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SMR.1303 - 6

Advanced Course:
CLIMATE CHANGE IN THE MEDITERRANEAN REGION
PART I: PHYSICAL ASPECTS
(12 - 16 March 2001)

"Aerosols in the Mediterranean, their origin and climatic effects"

Part II

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ATMOSPHERIC AEROSOLS AND CLIMATE

- scattering and absorption of incoming and outgoing radiation

- affects heat budgets of the lower troposphere and the ocean, e.g. semi-enclosed seas

- participation in cloud processes
  - CCN=cloud condensation nuclei
  - sulphate coating of particles

- heterogenous chemistry taking place on dust particles (sulphur cycle, greenhouse gases)

- marine biogeochemistry (iron, nutrients)

Formation

- break-up and agglomeration

- primary aerosols
  emissions
    -anthropogenic (urban, industrial, land use practice)
    -natural (volcanism, aeolian dust, sea-spray)

- secondary aerosols
  particle formation by gas reactions

Transformation

- Condensation
  CCNs and cloud forrming processes

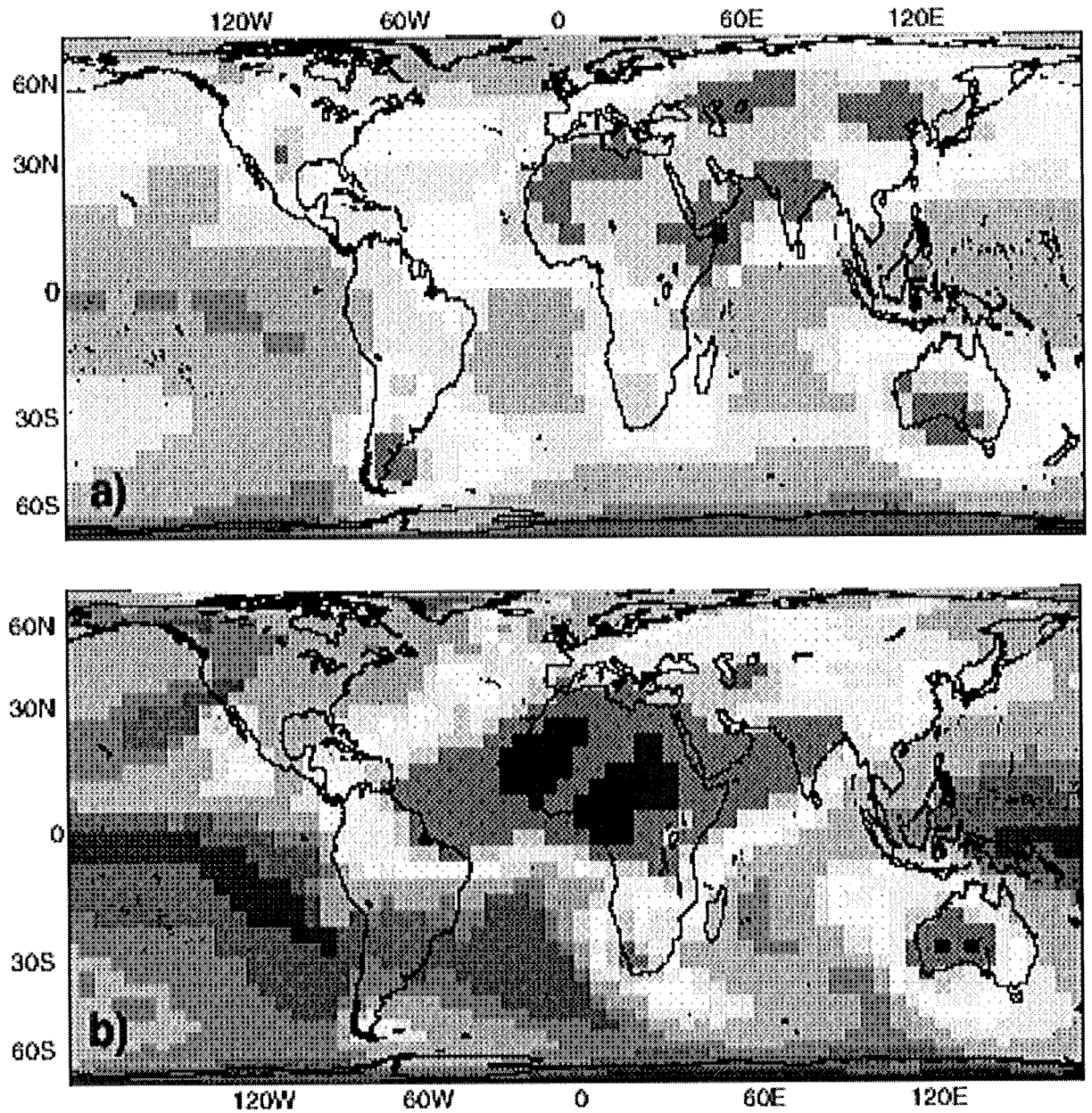
- Oxidation and Neutralization

- Coagulation

Removal

- Dry deposition

- Wet deposition


120W
60W
0
60E
120E
60N
30N
0
30S
60S
a)
b)
g/m2/yr
0 0.01 0.05 0.1 0.5 1 2 5 10 50 100

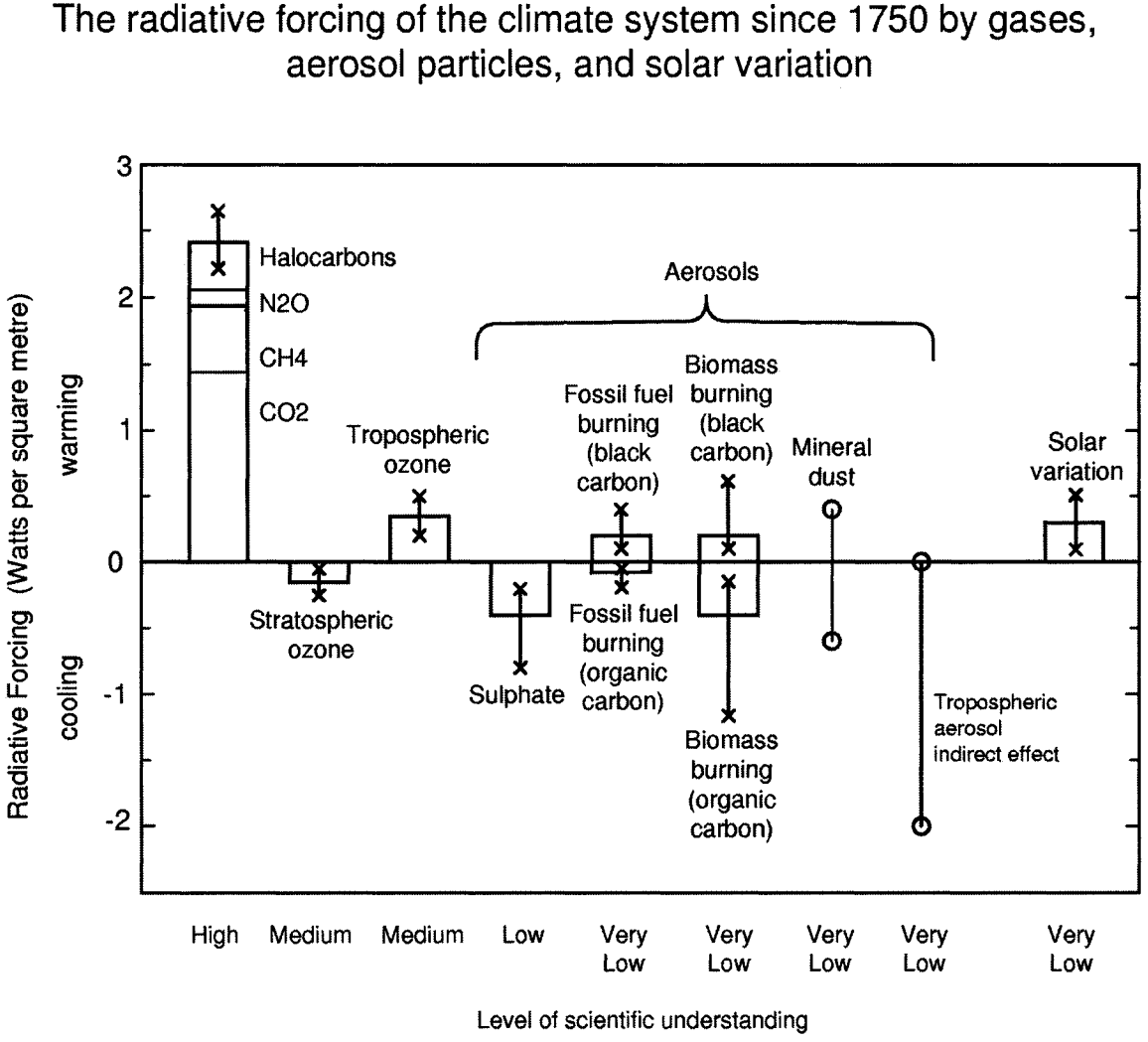


Figure 2: The contributors to climate change are quite varied. These include changes in the atmospheric concentrations of greenhouse gases and aerosols and variation in the output of the sun. All but the solar variation are strongly linked to some form of human activity. The bars represent the best estimates of the relative contributions of changes in these climate forcings - some yielding warming, some yielding cooling - from 1750 to the present. Some of the radiative forcing agents are well mixed over the globe, such as CO_2, thereby perturbing the global heat balance. Others represent only regional perturbations because of their limited spatial distribution, such as aerosols. Climate models include the better-characterized forcing agents. The simulations indicate that the estimated net effect of these perturbations is to have warmed the global climate since 1750, with the most perturbation and hence the most warming being in the past century.

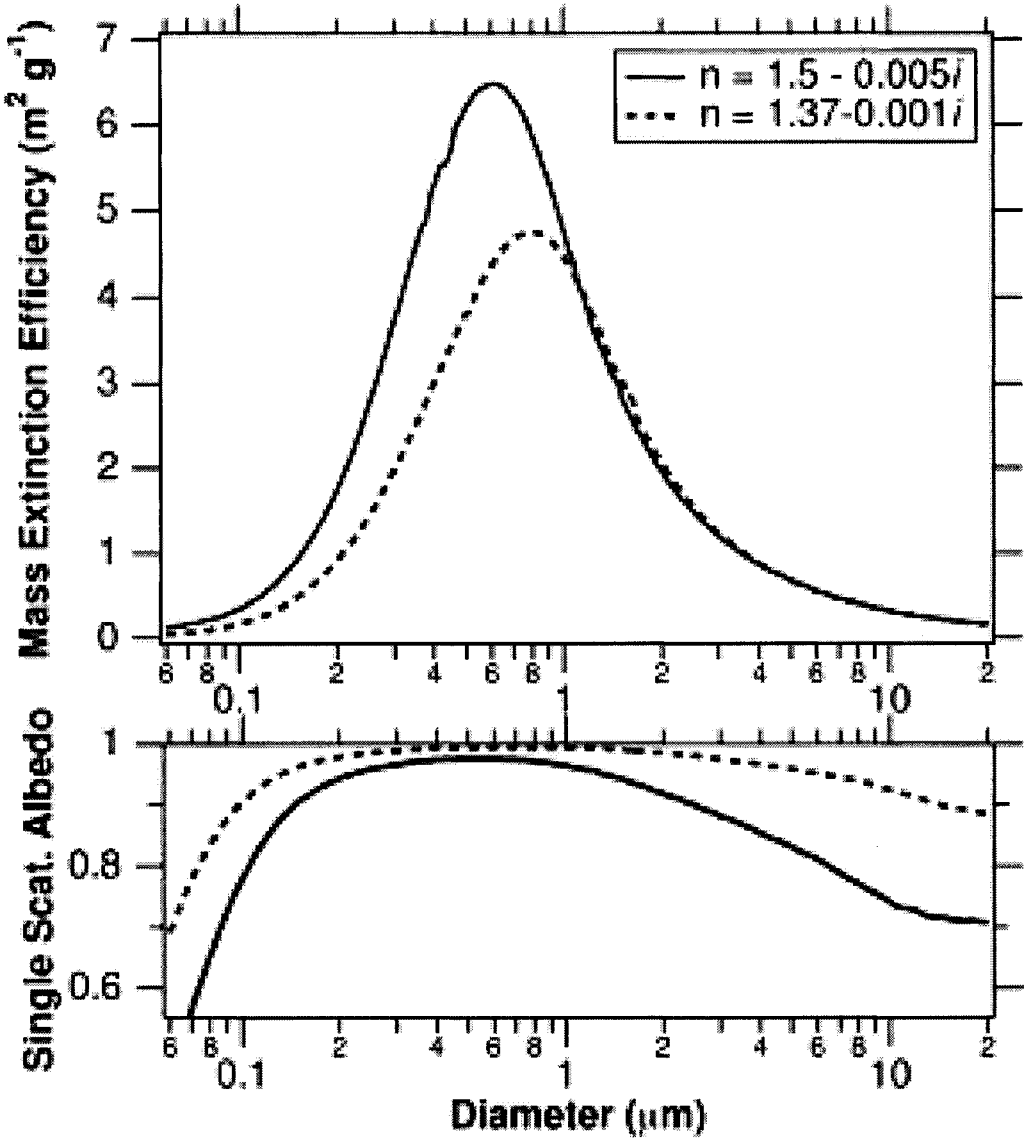


Figure 5.1. Extinction efficiency and single scattering albedo of aerosols. The calculations are integrated over a typical solar spectrum rather than using a single wavelength. Aerosols with diameters between about 0.1 and 2 scatter the most light per unit mass. Coarse mode aerosols have a smaller single scattering albedo even if they are made of the same material (i.e. refractive index) as accumulation mode aerosols. If the refractive index 1.37-0.001i viewed as that of a hydrated aerosol then the curve represents the wet scattering efficiency. The dry scattering efficiency would be larger and shifted to slightly smaller diameters.

