

# Size distribution and chemical composition of marine aerosols: a compilation and review

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## ABSTRACT

Some 30 years of physical and chemical marine aerosol data are reviewed to derive global-size distribution parameters and inorganic particle composition on a coarse  $15^\circ \times 15^\circ$  grid. There are large gaps in geographical and seasonal coverage and chemical and physical aerosol characterisation. About 28% of the grid cells contain physical data while there are compositional data in some 60% of the cells. The size distribution data were parametrized in terms of 2 submicrometer log-normal distributions. The sparseness of the data did not allow zonal differentiation of the distributions. By segregating the chemical data according to the major aerosol sources, sea salt, dimethylsulfide, crustal material, combustion processes and other anthropogenic sources, much information on mass concentrations and contribution of natural and anthropogenic sources to the marine aerosol can be gleaned from the data base. There are significant meridional differences in the contributions of the different sources to the marine aerosol. Very clearly, we see though that the global marine surface atmosphere is polluted by anthropogenic sulfur. Only in the case of sulfur components did the coverage allow the presentation of very coarse seasonal distributions which reflect the spring blooms in the appropriate parts of the oceans. As an example of the potential value in comparing the marine aerosol data base to chemical transport models, global seasonal meridional MSA distributions were compared to modelled MSA distributions. The general good agreement in mass concentrations is encouraging while some latitudinal discrepancies warrant further investigations covering other aerosol components such as black carbon and metals.

## 1. Introduction

Over the past decades, there have been numerous reviews of our knowledge of the marine aerosol. Each of these reviews had a specific focal point which was in line with the authors research interest or with specific issues at hand. Many attempts focused on global distributions of number concentration (Jaenicke, 1987) and related properties (Cobb and Wells, 1970; Hegg and Hobbs, 1992; Podzimek, 1980; Shiratory, 1934). A

general picture emerged of low number concentrations in the central parts of the oceans and increasing values towards the downwind coast of the continents. However, before the advent of commercial digital condensation particle counters in the 80s, such reviews were hampered by the incompatibility of sensors with different measuring principles and calibration standards. The size distribution of marine aerosols has been the subject of several reviews (Fitzgerald, 1991; O'Dowd et al., 1997). Inlet losses, particle size and concentration calibrations and compatibility of measuring principles are the prime problems of size distribution measurements in the atmospheric aerosol. Even

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today, they have not been overcome completely in either marine or continental aerosol experiments.

A first comprehensive physico-chemical review based on state of the art experimental techniques was given by Junge (1972). The best data coverage at that time (and still today) was reached in the North and South Atlantic. A first review of elemental composition with some marine coverage can be found in Rahn (1976). At that time, however, the sensitivity of many analytical methods precluded their use in the low concentration marine aerosol. Consequently, Rahn (1976) could not include many marine locations in his data base. In a series of research flights, wider marine coverage was reached with physical and chemical aerosol measurements (Hogan and Mohnen, 1979; Huebert and Lazrus, 1980; Patterson et al., 1980). However, with aircraft data, there are large and unresolved inlet and calibration issues. These issues, and the fact that those global flights concentrated on the free troposphere excluded them from the present surface aerosol review. Comprehensive reviews of the atmospheric aerosol system (Prospero et al., 1983) and of the organic aerosol (Duce et al., 1983) do not show any global maps of aerosol composition. The AEROCE\* (Arimoto et al., 1992) network of island stations supplied much information on the bulk composition of the marine aerosol. Consequently, many of the results are included in the present review which focuses on the surface marine aerosol and combines available physical and chemical information to produce global distributions of aerosol parameters which can be used in comparisons with chemical transport models and in assessments of global aerosol effects.

## 2. The data base

### 2.1 Physical data

Particle concentration and particle number-size distribution data were taken from reviewed literature or in some cases from similar but unpublished data from these same investigators to extend the global and seasonal coverage. Because of the very

limited extent of the physical data base, all sources of data were collected in Table 1.

Because particle size distributions are measured as the number concentrations within a series of finite diameter increments, the results are multivariate data sets in time and space with 10 to 100 concentrations at each location which represents on the order of 1-h average. Measurement and analysis techniques were not standardised, thus, the fundamental data are not comparable. To overcome this difficulty, the data were generally fit by a multimodal, log-normal function either by the original investigators or secondary analysis. In the marine boundary layer, the size distribution is generally confined to 3 distinct modes: an Aitken mode with a geometric number mean diameter around 50 nm, an accumulation mode with a mean diameter around 150 nm and a coarse or sea salt mode with a mean diameter above 400 nm. At times, there were other smaller modes below

Table 1. Sources of data on aerosol concentration and number-size distribution

Source	Geographical area/ experiment
Bates et al., 1998b	Tasman Sea, Southern Ocean, ACE 1
Covert et al., 1996b	Arctic, IAOE91
Covert et al., 1996a	Central Pacific, MAGE
Covert et al. (unpublished data)	Equatorial Western Pacific, CSP
Davison et al., 1996a	Southern Ocean
Heintzenberg and Leck, 1994	Arctic
Jaenicke et al., 1992	Southern Ocean
Jensen et al., 1996	North E Atlantic, ASTEX
Leaitch et al., 1996	NW Atlantic
Quinn et al., 1990	Central N Pacific, MAGE
Quinn et al., 1993	Central Eastern Pacific, MAGE
Quinn et al., 1995	Central Pacific, MAGE
Quinn et al., 1996	Central Pacific, MAGE
Raes et al., 1997	Tenerife
Van Dingenen et al., 1995	North Atlantic
Van Dingenen et al. (unpublished data)	Tenerife, ACE 2
Wiedensohler et al. (unpublished data)	Tasman Sea, Southern Ocean, ACE 1
Nowak et al. (unpublished data)	North and South Atlantic, Indic, AEROCRUISE 1999

\* AEROCE: atmosphere/ocean chemistry experiment.

