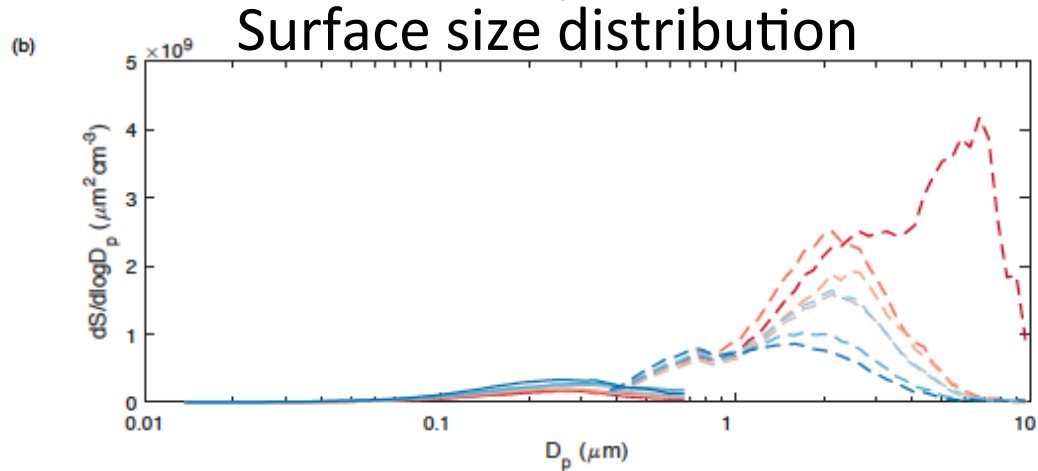
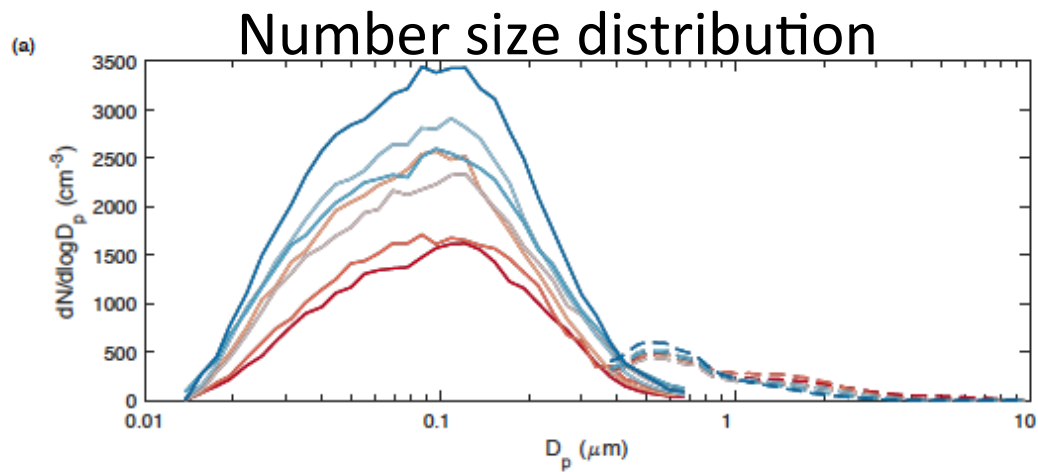


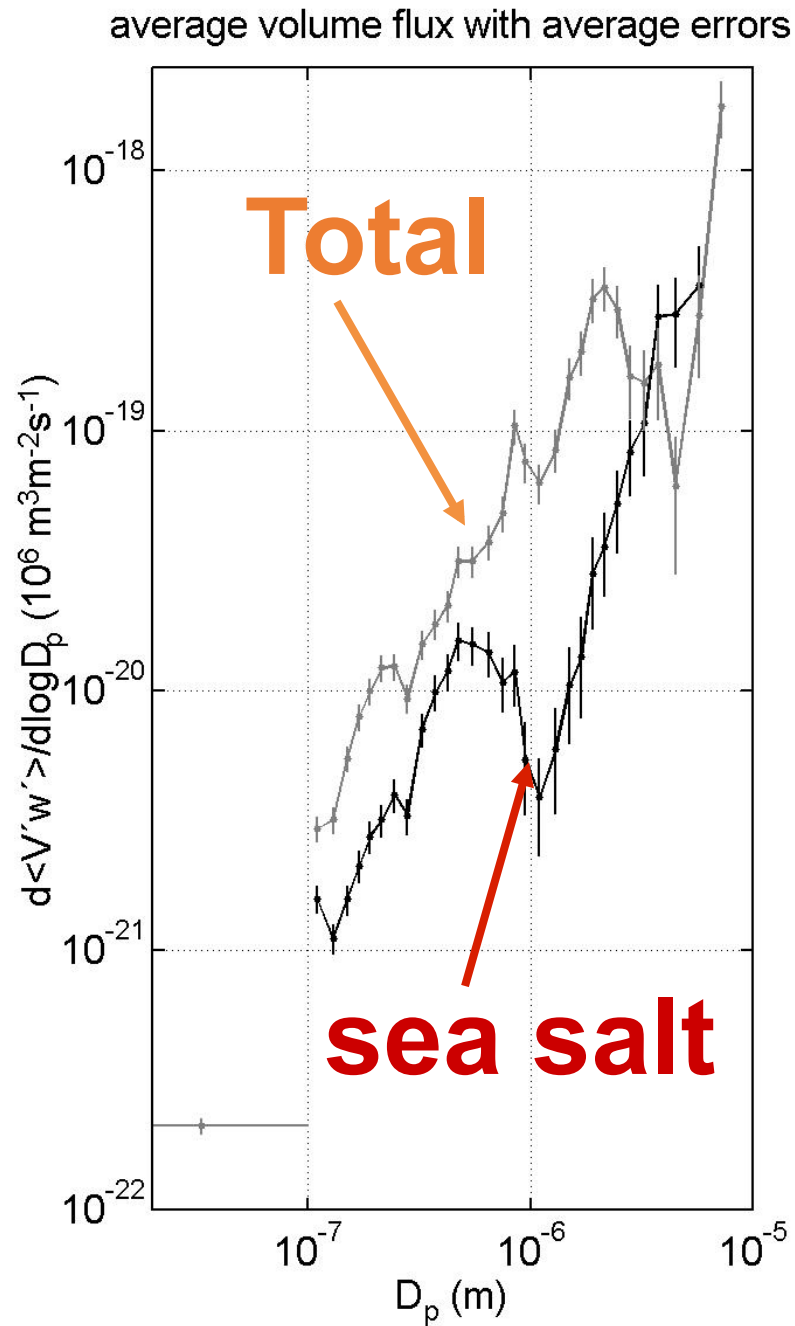
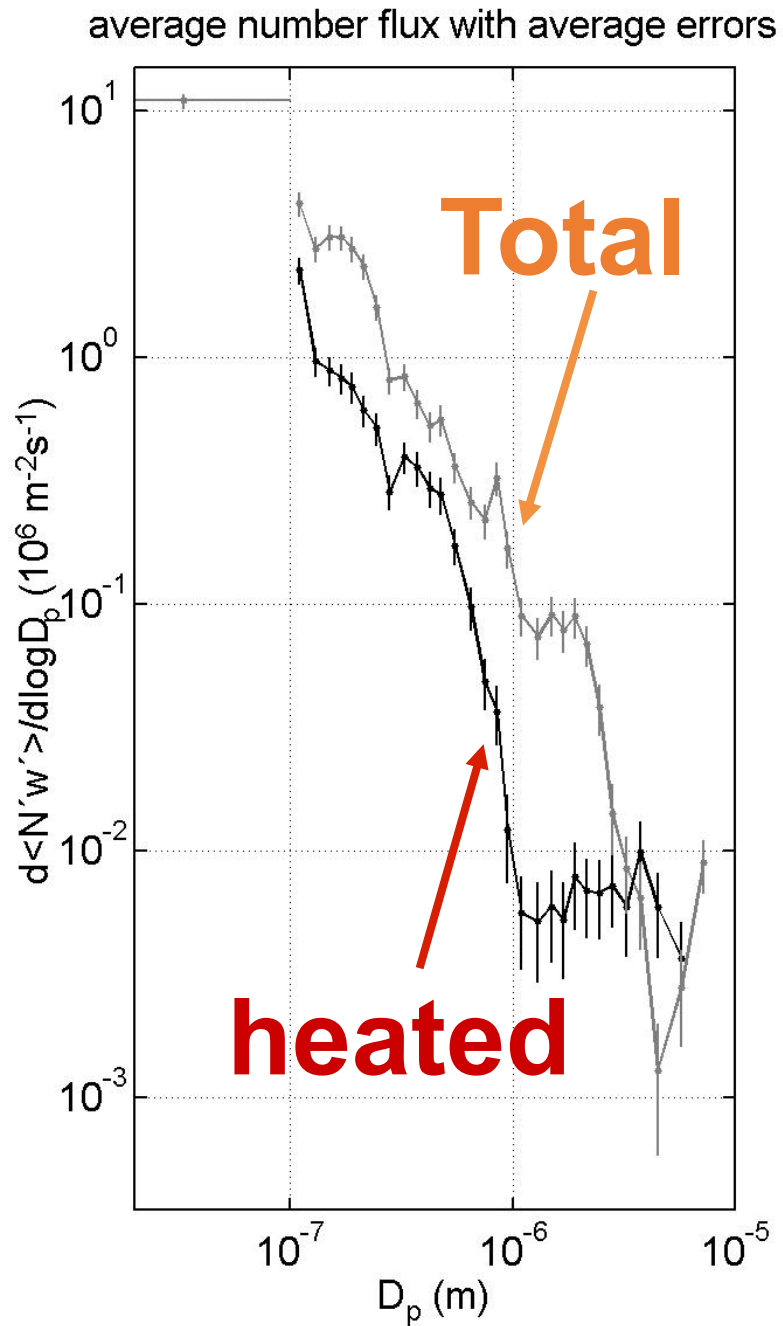
Figure 6. (a) Mean Ångström exponent, derived from MODIS Aqua observations over the period 2003–2016, versus surface wind speed and subdivided according to season; (b) MODIS Aqua fine mode adjusted median AOD (thin lines) and total adjusted median AOD (bold lines) obtained for different seasons of the period 2003–2016 versus surface wind speed.



Laboratory experiment simulating breaking waves with artificial sea water at different temperatures (-1 to +30 C)

The solid lines represent the DMPS measurements ( $D_p < 0,7 \mu\text{m}$  electrical mobility diameter), while the dashed lines show the OPS data ( $D_p > 0:35 \mu\text{m}$  optical equivalent diameter when  $m = 1:54-0i$ ).

Size dependent number flux



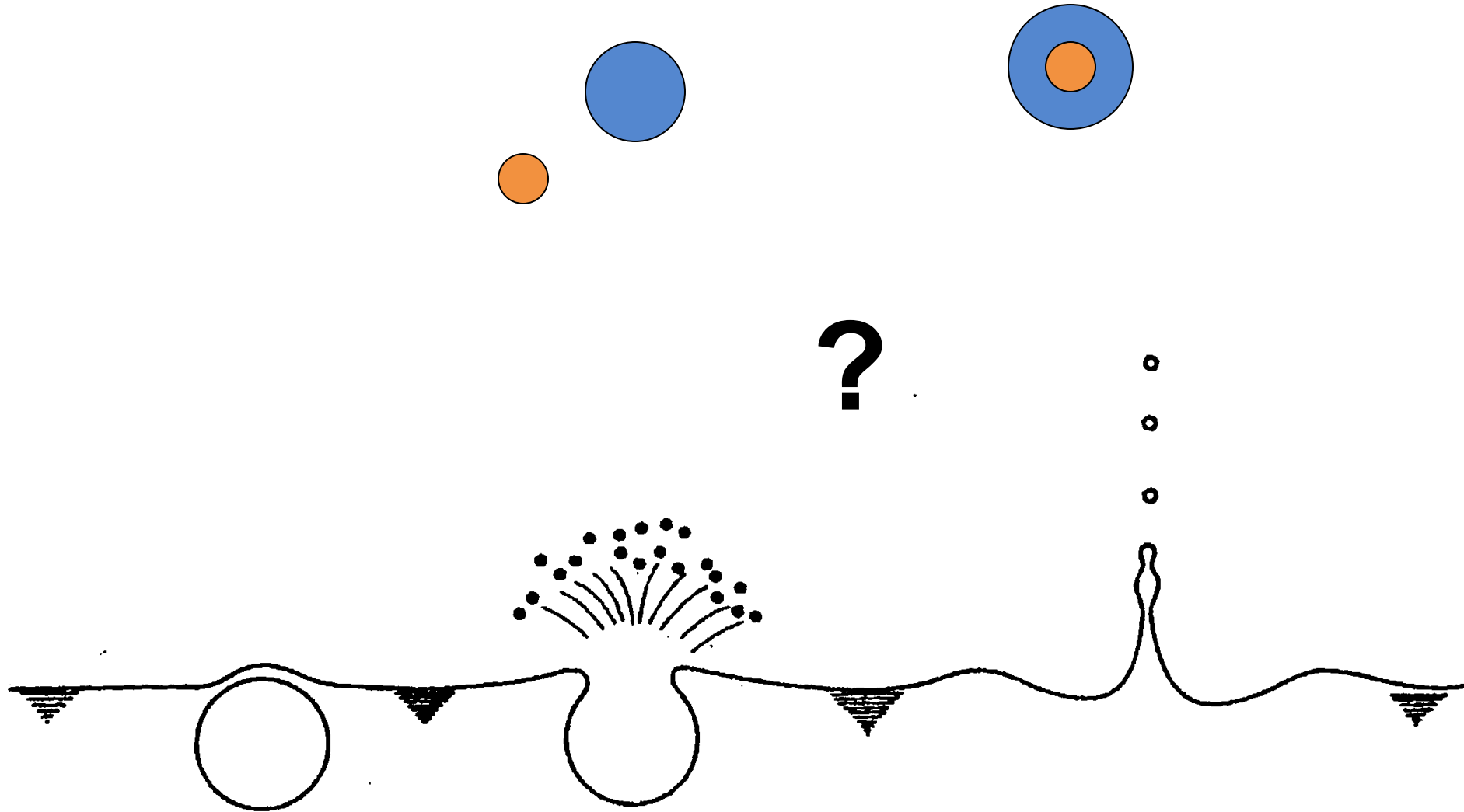
Size dependent volume flux

When heating the natural sea spray particles, many disappear or they shrink.

What is vaporized?

D. Nilsson, pers com.

**Is the internal/external mixing a result of the initial source processes? How?**



# What is happening with the droplet when injected into the air.

- Will it raise to higher altitudes? How big is it?
- It will dry, but does it form solid crystals?
- Interaction with gases, what gases?
- Don't forget the marine ecosystem, emits pre-cursor gases (DMS) and microorganisms

The oceans is not a uniform pool of water and the sea spray particles are shaped by a complex set of processes



More cloud  
condensation  
nuclei



Elevated  
 $\text{SO}_2$   
concentration



Elevated  
DMS  
concentration



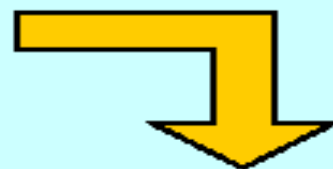
Enhanced  
DMS  
production



Enhanced  
phytoplankton  
growth



Ocean  
warms



Increased Earth albedo  
more sunlight reflected



Charlson, Lovelock, Andreae, Warren

# CLAW hypothesis Negative feedback loop

# Summary on sea spray

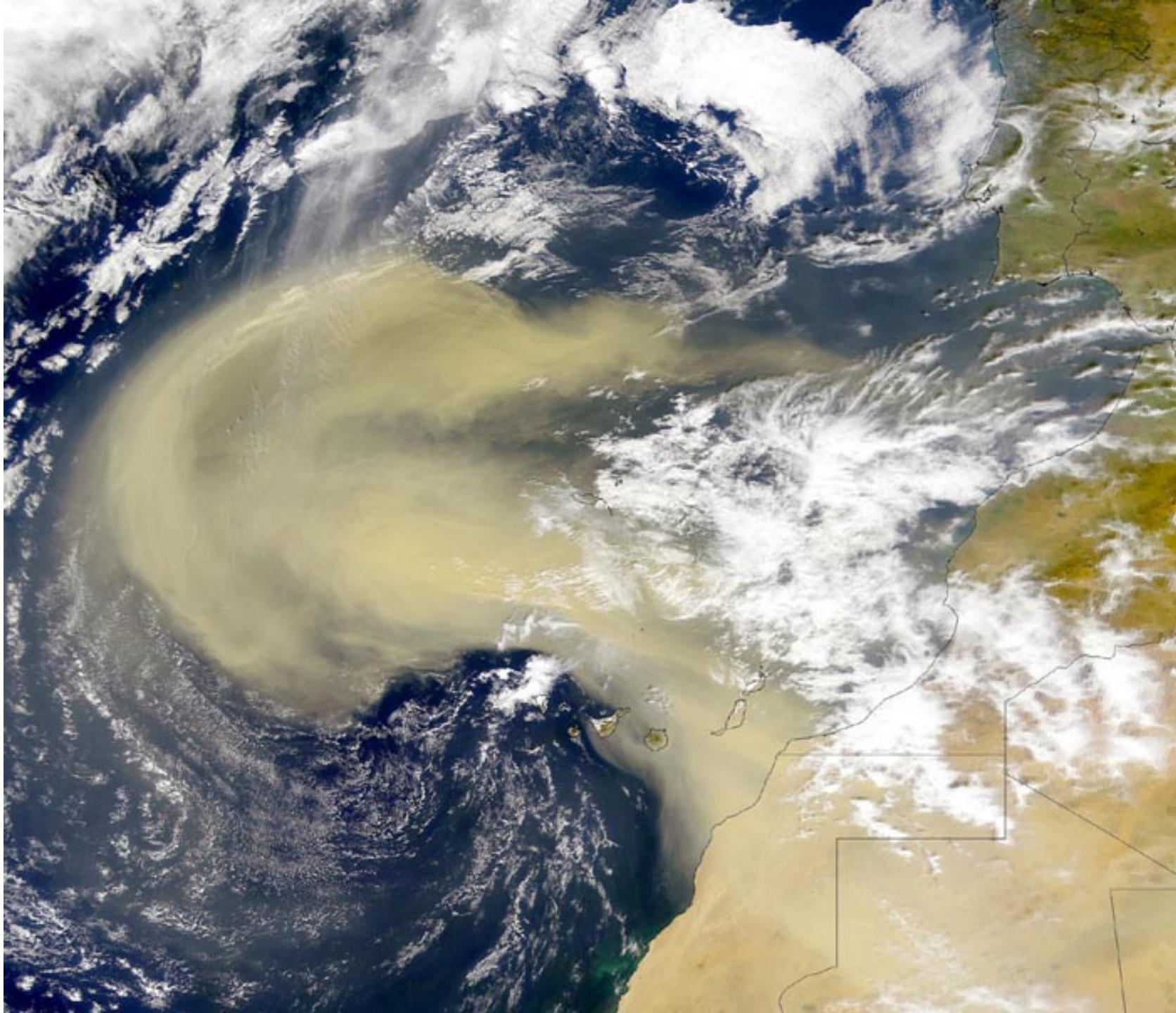
- The largest global source
- Several formation mechanisms, jet droplet, film droplet and tail droplets
- Number dominated by fine droplets, mass by large droplets
- Depend on salinity, temperature, windspeed and probably surface film (surface tension and viscosity)
- Contain more, e.g. organics, than sea salt
- Chemical mixture in the droplets different due to formation mechanism
- Gas absorption in the droplet, liquid phase reaction
- Source strength very difficult to estimate, (AeroCom show factor 10)