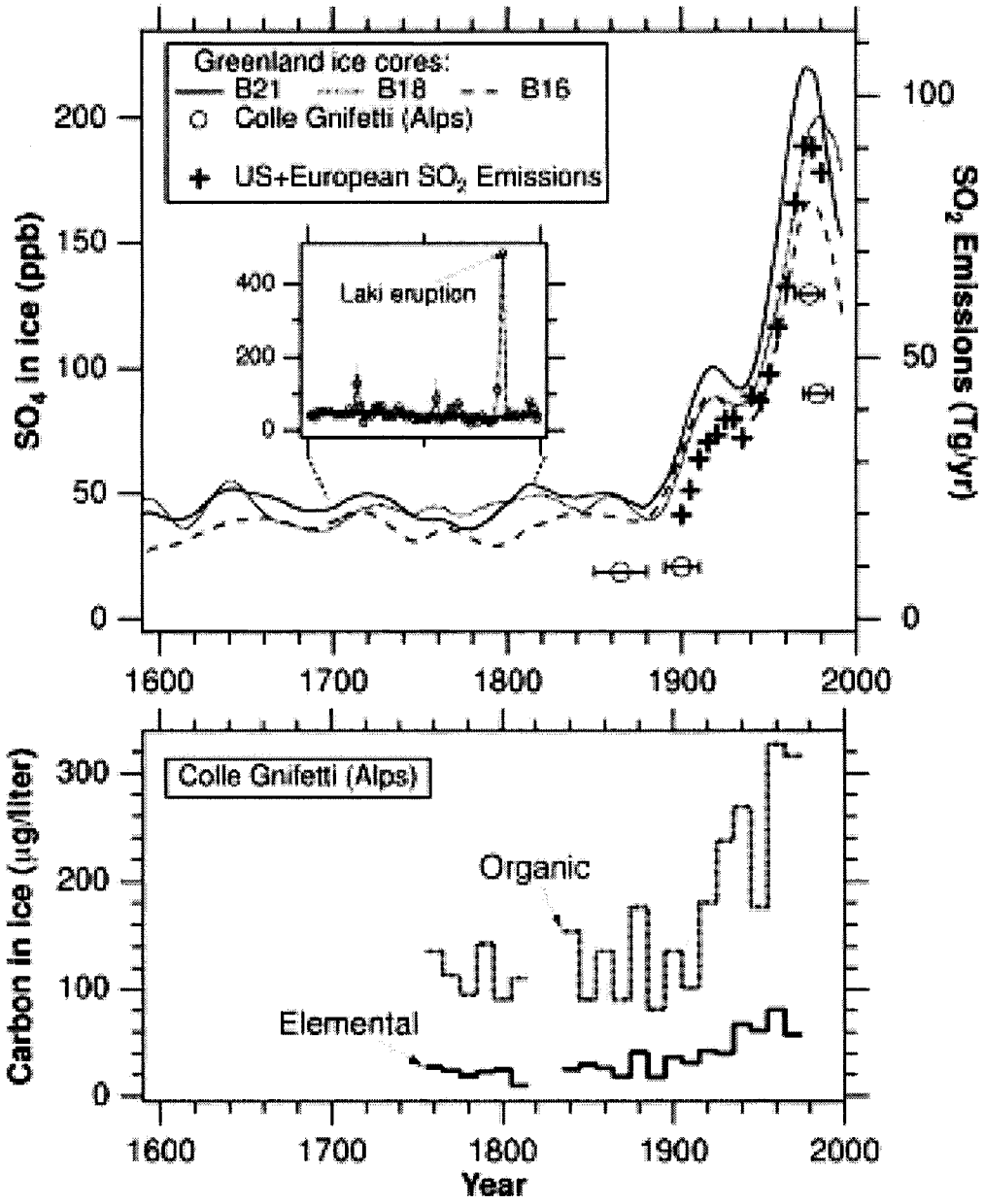


Figure 5.4a. Sulfate concentrations in several Greenland ice cores and an Alpine ice core (Fischer *et al.*, 1998; Dixcher *et al.*, 1995). Also shown are the total SO_2 emissions from sources from the US and Europe (Gschwandtner *et al.*, 1986; Mylona, 1996). The inset shows how peaks due to major volcanic eruptions have been removed by a robust running median method followed by singular spectrum analysis.

Figure 5.4b. Black carbon and organic carbon concentrations in alpine ice cores (Lavanchy *et al.*, 1999).

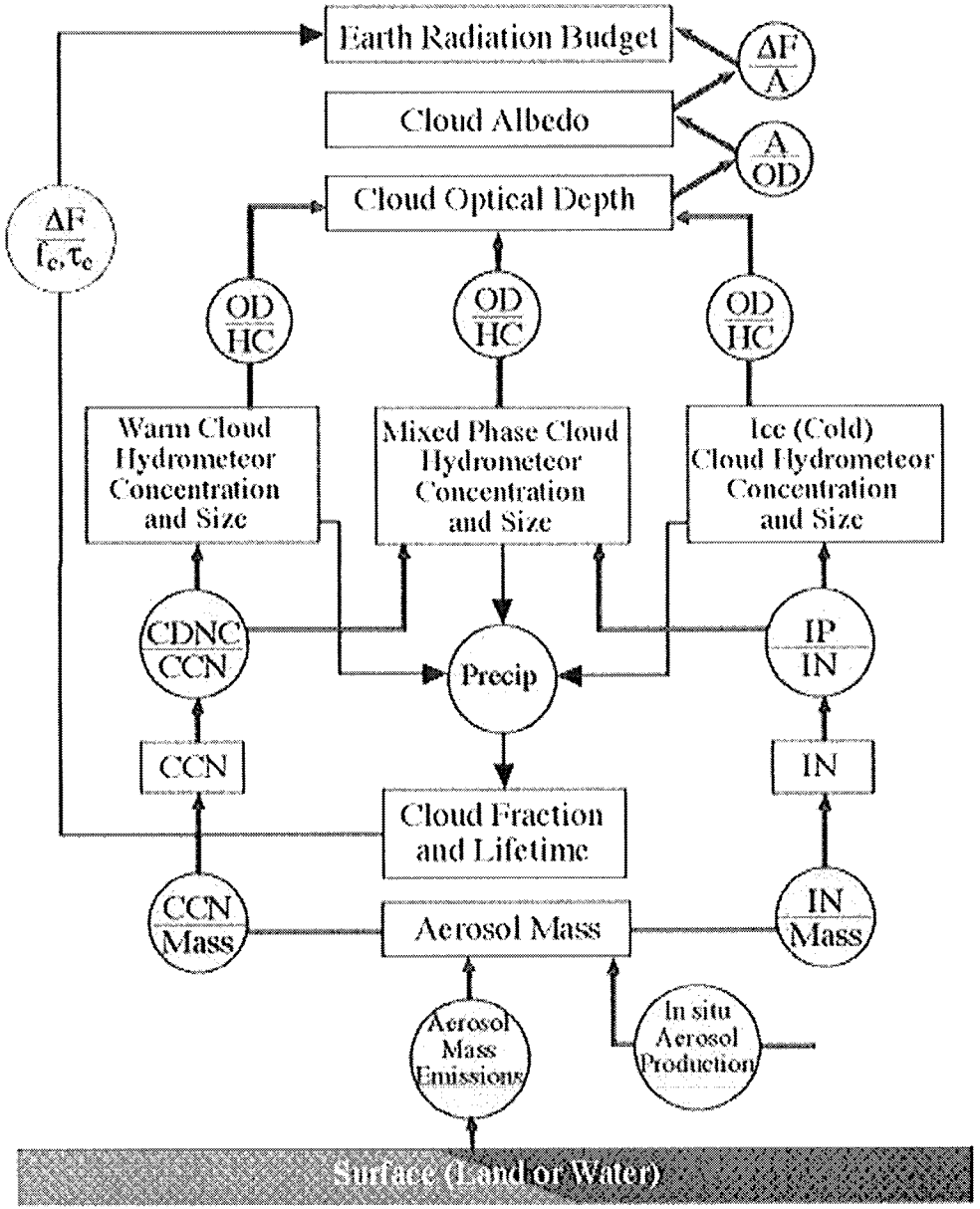
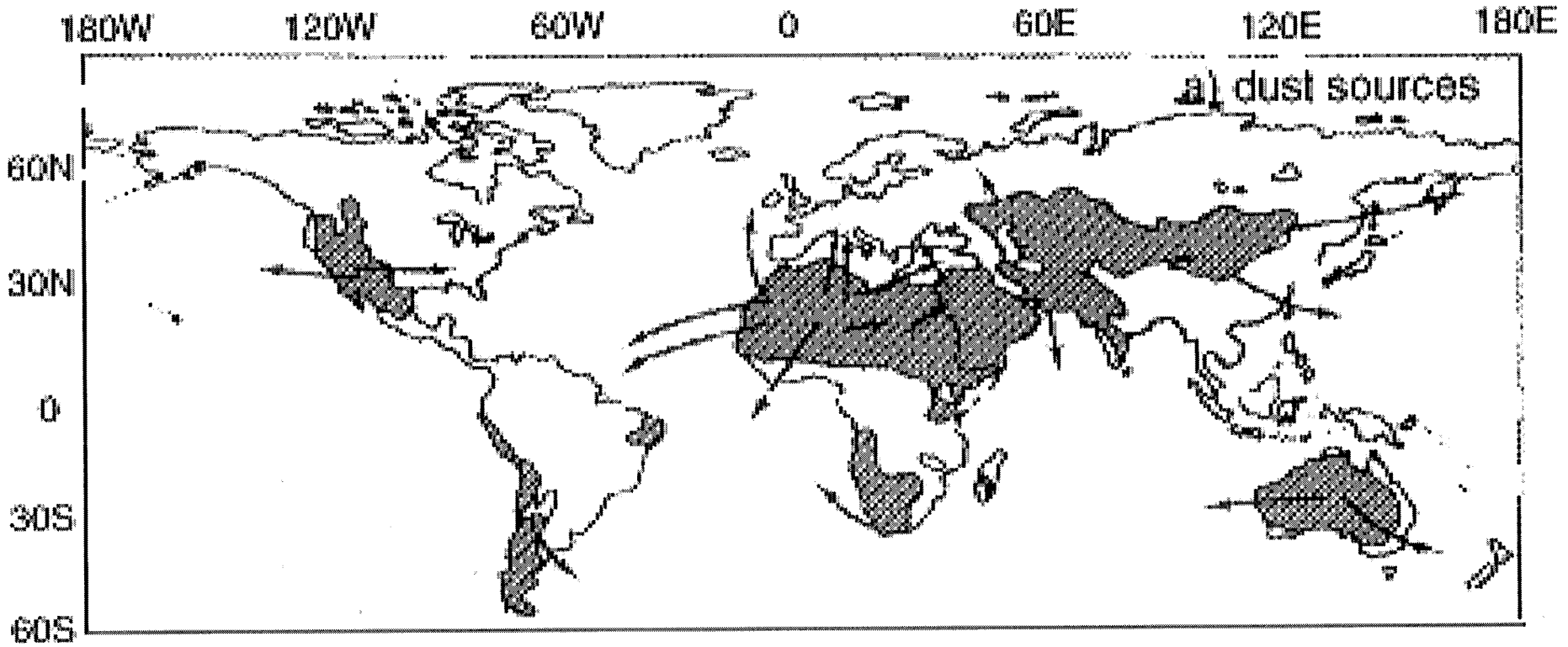
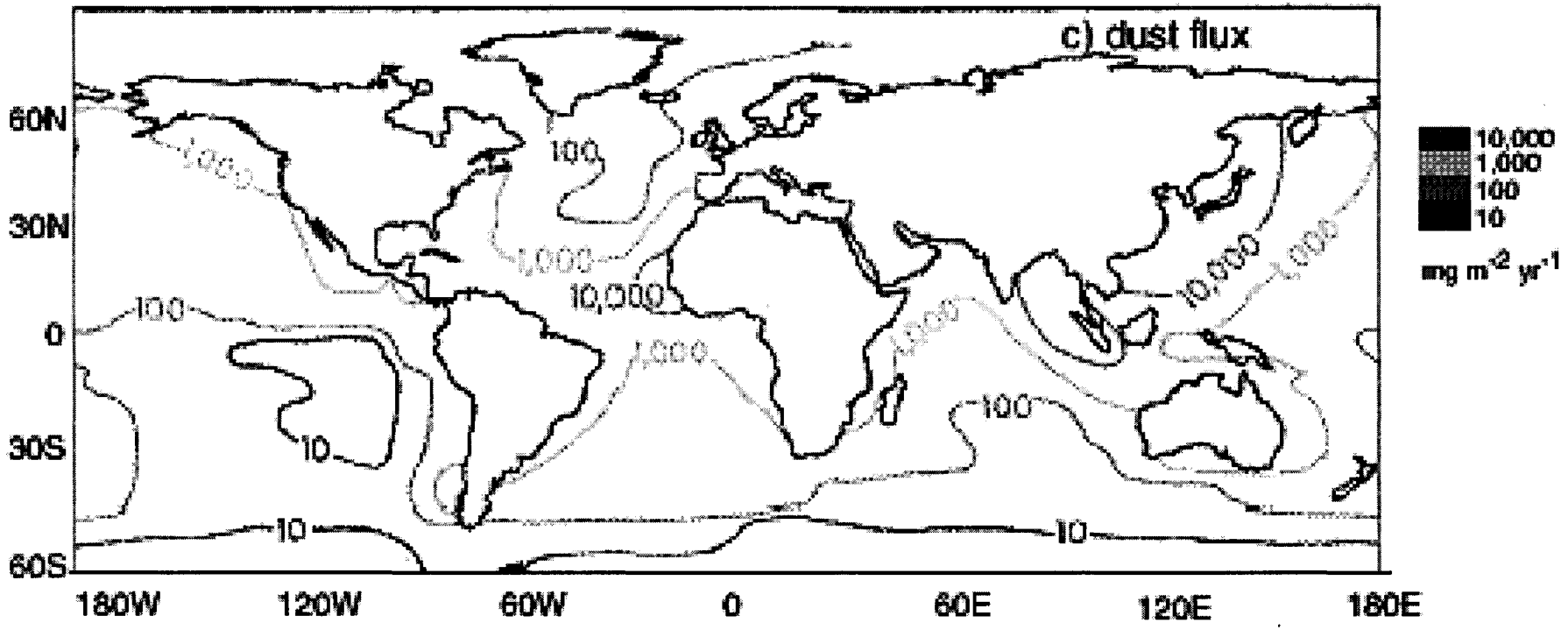
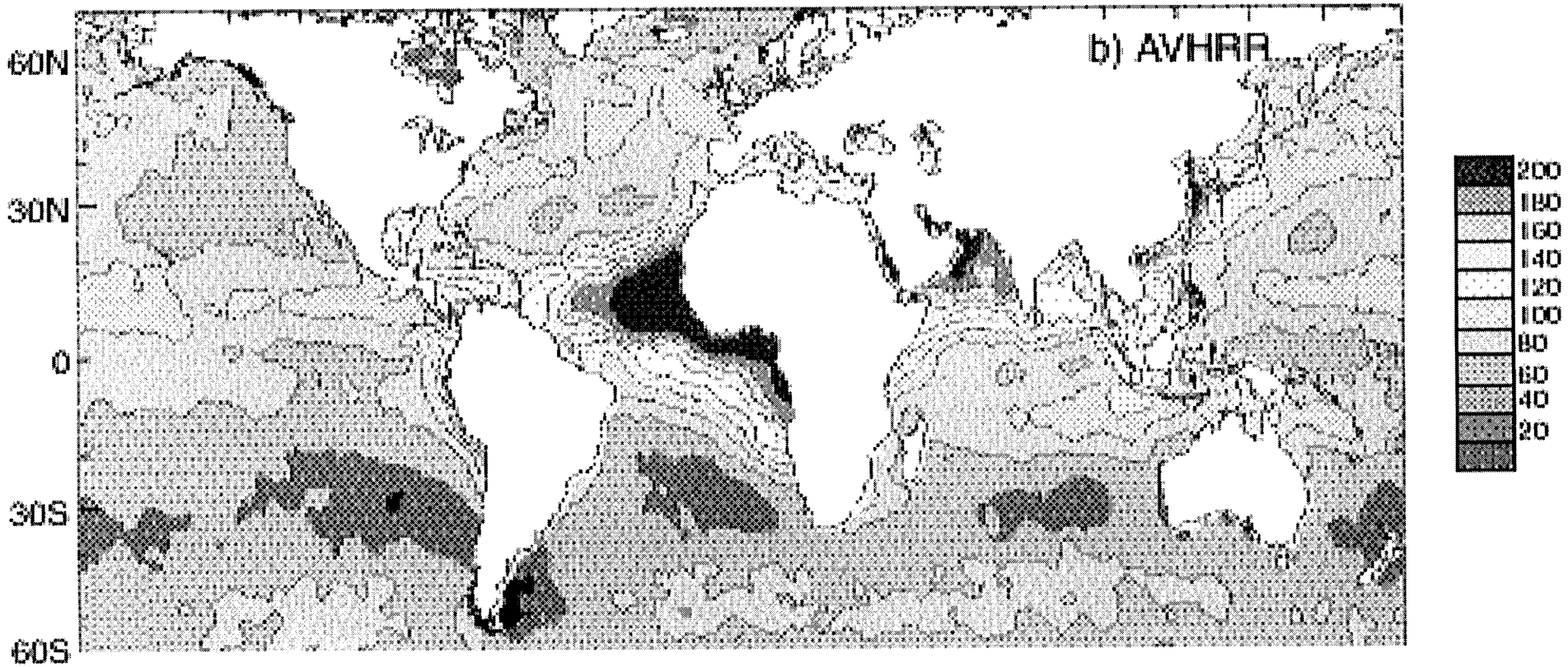