30 nm (ultrafine and small Aitken) or larger sea salt modes above 1  $\mu\text{m}$ . For the purposes of this review, 4 modes were considered, the 3 dominant ones and the ultrafine. This modal nature and its consistency facilitated analysis and simplification of the results. For each mode, a number concentration, geometric number mean diameter and standard deviation of the mode were calculated, giving up to 12 defining parameters. The total number concentration was also tabulated. Where data was available as hourly or less than hourly averages, 24-h averages were calculated and assigned to the cell statistics. Otherwise, some cells and experiments would be weighted more heavily with data that is not independent.

(1) *Completeness*. Only data covering at least 1 decade of size and at least 1 complete size mode were accepted.

(2) *Methodology*. Only data that were taken from instrumentation calibrated with respect to geometric size were included. Thus, much optical particle counter data was not included. Only data taken at a measured relative humidity of less than 40% RH were accepted as a matter of minimising the effect of hydration on the size distributions. This then reduced the data set largely to distributions determined via mobility or aerodynamic sizing techniques. (Differential Mobility Particle Sizers, DMPS (Hoppel, 1978) and Aerodynamic Particle Sizers, APS (Agarwal et al., 1982).)

(3) *Representativeness*. Only data covering at least two days within a given cell were accepted to minimise local, short-term meteorological and source influences.

(4) *Redundancy*. Only data with some dual measure of particle concentration were accepted, e.g., size independent number concentration or 2-particle counters which overlapped for part of their effective size range.

## 2.2. Chemical data

Compared to the physical data base, many more chemical data have been published since the sixties. Ideally, only ship data from dedicated aerosol experiments with rigorous sample control should go into a marine aerosol data base. However, in the data base, research cruises fulfilling these requirements covered only a small fraction of the world oceans. Four Arctic reports were included

(Lannefors et al., 1983; Leck et al., 1996; Leck and Persson, 1996; Maenhaut et al., 1996). Most research cruise data cover the North Atlantic region (Berresheim et al., 1991; Church et al., 1991; Church et al., 1990; Gravenhorst and Jendricke, 1974; Hansen et al., 1990; Kim et al., 1995; O'Dowd and Smith, 1993; Pszenny et al., 1990; Van Dingenen et al., 1995; Véron et al., 1992; Véron and Church, 1997; Whelpdale et al., 1990). For the summer of 1997, the ACE-2 experiment provided a large aerosol data base in the region Portugal, Canaries, North Africa (Andreae et al., 2000; Bates et al., 2000; Neusüß et al., 2000; Novakov et al., 2000; Putaud et al., 2000).

The data base contains 8 experiments on Tropical and South Atlantic cruises (Andreae et al., 1995; Berresheim et al., 1989; Bravo et al., 1982; Crozat et al., 1973; Pszenny et al., 1993; Pszenny, 1992; Putaud et al., 1993; Rahn, 1976). From the Western Pacific Ocean the data base contains the ship-borne experiments reported in (Andreae et al., 1983; Clarke, 1989; Clarke et al., 1987; Hirose et al., 1983; Huebert et al., 1993; Quinn et al., 1998; Tsunogai et al., 1988). The Eastern Pacific Ocean is covered by ten reports (Bates et al., 1992; Bravo et al., 1982; Hoffman et al., 1969; Maenhaut et al., 1983; Parungo et al., 1987; Parungo et al., 1986; Patterson and Duce, 1991; Quinn et al., 1998; Quinn et al., 1993; Saltzman et al., 1986a). In the Indic data from several Indian and US research cruises, were included (Clarke, 1989; Krishnamurti et al., 1998; Rhoads et al., 1997; Sadasivan, 1983; Savoie et al., 1987; Siefert et al., 1999). These results are complemented by unpublished data from the Indian Ocean Experiment (INDOEX) and other recent research cruises in the Indian Ocean area (A. Johansen, D. L. Savoie and J. M. Prospero, personal communication).

As a first complement, other cruise data from ships of opportunity were added. Most of these ships covered routes between Europe and Antarctica (Andreae, 1983; Bürgermeister and Georgii, 1991; Davison et al., 1996a; Davison et al., 1996b; Losno et al., 1992; Losno et al., 1989; Osadchii et al., 1980; Völkening and Heumann, 1990) and between Japan and Antarctica (Koga et al., 1991). As a further complement we added aerosol data from airborne marine boundary layer flights (Berresheim et al., 1990; Hansen et al., 1990; Harrison et al., 1996a).

Island data were added to increase geographical coverage in areas not covered by research cruises or by ships of opportunity. These data greatly enhanced the seasonal coverage, because they often included data from several years. The island data were complemented by coastal data taken at research stations with air mass analyses excluding local or land contamination. In the Arctic, these data cover the Svalbard region (Heintzenberg and Covert, 1987; Heintzenberg et al., 1981; Heintzenberg and Leck, 1994; Li et al., 1993; Maenhaut et al., 1994), Northern Greenland (Heidam et al., 1999; Heidam, 1983) and Point Barrow, Alaska (Li and Winchester, 1989), (Li et al., 1993; Li and Winchester, 1990). In the Atlantic region, data from Iceland (Prospero et al., 1995), the Faroes (Kemp, 1984; Prahm et al., 1976), the Shetland Islands (Keane and Fisher, 1968; Peirson et al., 1973), Frøya and Lista, Norway (Isakson et al., 1997), Scotland (Davison and Hewitt, 1992), Bermuda (Chen and Duce, 1983; Ellis Jr. et al., 1993; Hastie et al., 1988; Meinert and Winchester, 1977; Wolff et al., 1986), Mace Head, Ireland (Cooke et al., 1997; Francois et al., 1995) and Prospero and Savoie (personal communication), the Azores (Harrison et al., 1996b), Madeira Islands McGovern (personal communication) Portugal (Pio et al., 1996), Barbados (Ellis Jr. et al., 1993; Savoie et al., 1989; Zhu et al., 1997), and Sal Island (Savoie and Prospero, 1977), were included.