# Global aerosol sources (Tg/y)

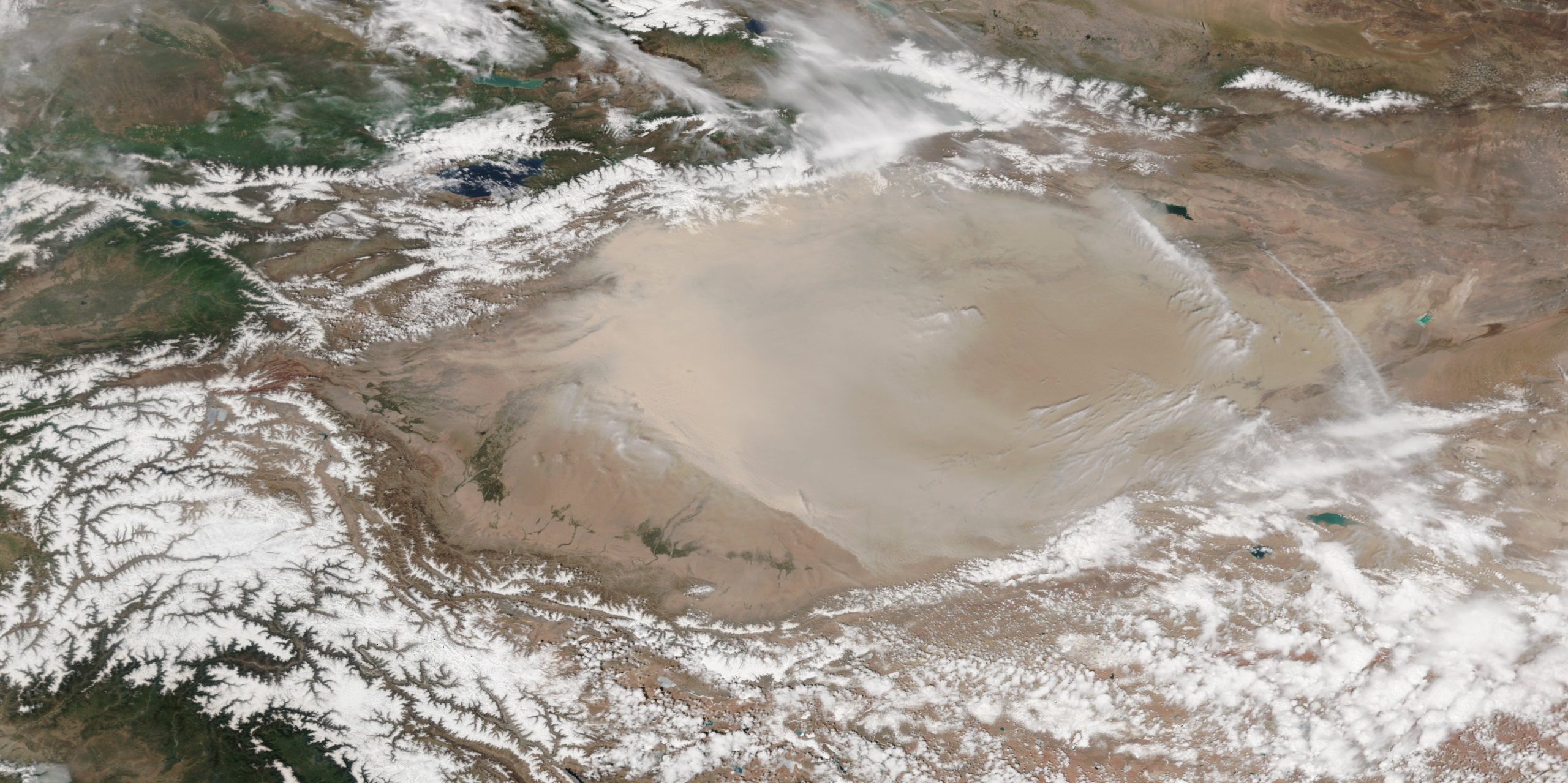
Andreae et al., 2005

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Dust

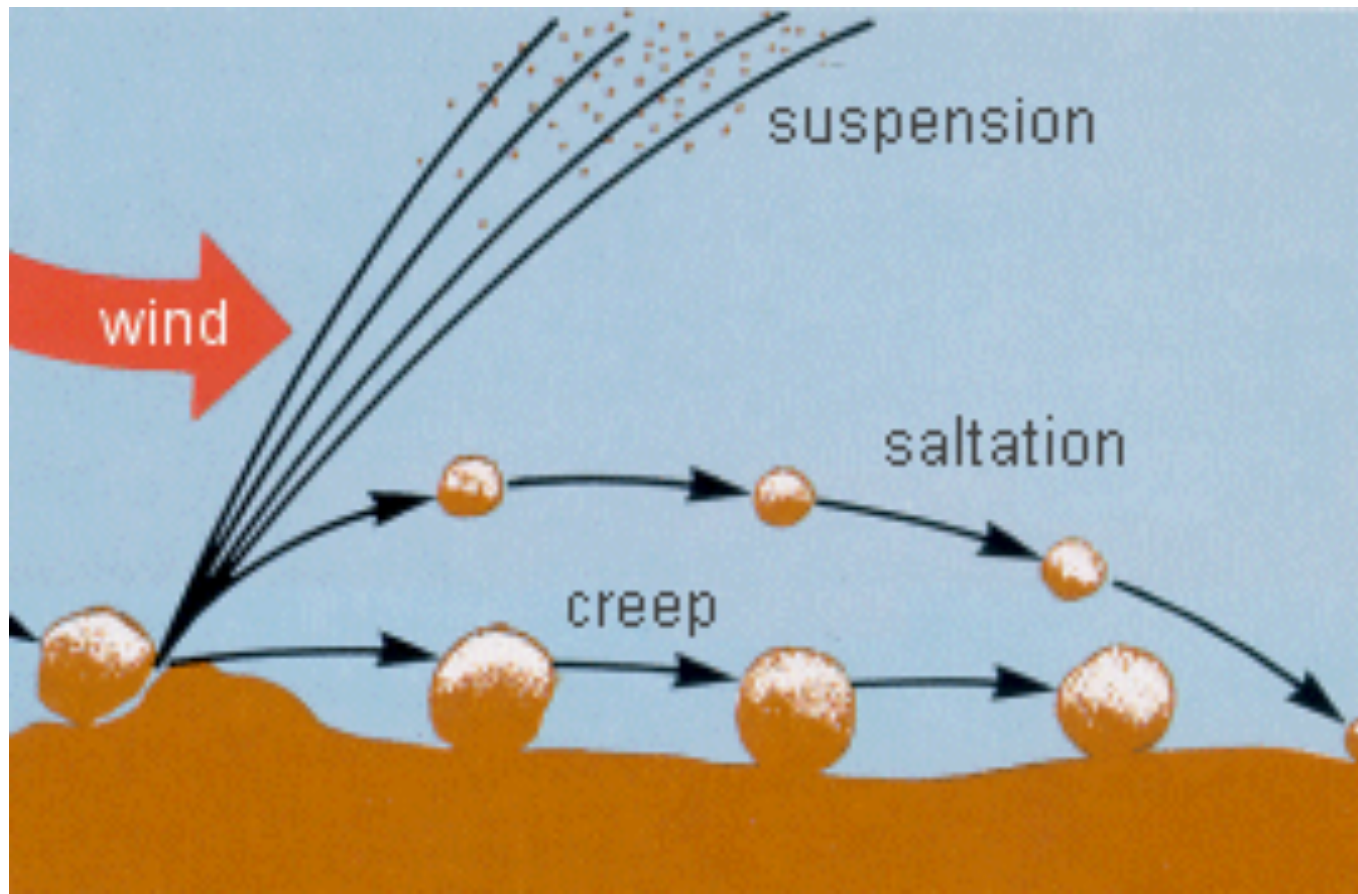






This true-color image of northwestern China's Taklimakan Desert from Suomi NPP shows a dust cloud over a light background. The Taklimakan is among the handful of well-known dust-producing regions around the globe.

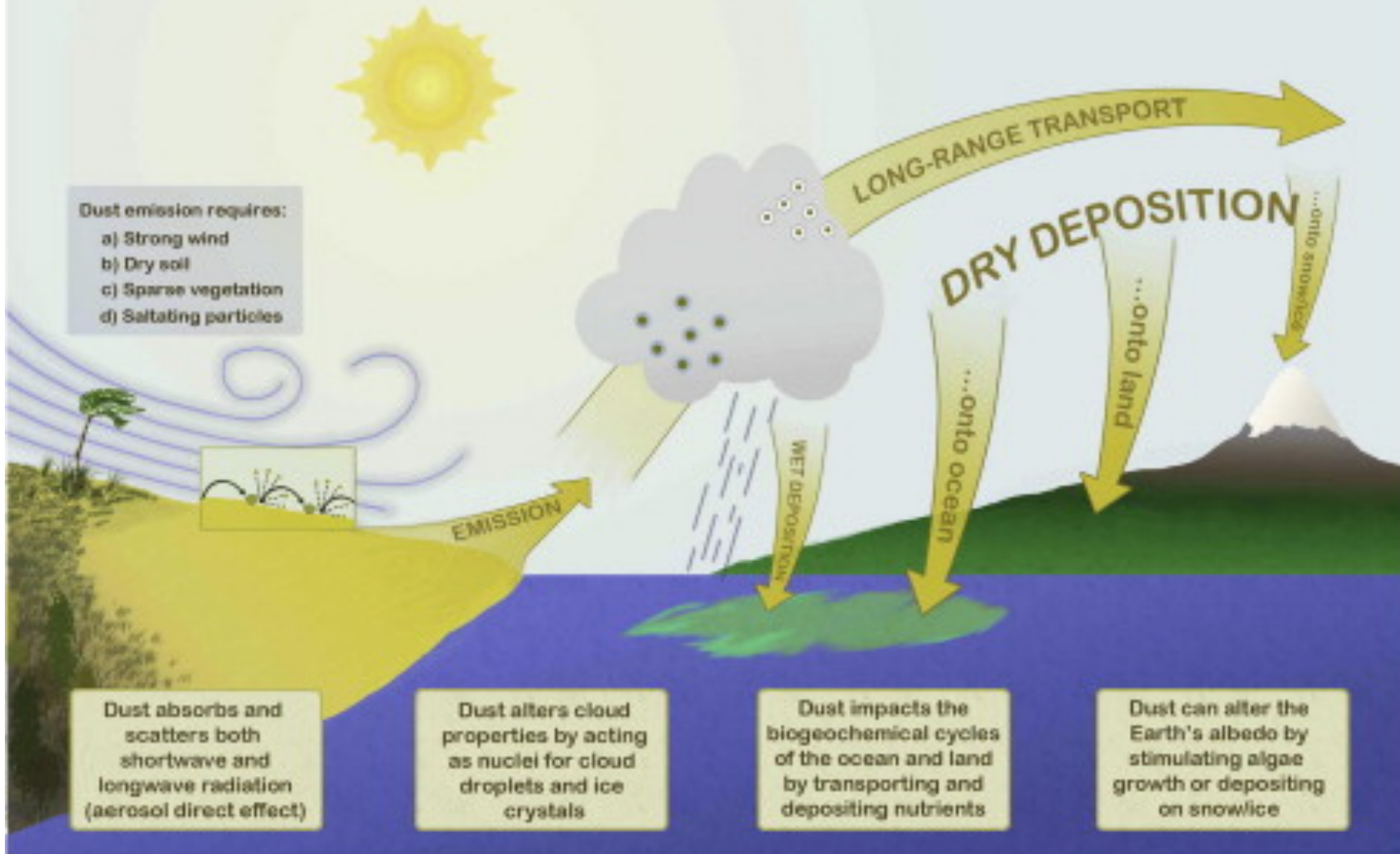






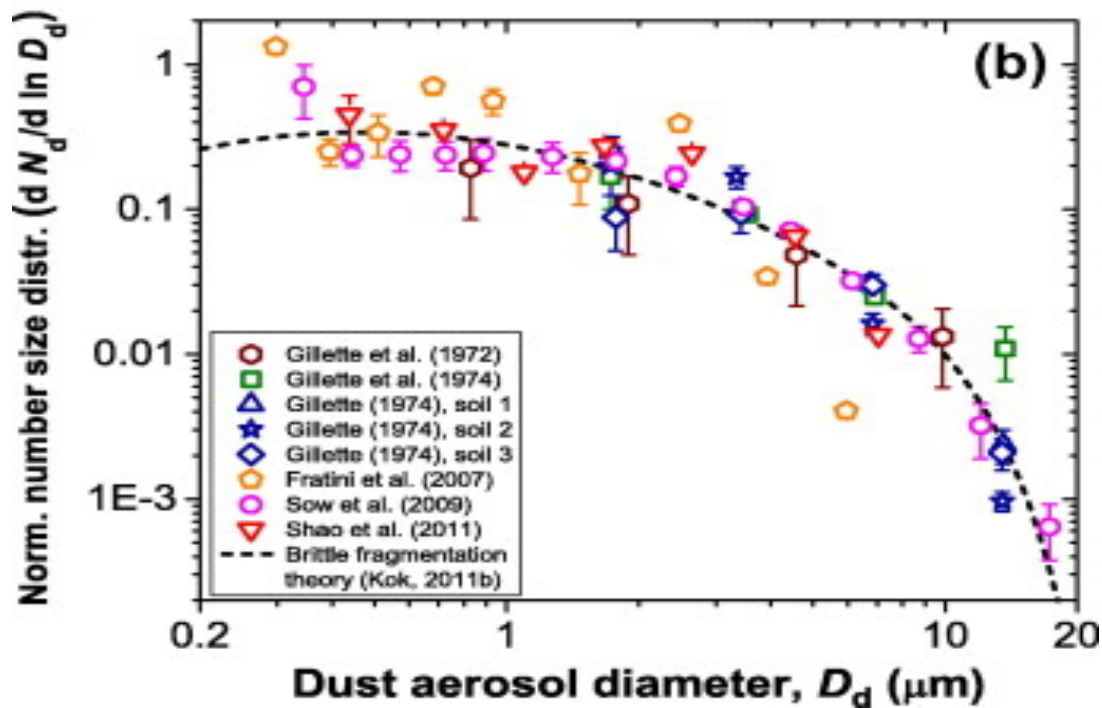
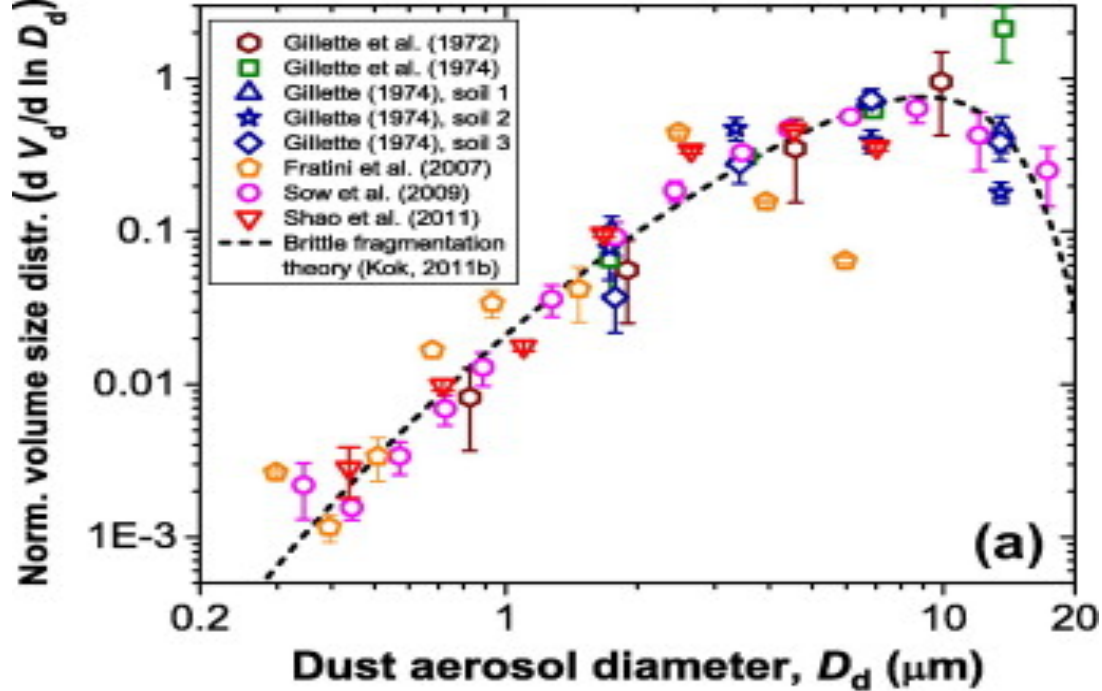
# Mineral Dust Impacts on Climate

Dust emission requires:  
a) Strong wind  
b) Dry soil  
c) Sparse vegetation  
d) Saltating particles



[Schematic](#) of interactions between dust and climate and [biogeochemistry](#).