Figure 5.5. Flow chart showing the processes linking aerosol emissions or production with changes in cloud optical depth and radiative forcing. Bars indicate functional dependence of the quantity on top of the bar to that under the bar. Symbols: CCN (Cloud condensation nuclei); CDNC (Cloud droplet number concentration); IN (Ice nuclei); IP (Ice particles); OC (Optical depth); HC (Hydrometeor concentration); A (Albedo); fc (Cloud fraction); τ_c (Cloud optical depth).


180W
120W
60W
0
60E
120E
180E
60N
30N
0
30S
60S
a) dust sources


c) dust flux
60N
30N
0
30S
60S
180W
120W
60W
0
60E
120E
180E
100
1,000
10,000
10
10,000
1,000
100
10
10
10
10,000
1,000
100
10
mg m-2 yr-1


b) AVHRR
60N
30N
0
30S
60S
200
180
160
140
120
100
80
60
40
20

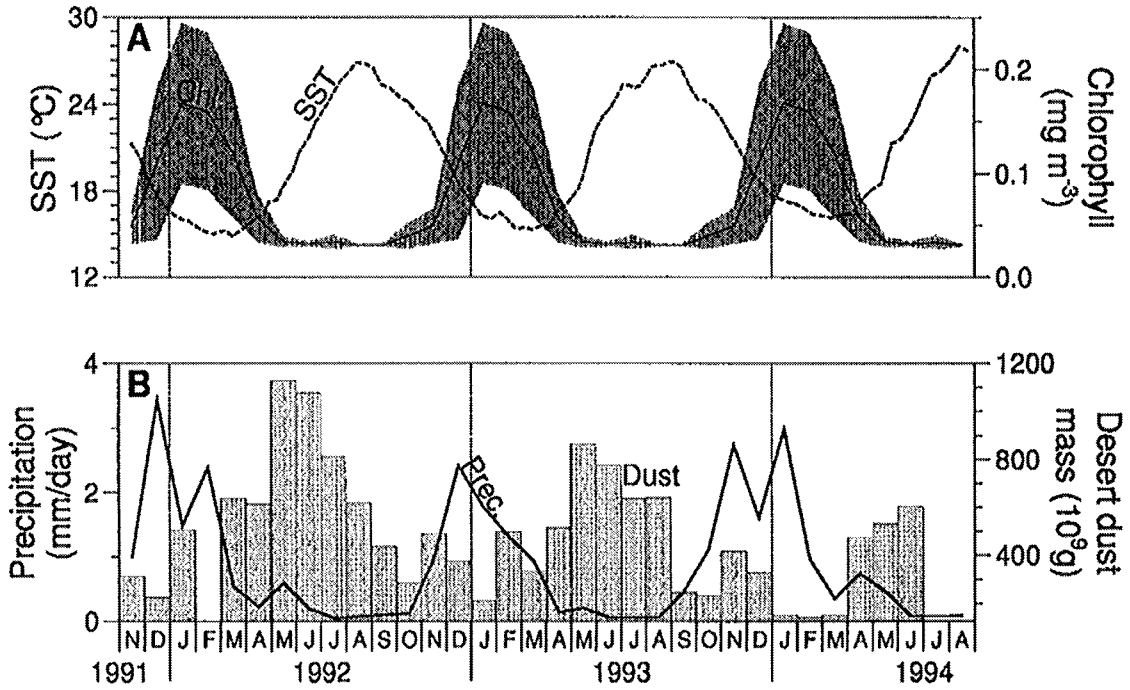


Fig. 6. Satellite observations: (a) Monthly averaged pigment concentrations observed for the period 1979–1985 derived from CZCS data (solid line) at the trap location, with the gray area indicating the 1 standard deviation for each month, and the weekly composite AVHRR sea surface temperature record at the trap location during the deployment period (dashed line). (b) Precipitation at the trap location (line) and desert dust mass over the Mediterranean (bars) during the deployment period. The dust data were redrawn after Dulac et al. (1996).

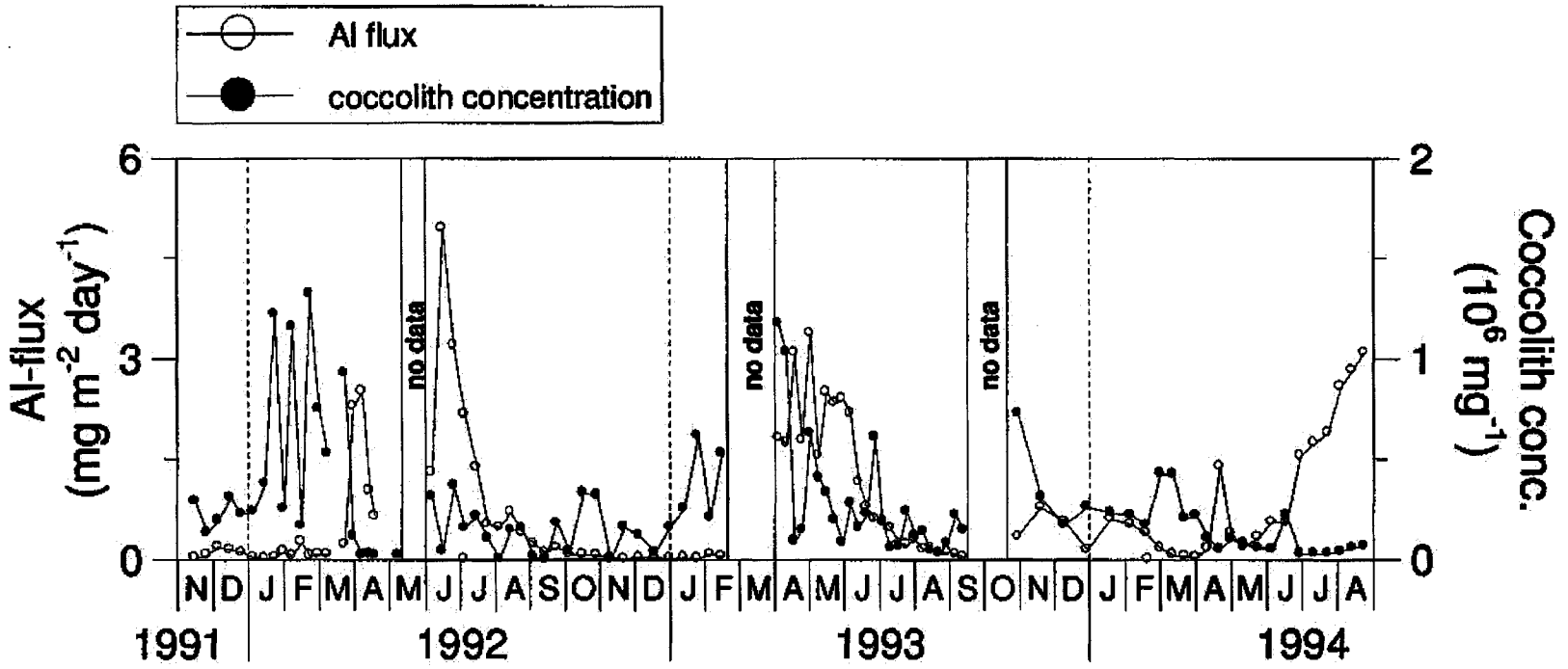
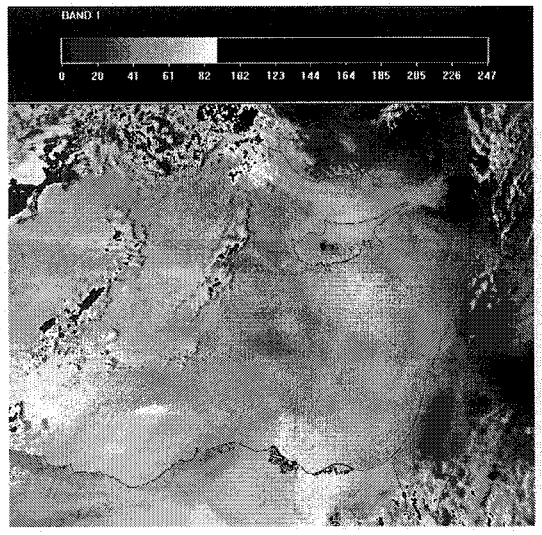
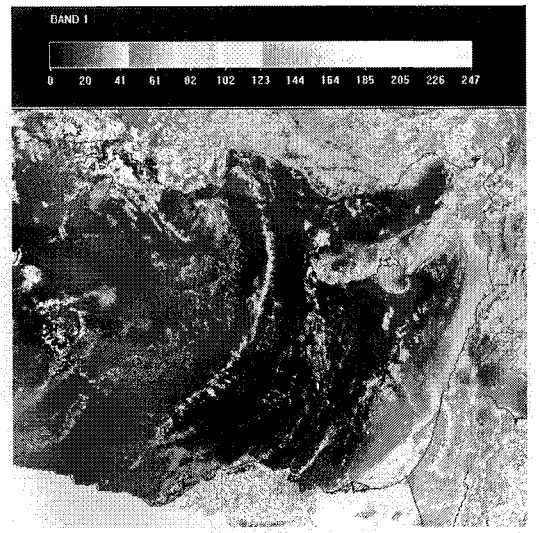


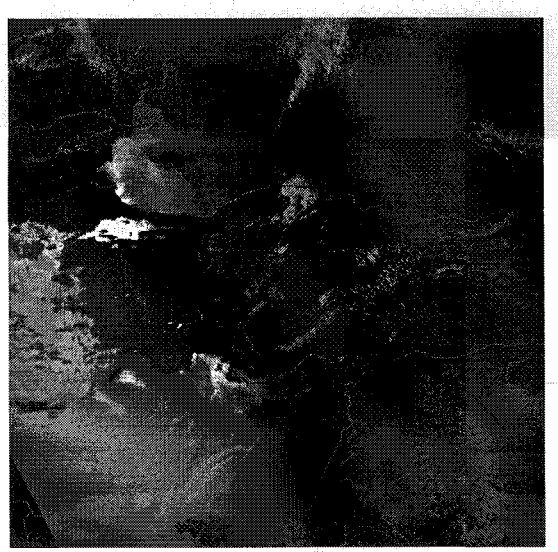
Fig. 5. Flux record for Al ($m^{-2}\,day^{-1}$) and coccolith concentration (number mg^{-1}).