In the Indic, a report from Amsterdam Island (Liousse et al., 1996), and from Kashidoo, Maldives (Prospero and Savoie, private communication) was used. From the Western Pacific, we added data reported for Cheju Island (Arimoto et al., 1996; Carmichael et al., 1996; Chen et al., 1997; Kim et al., 1998), the Ogasawara Islands (Matsumoto et al., 1998), the islands Onna, Chichijima and Hachijojima (Suzuki and Tsunogai, 1988), Enewetak (Duce et al., 1980; Settle and Patterson, 1982), Cape Grim, Tasmania (Ayers et al., 1986; Heintzenberg and Bigg, 1990; Huebert et al., 1998), New Zealand (Allen et al., 1997; Wylie and Mora de, 1996), and for Christmas Island (Huebert et al., 1996),

Time series reaching from Shemya, North Pacific to New Caledonia, South Pacific reported by Saltzman et al. (1986b) and Arimoto et al. (1996) were used. For the Eastern Pacific, there are several reports of Hawaiian coastal data (Darzi

and Winchester, 1982; Hoffman and Duce, 1972; Hoffman et al., 1972; Hoffman et al., 1969; Patterson and Duce, 1991) and Samoa data presented in Maenhaut et al. (1981) and Savoie et al. (1994). Marine metal data from the north-western coast of the US were taken from Vong et al. (1997). Antarctic coastal and island data from 9 reports were used (Artaxo et al., 1991; Artaxo et al., 1992; Dick, 1991; Harvey et al., 1991; Koga et al., 1999; Legrand et al., 1998; Minikin et al., 1998; Savoie et al., 1993; Wolff and Cachier, 1998).

Marine areas which are mostly surrounded by land, such as the Baltic, the Mediterranean and the Sea of Japan were excluded from the review. Furthermore, marine samples from the heavily trafficked European waters and from coastal stations on the central European mainland east of 5°W were not used. The total data base spans a period of more than 30 years, beginning in 1966, and ending in spring 1998.

Additionally, data sets from the AEROCE, ACE\*-1 (Bates et al., 1998a), ACE-2 (Raes et al., 2000) and INDOEX experiments were made available by the respective investigators. Some of these had not been published before or only had been published in summary form. Of the many chemical components which have been analysed in marine aerosol samples, a subset was selected which fulfilled the following requirements.

(1) *Completeness.* Only components with data in both hemispheres were accepted.

(2) *Specific, calibrated analytical methodology.* Results of ion chromatography, capillary electrophoresis, flame photometry, Particle-induced X-ray emission and Instrumental Neutron Activation Analysis were accepted. Results of filter ashing were not used. A compromise was made with black carbon (BC), the most important tracer of combustion sources in order to achieve better global coverage. Results from aethalometer (Hansen et al., 1982) and other photometric methods (Andreae et al., 1983; Bond et al., 1999; Heintzenberg, 1982; Heintzenberg, 1988) were accepted even though there are calibration discrepancies between these methods (Heintzenberg et al., 1997).

(3) *Representatives of all major natural and anthropogenic aerosol sources.* Tracers for sea

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\* ACE: international global atmospheric chemistry's aerosol characterization experiment.

Table 2. List of chemical components in the data base

#	1	2	3	4	5	6	7	8	9	10
Component	Na	Mg	Cl	nssSO <sub>4</sub>	MSA	BC	Fe	Si	nssCa	nssK
<i>n</i>	727	319	575	1164	847	1413	208	55	373	310
<i>m</i> (ng m <sup>-3</sup> )	3600	440	5800	1010	36	150	550	770	210	58
sea salt	X	X								
crustal							X	X	X	
anthropogenic				X		X				

#	11	12	13	14	15	16	17	18	19	20
Component	Al	ncMn	V	Cr	Ni	Cu	Zn	Cd	Pb	BaP
<i>n</i>	182	106	91	58	65	76	124	49	131	14
<i>m</i> (ng m <sup>-3</sup> )	630	1.6	1.0	1.1	2.1	3.9	7.6	0.10	1.4	0.0012
crustal	X									
anthropogenic		X	X	X	X	X	X	X	X	X

BaP = benz(a)pyrene; BC = black carbon; *m* = mass concentration; MSA = methanesulfonate; *n* = number of samples in data base; nc = non-crustal; nss = non-sea-salt; nssSO<sub>4</sub> = non-sea-salt sulfate; X indicates which component was used as tracer for sea salt, crustal and anthropogenic sources.

salt were Na, Mg, Cl, for marine biogenic (Methanesulfonate, MSA), for crustal material (Fe, Si, nss\*Ca). For anthropogenic particle sources BC, V, Benz(a)pyrene (BaP), nc\*\*Mn, Cr, Ni, Cu, Zn, Cd, and Pb were available. Non-sea-salt sulfate (nssSO<sub>4</sub>) is a mixed tracer, the concentration of which is affected by marine biogenic emissions and by the combustion of biomass and fossil fuel. Non-sea-salt potassium (nssK) is also a mixed tracer of marine and terrestrial biological material. Aerosol components with strong and variable partitioning between gas phase and condensed phase such as nitrate and organics were excluded from the review.

(4) *Redundancy*. In order to increase internal consistency and global coverage, more than one tracer for each aerosol source was incorporated. However, tracers with less than 10 samples were excluded.

Table 2 lists the 20 chemical aerosol components which were included in the data base.

The 4264 geographical positions of all 6860 chemical results in the data base are shown in Fig. 1b. Each entry in Fig. 1b stands for mass concentrations of one or several of the components listed in Table 2.

\* nss: non-sea-salt.