[Mahowald et al.,  
Aeolian Research,  
15, 2014](#)



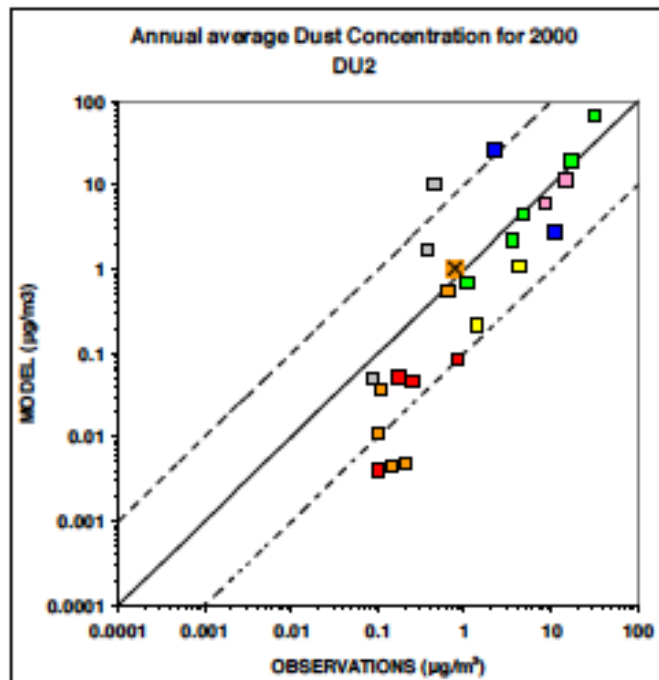
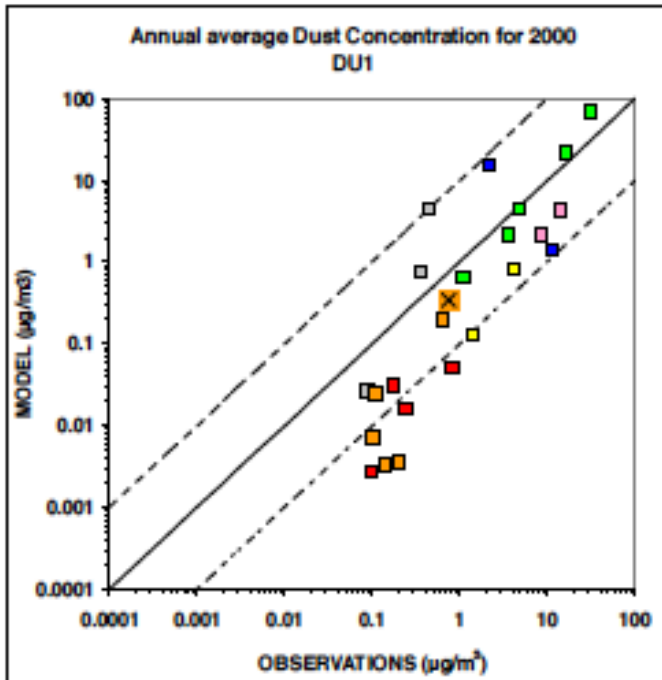
Compilation of measurements of the volume (A) and number (B) dust size distribution at emission.

Measurements by Gillette and colleagues ([Gillette, 1974](#), [Gillette et al., 1972](#), [Gillette et al., 1974](#)) used optical microscopy, and were taken in Nebraska and Texas. Conversely, measurements by [Fratini et al., 2007](#), [Sow et al., 2009](#), and [Shao et al. \(2011a\)](#) used optical [particle counters](#). These measurements were made in China, Niger, and Australia, respectively. All measurements were normalized following the procedure described in [Kok \(2011b\)](#).  
[Mahowald et al., Aeolian Research, 15](#), 2014

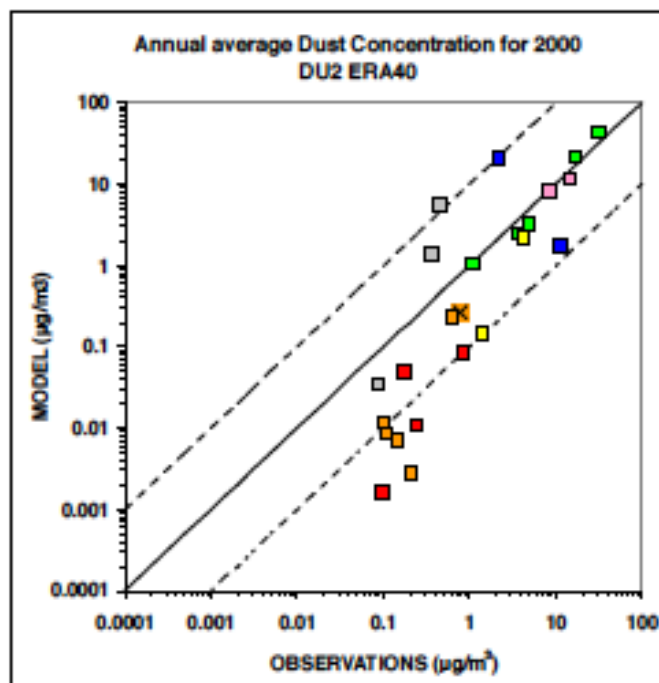
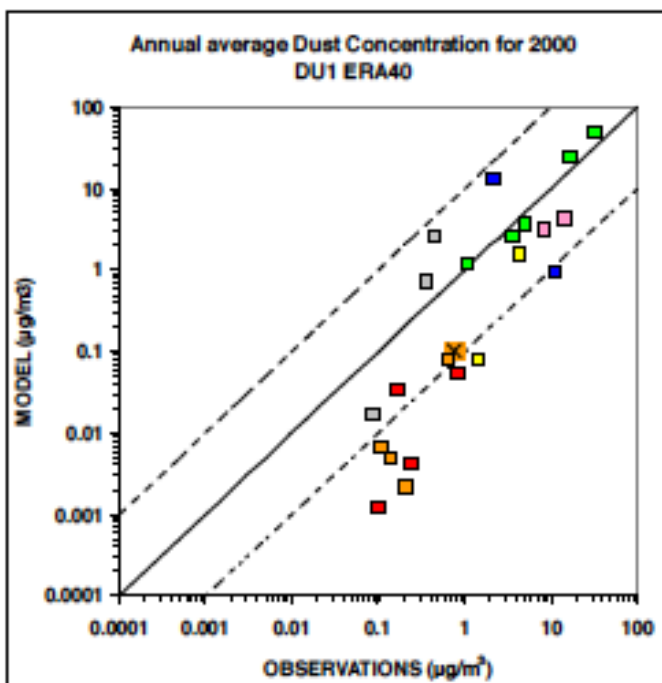
**Table 6.** Regional dust emissions ( $\text{Tg yr}^{-1}$ ) (regions shown in Fig. 3c) for the different simulations.

	N. Africa ( $\text{Tg yr}^{-1}$ )	S. Africa ( $\text{Tg yr}^{-1}$ )	Middle East ( $\text{Tg yr}^{-1}$ )	Asia ( $\text{Tg yr}^{-1}$ )	N. America ( $\text{Tg yr}^{-1}$ )	S. America ( $\text{Tg yr}^{-1}$ )	Australia ( $\text{Tg yr}^{-1}$ )
AeroCom Median	792	11.8	128	137	2	9.8	30.7
DU1	659	57.4	244	395	30	367	34.7
DU2	611	99.4	325	934	65	681	47.8
DU1_ERA40	528	54.1	182	283	22	314	35.7
DU2_ERA40	460	93.2	233	639	48	569	49.7

Model



Model



Observation

Observation

Annual mean dust concentrations from the simulation of year 2000 compared to measured multi-annual means at 24 stations.

The colours correspond to the location of each station.

E-Pacific=red,

W-Pacific=orange,

S-Africa=blue,

Atlantic=green,

Australia=yellow,

Asia=pink,

S-Ocean=grey

The dotted lines denote the 1 : 10 to 10 : 1 range.

# What about the dust in the atmosphere?

- Source strength and atmospheric burden very difficult to estimate
- A minor amount of the mass but more than half of the number is found in the deposited long range transported dust clouds.
- Exposed to gases incl water vapor
- Easy to see from space, i.e. strongly affect the radiation balance
- Important ice nuclei, important in inducing precipitation
- Does it bring any microbes?
- Can contribute substantially to deposition of nutrients to far away ecosystems



# Global aerosol sources (Tg/y)

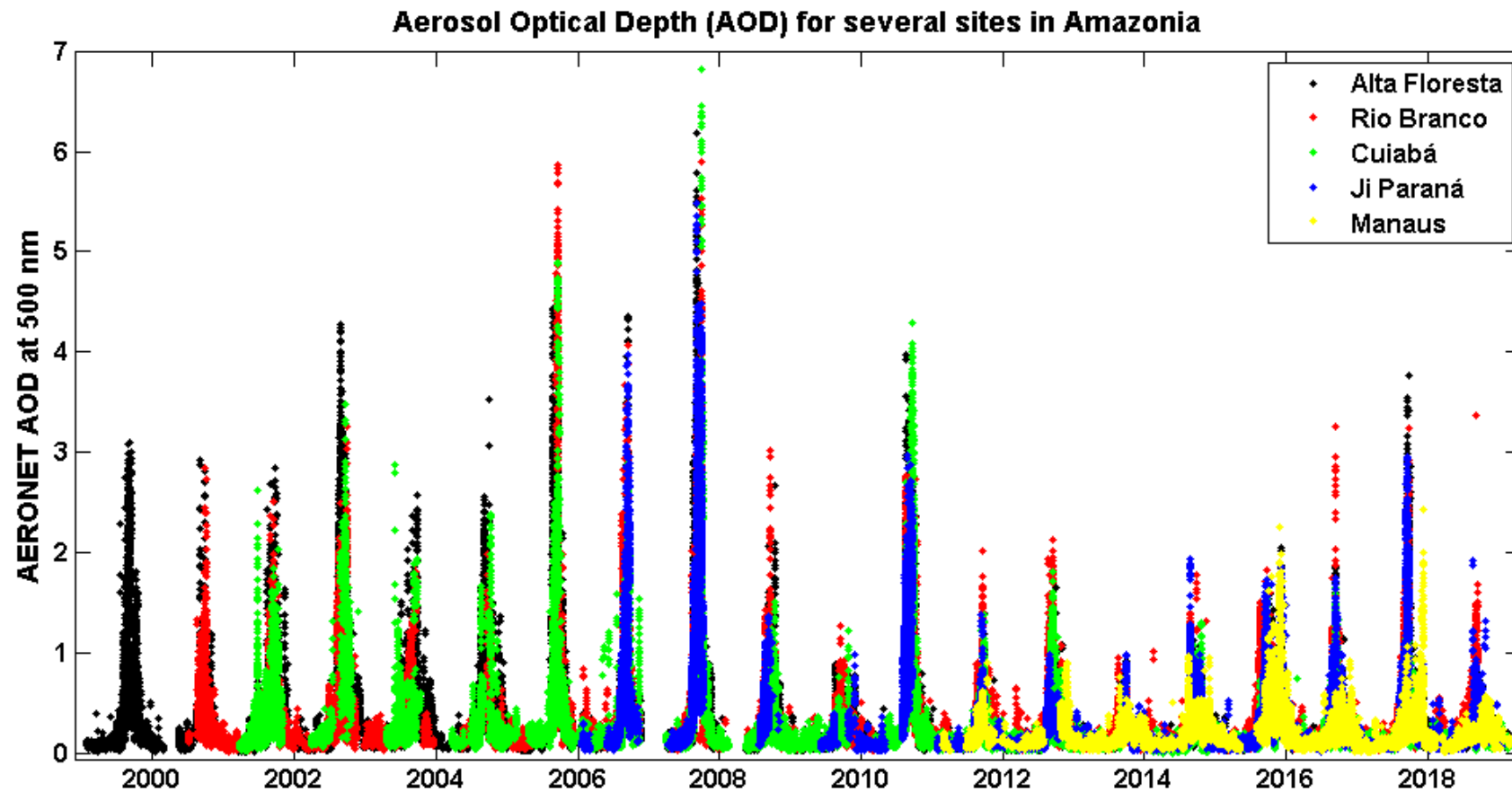
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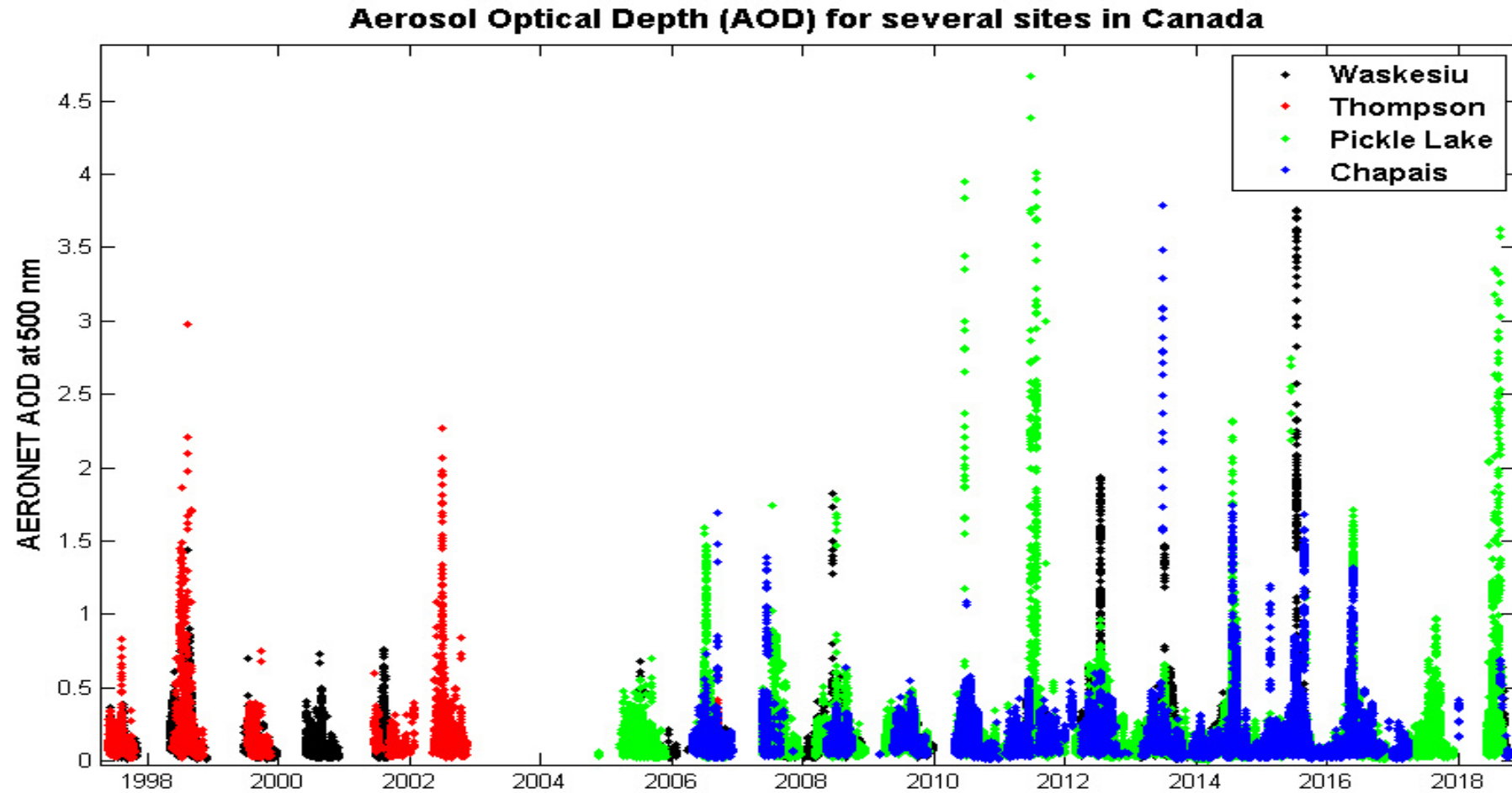
Biomass burning



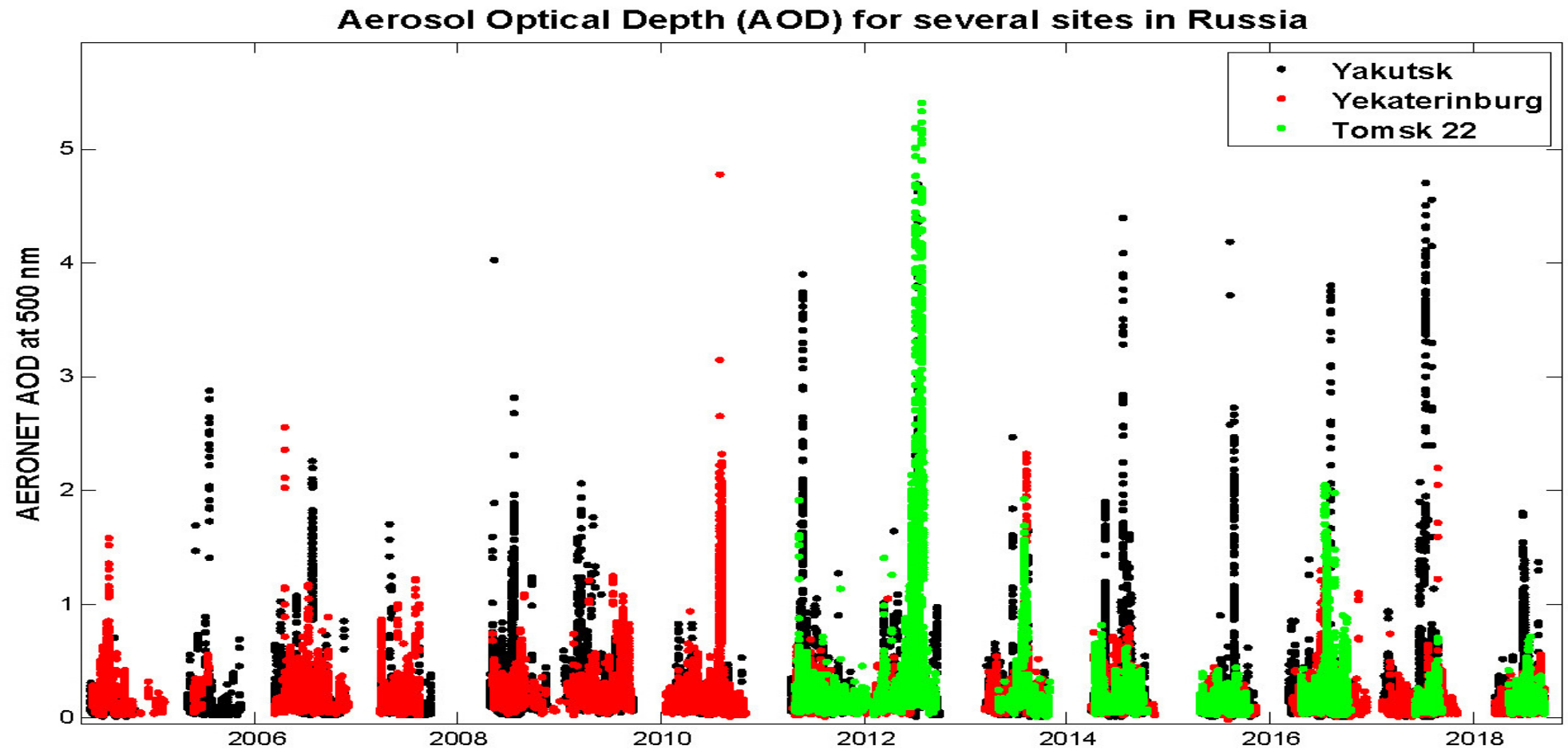
- Time series of aerosol optical depth for several sites in Amazonia from 2000 to 2019 measured with AERONET sun photometers.