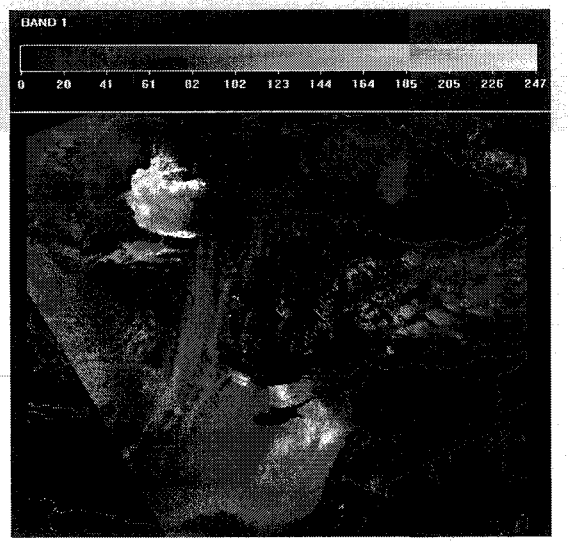
NOAA-11 AVHRR Ch 4
6 April 1994



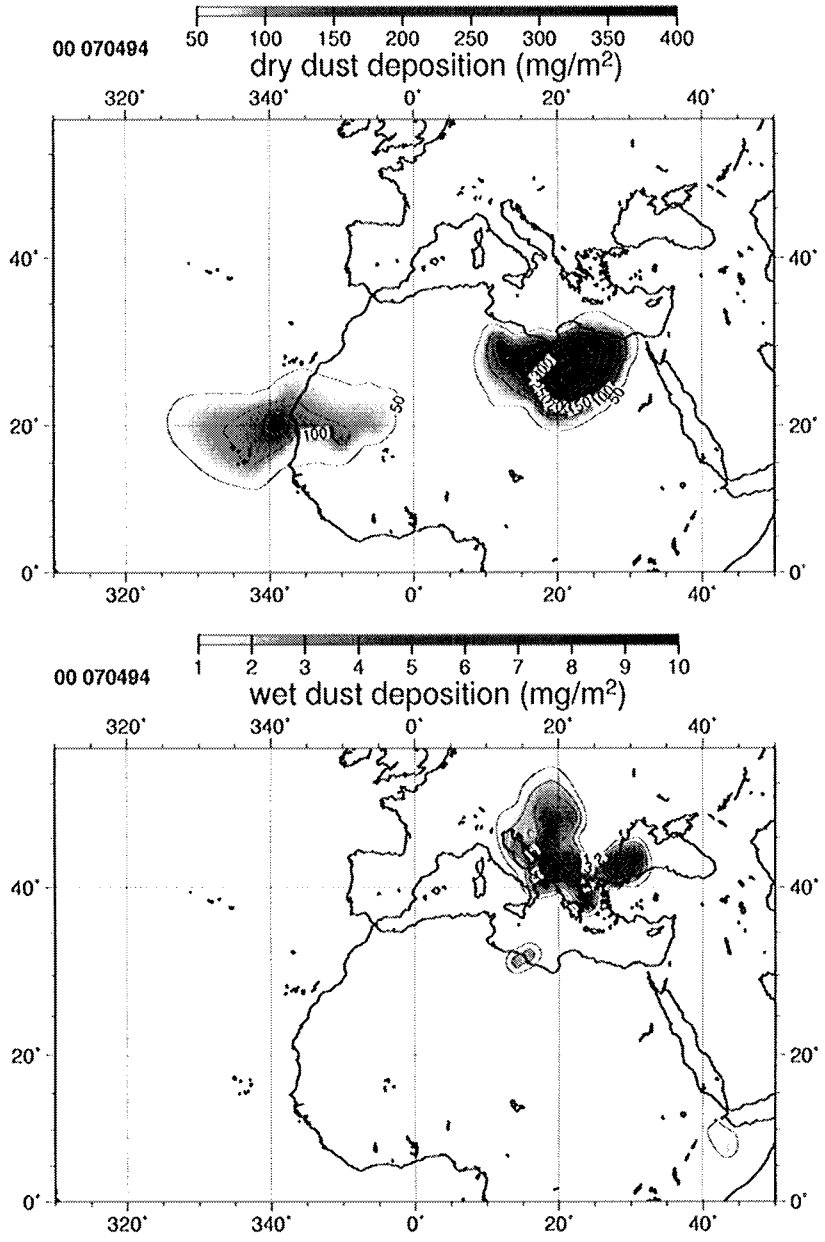
NOAA-11 AVHRR ch. 4
7 April 1994



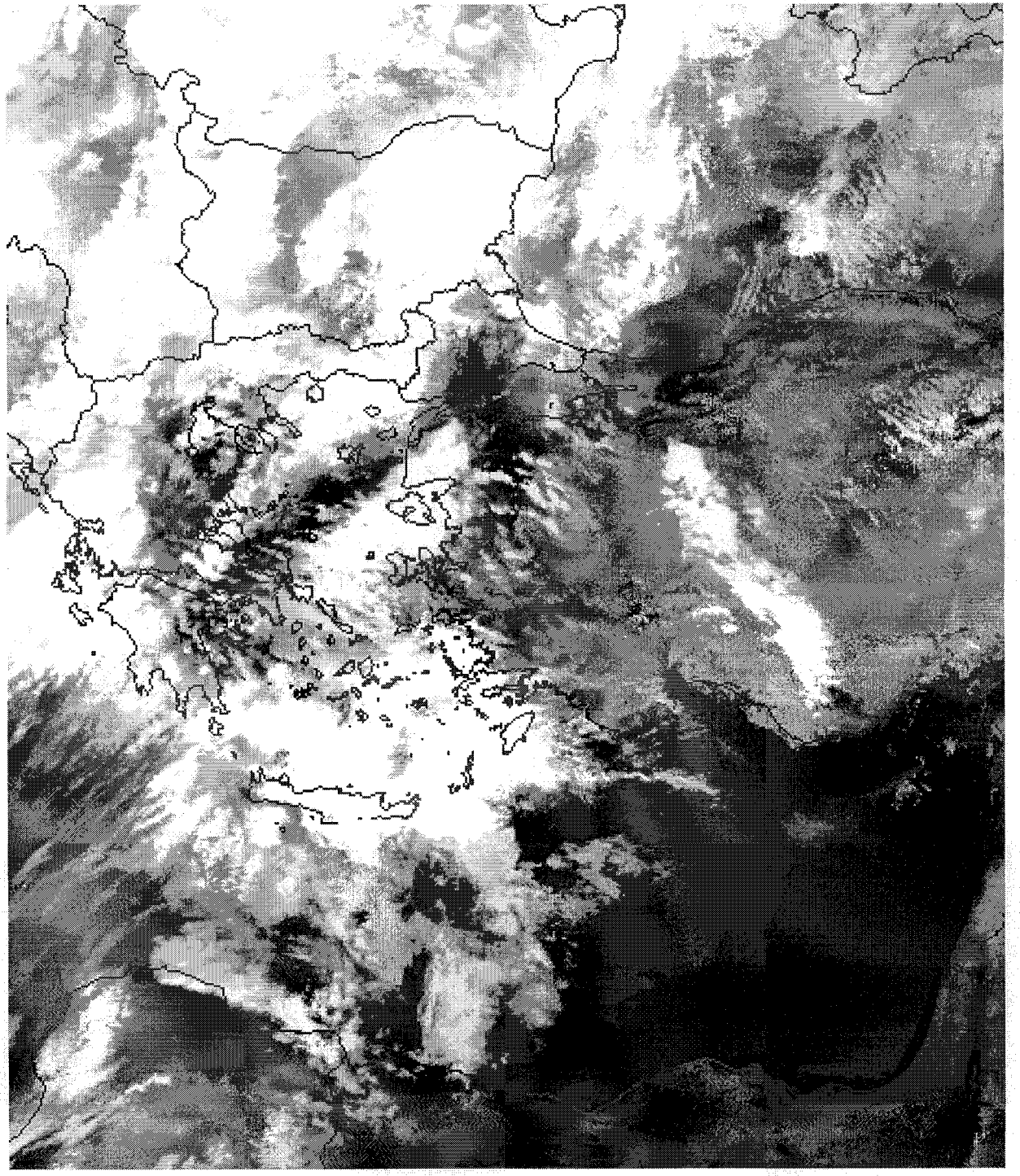
NOAA-11 AVHRR Ch. 1
14 April 1994

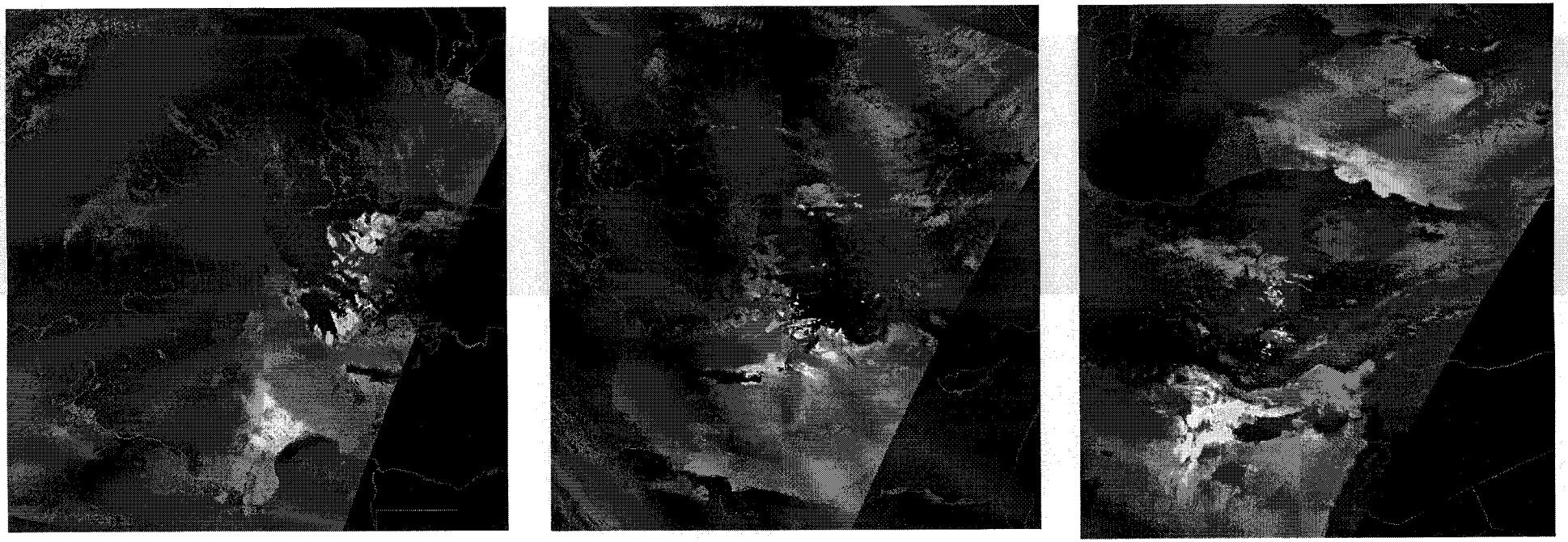


NOAA-11 AVHRR Ch. 1
15 April 1994

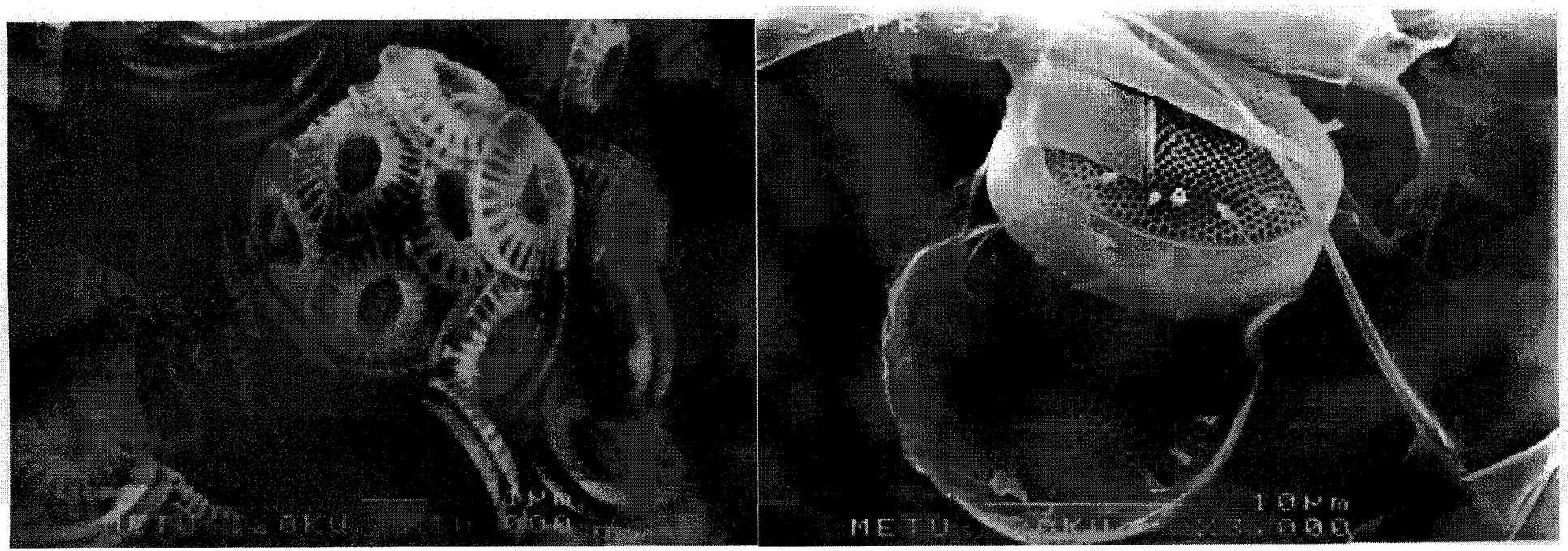

00 070494
50 100 150 200 250 300 350 400
dry dust deposition (mg/m2)
320° 340° 0° 20° 40°
40°
20°
0°
50
100
150
200
00 070494
1 2 3 4 5 6 7 8 9 10
wet dust deposition (mg/m2)
320° 340° 0° 20° 40°
40°
20°
0°