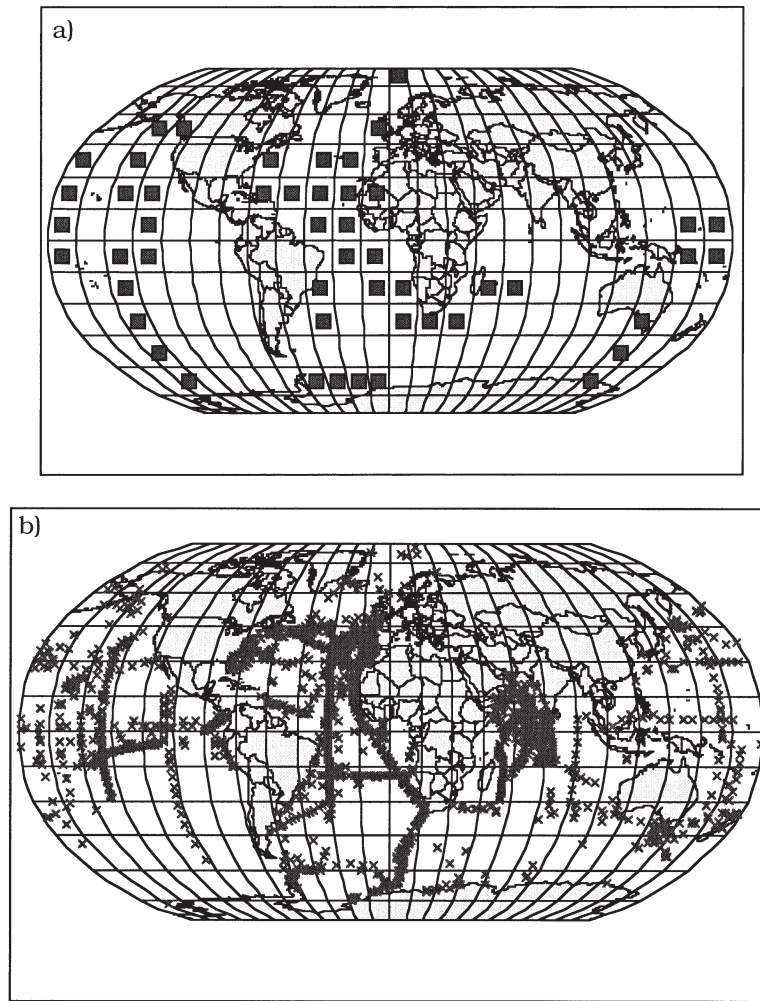
\*\* nc: non-crustal.

### 3. Data processing

Because the prime objective of the exercise was the generation of marine aerosol data for aerosol modelling purposes, the input data were to be aggregated in a global grid. Due to the sparseness of data, a 15° × 15° grid was chosen (cf. Fig. 1) for which simple aerosol statistics were calculated. The aerosol data came from many experiments of very different character. Ship cruises usually last from 3 to 6 weeks while island and coastal data cover weeks to years. Ideally, each cruise sample is characterised by start and stop position and date. Many results, however, were reported at single sample positions (even if the ship was moving). Besides grid-average concentrations, standard deviations were calculated.

According to its date, each sample was sorted into the 4 seasonal periods: January–March, April–June, July–September and October–December. However, because of the sparseness of the data, most aerosol parameters did not allow a discussion of seasonal variabilities.

All mass concentrations are given in ng/m<sup>3</sup>. Non-sea-salt fractions of sulfate and potassium were calculated with sodium as reference element from average sea water composition (Wilson, 1975). With the average crustal composition taken from Taylor (1964), non-crustal fractions of calcium were calcu-



*Fig. 1.* (a) Grid cells with physical data at marine positions marked by a square in the center of each cell. Superimposed is a  $15^\circ \times 15^\circ$  latitude, longitude scale. Coastal data which met the acceptance criteria sometimes appears inconsistently to be a continental grid point. (b) Positions of all samples in the chemical data base. Superimposed is a  $15^\circ \times 15^\circ$  latitude, longitude scale.

lated with either iron, silicon or aluminum as reference element (depending on availability in the specific sample). Total sea salt and total crustal mass concentrations were calculated from the same compositional tables with the mass concentrations of the respective tracers marked in Table 3. Averages were formed whenever more than one tracer was available for sea salt or crustal material in any individual sample. We are aware of the limitations of this simplistic global usage of sea salt and crustal elemental signatures. Marine aerosol studies have

shown that the abundancies of many of the soil elements in aerosol samples can deviate substantially from that of bulk soil (Holmes and Zoller, 1996). However, such elemental information is available at a few marine locations only. Thus, we refrain from applying individual soil signatures in our global review.

Chlorine was excluded from the calculation of total sea salt concentrations because of potentially large losses due to displacement reactions in acidic aerosols or on the aerosol filters during sampling

Table 3. Global meridional statistics of the physical aerosol data

Latitude range	Aitken mode number	Geometric mean diameter	Geometric standard deviation	<i>N</i>	Accumulation mode number	Geometric mean diameter	Geometric standard deviation	<i>N</i>
75–90	160	45	1.5	2	60	170	1.6	1
60–75				0				0
45–60	230	31	1.5	4	110	200	1.6	4
30–45	210	45	1.5	9	250	180	1.4	10
15–30	250	44	1.4	10	170	170	1.4	10
0–15	280	46	1.6	7	240	160	1.5	7
0–15	150	47	1.5	10	160	170	1.5	10
–15–30	390	40	1.4	7	220	160	1.4	7
–30–45	600	36	1.4	7	200	150	1.4	7
–45–60	300	31	1.4	3	110	140	1.5	3
–60–75	310	35	1.4	9	70	150	1.6	5
–75–90				0				0

Number concentrations in  $\text{cm}^{-3}$ . *N* = number of cases.

(Hitchcock et al., 1980; Klockow et al., 1979; Martens et al., 1973). The global average chlorine/sodium ratio in Table 2 illustrates this problem. For sea water this ratio should be 1.8 but a value of only 1.6 was found on average.