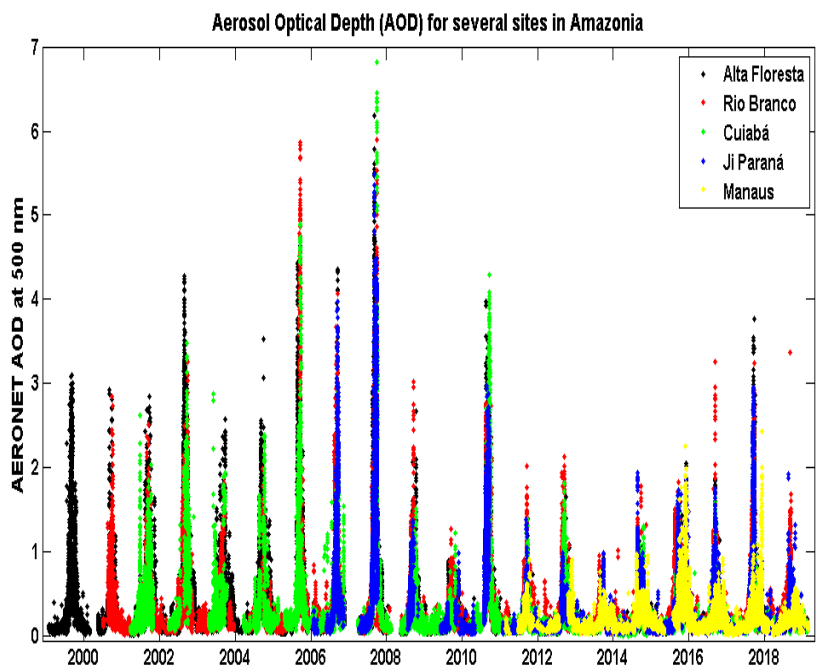
Time series of AOD at 500nm from 1997 to 2018 for 4 AERONET sites in the Canadian boreal forest region: Waskesiu, Thompson, Pickle Lake and Chapais. AOD values larger than 2 were observed during almost all years.



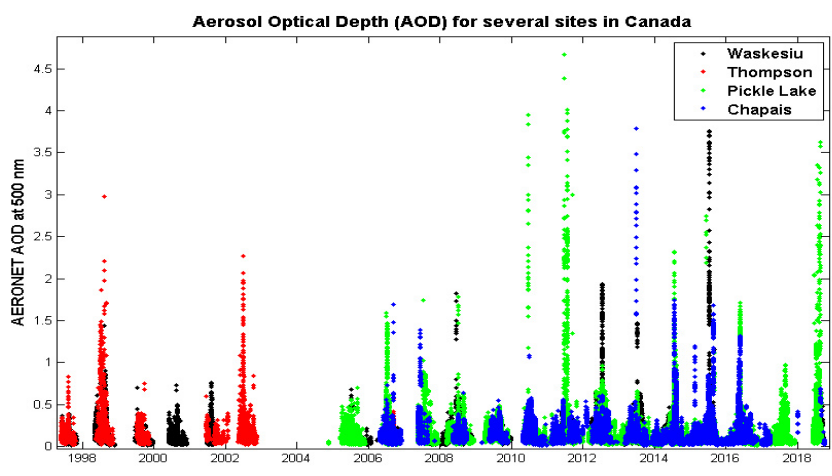
Time series of aerosol optical depth from 2005 to 2018 for 3 AERONET stations over Eurasian Boreal forests: Yakutsk, Yekaterinburg and Tomsk.



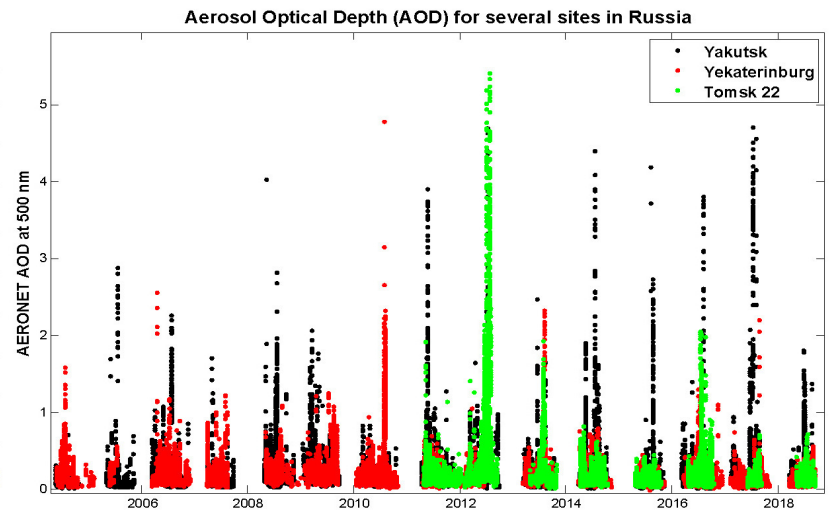
# Amazonas



# Canada



# Russia



Groundbased AOD measurements from three major forested areas

Table 1: Emission factors for pyrogenic species emitted from various types of biomass burn<sup>a</sup>

Species	Savanna and grassland			Tropical forest			Temperate forest			Boreal forest			Peat Fires		Agricultural residues (open)			
	average	std. dev.	N	average	std. dev.	N	average	std. dev.	N	average	std. dev.	N	average	std. dev.	N	average	std. dev.	N
MCE	0.94	0.02	48	0.91	0.03	15	0.90	0.05	45	0.89	0.04	21	0.79	0.02	5	0.92	0.06	30
CO <sub>2</sub>	1660	90	31	1620	70	9	1570	130	39	1530	140	14	1530	130	5	1420	240	25
CO	69	21	49	105	40	15	113	30	46	121	47	22	290	23	5	74	50	34
CH <sub>4</sub>	2.5	1.0	47	6.2	2.0	13	5.2	2.8	36	5.5	2.5	20	9.3	1.5	5	5.5	5.7	17
Total non-methane hydrocarbons	3.5	1.2	13	5.6	1.5	4	13.4	11.8	13	6.0	2.9	8	7.9	-	0	5.8	5.1	11
C <sub>2</sub> H <sub>6</sub>	0.32	0.29	28	0.35	0.39	6	0.31	0.09	20	0.28	0.13	12	0.11	0.05	3	0.29	0.24	10
C <sub>3</sub> H <sub>8</sub>	0.85	0.38	25	1.11	0.24	5	1.12	0.30	20	1.54	0.66	7	1.47	0.72	3	0.96	0.52	12
C <sub>4</sub> H <sub>10</sub>	0.41	0.32	28	0.88	0.23	7	0.69	0.57	20	0.97	0.37	14	1.85	1.5-2.2	2	0.65	0.46	10
C <sub>5</sub> H <sub>12</sub>	0.071	0.111	8	0.013	-	1	0.05	0.02	7	0.062	0.031	3	0.006	-	1	0.17	0.01-0.34	2
C <sub>6</sub> H <sub>14</sub>	0.46	0.45	26	0.86	0.41	5	0.63	0.40	19	0.67	0.45	7	1.14	1.07-1.21	2	0.41	0.26	15
C <sub>7</sub> H <sub>16</sub>	0.13	0.18	20	0.53	0.15-0.91	2	0.27	0.19	14	0.29	0.10	8	0.99	-	1	0.17	0.07	9
1-Butene	0.082	0.049	13	0.073	0.020-0.125	2	0.12	0.061	9	0.16	0.143	4	0.46	0.18-0.74	2	0.083	0.043	8
i-Butene	0.041	0.019	6	0.109	-	1	0.086	0.074	9	0.052	0.032	3	0.31	-	1	0.079	0.040	3
trans-2-Butene	0.020	0.012	11	0.033	0.016-0.050	2	0.037	0.031	9	0.030	0.018	3	0.78	-	1	0.036	0.014	6
cis-2-Butene	0.017	0.010	11	0.031	0.020-0.042	2	0.038	0.039	9	0.023	0.016	3	0.62	-	1	0.027	0.010	6
Butadiene	0.095	0.057	13	0.15	-	0	0.125	0.068	12	0.089	0.030	4	0.22	0.19-0.26	2	0.16	0.24	10
n-Butane	0.021	0.011	14	0.041	-	1	0.081	0.060	11	0.111	0.059	7	0.32	-	1	0.043	0.029	7
i-Butane	0.007	0.005	13	0.015	-	1	0.031	0.026	11	0.052	0.051	6	0.90	-	1	0.016	0.017	7
1-Pentene	0.022	0.009	6	0.058	-	1	0.048	0.024	7	0.046	0.025	3	0.110	-	1	0.015	0.011	5
2-Pentenes	0.014	0.020	4	0.026	-	0	0.043	0.023	5	0.011	0.006-0.016	2	0.62	-	1	0.023	0.005	4
n-Pentane	0.007	0.008	11	0.014	-	1	0.034	0.026	10	0.050	0.015	6	0.24	-	1	0.042	0.057	7
2-Methyl-butanes	0.025	0.037	7	0.075	-	1	0.056	0.045	6	0.057	-	0	0.125	-	1	0.026	0.013	5
2-Methyl-butane	0.008	0.009	10	0.008	-	1	0.017	0.011	8	0.032	0.016	6	0.123	-	1	0.019	0.014	5
n-Pentadienes	0.048	-	1	0.042	-	0	0.035	0.016	4	0.049	-	0	0.10	-	0	0.030	-	0
Isoprene	0.101	0.158	10	0.22	0.016-0.42	2	0.10	0.05	9	0.074	-	1	0.52	0.05-0.98	2	0.17	0.26	7
Cyclopentene	0.019	0.016	4	0.022	-	0	0.041	0.019	5	0.08	-	0	0.025	-	1	0.007	0.002	3
Cyclopentadiene	0.026	-	1	0.036	-	0	0.027	0.025-0.029	2	0.047	-	0	0.10	-	1	0.001	-	1
4-Methyl-1-pentene	0.049	-	1	0.049	-	1	0.047	-	0	0.044	-	0	0.09	-	0	0.005	0.007	4
2-Methyl-1-pentene	0.018	0.032	4	0.037	-	0	0.058	0.027	3	0.043	-	0	0.11	-	1	0.026	-	0
1-Hexene	0.043	0.018	6	0.065	-	1	0.084	0.022	3	0.109	-	1	0.14	-	0	0.012	0.005	3
Hexadienes	0.006	-	1	0.007	-	0	0.006	0.006-0.006	2	0.008	-	0	0.077	-	0	0.005	-	0
n-Hexane	0.018	0.028	10	0.032	-	0	0.032	0.040	10	0.054	0.035	3	0.14	-	1	0.032	0.059	4
Isohexanes	0.019	0.028	3	0.048	-	0	0.026	0.038	8	0.013	0.008-0.018	2	0.54	-	1	0.067	0.115	4
Heptanes	0.016	0.019	6	0.024	-	0	0.029	0.026	8	0.021	0.018-0.024	2	0.112	-	1	0.031	0.033	4
Octanes	0.021	0.027	3	0.012	-	1	0.036	0.023	5	0.027	-	0	0.065	-	1	0.003	-	1
Terpenes	0.104	0.096	5	0.15	-	0	1.17	1.95	9	1.53	-	1	0.08	0.005-0.16	2	0.029	0.031	3
Benzene	0.33	0.22	18	0.38	0.05	4	0.39	0.20	16	0.57	0.21	7	0.87	0.78-0.95	2	0.28	0.20	15
Toluene	0.20	0.14	16	0.23	0.04	4	0.25	0.17	15	0.35	0.11	6	0.45	0.37-0.52	2	0.16	0.10	15
Xylenes	0.086	0.077	8	0.086	0.049	3	0.16	0.090	9	0.11	0.016	3	0.23	-	1	0.09	0.11	9
Ethylbenzene	0.022	0.010	8	0.043	0.034	3	0.041	0.018	10	0.038	0.011	3	0.042	-	1	0.045	0.049	7
Styrene	0.056	0.029	6	0.028	-	0	0.066	0.028	8	0.13	-	0	0.55	0.027-0.082	2	0.043	0.029	6
PAHs	0.012	0.016	4	0.14	-	0	0.017	0.019	6	0.72	-	1	0.39	-	0	0.033	0.017	4
Methanol	1.35	0.47	14	2.8	0.5	4	2.2	0.9	19	2.33	1.45	13	2.5	0.4	3	2.6	1.4	8
Ethanol	0.036	0.017-0.055	2	0.067	-	0	0.076	0.089	7	0.058	0.063	3	0.16	-	0	0.05	-	0
1-Propanol	0.025	-	1	0.038	-	0	0.047	-	0	0.044	-	0	0.090	-	0	0.027	-	0
2-Propanol	0.06	-	0	0.12	-	0	0.13	-	0	0.14	-	0	0.29	-	0	0.09	-	0
Butanols	0.11	0.008-0.21	2	0.009	-	1	0.064	0.029-0.098	2	0.072	-	0	0.15	-	0	0.011	-	1
Cyclopentanol	0.033	-	1	0.032	-	1	0.038	-	0	0.040	-	0	0.083	-	0	0.016	-	1
Phenol	0.43	0.19	7	0.23	0.006-0.45	2	0.25	0.09	3	0.63	-	0	0.47	0.42-0.51	2	0.50	0.49	4
Formaldehyde	1.23	0.65	16	2.40	0.63	3	2.08	0.70	15	1.75	0.40	4	1.07	0.44	3	1.8	0.7	7
Acetaldehyde	0.84	0.65	9	2.26	1.55-2.97	2	1.07	0.62	13	0.81	0.23	4	1.16	0.70-1.63	2	1.7	1.2	4
Hydroxyacetaldehyde (glycolaldehyd.)	0.21	0.18	5	0.42	-	0	0.39	-	1	0.48	-	0	0.11	-	1	3.2	2.3-4.1	2
Glyoxal	0.40	-	0	0.60	-	0	0.65	-	0	0.69	-	0	1.4	-	0	0.23	-	1
Methylglyoxal	0.40	0.15-0.64	2	0.52	-	0	0.27	-	1	0.67	-	0	0.23	-	1	0.55	-	1
Acrolein (Propenal)	0.48	0.25	6	0.65	-	1	0.34	0.13	7	0.33	-	1	0.27	-	1	0.65	0.45	4
Propenal	0.053	0.009-0.097	2	0.10	-	1	0.087	0.040	4	0.24	-	1	0.33	-	0	0.18	-	1
Butanals	0.11	0.054-0.220	2	0.13	0.073-0.18	2	0.11	0.07	5	0.16	-	0	0.02	-	1	0.17	0.02-0.32	2
Methacrolein	0.17	-	0	0.15	-	1	0.14	0.18	5	0.11	0.12	3	0.38	-	0	0.28	-	1
Crotonaldehyde	0.25	-	0	0.24	-	1	0.40	-	0	0.43	-	0	0.88	-	0	0.42	-	1

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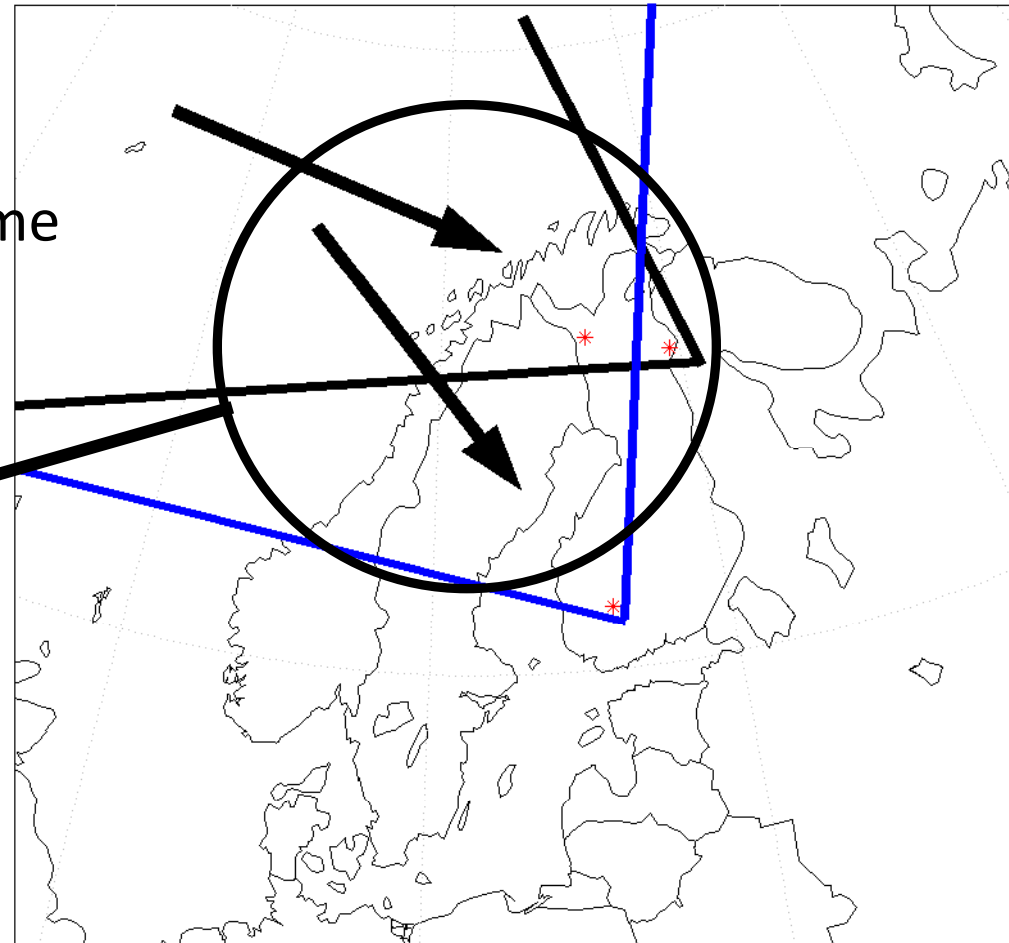