NOAA-099
AVHRR Ch.4
on 28 March 1994





NOAA-12 AVHRR Ch. 4 images on 4, 5 and 6 April 1995



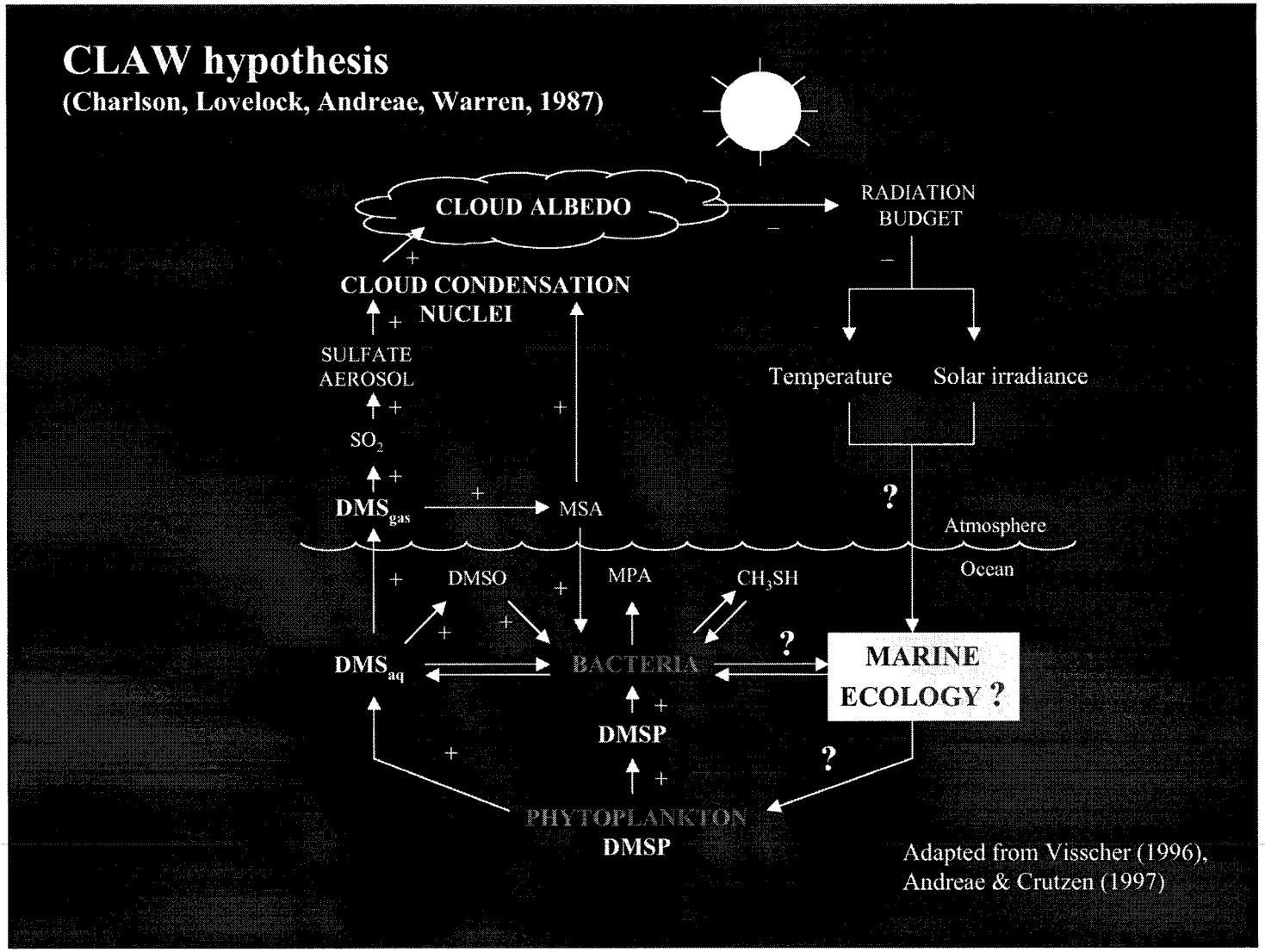
SEM images of sea-water samples collected from 32°E 35°-36°N by 32°E on 6 April 1995

Coccolith *Emiliana Huxleyi aestivalis*

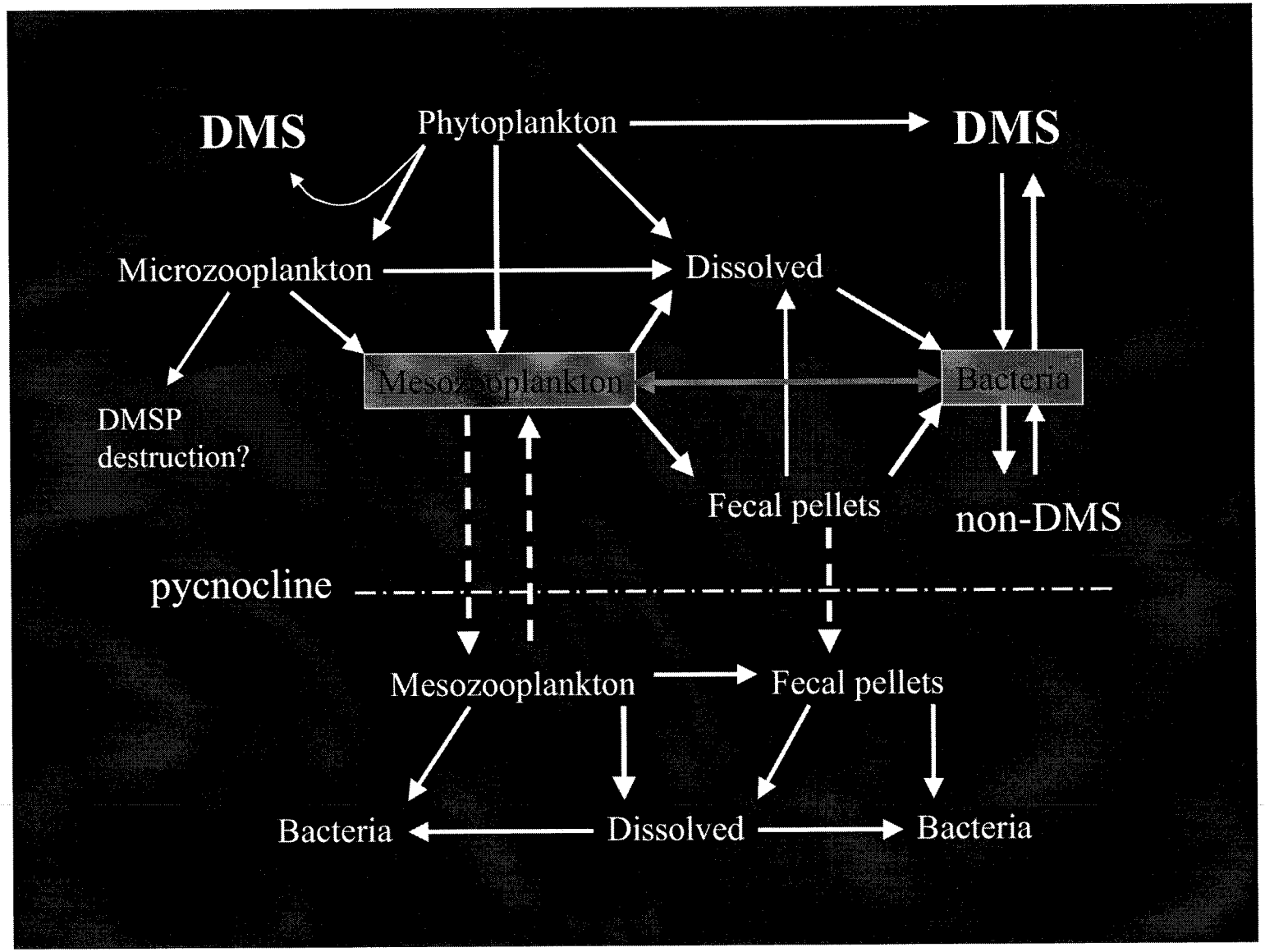
Thalassiosira

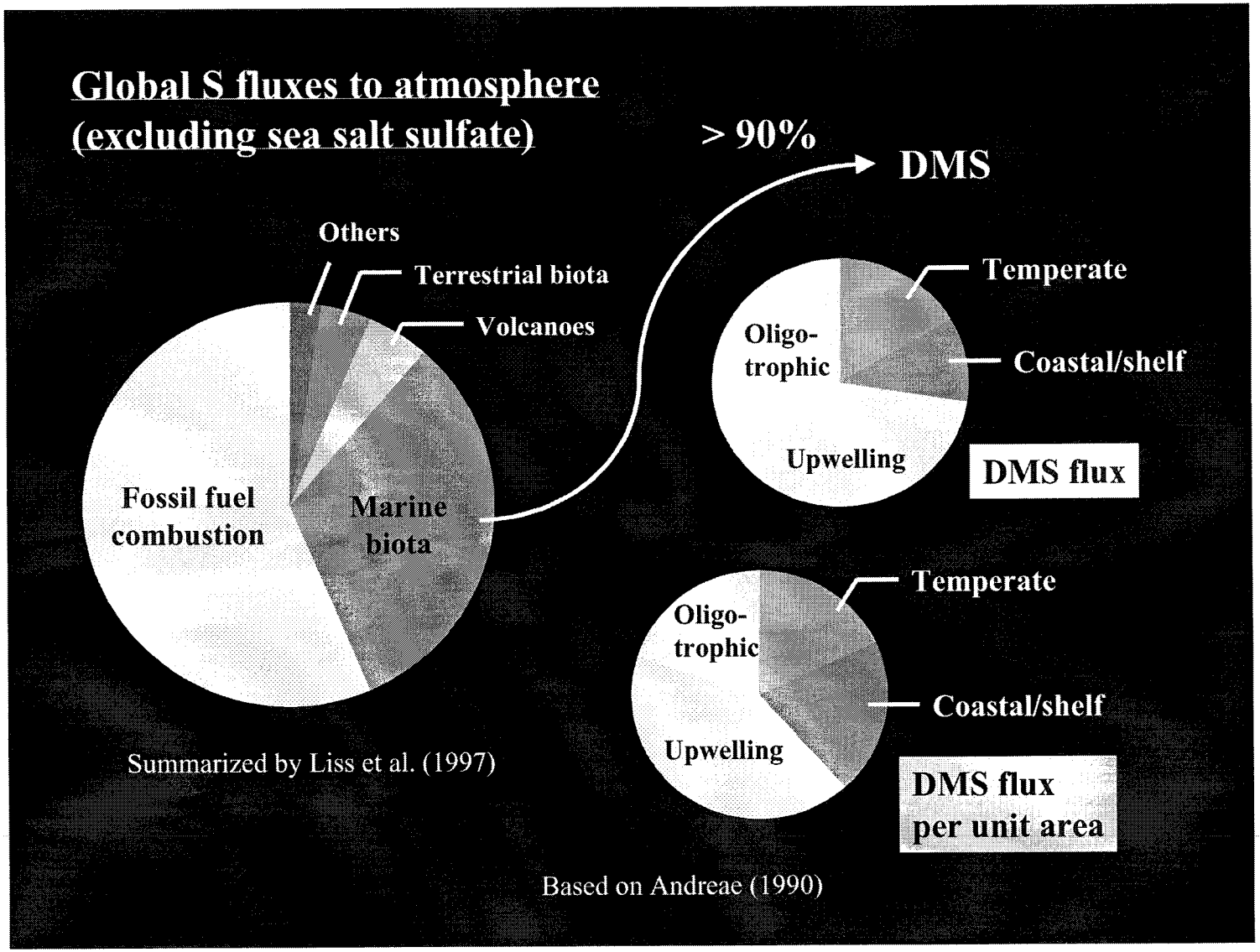
CLAW hypothesis

(Charlson, Lovelock, Andreae, Warren, 1987)



Adapted from Visscher (1996), Andreae & Crutzen (1997)


DMS
Phytoplankton
DMS
Microzooplankton
Dissolved
Mesozooplankton
Bacteria
DMSP
destruction?
Fecal pellets
non-DMS
pycnocline
Mesozooplankton
Fecal pellets
Bacteria
Dissolved
Bacteria


Global S fluxes to atmosphere
(excluding sea salt sulfate)
> 90%
DMS
Others
Terrestrial biota
Volcanoes
Fossil fuel combustion
Marine biota
Summarized by Liss et al. (1997)
Temperate
Oligo-trophic
Coastal/shelf
Upwelling
DMS flux
Temperate
Oligo-trophic
Coastal/shelf
Upwelling
DMS flux per unit area
Based on Andreae (1990)