## 4. Results

### 4.1. Physical data

Aerosol concentration and number-size distribution data which fit the acceptance criteria could be found for only 28% of the  $15^\circ$  by  $15^\circ$  grid cells (cf. Fig. 1a). Major gaps occur in the Indian Ocean and the latitude band  $60^\circ$  to  $75^\circ\text{N}$ , where no data were available. The ultrafine and sea-salt size ranges were less well represented and could not be included in a meaningful global distribution. Concentration and size parameters are presented for those modes where they were available but latitudinal distributions are not presented. Otherwise, the resulting latitudinal profile of longitudinally averaged data is not biased by large, unequal geographical weighting. There are no obvious biases from continentally or locally influenced data. Figs. 2–4 show the longitudinal profiles of total number concentration and, for the Aitken and accumulation sizes, modal number concentration and geometric number-mean diameter. A one standard deviation range of variability is shown for each meridional set. Table 3 presents the size distribution parameters for all

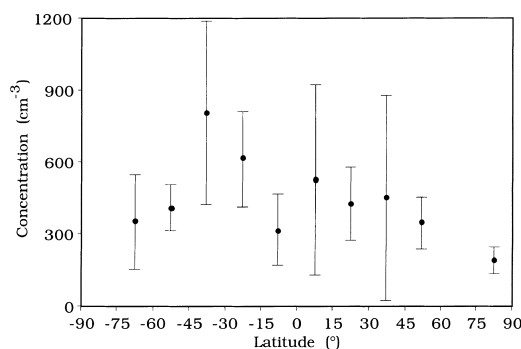


Fig. 2. Global annual average, meridional distribution of total particle number concentrations in units of particles per  $\text{cm}^3$ . A one standard deviation range of variability is shown for each meridional set.

four modes for each latitudinal cell where data was available.

The total particle number concentration varied from about 200 to  $800 \text{ cm}^{-3}$  and there was no significant interhemispheric difference (cf. Fig. 2). There were maxima in concentration in the mid-latitudes, a general minimum in the equatorial region and again toward the poles. However, this picture of total number concentration is not a good representation of the aerosol since it is the sum of the Aitken and accumulation modes which are largely independent in terms of source, transport and removal processes. The longitudinal average of the separate Aitken and accumulation modes (cf. Figs. 3, 4) illustrates the difference in the

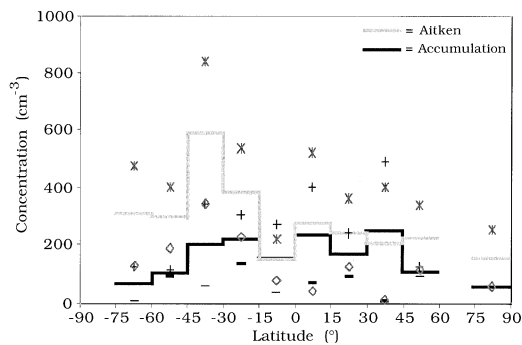


Fig. 3. Global annual average, meridional distributions of particle number concentrations for Aitken and accumulation mode in units of particles per  $\text{cm}^3$ . A one standard deviation (stdev) range of variability is shown for each meridional set. \* = Aitken + stdev;  $\diamond$  = Aitken - stdev; + = accumulation + stdev; - = accumulation - stdev.

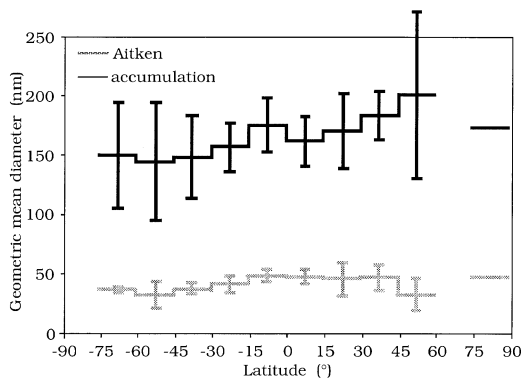


Fig. 4. Global annual average, meridional distributions of modal diameters for Aitken and accumulation mode in units of nm. A one standard deviation range of variability is shown for each meridional set.

latitudinal profiles of the parameters of these modes. In this view of the aerosol parameters, there are hemispheric differences and latitudinal trends in both concentration and mean size. Aitken mode concentration in the southern hemisphere is 300 to 600  $\text{cm}^{-3}$  compared to 150 to 300  $\text{cm}^{-3}$  observed in the northern hemisphere. By contrast, accumulation mode concentration is similar in the northern and southern hemispheres. Maxima were observed in the mid- and tropical- North Atlantic and Indic Oceans downwind of continental sources. Similar maxima can be expected in the north-western Pacific. Although there is no size

distribution data west of the date line, measurements of total concentration along 170°W showed a midlatitude maximum presumably from Asian sources.

The geometric mean diameter of both the Aitken and accumulation modes is larger in the northern hemisphere by about 25% (cf. Fig. 4). The combination of increased concentration and increased mean diameter for the accumulation mode results in a volume concentration ratio of about 2:1 between the northern and southern hemispheres. This is similar to the ratio of non-seasalt sulfate reported below. Other chemical components such as submicrometric seasalt, MSA and organic compounds that are more evenly distributed latitudinally may make the total hemispheric mass difference less than that of sulfate. The size distribution data do not reflect the large sulfate mass in the chemical data in the equatorial region. This results from the lack of size data in the equatorial Atlantic associated with biomass burning. Table 3 gives summary statistics of the physical data.

These latitudinal trends may be due to natural and anthropogenic continental sources. From the point of view of aerosol dynamics, an increased particulate volume or surface area in the northern hemisphere from either source may reduce new particle formation and thus have a controlling effect on the particle concentration and modal diameter of the Aitken mode.

While the "goodness of fit" of the lognormal parameters to the original data and the uncertainty in the original data cannot in general be quantified due to the multitude of data sources, the uncertainty in data derived from Bates et al., Covert et al., Quinn et al. and Van Dingenen et al. can be estimated. The uncertainty in number and mean diameter have been estimated as  $\pm 10\%$  and  $\pm 2.5\%$  respectively. The uncertainty in the geometric standard deviation of the modes due to instrumental and fitting errors are on the order of  $\pm 0.1$ . Except for the standard deviation the errors in the lognormal parameters are small compared to the uncertainty of the underlying data. The differences between the fitted results and the original data may be much larger for higher moments of the size distribution.