Species	Savanna and grassland			Tropical forest			Temperate forest			Boreal forest			Peat Fires			Agricultural residues (open)		
	average	std. dev.	N	average	std. dev.	N	average	std. dev.	N	average	std. dev.	N	average	std. dev.	N	average	std. dev.	N
Hexanals	0.048	0.068	3	0.021	0.010-0.031	2	0.038	0.033	4	0.038	-	0	0.06	-	0	0.020	0.011-0.03	2
Heptanals	0.003	0.001-0.005	2	0.004	-	1	0.005	-	0	0.005	-	0	0.010	-	0	0.001	-	1
Acetone	0.47	0.18	7	0.63	-	1	0.58	0.42	11	1.59	1.61	7	0.91	0.69-1.12	2	0.61	0.47	4
2-Butanone	0.13	0.1	6	0.50	-	1	0.23	0.21	9	0.16	0.04	5	0.34	0.14-0.54	2	0.60	0.29-0.90	2
2,3-Butanedione	0.35	0.2	4	0.73	-	1	0.89	0.86	5	0.34	-	1	0.32	-	1	1.23	1.15-1.31	2
1-Butene-3-one (Methylvinyl ketone)	0.23	-	1	0.39	-	1	0.165	0.109	5	0.099	0.097-0.10	2	0.057	-	1	0.48	0.25-0.70	2
Pentanones	0.015	0.006	3	0.039	0.028-0.090	2	0.066	0.033	5	0.075	-	0	0.075	-	1	0.10	0.007-0.20	2
Hexanones	0.048	-	1	0.057	-	0	0.045	0.043-0.046	2	0.066	-	0	0.14	-	0	0.040	-	0
Heptanones	0.006	-	1	0.002	-	1	0.005	-	0	0.005	-	0	0.011	-	0	0.002	-	1
Octanones	0.015	-	1	0.019	-	1	0.023	-	0	0.024	-	0	0.030	-	0	0.025	-	0
Benzaldehyde	0.102	0.097	4	0.027	-	1	0.132	0.077	3	0.096	-	0	0.056	-	1	0.039	0.008-0.07	2
Acetol (hydroxyacetone)	0.56	0.3	3	1.36	-	0	1.13	-	1	1.6	-	0	0.64	0.42-0.86	2	1.74	0.29	3
Furan	0.29	0.14	8	0.33	0.25-0.41	2	0.41	0.26	8	0.36	0.28-0.44	2	1.07	0.74-1.4	2	0.50	0.47	3
2-Methyl-furan	0.20	0.14	6	0.28	0.28	3	0.34	0.21	5	0.43	-	0	0.31	0.12-0.50	2	0.53	0.519	3
3-Methyl-furan	0.010	0.004	3	0.055	0.030-0.080	2	0.034	0.016	3	0.052	-	0	0.11	-	0	0.076	0.003-0.15	2
2-Ethylfuran	0.005	0.001-0.009	2	0.003	-	1	0.016	0.012	5	0.008	-	0	0.026	-	0	0.0005	-	1
2,4-Dimethyl-furan	0.008	-	1	0.024	-	1	0.012	-	0	0.013	-	0	0.026	-	0	0.002	-	1
2,5-Dimethyl-furan	0.063	0.067	4	0.086	-	0	0.070	0.070	5	0.10	-	0	0.14	-	1	0.098	-	1
Tetrahydrofuran	0.009	0.002-0.016	2	0.017	-	1	0.001	0.0005-0.0017	2	0.011	-	0	0.023	-	0	0.006	-	1
2,3-Dihydrofuran	0.014	0.013-0.015	2	0.014	-	1	0.003	0.001-0.004	2	0.013	-	0	0.026	-	0	0.005	-	1
Benzofuran	0.045	0.040	4	0.016	-	1	0.094	0.071	3	0.067	-	0	0.032	-	1	0.024	0.004-0.044	2
Furfural (2-Furaldehyde)	0.73	0.74	3	0.78	-	0	0.52	0.81	7	0.61	-	1	1.10	0.12-2.1	2	1.03	-	1
Methyl formate	0.073	-	1	0.051	-	0	0.024	0.022-0.027	2	0.024	-	1	0.12	-	0	0.04	-	0
Methyl acetate	0.159	0.059-0.26	2	0.13	-	0	0.095	0.058	5	0.087	-	1	0.32	-	0	0.09	-	0
Acetonitrile	0.17	0.07	9	0.49	0.14	3	0.23	0.18	13	0.31	0.10	6	0.60	-	1	0.23	0.26	6
Acrylonitrile	0.037	0.009	3	0.04	-	1	0.031	0.014	6	0.068	-	0	-	-	0	0.094	0.061	3
Propionitrile	0.027	0.012-0.042	2	0.09	-	1	0.011	0.011-0.012	2	0.11	-	0	-	-	0	0.17	-	1
Pyrole	0.013	-	1	0.12	-	1	0.062	0.085	3	0.15	-	0	-	-	0	0.22	-	1
Trimethylpyrazole	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0
Methylamine	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0
Dimethylamine	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0
Ethylamine	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0
Trimethylamine	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0
n-Pentylamine	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0
2-Methyl-1-butylamine	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0
Formic acid	0.30	0.21	14	0.88	0.63	4	1.11	1.27	12	1.91	2.24	8	0.29	0.14	3	0.86	0.89	7
Acetic acid	2.31	1.8	13	3.3	0.8	3	2.74	1.60	11	3.80	2.04	4	4.9	0.97	3	4.6	3.4	7
H <sub>2</sub>	0.97	0.35	6	3.1	0.7	5	2.1	0.4	4	1.6	0.4	8	1.2	-	1	2.6	2.6-2.7	2
NO <sub>x</sub> (as NO)	2.5	1.3	18	2.8	1.3	7	3.0	1.8	16	1.18	0.86	11	1.2	0.31-2.2	2	2.6	1.1	18
HONO	0.47	0.21	6	0.85	-	1	0.33	0.17	5	0.41	-	1	0.35	0.21-0.49	2	0.37	0.04	3
N <sub>2</sub> O	0.18	0.09	11	0.20	-	0	0.25	0.12	3	0.24	0.06	5	-	-	0	0.09	0.04	5
NH <sub>3</sub>	0.90	0.49	16	1.34	0.78	4	0.98	0.69	22	2.5	1.75	4	4.2	3.2	3	0.93	0.62	13
HCN	0.44	0.26	16	0.44	0.21	5	0.64	0.39	12	0.53	0.30	11	4.4	1.2	3	0.43	0.19	6
Cyanogen, (CN) <sub>2</sub>	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0	-	-	0
N <sub>2</sub>	2.6	-	0	2.6	-	0	2.6	-	0	2.6	-	0	-	-	0	2.6	-	0
SO <sub>2</sub>	0.47	0.44	12	0.77	0.37	3	0.70	0.48	5	0.75	0.14-0.31	2	4.3	-	1	0.80	0.71	10
Dimethyl sulfide (DMS)	0.008	0.011	5	0.0022	-	1	0.014	0.015	3	0.0023	-	1	0.045	0.003-0.088	2	0.07	-	1
COs	0.038	0.045	4	0.078	0.046	3	0.035	0.044	6	0.038	0.031	3	0.110	-	1	0.059	0.070	4
HCl	0.13	0.10	3	0.13	-	0	0.039	0.031	3	0.13	-	0	0.008	-	1	0.18	0.255	3
CH <sub>2</sub> Cl	0.064	0.067	15	0.029	0.02-0.04	2	0.042	0.056	8	0.060	0.033	4	0.15	-	1	0.16	0.13	4
CH <sub>2</sub> Br	0.0029	0.0052	13	0.0078	0.005-0.010	2	0.0015	0.0010	3	0.0029	0.0011	4	0.010	-	1	0.0011	-	1
CH <sub>2</sub> I	0.0007	0.0006	9	0.0068	-	1	0.0005	0.0004-0.001	2	0.0004	-	1	0.012	-	1	0.0002	-	1
Hg <sup>0</sup>	4.8E-05	4.2E-05	4	1.0E-04	4.7E-5-1.7E-4	2	2.0E-04	1.8E-04	6	2.3E-04	3.0E-04	6	-	-	0	5.1E-05	5.0E-05	3
PM <sub>2.5</sub>	6.7	3.3	20	8.3	3.3	9	18.1	14.5	28	18.7	15.9	5	17.3	-	1	8.2	4.4	18
TPM	8.7	3.1	11	10.9	5.3	4	18.4	8.3	11	15.3	12.3-18.3	2	26.2	-	0	11.6	8.1	5
TC	3.2	1.5	10	5.5	1.6	4	8.4	2.2	3	9.9	-	0	12.6	-	0	4.9	3.9	18
OC	2.8	1.3	13	4.4	1.9	5	9.4	5.6	12	7.5	-	1	12.4	-	1	4.5	3.6	16
BC	0.53	0.37	17	0.51	0.34	8	0.55	0.36	14	0.53	0.08	3	0.19	-	1	0.42	0.27	20
Levoglucosan	0.05	-	1	0.42	-	1	1.32	1.21	6	1.3	-	1	0.57	-	1	0.73	0.66	6
K	0.40	0.24	12	0.32	0.22	4	0.17	0.16	4	0.17	-	0	0.004	-	1	0.49	0.46	8
CN	2.7E+16	2.4E+16	4	3.9E+15	1.3E+15	3	9.9E+15	-	0	4.2E+15	-	1	-	-	0	5.4E+15	2.2E+15	3
CN (0.3% SS)	7.9E+14	-	1	1.7E+15	1.65E+15-1.68E+15	2	2.0E+15	3.4E+15	3	1.6E+15	-	0	-	-	0	1.6E+15	-	0
NC>= 0.12 µm diameter	9.1E+14	6.1E+14	3	2.7E+15	-	1	7.0E+15	-	0	1.6E+15	-	0	-	-	0	1.6E+15	-	0

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# Natural sources??

- Cleanest possible sectors selected (W-N)
- Trajectories describing transport within sectors more than 90% of the time selected

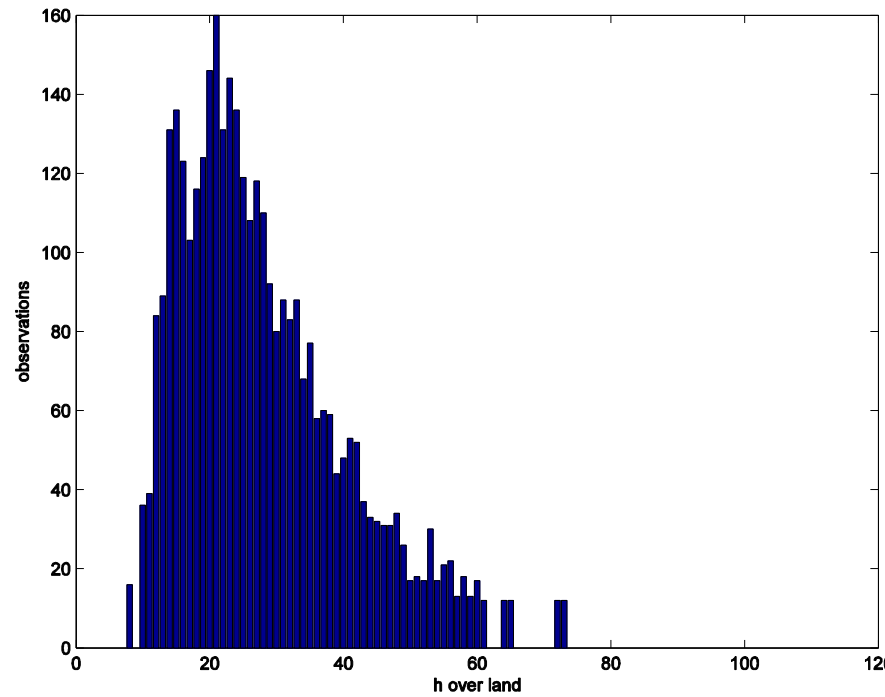


Non-zero,  
but very low  
population density  
=

Small anthropogenic contributions

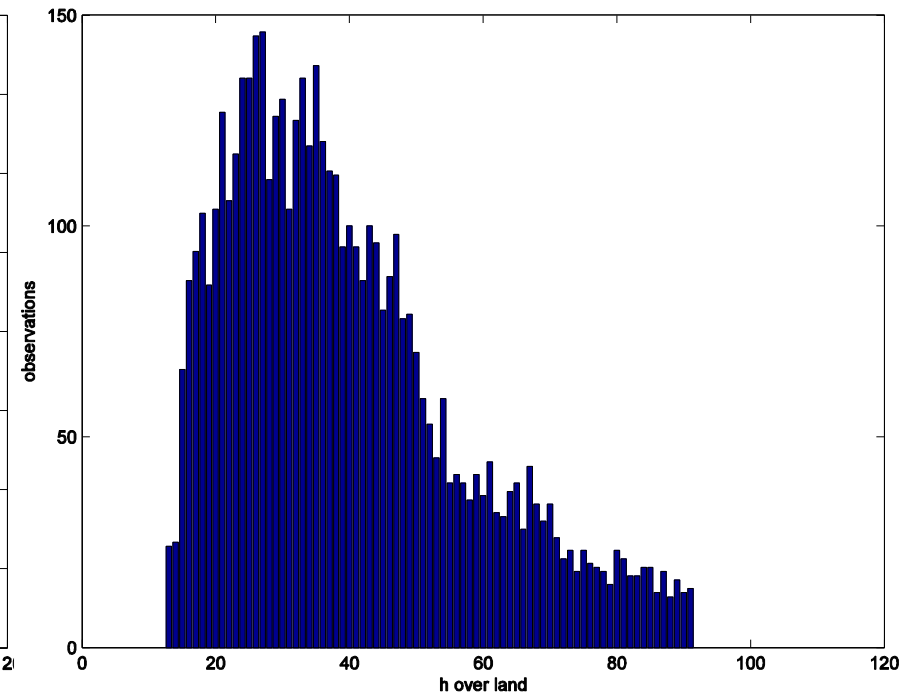
# Distribution of trajectories

Värriö and Pallas



~4500 observations

Hyytiälä



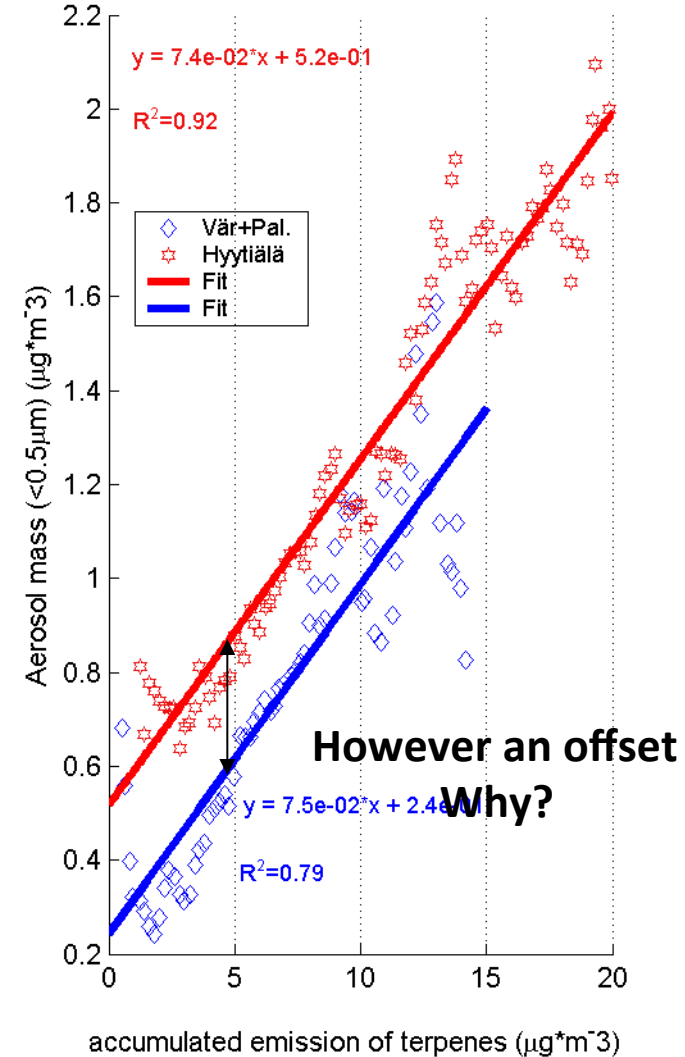
~5200 observations



# Mass evolution and emitted terpenes

- Better agreement between northerly and southerly stations using emitted terpenes as variable
  - $0.074 \mu\text{g} \cdot \text{m}^{-3} \cdot \mu\text{g}_{\text{terp}}^{-1} \cdot \text{m}^{-3}$  *Hyytiälä*
  - $0.075 \mu\text{g} \cdot \text{m}^{-3} \cdot \mu\text{g}_{\text{terp}}^{-1} \cdot \text{m}^{-3}$  *Vär. & Pal.*
- Assuming that lifetimes of terpenes are short (e.g. Hakola et al., 2003) → fraction of reacted terpenes found in the particle phase: an indirect measure of the **yield**