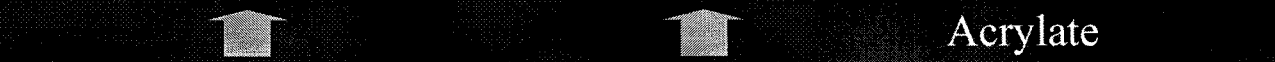
DMS and sulfur cycle

Lovelock et al. (1972) Nature 237: 452–453

- Missing flux of oceanic volatile sulfur compounds
- H_2S ?
- Dimethyl sulfide: $H_3C\text{-}S\text{-}CH_3$
  - Ubiquitous in surface ocean
  - "Smell of the sea"
  - Major biogenic sulfur compound for sea-air exchange

DMSP-to-DMS conversion in the ocean

DMSP-lyase (phytoplankton, bacteria)



Acrylate

Dimethylsulfoniopropionate (DMSP)

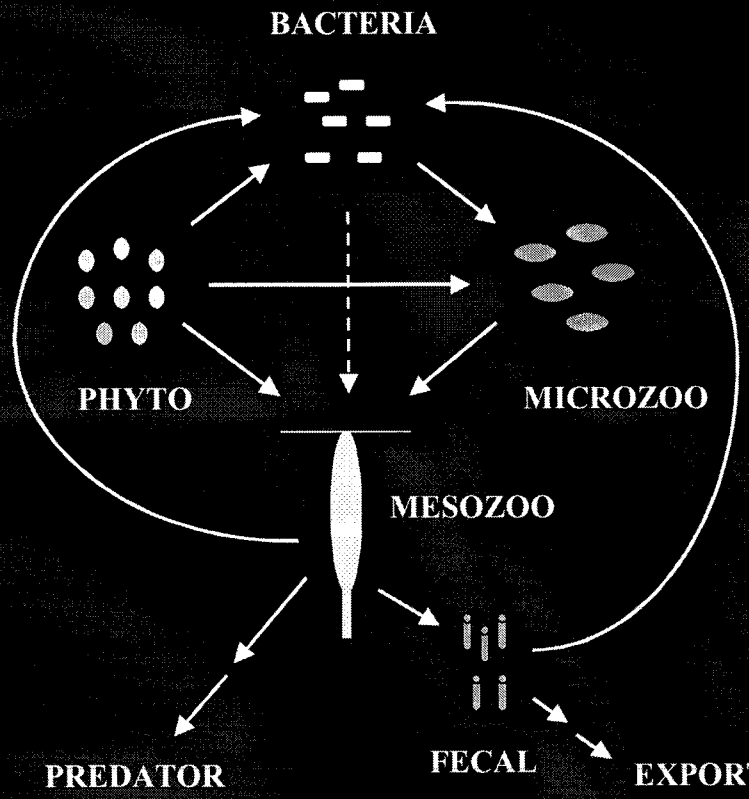
Dimethyl sulfide (DMS)

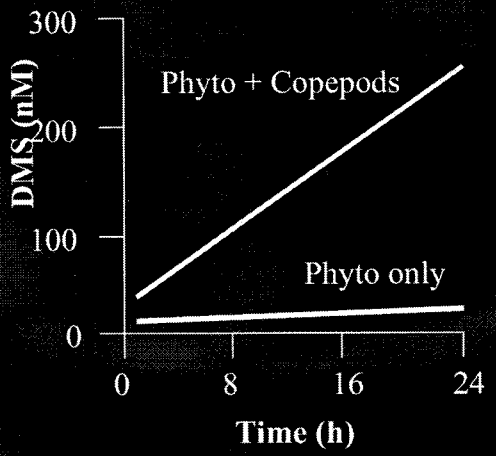
osmolyte / cryoprotectant in marine algae

> 90% sea-to-air biogenic sulfur flux; up to 23% global sulfur flux to atmosphere

(Nguyen et al. 1978, 1983, Barnard et al. 1982, Andreae & Raemdonck 1983, Malin 1996, Visscher 1996)

COPEPODS: Gate-keepers of the pelagic microbial food web





(Dacey & Wakeham, 1986)

Iron speciation in the eastern Mediterranean precipitation from the perspective of Sahara dust effects on the marine ecosystem

Türkan Özsoy[1] and A. Cemal Saydam[2]
[1]*Mersin University, Department of Environmental Engineering, Mersin, Turkey*
[2]*Turkish Scientific and Technical Research Council (TÜB_TAK), Ankara*

Table 1. Statistical results of the measured parameters; pH, conductivity (μS cm^{-1}), soluble Fe, particulate Fe and particulate Al concentrations (μM) in the precipitation samples for the whole sampling period at Erdemli. The number of the samples are given in parenthesis.

Parameter	Arit. mean	Geo. Mean	VWM	Min.-Max.
pH (87)	5.6±0.9	5.5	4.95	3.5-7.6
Conductivity (41)	74.6±79.6	51.4	-	12.9-391.0
Fe(II) (83)	0.110±0.108	0.062	0.054	BDL-0.422
Fe(III) (83)	0.028±0.079	0.008	0.014	BDL-0.664
Fe_{Ts} (84)	0.141±0.150	0.084	0.069	BDL-0.994
Fe_{filt} (77)	1.469±3.650	0.350	1.030	0.028-26.6
Fe_{par} (84)	22.60±68.60	2.50	16.32	0.07-534.0
Total Fe (80)	24.31±69.83	3.99	17.33	0.12-534.0
Al_{par} (84)	88.77±247.50	15.28	56.31	0.37-1843.0

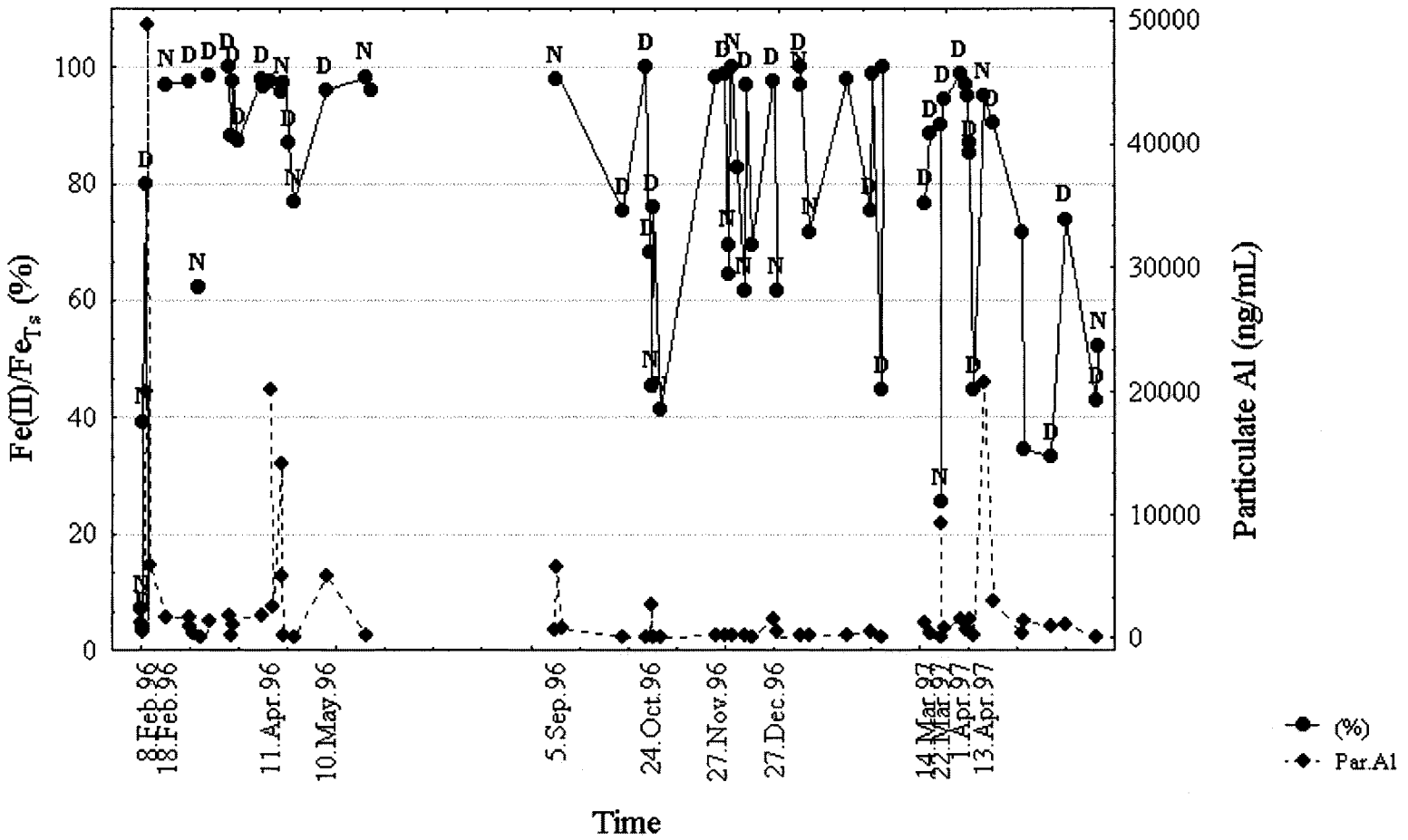
total iron: $Fe_{tot} = Fe_{filt} + Fe_{par}$

particulate iron: Fe_{par}

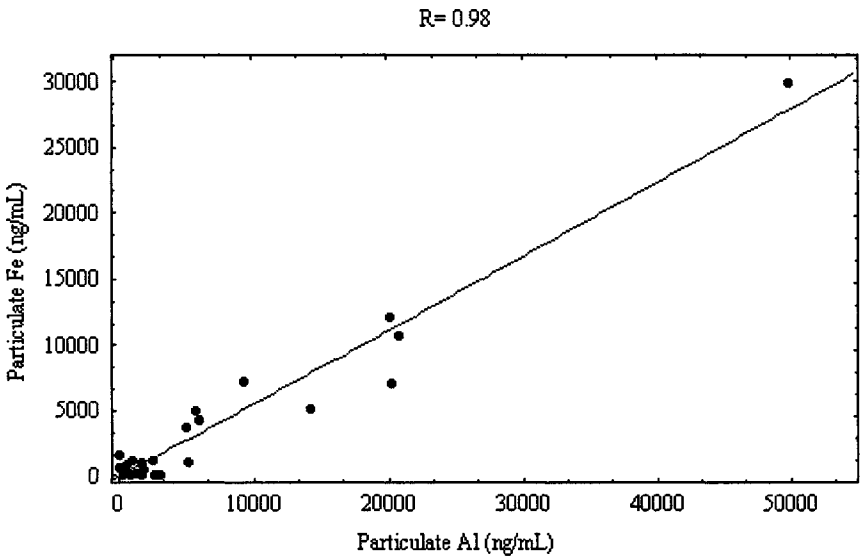
filterable iron: Fe_{filt} = colloidal + soluble

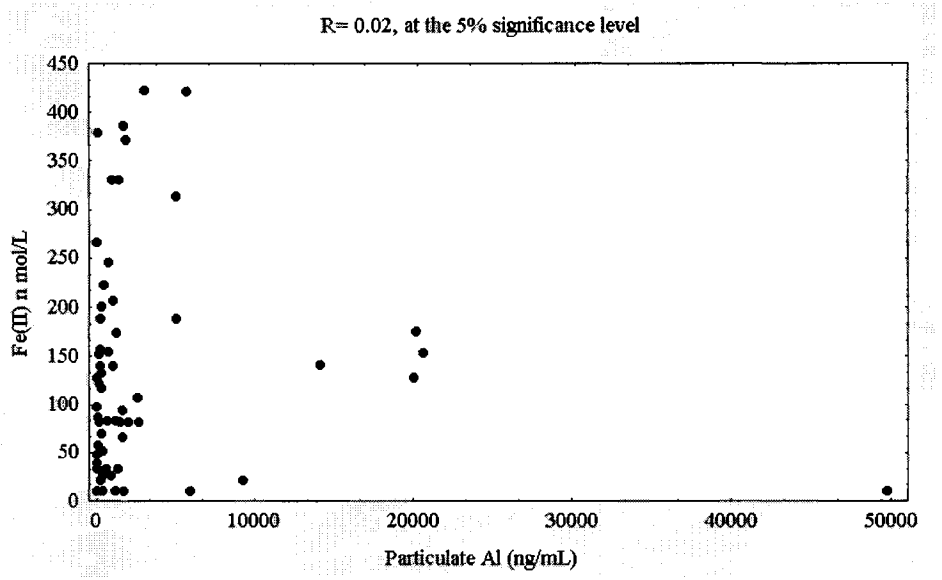
soluble iron: $Fe_{Ts} = Fe(II) + Fe(III)$

kjhkjh



Time series of the ratio of dissolved Fe(II) to Total dissolved Fe and of mineral dust in precipitation samples. Values from daytime samples are denoted with letter D and night samples by letter N.





A strong correlation (R=0.98) exists between particulate Fe and Al fractions, as both are mainly of crustal origin.

Very weak correlation (R=0.02) between particulate Al (i.e. mineral dust) and Fe(II) concentrations suggests there is no link between soluble Fe(II) and mineral dust load. Most soluble iron appears to be present without dust, except for some outlyers with high dust load.

Conclusions:

1. A strong correlation exists between particulate Fe and Al fractions of precipitation, both of crustal origin. No correlation was observed between the soluble and insoluble fractions of iron as well as between the soluble species of Fe(II) and Fe(III) and of Fe_{Ts} and Fe_{filt} concentrations. It was found that the Fe_{filt} fraction, measured by AAS after filtering, and frequently (and incorrectly) interpreted to be the soluble iron fraction in the literature, represents mostly colloidal iron. The correct measure of soluble iron is Fe_{Ts}.