#### 4.2. Chemical data

Of the available marine 15° by 15° data cells, adequate chemical data could be found for 65%



of them. Major gaps occurred in all oceans south of 15°S and in the eastern Pacific region where few data were found. Many questions can be posed of the data base of processed chemical information about the marine aerosol. Beginning with the distribution of natural components in the aerosol we face the first problems with sea salt. In many studies, sea-salt components were analysed and used in order to derive non-sea-salt sulfate but then not explicitly reported. The relaxation of the requirement of defined coarse particle inlet characteristics for the sake of increased data coverage leads to great uncertainties concerning the sampled mass fraction of sea salt. Also several data sets appear to be strongly affected by site-specific characteristics. On ships, very high sea-salt concentrations can be measured during stormy weather or whenever bow-generated sea spray affects the samples. Similar problems occur at coastal sites within a few hundred meters of the shoreline, which can be strongly affected by the local surf conditions. With all these caveats, we present in Fig. 5 global average meridional distributions of total sea salt concentrations. Within a factor of two the data agree with the results of a recent sea salt model study (Gong et al., 1997b). The sea-salt maximum in the southern hemispheric "roaring forties and sixties" is not seen as strongly as expected from the models. It is suspected that this is partly due to the lack of data and also because aerosol samples in this latitudinal belt often have been taken during fair weather conditions only.

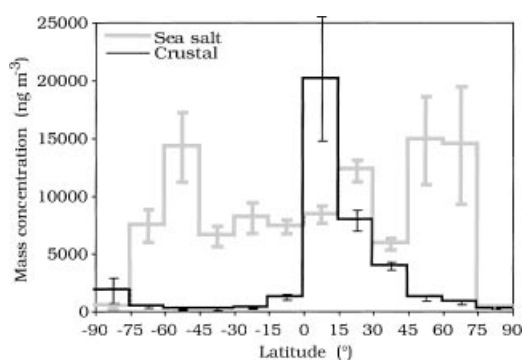


Fig. 5. Global annual average meridional distributions of total sea salt and crustal aerosol mass concentrations in  $\text{ng m}^{-3}$ . The bars indicate the variability of the data by  $\pm$  one standard deviation.

A global meridional distribution of crustal concentrations in the marine aerosol is also shown in Fig. 5. As a result of the major African subtropical source region and related wind regime, the aerosol in the latitudinal belt between 0° and 30°N is strongly affected. Only relatively few samples with crustal components were available in the Indic (Sadasivan, 1983; Siefert et al., 1999) and in the Western Pacific (Arimoto et al., 1996; Chen et al., 1997) where the spring time Kosa dust phenomenon in the region 30°N to 45°N is a regular source of continental aerosol material (Hirose et al., 1983). Significant crustal aerosol concentrations were found in the marine aerosol in the Antarctic as well as in the tropical and subtropical regions. The overall latitudinal distribution, which strongly differs from that for sea salt, reflects the distribution of the major continental dust sources in the northern hemisphere.

MSA is another natural component of the marine aerosol which has been analysed in many marine studies of the global sulfur cycle. Its precursor dimethylsulfide (DMS) is produced by marine micro-organisms and thus has a strong seasonal cycle as well as latitudinal trends. After sulfate and Black carbon (BC) MSA has the highest number of data points (cf. Table 2). Consequently, we display in Fig. 6 seasonal averages of global meridional MSA distributions which reflect the spring blooms in the appropriate parts of the oceans. The northern hemispheric spring blooms appear to yield highest MSA concentrations. Storms and surface cooling mix the surface water in this region, replenishing the nutrient supply from deeper waters.

A first comparison of the experimental MSA data was made with model results for MSA derived with a global chemical transport model (Mian Chin, personal communication). For 1990 emissions and meteorological fields, this model calculates particulate sulfur components in a  $2^\circ \times 2^\circ$  grid (reduced to  $1^\circ \times 1^\circ$  at the last  $1^\circ$  at each pole). Average zonal and seasonal means of gridded MSA model results are plotted in Fig. 6. For seasons 2 and 3 (April through September) concentration levels and meridional distributions of model and experimental results agree very well. For the months October through March, the southern hemisphere maxim is shifted north by about 30° compared to the experimental findings