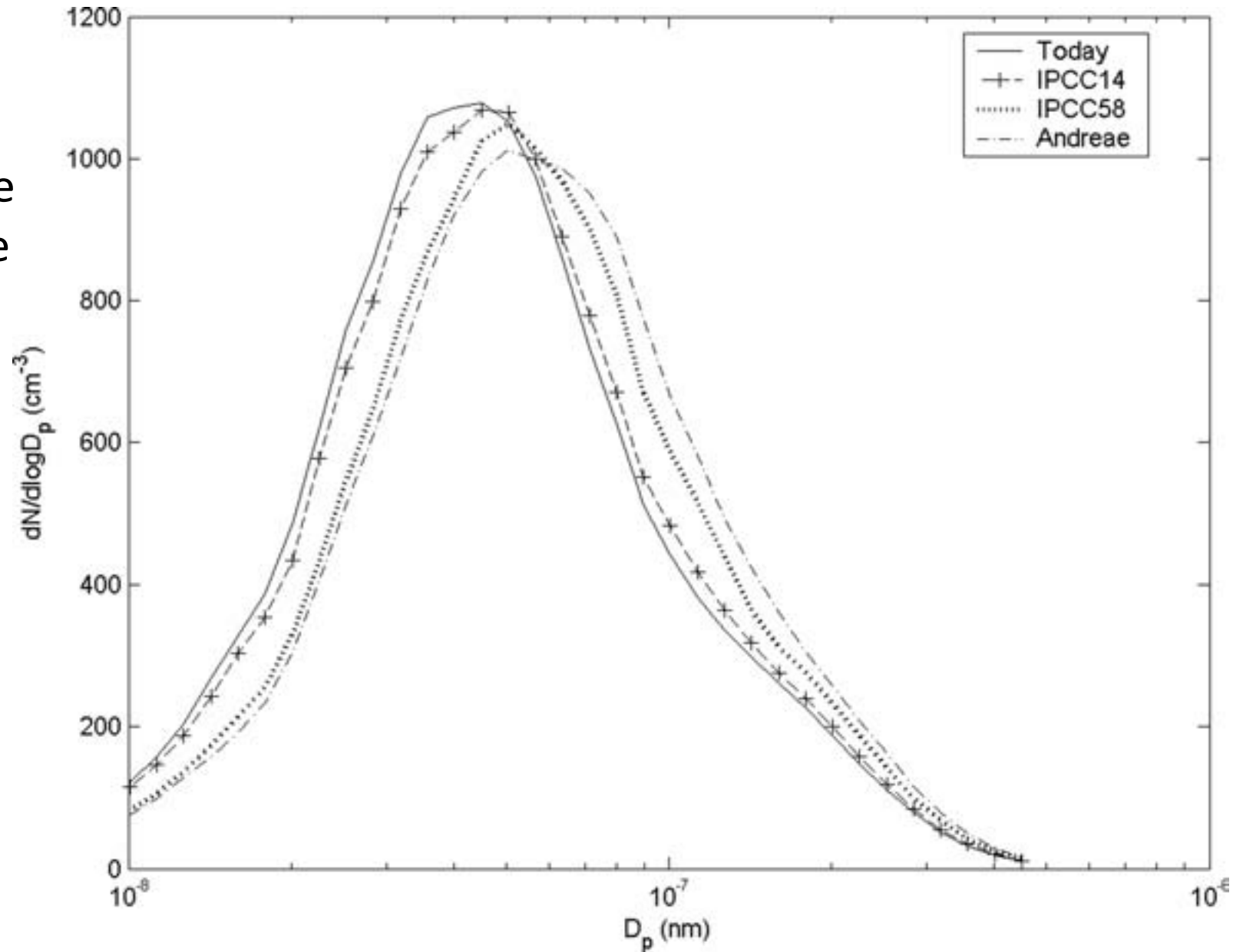
However: only "apparent yield" since we omit sinks and possibly other sources of aerosol mass



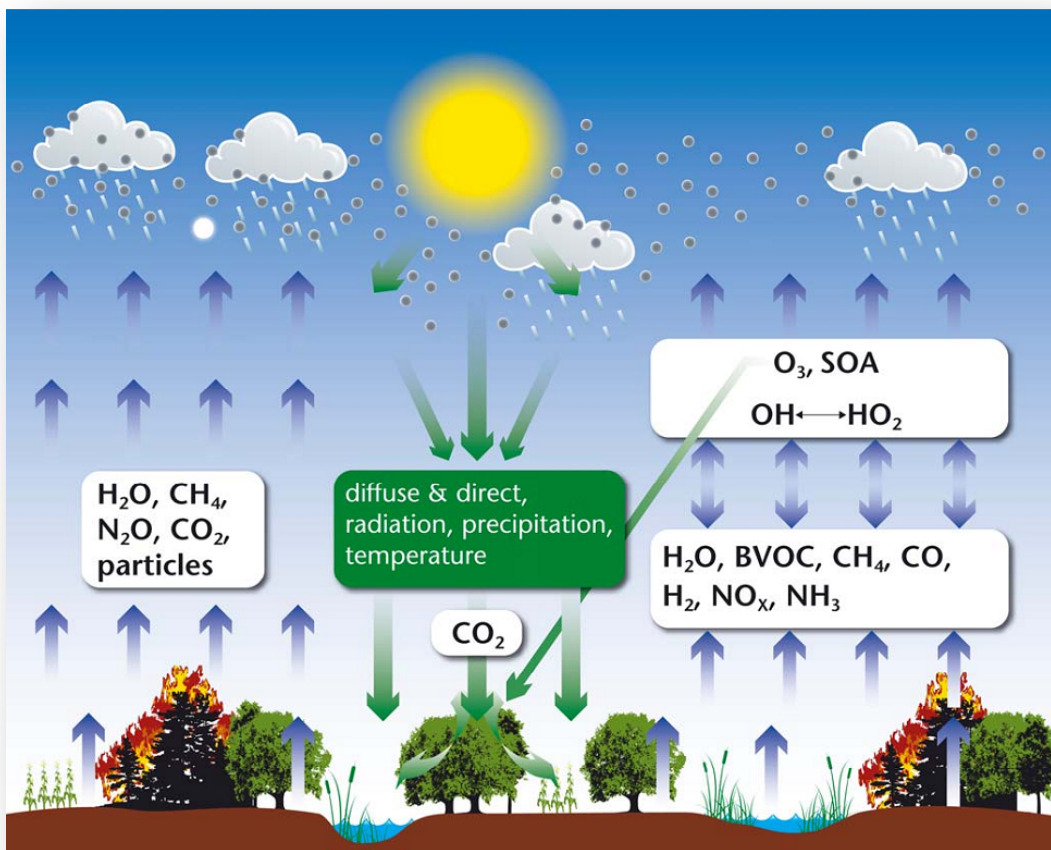
Estimated regional average size distribution resulting from needle leaf forest sources alone over the northern Fenno Scandinavian region.

Solid line represents the typical size distribution at current.

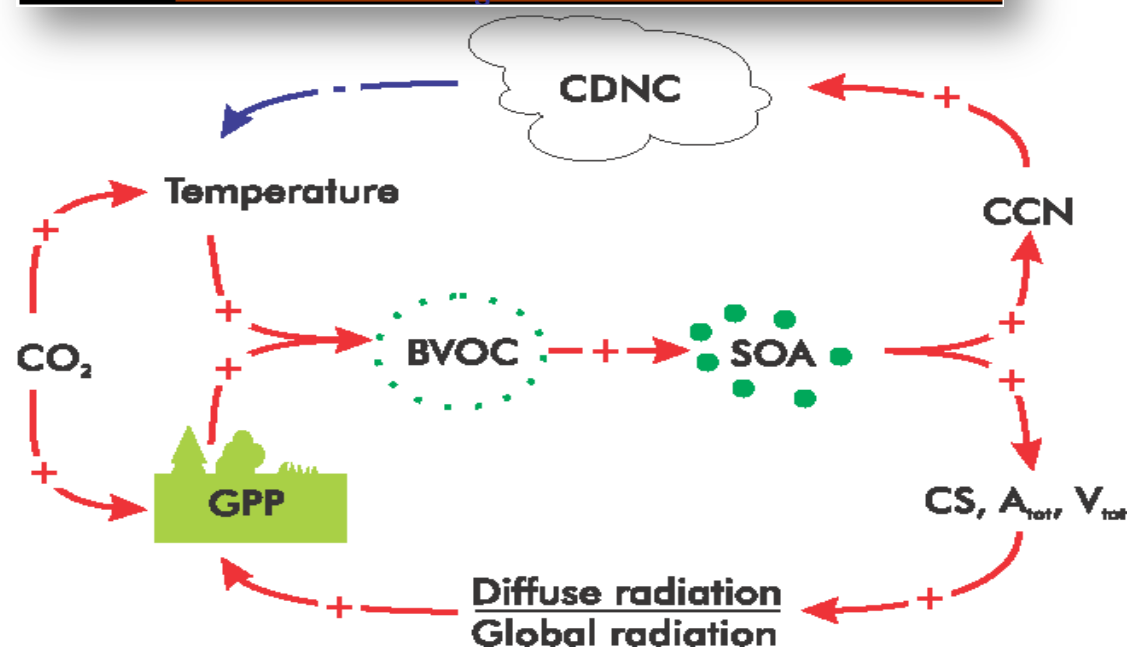
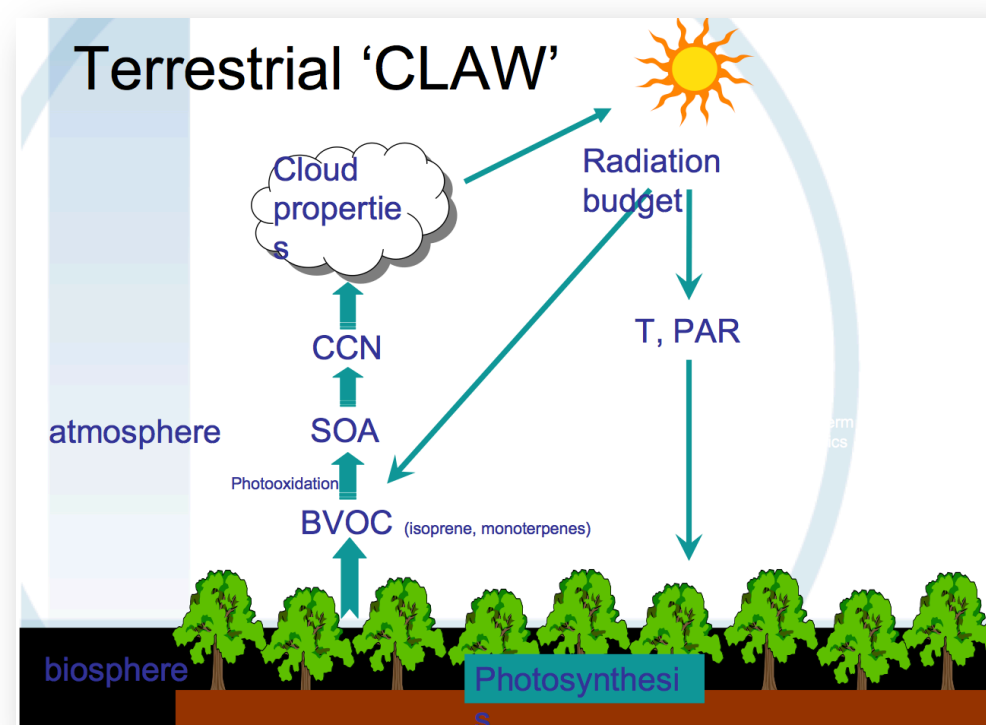
Increase of temperature by 1.4° (+-signs), 5.8° (dotted) and 8° (dash-dotted). considered.



# Conceptual overview of terrestrial carbon cycle – chemistry – climate interactions



Arneeth et al., 2011



Kulmala et al., 2013

# What about the biogenic source?

- Source strength and atmospheric burden very difficult to estimate
- Very many different compounds, primary particles and precursor gases emitted to the atmosphere.
- Important components in the atmospheric chemistry
- Organics dominate!
- Dominated by secondary aerosol and fine particles.
- Feedback loops

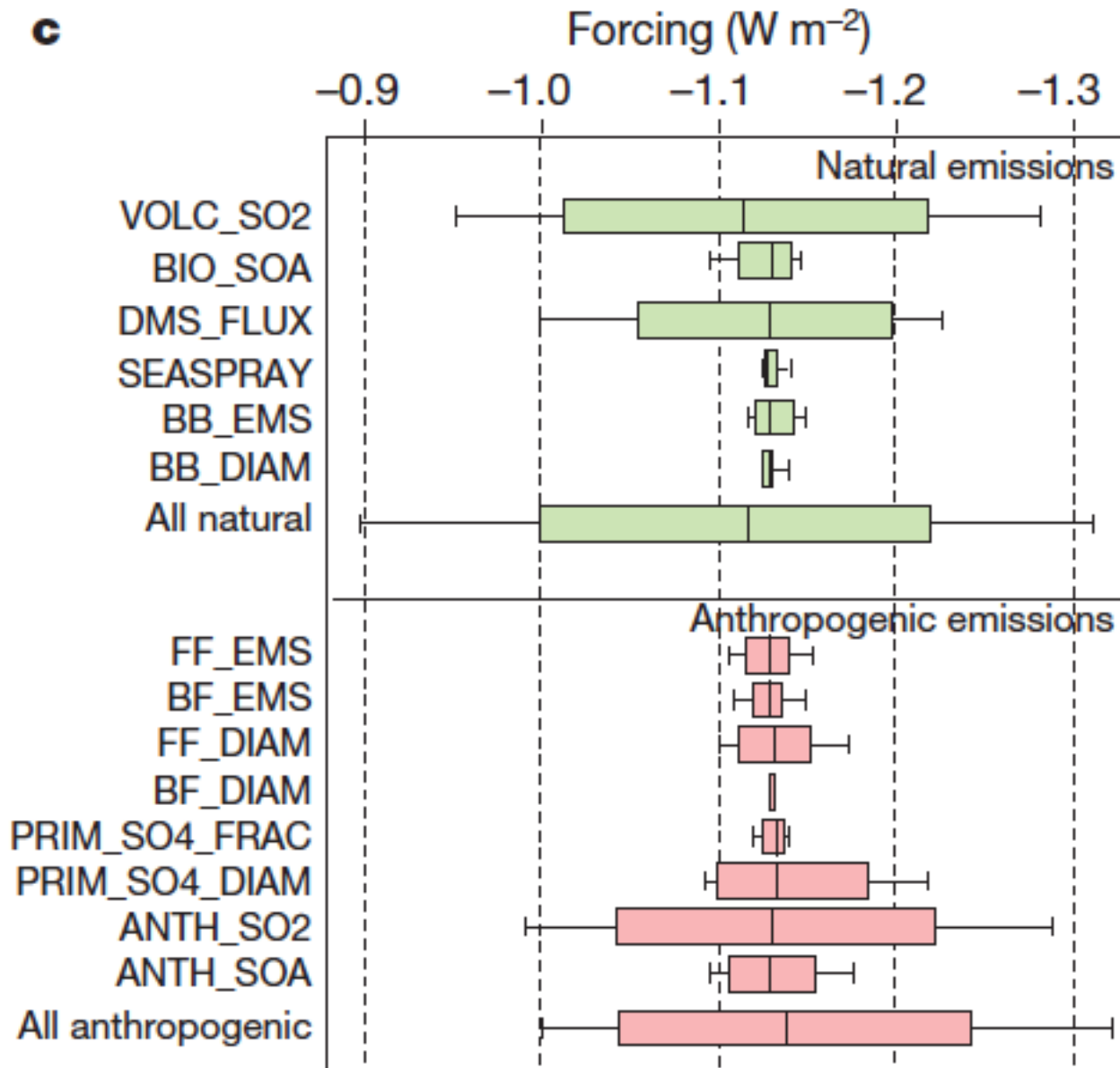
Our knowledge on natural sources is quite limited!

Source	Natural Global	
	Min	Max
Sea spray	1400	6800
including marine POA	2	20
Mineral dust	1000	4000
Terrestrial PBAPs	50	1000
including spores		28
Dimethylsulphide (DMS)	10	40
Monoterpenes	30	120
Isoprene	410	600
SOA production from all BVOCs	20	380