2. The soluble Fe(II) concentration of precipitation varied independently of the concentration of particulate Fe, hence of the mineral dust load scavenged from the local atmosphere. The volume weighted mean Fe_{filt} concentration of the precipitation samples collected during the episodic **"red dust"** events were found to be relatively higher. The geometric mean ratio of soluble Fe(II) and Fe_{Ts} to Total Fe were found to be 1.6% and 2.1% respectively, while the mean ratio of Fe_{filt} to Total Fe was 9.6%. It is noteworthy that the lowest ratios for both species obtained from the precipitation samples thus mineral dust concentrations were relatively high. The solubility of Fe(II) obtained from this study is exactly the same as the value obtained from mineral aerosol samples collected at Barbados (*Zhu et al.*, 1997).

3. During spring months, the flux of soluble Fe(II) fraction in most atmospheric wet deposition events is sufficient to support the maximum primary production rate expected in the Eastern Mediterranean Sea.

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A CASE STUDY OF ATMOSPHERIC SULFATE AEROSOLS AND THEIR POSSIBLE BIOGENIC SOURCES

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Among the biogenic sources of sulfate aerosols, reduced sulfur gases such as dimethylsulfide (DMS) constitute and control a significant fraction of the atmospheric nss-sulfate budget (*Charlson et al.*, 1987, e.g.).

Certain species of phytoplankton, e.g:

-*Phaeocystis pouchetii* (dinoflagellate)

-*Emiliana huxleyi* (coccolithophore)

are known to be important producers of DMS (*Matrai and Keller*, 1993, e.g.). Biogenic sulfur, created mainly by seasonal phytoplanktonic activity, could be particularly rich sources in certain regions and dominate the atmospheric sulfur cycle.

Daily samples of aerosol collected at the Turkish coast of the Northeastern Mediterranean Sea, Erdemli during October 1991-December 1992 were analyzed by IC to determine the major anions; sulfate, nitrate and chloride concentrations.

The mean nss-sulfate concentration was found to be one of the highest among the reported values for rural areas worldwide.

Table 1. Comparison of the major anion concentrations of aerosol samples (all species in μg/m^3) with the data reported from various locations around the world.

Location	Cl^-	NO_3^-	nss-SO_4^{2-}
Erdemli[a](303)	5.55±11.3	2.74±2.3	6.57±6.54
Eastern Med. Finokalia[b] (49)	1.38±1.69	1.51±1.13	8.20±4.14
Eastern Med. Etzion, Israel[c] (169)	-	-	7.43±6.91#
Eastern Med. Etzion, Israel[c] (93)	-	-	6.05±3.89#
Western Med.[d]	-	-	4.63*
Western Med. Blanes[e]	-	-	1.28±0.88
Western Med. Mallorca[f] (10)	0.97±0.75	1.03±0.36	1.52±0.60*
Eastern Black Sea[g] (4)	-	2.10±0.80	4.30±1.60
Western Black Sea[g] (14)	-	9.10±2.60	3.10±0.80
U.K., Hazelrigg[h] (65)	4.22±2.45	5.27±5.02	8.29±8.11*
U.K., Hemsby[i] (302)	4.58±3.53	6.82±6.82	5.22±4.66
Northwestern Indian Ocean[j] (96)	-	0.60±0.55	1.60±1.50
Bermuda[k] (78)	3.90	-	0.91±1.70
Barbados[l] (343)	-	-	0.82±0.63
Iceland[m]	-	0.24	0.64
Pacific[n] Oahu (56)	-	0.35±0.18	0.37±0.34
Mid Pacific[n] Fanning (48)	-	0.18±0.08	0.64±0.15

[a]Present study; [b]Mihalopoulos et al. (1997); [c]Luria et al. (1996); [d]Bergametti et al. (1989); [e]Alarcon and Cruzado (1988); [f]Simo et al. (1991); [g]Hacısaliho_lu et al. (1992); [h]Harrison and Pio (1983); [i]Yaaqub et al. (1991); [j]Savoie et al. (1987); [k]Chen and Duce (1983); [l]Arimoto et al. (1992); [m]Prospero et al. (1995); [n]Prospero et al. (1985).

* Calculated from reported SO_4^{2-} and Na^+ concentrations.

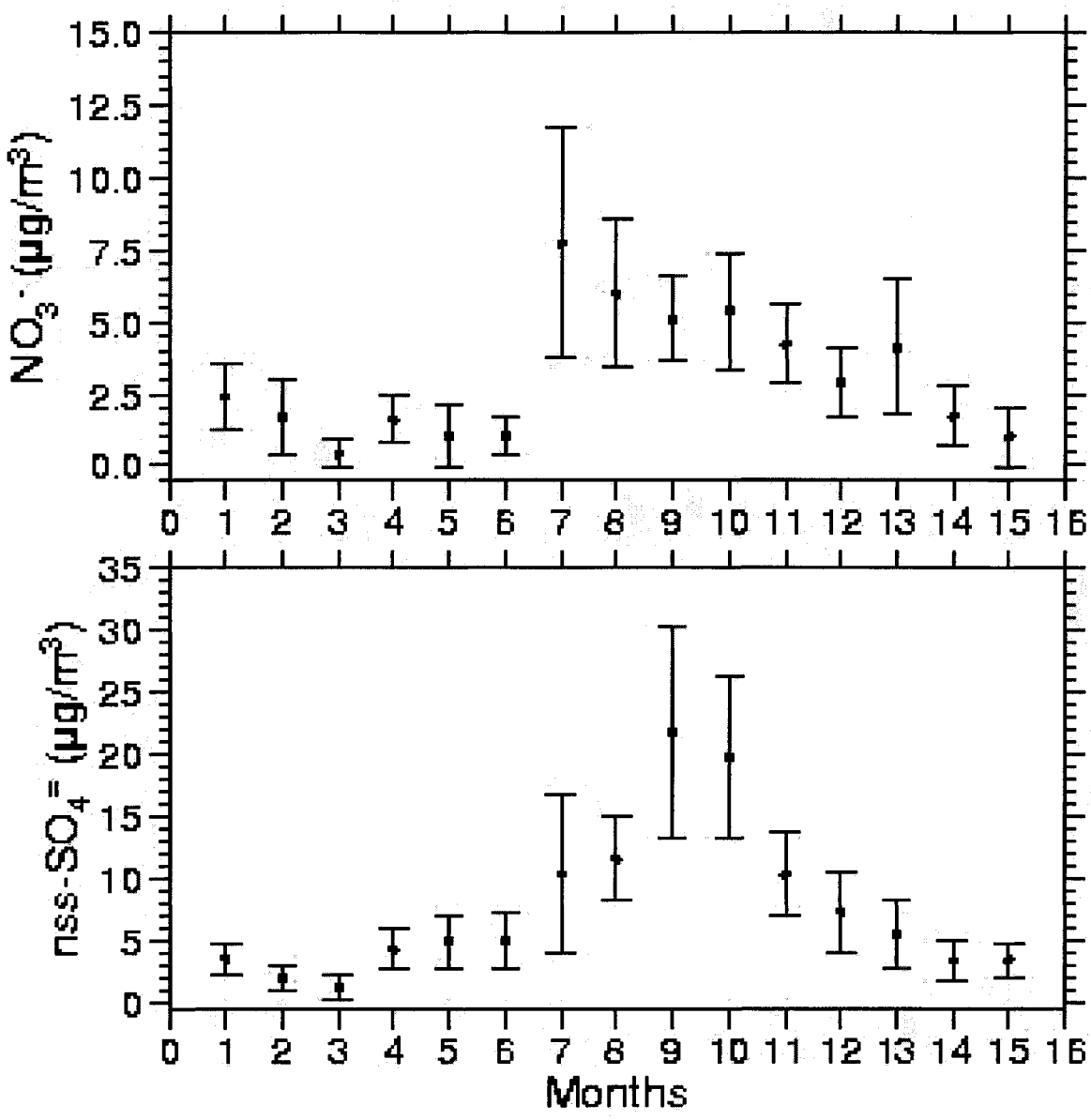
#Calculated from reported mean particulate SO_4^{2-} concentration, by accounting for about 10% of total particulate SO_4^{2-} is sea salt contribution.

Table 2. The comparison of the mean trace element concentrations (ng/m^3) and EF_{crust} values for Erdemli aerosols with those reported from several western Mediterranean sites (After *Guerzoni et al.*, 1999).

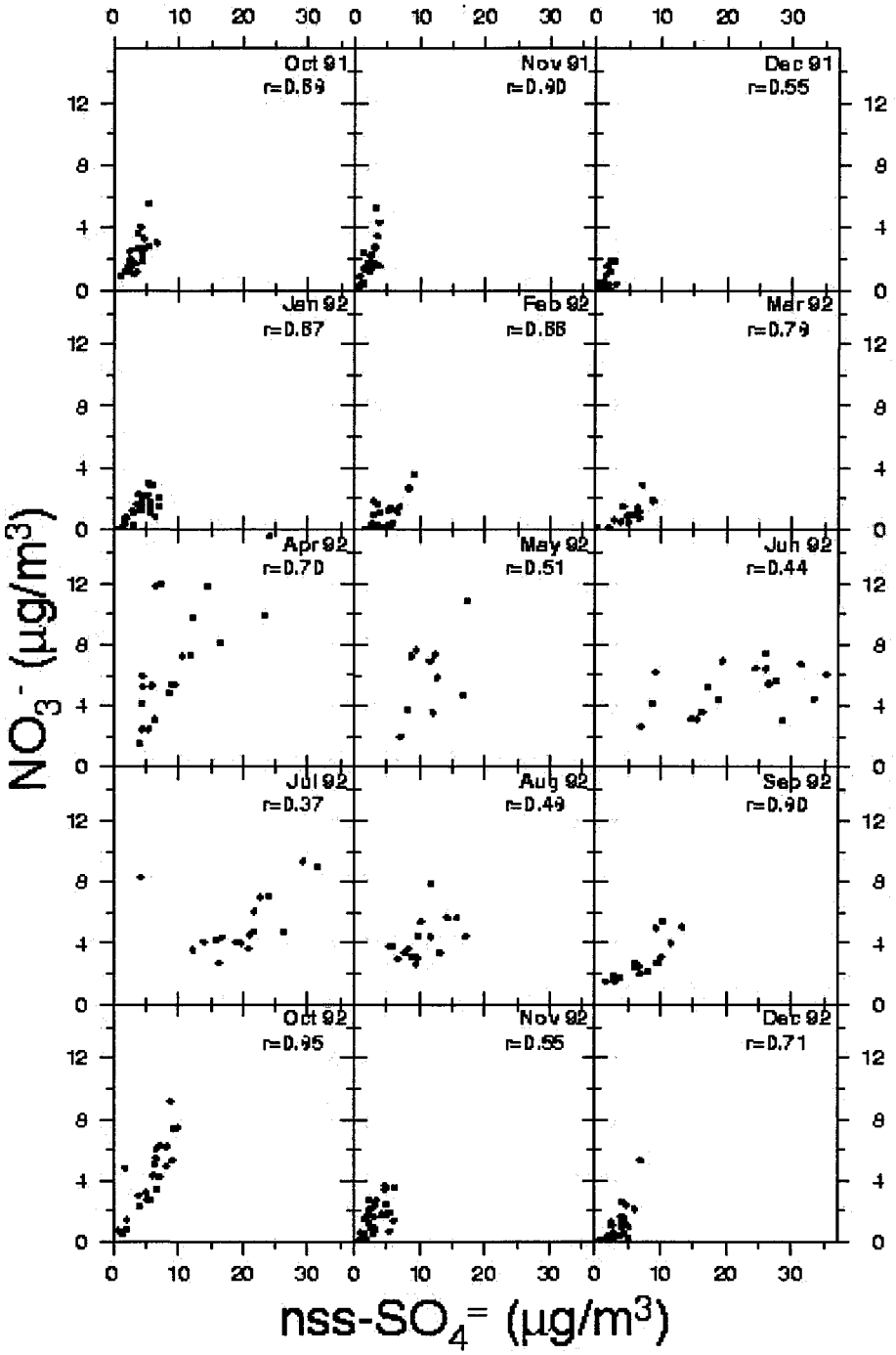
Station	Al	EF_{Al}	Fe	EF_{Fe}	Mn	EF_{Mn}	Zn	EF_{Zn}	Cd	EF_{Cd}	Pb	EF_{Pb}
Erdemli[a]	680	1	685	1.5	12.6	1.6	19	33	0.19	116	30	295
Blanes[b]	390	1	316	1.3	10	2.3	50	151	0.60	628	50	843
Corsica[c]	168	1	144	1.25	4.3	2.7	19	133	0.66	1633	16	635
Sardinia[d]	480	1	278	0.85	7.4	1.3	21	52	0.30	260	14	194
Cap Ferrat[e]	370	1	320	1.3	11	2.6	41	130	0.60	676	58	1045
Vignola[f]	109	1	-	-	1.7	1.4	12	130	0.11	423	9	550
Tour duValat[g]	380	1	275	1.1	13	2.9	60	186	0.51	-	56	982

[a]Kubilay (1996); [b]Chester et al. (1993b); [c]Bergametti et al. (1989); [d]Keyse (1995); [e]Chester et al. (1990); Migon et al. (1993); [g]Guieu (1991a); (-) no data reported.