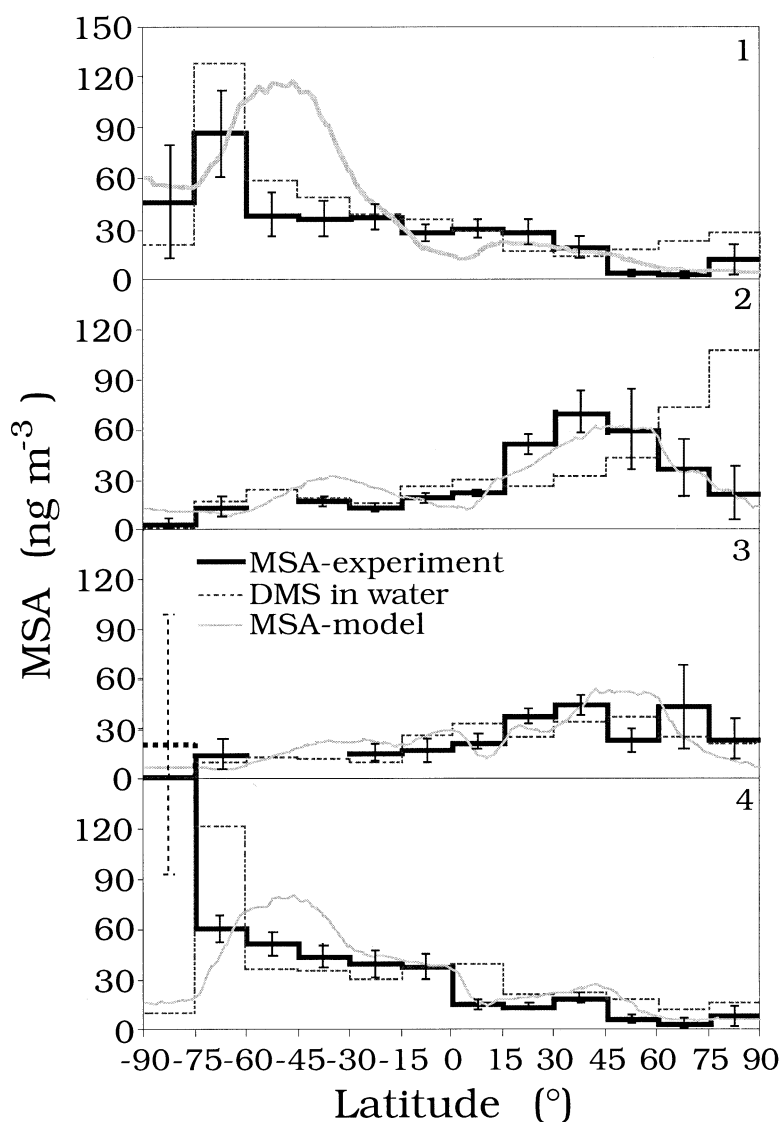


Fig. 6. Seasonal averages of global meridional data of methanesulfonate (MSA) and from a global chemical transport model for 1990 (Chin et al., 2000) in  $\text{ng m}^{-3}$ , DMS in surface water in rel. units (from Kettle et al., 1999), see text for details. 1 = January–March, 2 = April–June, 3 = July–September, 4 = October–December. The dashed MSA data in the southernmost cell of season 3 mark offscale MSA values in season 4. The bars indicate the variability of the data by  $\pm$  one standard deviation.

but the concentration levels are still in good agreement.

A second comparison can be made with the global fields of the MSA precursor DMS for which a recently compiled data gives monthly values in surface sea water on a  $1^{\circ} \times 1^{\circ}$  grid (Kettle et al.,

1999). Zonal and seasonal averages of these DMS values were calculated for the  $15^{\circ}$  resolution grid resolution of our aerosol data base. There is no simple relationship between the amount of DMS in sea water and corresponding concentrations in surface air. For the comparison with aerosol con-

centrations, we calculated a least square fit of the annual mean meridional distributions of DMS and MSA with which we scaled the DMS distributions of the four seasons, the result of which we added to Fig. 6.

With the exception of the polar regions, latitudinal distributions of DMS and MSA track well. In the Antarctic region, both MSA and DMS data are very patchy and highly variable (cf. also Fig. 16 in Kettle et al., 1999), in particular during the productive austral summer half year (seasons 1 and 4). During the productive season of the northern hemisphere, (seasons 1 and 2), there is an increasing disagreement between measured and modelled MSA data on one hand and the DMS field with latitude north of 45°.

Several tracers in the data base can be used to illustrate the influence of anthropogenic emissions on marine aerosol composition. Non-sea-salt sulfate ( $\text{nssSO}_4$ ) is a mixed tracer with a large anthropogenic fossil and biomass fuel component and a biological oceanic component. The interpretation of its information content is further complicated by the findings of Li-Jones and Prospero (1999) indicating chemical reactions of  $\text{SO}_2$  on dust particles yielding sulfate. Non-sea-salt sulfate has regionally and seasonally varying marine biogenic DMS-derived components. For want of biogenic sulfate data  $\text{nssSO}_4$  is plotted in Fig. 7 together with biogenic sulfate for the modelled year 1990 (Mian Chin, personal communication). It is obvious that the global marine boundary layer is polluted by anthropogenic sulfate. We recall here that the data set includes coastal experiments with the restriction to samples with on-shore air flow. The global average contribution of the modelled biogenic sulfate is 31%.

The consistency of physical and chemical meridional distributions was checked after calculating total mass concentrations from the sum of the third moments of the two submicrometer modes using the modal parameters in Table 3. They are displayed as dashed line ( $M_{\text{sub}\mu}$ ) in Fig. 7 after multiplying the volume concentrations with an assumed particle density of  $1 \text{ g cm}^{-3}$ . In the northern hemisphere a good agreement between  $M_{\text{sub}\mu}$  and  $\text{nssSO}_4$  is found while in the southern hemisphere the physical data indicate more mass than explained by  $\text{nssSO}_4$  between 75°S and 15°S. A total of 82% of the meridional sum of  $M_{\text{sub}\mu}$  is explained by the corresponding sum of  $\text{nssSO}_4$ .

We note that particle densities between  $1.5 \text{ g cm}^{-3}$  and  $2 \text{ g cm}^{-3}$  are more frequently applied to the atmospheric aerosol. Assuming these densities would indicate that on meridional average  $\text{nssSO}_4$  only explains between 61% and 46% of the submicrometer mass, respectively, leaving room for other aerosol components such as sea salt and carbonaceous material.

BC is a good tracer of both biomass and fossil fuel combustion (Cachier et al., 1985). Vanadium is mostly due to oil combustion (Rahn and Lowenthal, 1984). Several metals in the data base can also be used as more general tracers of anthropogenic emissions (components 13–19 in Table 2). However, their global average concentrations vary over several orders of magnitude (cf. Table 2). Therefore we normalised their global meridional distributions by their respective meridional maximum concentrations and plotted their average relative meridional distribution in Fig. 7. Chemically related to the polyaromatic character of BC is BaP which also is emitted mainly from fossil fuel combustion. The distribution given in Fig. 7 stems from two meridional profiles derived during one cruise from Europe to Antarctica and back, January through April 1977 (Osadchii et al., 1980). No other data on this unique tracer were found in the literature.

The distribution of  $\text{nssSO}_4$  is broadened by biogenic contributions, mainly in the southern hemisphere but to a small extent also in the Arctic/subarctic region. The concentrations in the northern hemisphere maxima are within a factor of two of the summer North Atlantic concentrations given in a recent global aerosol climatology (Koepke et al., 1997) which does not show any marine details in the southern hemisphere. The relative metal distribution shows 2 maxima in the northern hemisphere. The southern maximum may have some contribution from biomass burning while the northern one is due to fossil fuel combustion in the midlatitude industrial regions. We note though the large variability of the results in the respective meridional columns.