IPCC, 2014

# Which natural aerosol source is the "most" important?

c



Estimated contribution to the uncertainty in aerosol climate forcing

Carslaw et al., 2013, Nature





Thank you all for your attention

See you Wednesday  
Same time and same place

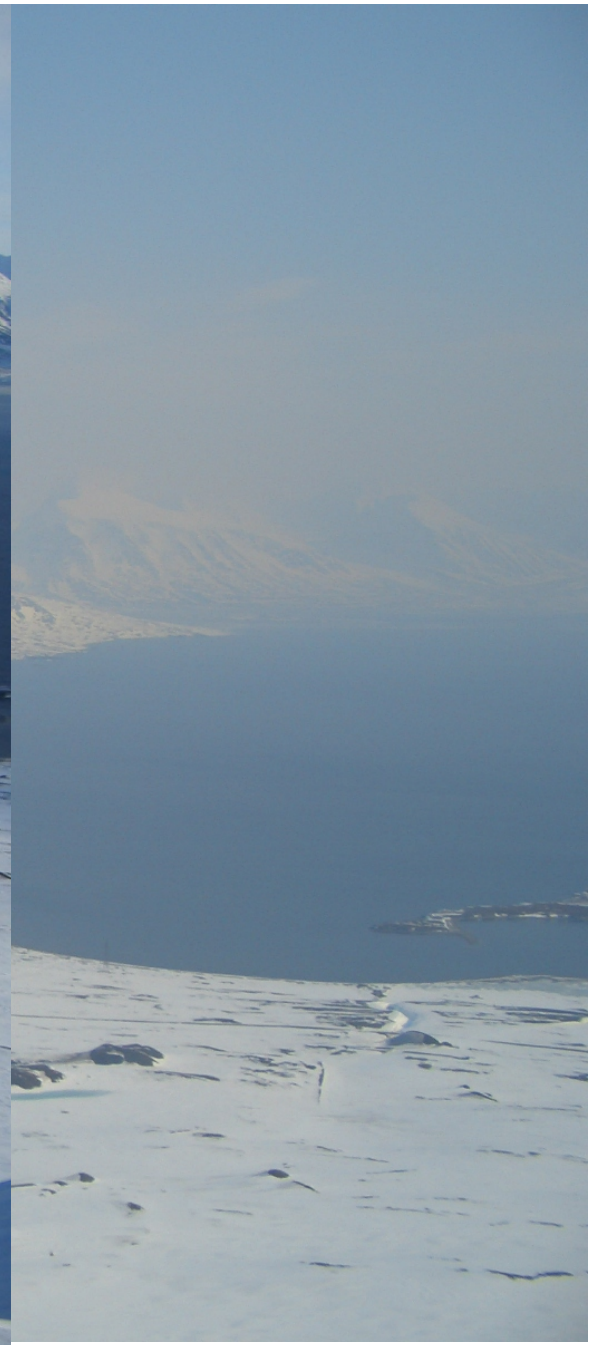
# Arctic aerosols

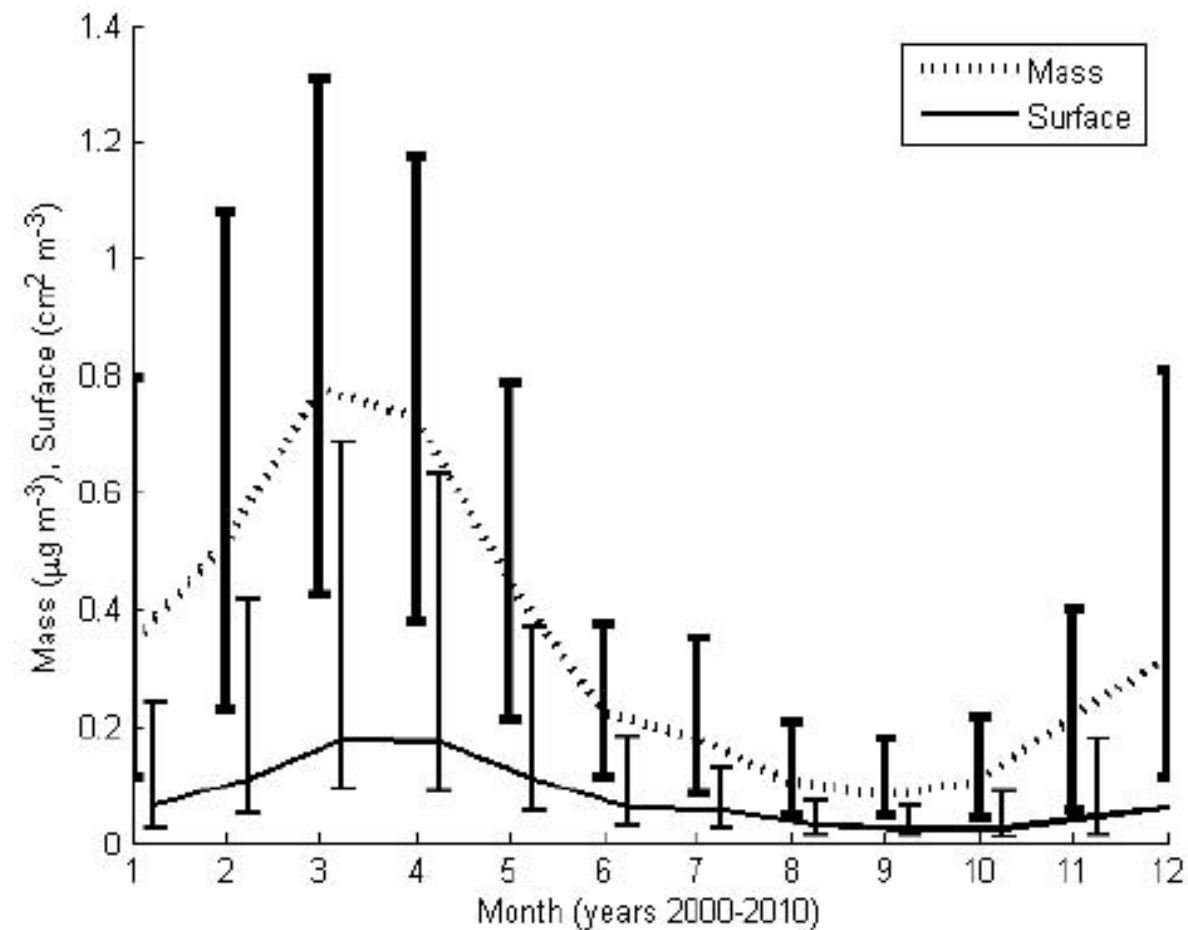
Pristine areas, do they exist?



Zeppelin station, Ny Ålesund, Svalbard

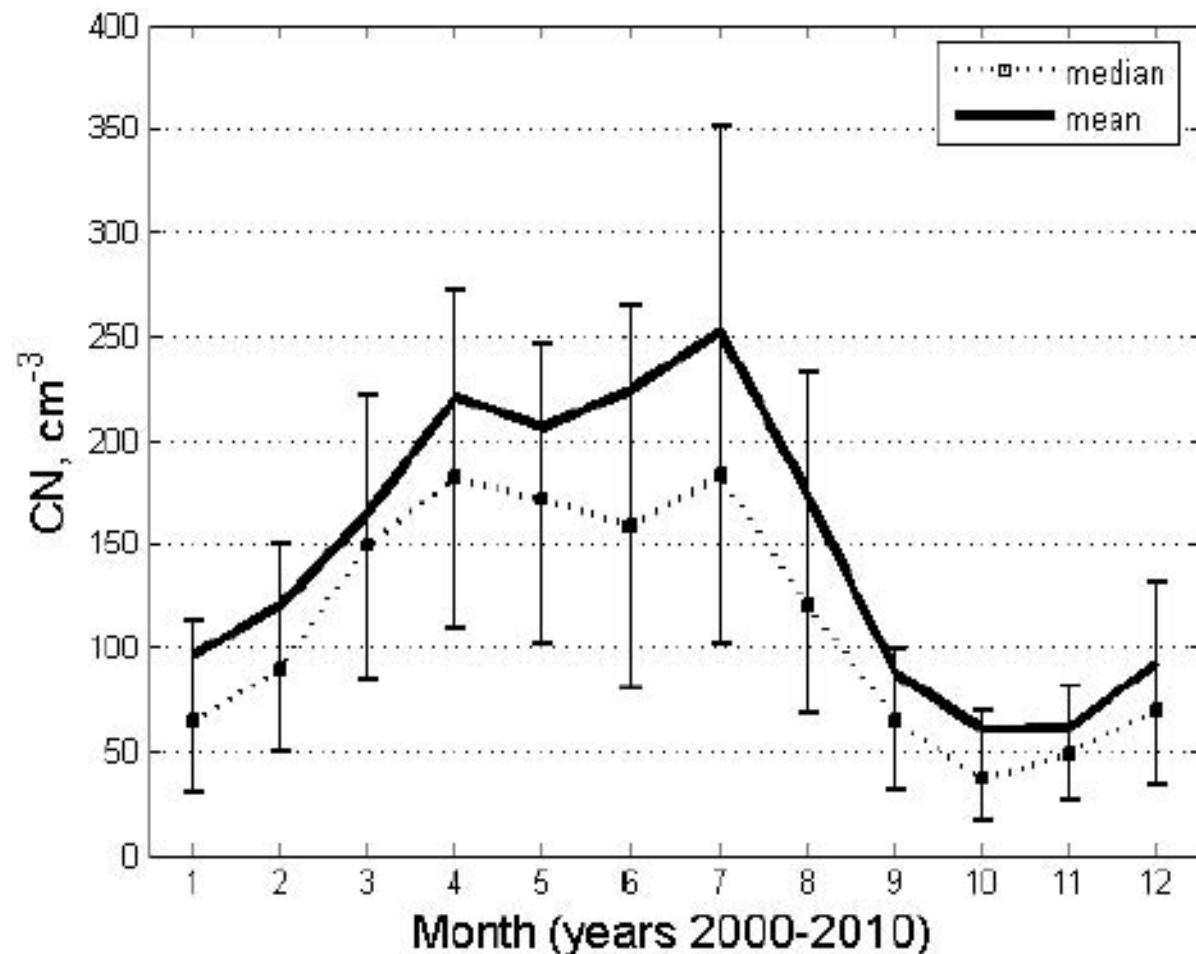






Annual average variation of integrated surface and mass, March 2000–March 2010.

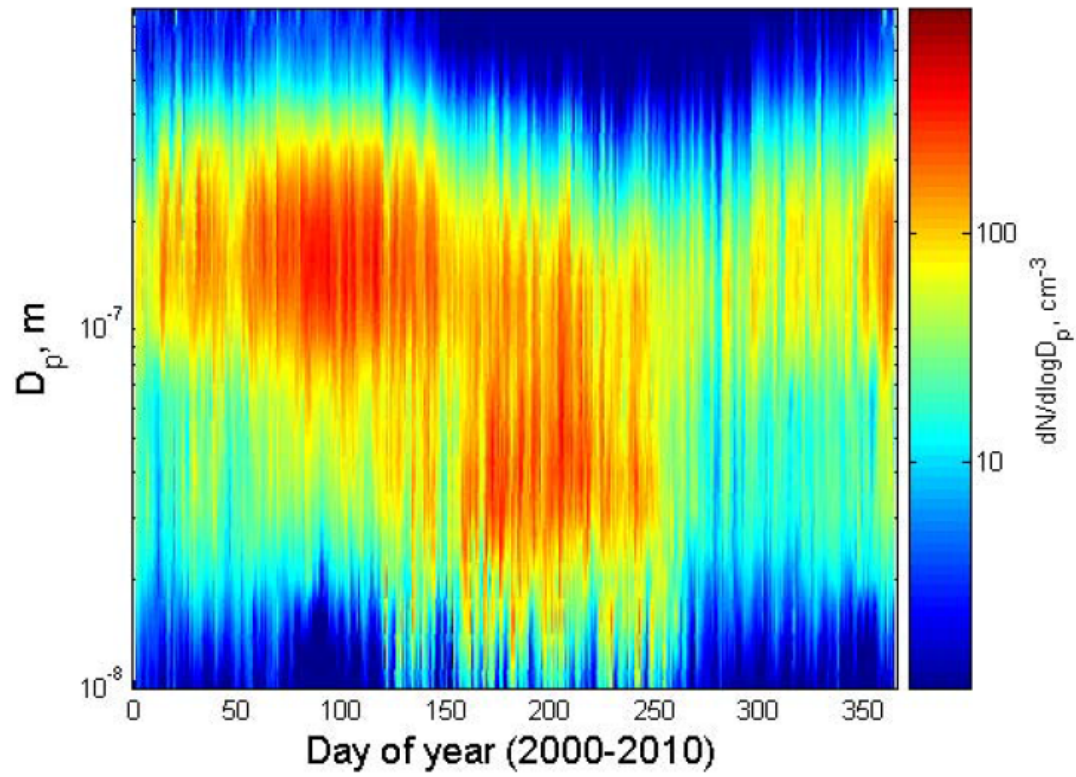
Mass data calculated from aerosol number size distribution assuming a density of  $\rho = 1 \text{ g cm}^{-3}$ . 25–75th percentile ranges indicated by errorbars.



Annual average variation of median and mean integrated number concentration per month March 2000–March 2010.

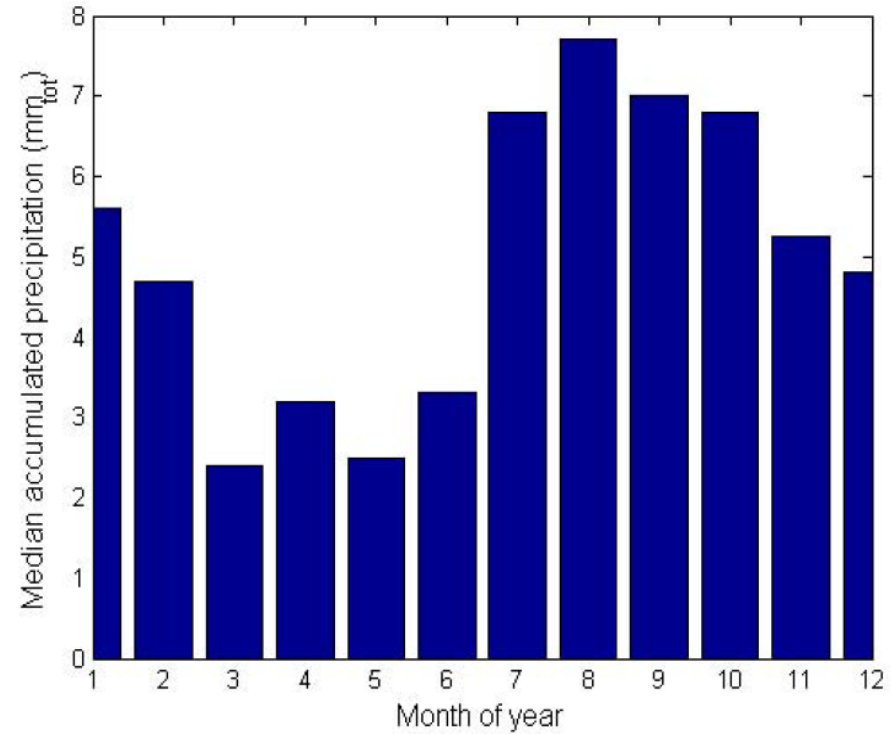
25–75th percentile ranges indicated by vertical “error bars”.





Spectral plot of daily average aerosol number size distributions, March 2000–December 2010.

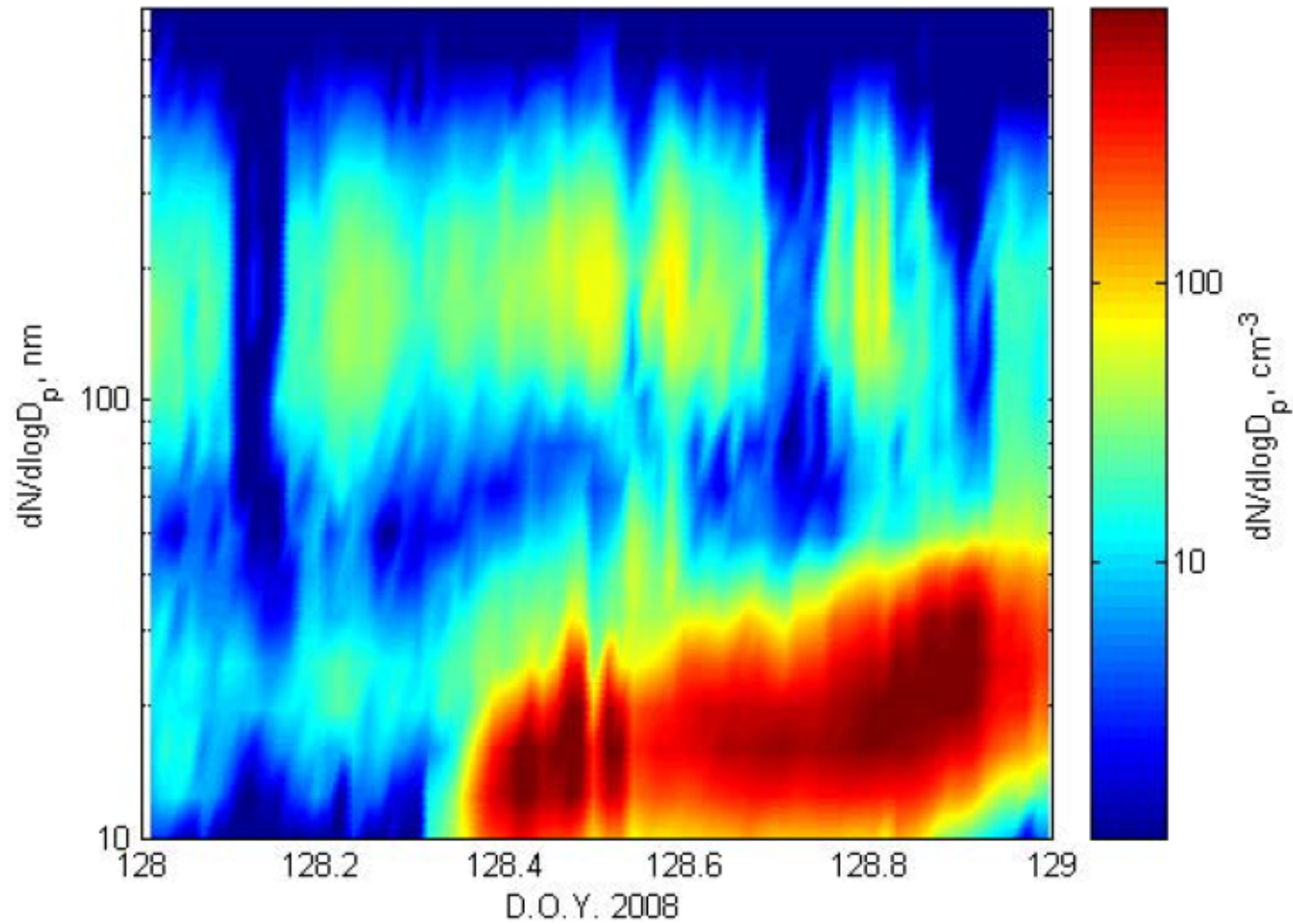
Units on x-axis as day of year.



Monthly median of accumulated precipitation experience by trajectories arriving at Svalbard during the period of 2000–2010.