$$EF_{crust} = (X/Al)_{aerosol} / (X/Al)_{crust}$$

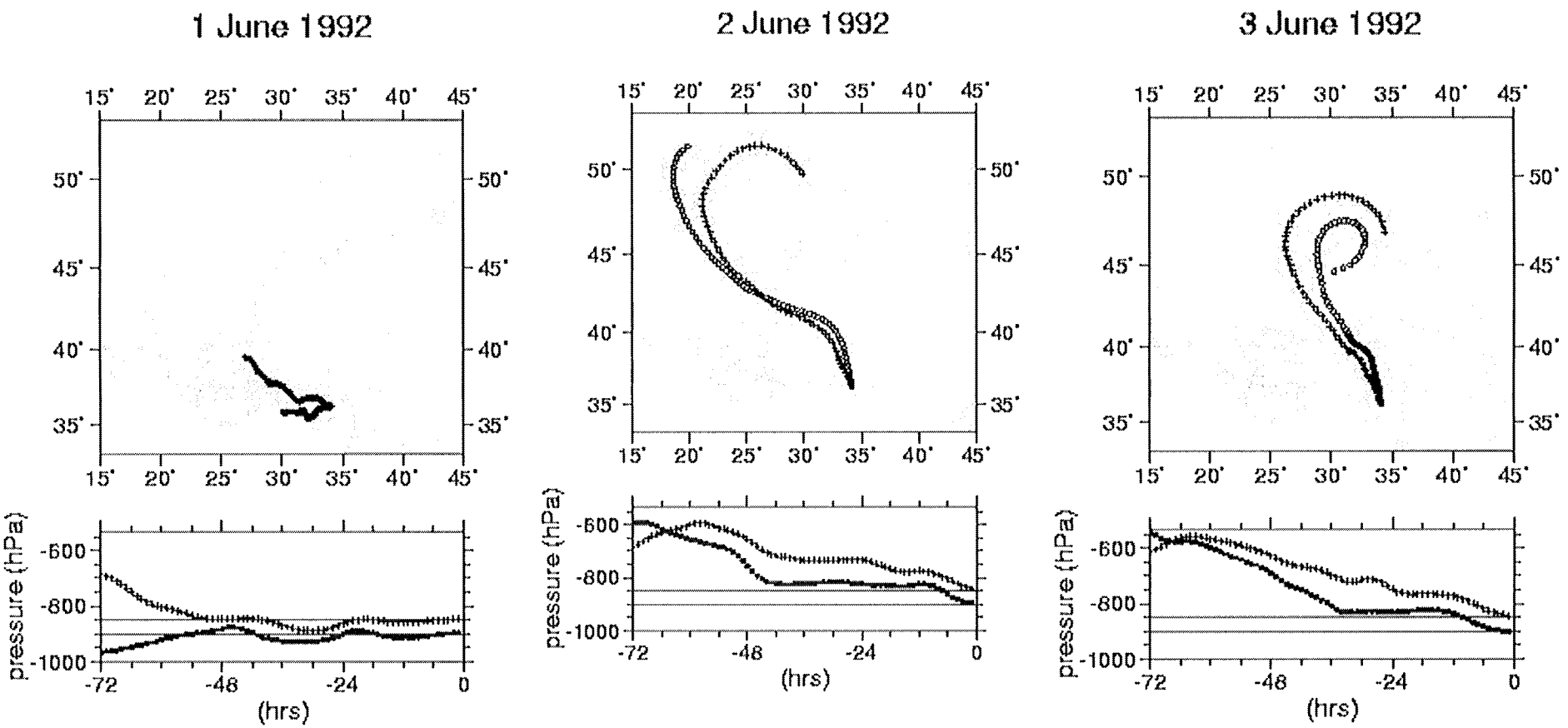


Monthly mean aerosol concentrations and standard deviations for aerosol nitrate and nss-sulfate at Erdemli.



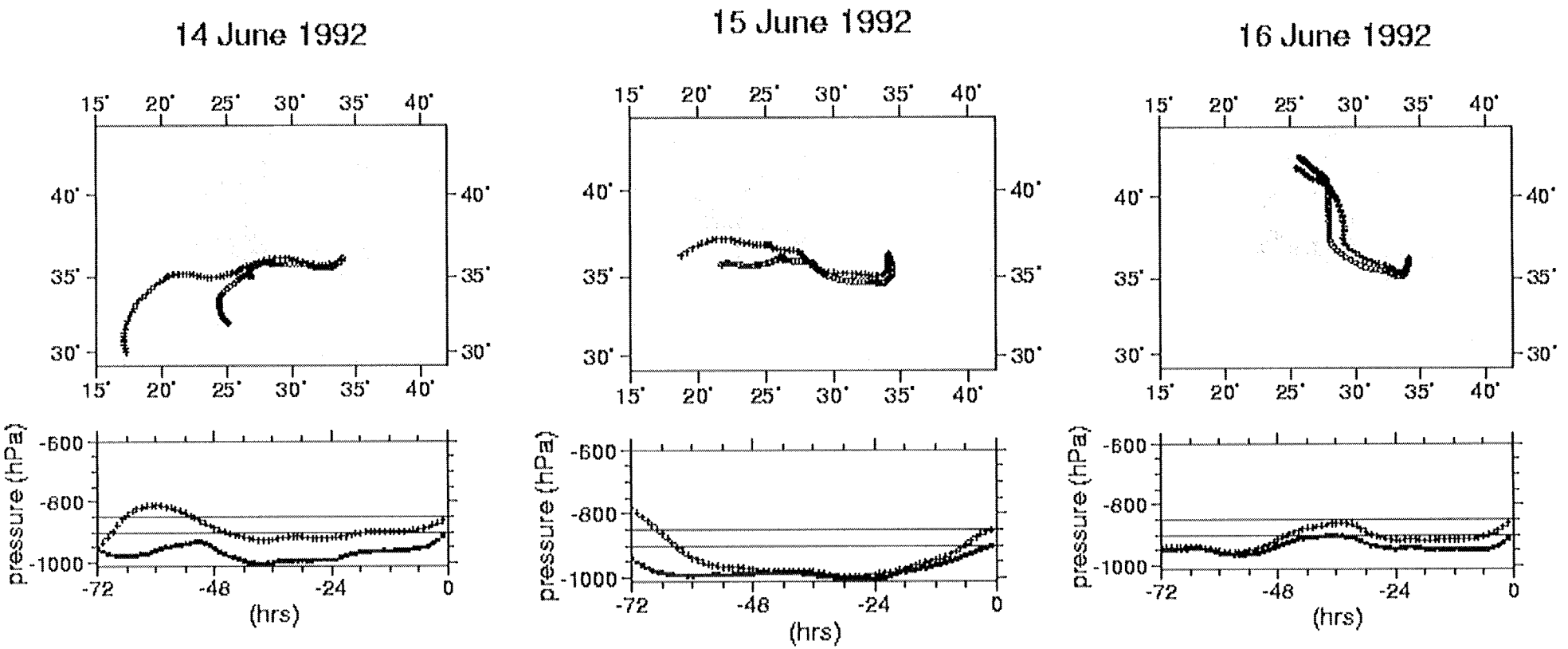
The monthly relation in between NO_3^- and nss-$SO_4^=$ concentrations in the atmosphere over Erdemli.

Aerosol nitrate and nss-sulfate concentrations are highly correlated during the autumn, winter and early spring months. This good correlation is disturbed in summer (May, June, July and August 1992) most probably due to the contributions from some extra and uncommon sources



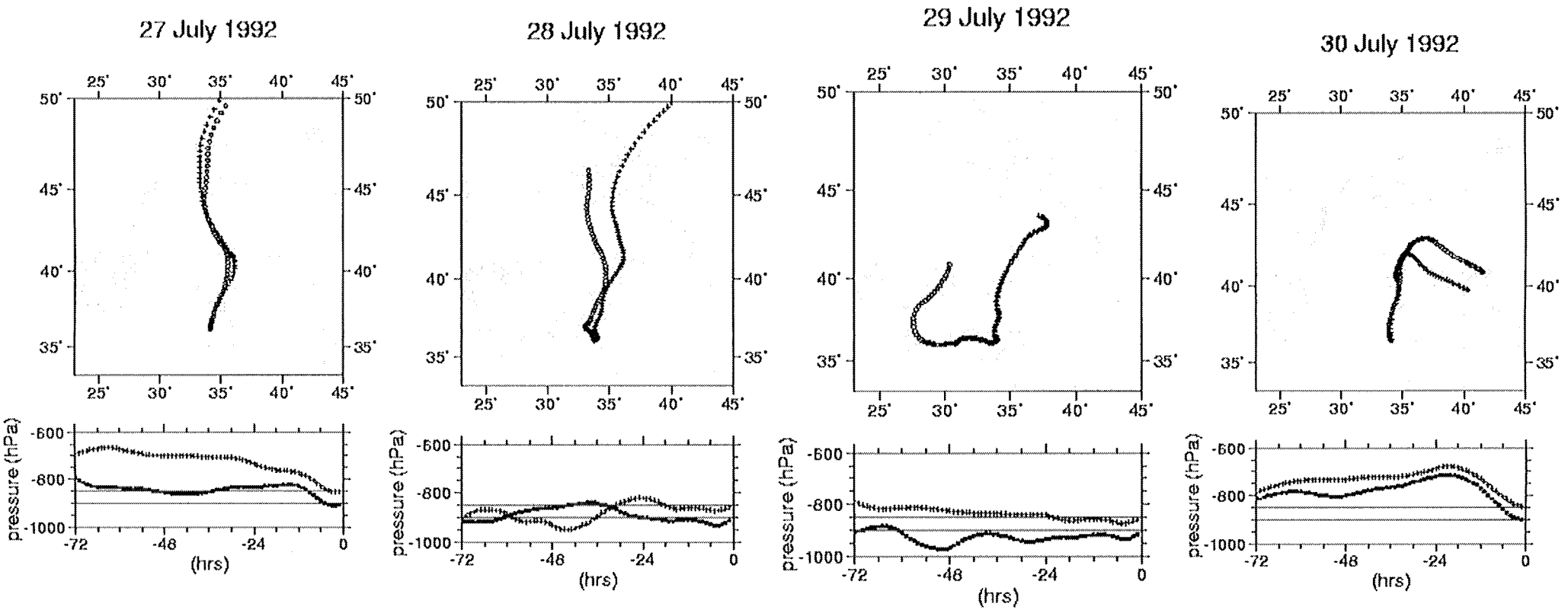
	1 June 1992	2 June 1992	3 June 1992
nsss ($\mu g/m^3$):	**28.68**	**33.40**	**35.28**
nitrate ($\mu g/m^3$):	**3.05**	**4.46**	**6.05**

Horizontal and vertical projections of air-mass back trajectories arriving at 900 and 850 hPa levels (a) 1 June 1992; (b) 2 June 1992; (c) 3 June 1992.



nsss ($\mu g/m^3$):	**14.67**	**16.30**	**8.67**
nitrate ($\mu g/m^3$):	**3.15**	**3.59**	**4.15**

Horizontal and vertical projections of air-mass back trajectories arriving at 900 and 850 hPa levels (a) 14 June 1992; (b) 15 June 1992; (c) 16 June 1992.



nsss	**29.29**	**31.44**	**4.24**	**21.79**
nitrate	**9.34**	**8.99**	**8.29**	**6.06**

Horizontal and vertical projections of air-mass back trajectories arriving at 900 and 850 hPa levels (a) 27 July 1992; (b) 28 July 1992; (c) 29 July 1992; (d) 30 July 1992.

Comparison of the aerosol nss-sulfate and nitrate concentrations ($\mu g/m^3$), Aluminum, Zinc, Cadmium, Lead concentrations (ng/m^3) and crustal enrichment factors of anthropogenic elements (EF_{Zn}, EF_{Cd} and EF_{Pb}) of the consecutively collected samples with the summer geometric mean values of the corresponding parameters in Erdemli.

Date	nss-SO_4^{2-}	NO_3^-	Al	Zn	Cd	Pb	EF_{Zn}	EF_{Cd}	EF_{Pb}
Air mass trajectories crossing over the eastern Mediterranean									
1 June	28.68	3.05	1957	15.0	0.58	21.0	9.01	122.0	70.6
2 June	33.40	4.46	1628	7.5	0.52	19.0	5.42	131.4	76.8
3 June	35.28	6.05	1751	6.0	0.52	43.0	4.03	122.2	161.7
14 June	14.67	3.15	2297	13.7	0.47	17.0	7.01	84.2	48.7
15 June	16.30	3.59	1287	10.1	0.24	15.0	9.23	76.7	76.7
16 June	8.67	4.15	1069	8.6	0.19	102.0	9.46	73.1	628.2
S.G.M.	**12.57**	**4.06**	**1213**	**31.8**	**0.42**	**35.7**	**30.8**	**142.4**	**193.4**
Air mass trajectories crossing over the Black Sea and Anatolian mainland									
27 July	29.29	9.34	1005	83.2	0.57	16.0	97.3	233.4	104.8
28 July	31.44	8.99	1082	86.1	0.65	55.0	93.6	247.2	334.7
29 July	4.24	8.29	1170	205.6	1.54	24.0	206.6	541.6	135.0
30 July	21.79	6.06	1630	90.1	0.69	25.0	65.0	174.2	101.0

S.G.M: Summer Geometric Mean

CONCLUSIONS

The mean nss-sulfate concentration of Erdemli aerosol was found to be one of the highest among the reported values for rural areas worldwide.

Both aerosol nitrate and nss-sulfate concentrations exhibit a clear seasonal variation and show a good correlation with local wet deposition.

Significant correlation ($r=0.75$) between aerosol nitrate and nss-sulfate concentrations during autumn, winter and early spring months is most likely disturbed by the contribution of nss-sulfate from biogenically produced sulfur sources especially during summer months ($r=0.34$).

Nitrate concentrations along with the crustal enrichment factors of Zn, Cd and Pb have revealed that the anthropogenic influence on the aerosols associated by the trajectories crossing the eastern Mediterranean and local coastal waters was much less pronounced than on samples associated with trajectories crossing over the Black Sea.

However, only one, unique case study (27, 28, 29, 30 July 1992) has demonstrated the magnitude of this anthropogenic contribution for the Black Sea originated aerosol samples to be insignificant compared to the contribution from the enhanced coccolithophore bloom detected within the same time period.

Black Sea is particularly likely to be an important biogenic DMS source and the contribution from this prolific source to the atmospheric nss-sulfate levels might be as high as 85%, depending on the scale and intensity of the bloom.

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