Some information on source distributions can be gleaned from the hemispheric distribution of the tracers displayed in Fig. 7. The broad distribution of  $\text{nssSO}_4$  is reflected by a ratio of southern-to-northern hemispheric concentration integrals of 0.42 (cf. the corresponding ratio of

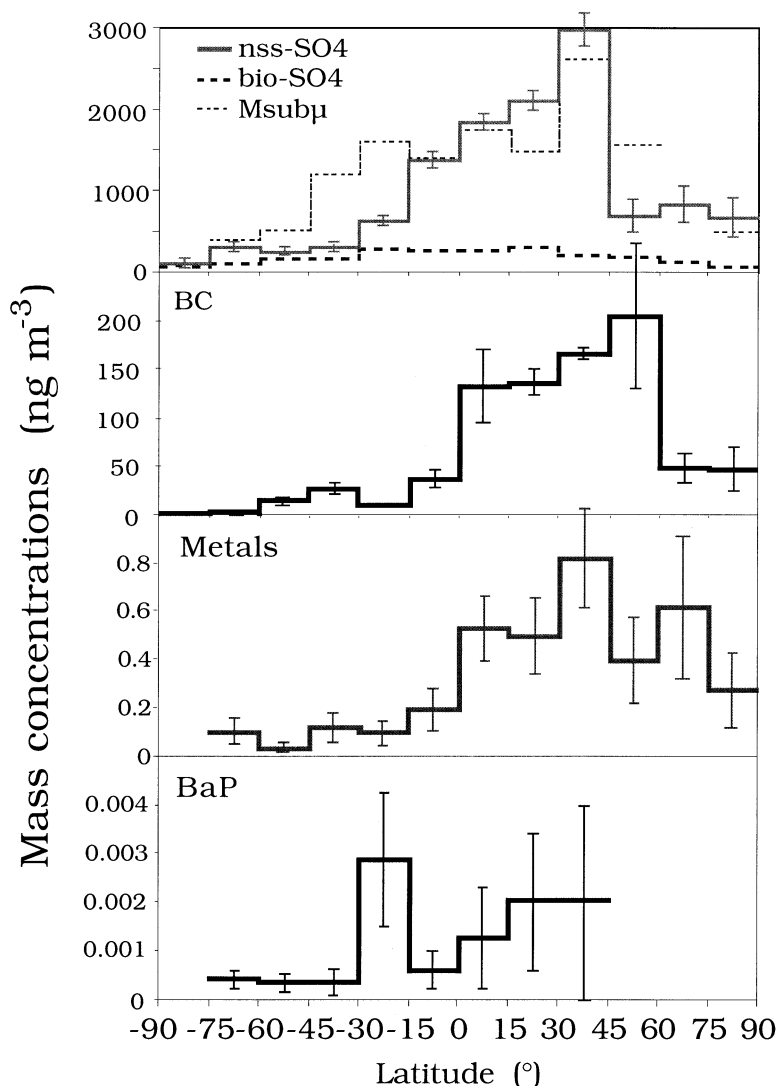


Fig. 7. Global annual average meridional distributions of non-sea-salt sulfate ( $\text{nssSO}_4$ ), modelled biogenic sulfate from a global chemical transport model for 1990 (Mian Chin, personal communication), black carbon (BC) anthropogenic metals (average relative meridional distribution of components 13–19 in Table 2) and Benz(a)pyrene in the marine aerosol in  $\text{ng m}^{-3}$ . The bars indicate the variability of the data by  $\pm$  one standard deviation. For comparison, the dashed line  $\text{Msub}_\mu$  shows the total mass concentration that was calculated from the physical parameters of the two submicrometer modes of the aerosol using an assumed particle density of  $1 \text{ g cm}^{-3}$ .

land surfaces of 0.41). The corresponding integral concentration ratio for BC is only 0.13. While both tracers come from combustion sources BC is more specific for diesel-powered surface traffic which for the past decades was

largely concentrated in northern midlatitudes. The hemispheric concentration ratio for metals (taken from their average relative distribution) is 0.26, again indicating a somewhat broader latitudinal source distribution than that for BC.

## 5. Conclusions

A review of some 30 years of marine aerosol data reveals that there are large gaps in geographical and seasonal coverage and physical and chemical characterisation. Nevertheless, on a coarse  $15^\circ \times 15^\circ$  grid much information on submicrometer size distribution, mass concentrations and contribution of natural and anthropogenic sources to the marine aerosol can be gleaned from the data. The lack of adequate sampling in most of the existing marine aerosol studies prevented a compilation of representative supermicrometer data. This shortcoming could affect the concentrations of several species marine and crustal species including  $\text{nssSO}_4$ . Other major gaps in marine aerosol data that are indicated by their absence in this review are size-differentiated chemical particle composition and aerosol data under high wind conditions, which are prevalent in the northern and southern mid latitude regions.

There are significant meridional differences in the contributions of the different sources to the marine aerosol. Very clearly we see though that the global marine surface atmosphere is polluted by anthropogenic sulfur.

Only in the case of sulfur components the coverage allowed the presentation of very coarse seasonal distributions which reflect the spring blooms in the appropriate parts of the oceans. As an example of the potential value in comparing the marine aerosol data base to chemical transport models global seasonal meridional MSA distribu-

tions were compared to modelled MSA distributions (Mian Chin, personal communication). The general good agreement in mass concentrations is encouraging while some latitudinal discrepancies warrant further investigations.

In similar ways, the gridded aerosol data should be compared to results of other global chemical transport models such as for sea salt (Erickson et al., 1986; Gong et al., 1997a), crustal material (Tegen and Fung, 1994), sulfur (Langner and Rodhe, 1991), and black carbon (Liousse et al., 1996) and global aerosol climatologies such as the one by Koepke et al. (1997). The metal data in particular can serve to validate models such as that presented by Pacyna et al. (1984) for metal emissions over Europe the results of which then only were compared to atmospheric data at a single Norwegian station or global emission inventories such as given by Nriagu (1979) which was not validated by atmospheric data.

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