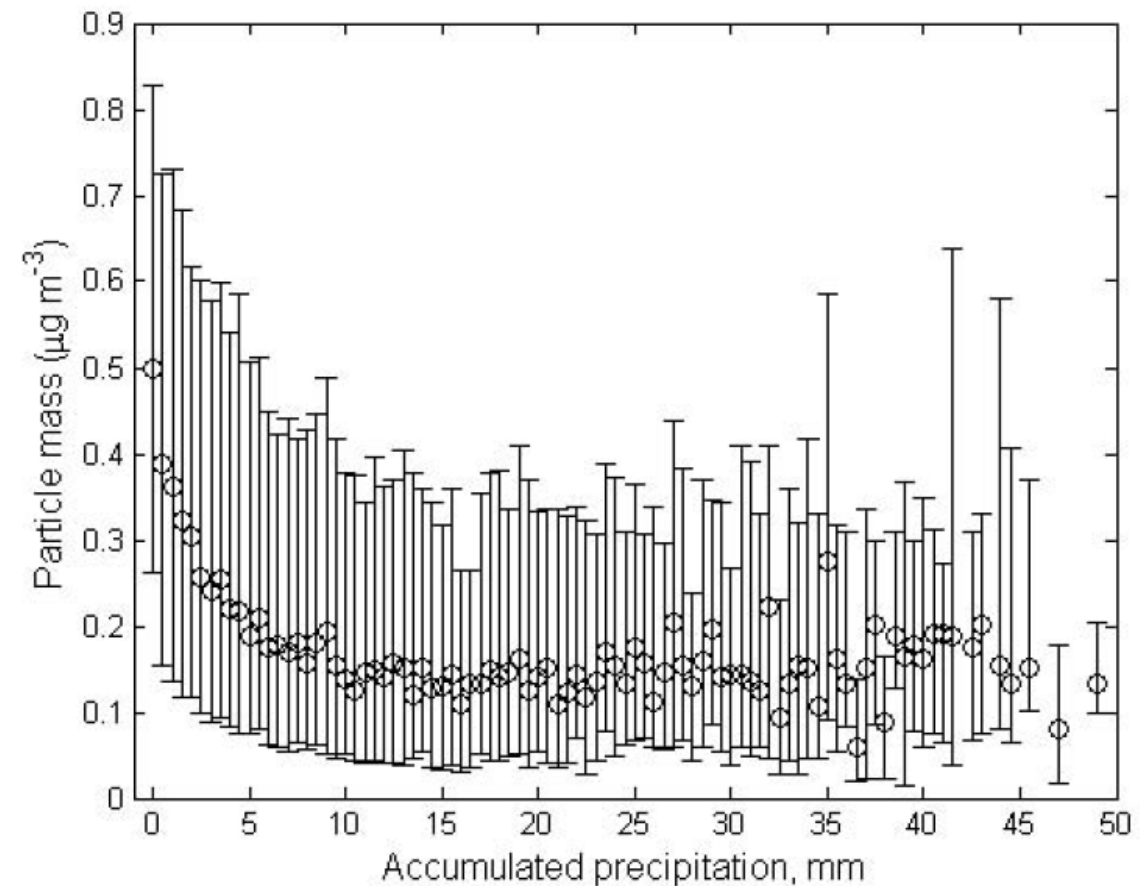
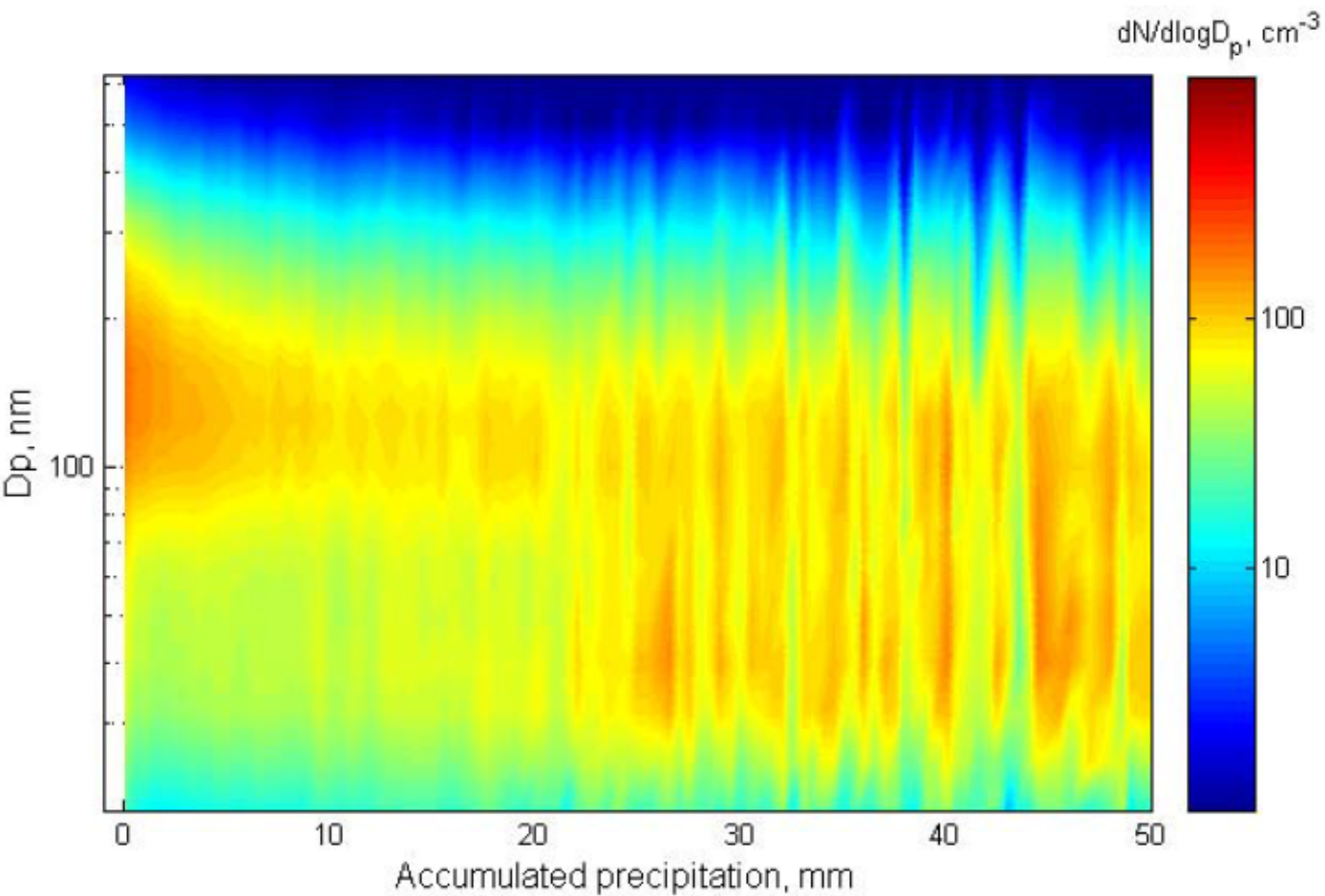


20080507



Example of an Arctic nucleation event as observed 7 May 2008.

$dN/d\log D_p$  (cm<sup>-3</sup>) versus decimal day of year (D.O.Y.).



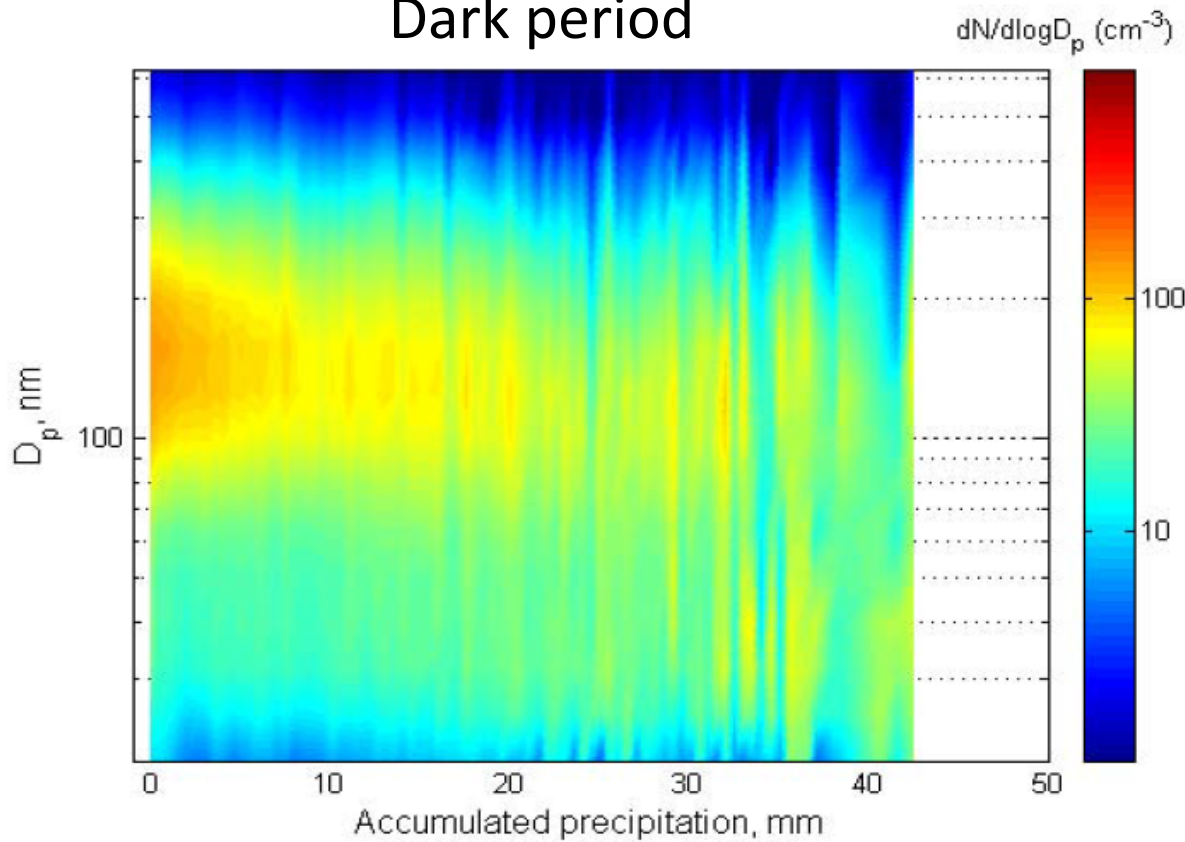
Evolution of aerosol number size distribution as a function of accumulated precipitation (mmtot) along 240 h trajectories.

Submicron aerosol mass (10–630 nm;  $1 \text{ g cm}^{-3}$ ) as a function of accumulated precipitation along the trajectories. All data collected between 2000 and 2010.

Data are shown binned over a step size of  $0.5 \text{ mmtot}$ , and the corresponding size distributions over this ranges of precipitation is presented as median values.

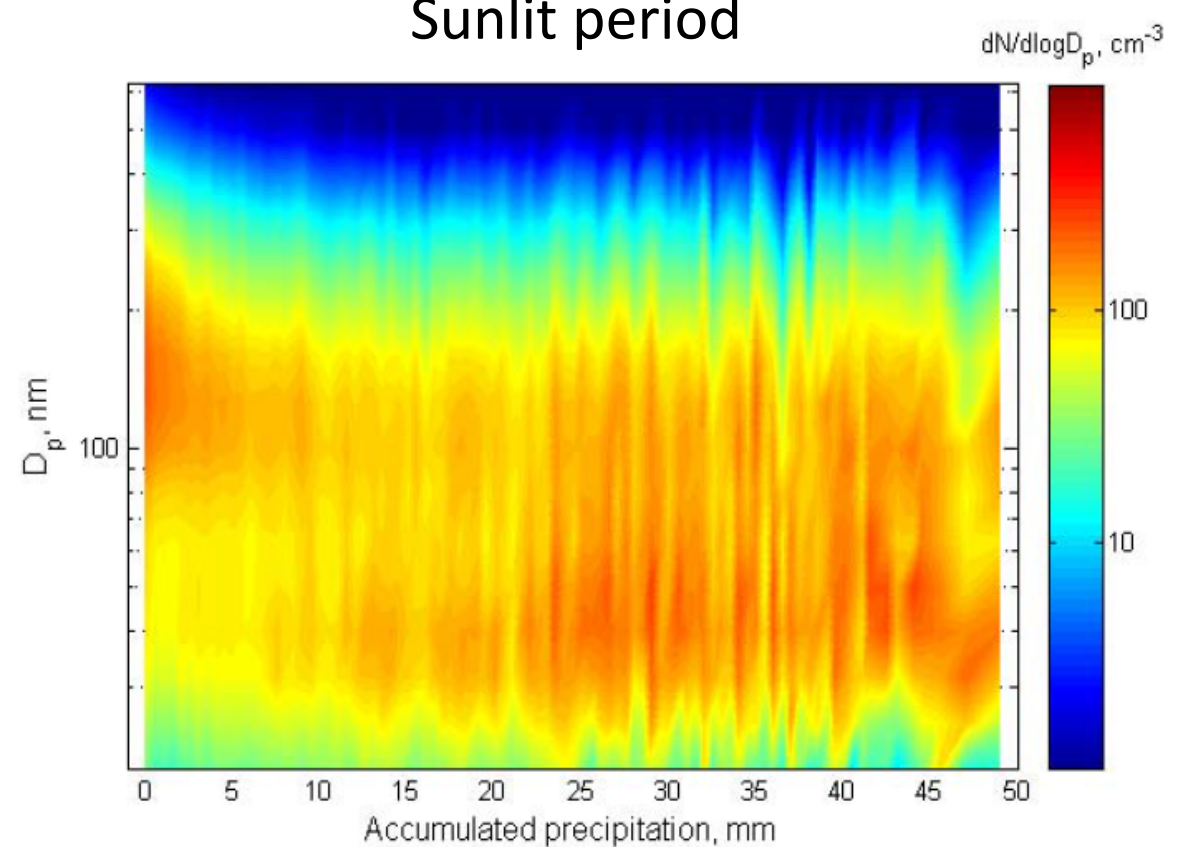
Data are shown as 25–75th percentile ranges per bin (solidlines) and median (circles).

## Dark period



Evolution of aerosol number size distribution as a function of accumulated precipitation (mmtot) along 240 h trajectories for the dark period (October–February). Data from 2000–2010

## Sunlit period



Evolution of aerosol number size distribution as a function of accumulated precipitation (mmtot) along 240 h trajectories for the sunlit period (March–September). Data from 2000–2010.

# Is the nucleation particles a natural source?

- Where do the precursor gases come from?
- The Arctic sea?
- The Arctic ecosystem?
- Subsiding long distant transported gases from the continents?

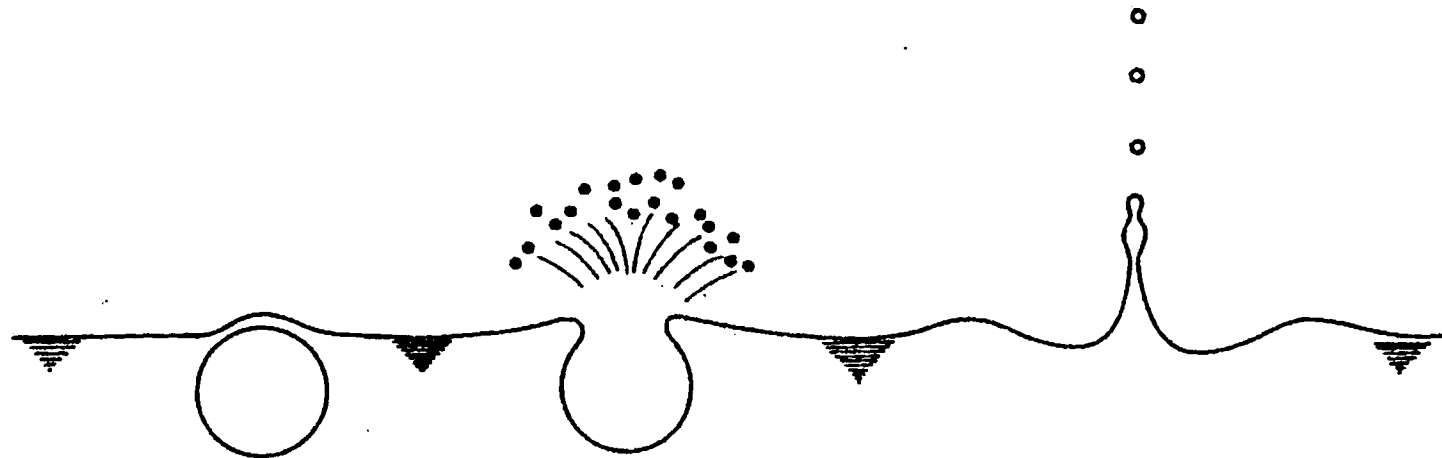
# Arctic sources and sinks

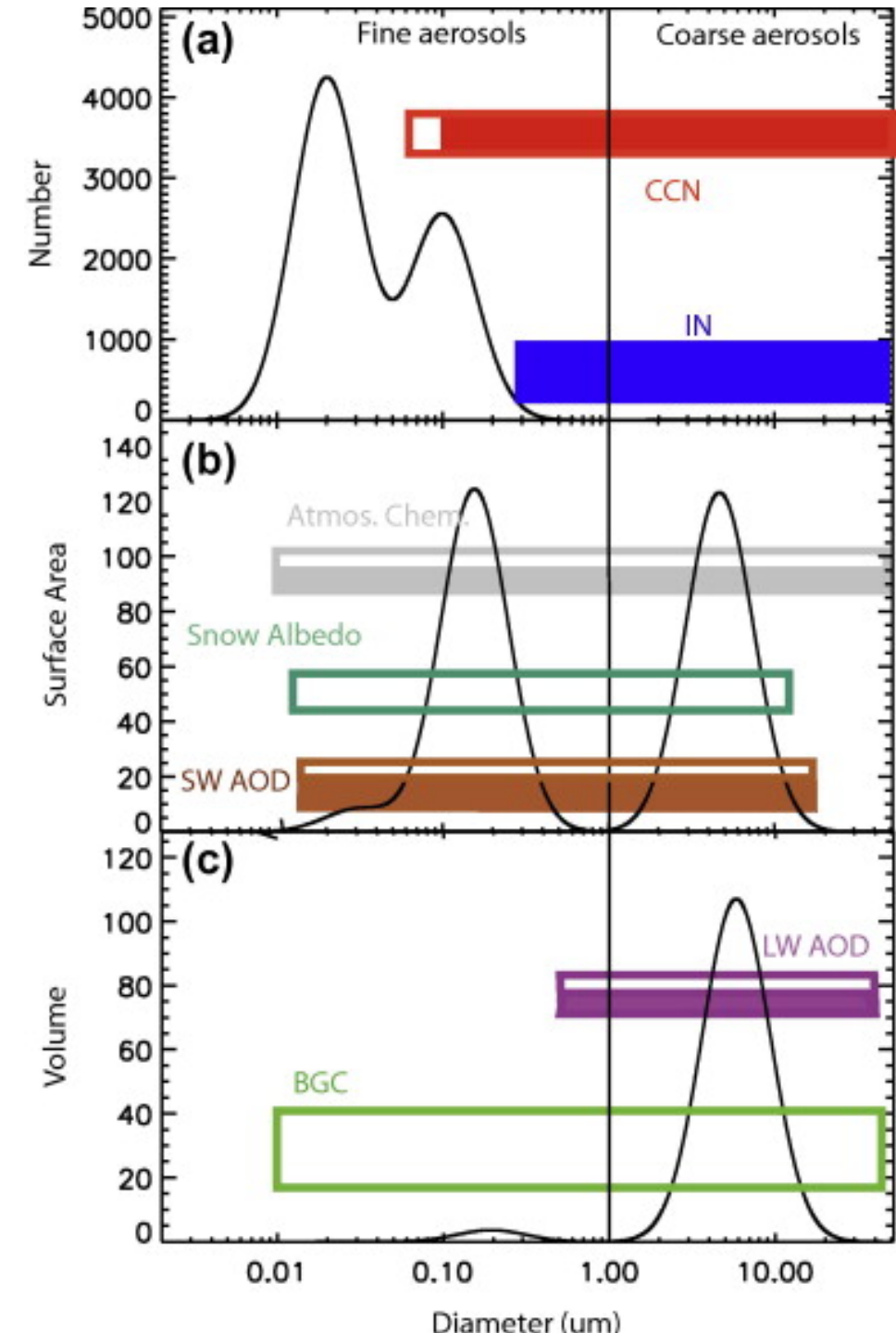
- Seasonal variations in size reveal different source processes
- Long distant transport can be facilitated by meteorology
- Precipitation dominate deposition process
- Sink processes can induce formation processes
- The nucleation observed is probably natural but how is still unclear





# The Primary Marine Aerosol Source





- (a) Aerosol number,  
 (b) surface area, and  
 (c) volume for a typical trimodal aerosol distribution

Abbreviations:

CCN, cloud condensation nuclei (*red*);

IN, ice nuclei (*blue*);

SW AOD, shortwave aerosol optical depth (*brown*);

LW AOD, longwave aerosol optical depth (*purple*); and

BGC, biogeochemically relevant species (*green*).

Solid boxes represent only size-dependent processes, and the outlined boxes represent the part of the impact that is composition dependent.