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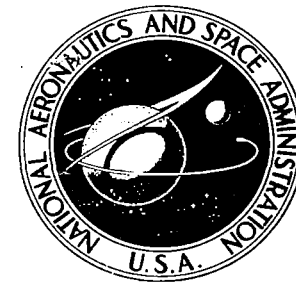

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ATMOSPHERIC AEROSOLS: A LITERATURE SUMMARY OF THEIR PHYSICAL CHARACTERISTICS AND CHEMICAL COMPOSITION

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ATMOSPHERIC AEROSOLS: A LITERATURE SUMMARY OF THEIR PHYSICAL CHARACTERISTICS AND CHEMICAL COMPOSITION

by

Franklin S. Harris, Jr.[1]

SUMMARY

This report contains a summary of 199 recent references on the characterization of atmospheric aerosols with respect to their composition, sources, size distribution, and time changes, and with particular reference to the chemical elements measured by modern techniques, especially activation analyses.

INTRODUCTION

A review of the literature has been made as a guide to the characterization of aerosols which would be useful in air quality measurements by NASA-LaRC and Old Dominion University in cooperation with Region VI, Virginia State Air Pollution Control Board. The purpose of the joint measurements is to learn what types of variables it would be useful to determine, in addition to the usual mass loading and meteorological factors. Some activation analysis (78, 171) has been made but it was desired to see what elements, size distribution, and other factors would be helpful in planning measurements and interpreting the atmospheric aerosol. The joint program is not intended to duplicate the present Region VI monitoring program, but rather to explore the usefulness of new type instrumentation techniques. The aerosols collected by many techniques described briefly elsewhere (78, 171) can be analyzed to obtain geographical, temporal, elemental, chemical, and source information with respect to size distribution. The examination of recent literature, particularly with respect to the results of elemental analysis and other variables of interest, may be of use to others working in this field and so is made available in this report.

For the varied experimental conditions and techniques employed reference must be made to the original articles. No attempt has been made to explain differences which result from different conditions and techniques. Some general agreement can be seen, offering useful suggestions for application elsewhere.

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The coverage of the literature is extensive but not exhaustive. In some references only the conclusions are given in abstract form. Papers presented at recent meetings may be available as abstracts only, preprints, or in extended journal articles. References to an abstract journal for some not readily available articles have been given in square brackets []. The "N" numbers represent a listing of report literature in the National Aeronautics and Space Administration *Scientific and Technical Aerospace Reports (STAR)*, which is obtainable from the Superintendent of Documents, U. S. Government Printing Office, Washington, DC 20402. The "APA" refers to *Air Pollution Abstracts*, published by the Environmental Protection Agency, and obtainable from the Superintendent of Documents. The "A" numbers refer to books and journal articles listed in the *International Aerospace Abstracts*, published by the American Institute of Aeronautics and Astronautics (AIAA), 750 Third Ave., New York, NY 10017. "MGA" refers to *Meteorological and Geoastrophysical Abstracts*, published by the American Meteorological Society, 45 Beacon St., Boston, MA 02108.

PHYSICAL CHARACTERISTICS

Size Distribution

The variation of the size distribution of the aerosol particles with the components and conditions is one of the best diagnostic tools to identify sources and history of the aerosol. There is a variation of the total mass loading with time and location, but more important is the varied distribution of components throughout the particle size range, either as complete particles or smaller particles collected in or on particles, or a mixture. In most cases the measurements of particle sizes are crude and only in rather large size intervals, though still significant. More convenient and better measurements of size distribution with the separation of the particles into narrower sizes is needed for better determination of size distribution effects.

There have been many types of mathematical functions used to represent size distributions. For the overall size distribution of an aged aerosol the power law suggested by Junge (96) has been valuable. Another type, in various forms, is characterized by a maximum, a spread parameter and a total number, such as the modified gamma function or a log Gaussian. The distribution may be of two (bimodal) (89, 117, 139, 143, 159), or multiple components (140, 178, 182). The size distribution can be plotted in several ways, such as the log of the size against the differential log of the number, or against a linear surface or volume. Log probability plots are useful, and sometimes cumulative mass percent against particle size is used (117, 141). Some rough classification of types of distributions have been suggested such as clean continental, average western continental, urban and oceanic background (140, 182). However, an aerosol should be viewed as a collection of size distributions of individual components with some interaction between them. The variation in chemical elements as a function of

time and size distribution has been noted (13, 26, 85, 106, 110, 145, 169, 182). An overall Junge power law with an exponent of about 3 (2) fits many situations, but it varies with altitude and location (3, 14, 159). It is known that the temperature inversion influences the distribution (2), and with a drop in relative humidity with an increase in temperature, the larger particles become less concentrated (2, see also section on relative humidity). An increase in vapor pressure increases particle size (106). A Kraft mill broadens the aerosol droplet distribution and a forest fire narrows it (37, 38). The same element may be on different size particles in residential, industrial, and near pollution sources (98). In an oceanic aerosol the distribution of elements is not the same as city produced (9). Auto exhaust particles grow and agglomerate with time, especially and first two hours (80). In some measurements no size effect has been found at that time and for those conditions for Na, Cr, Co, and Cs (118), Bi, Sn, Cu, Co, Ti and K from coal-fired plant fly ash (131), Co, Mn, and Cl (mixed) (50), and Fe and S (92).

Reported measurements show that the following are found primarily on the <u>small</u> sizes: water soluble components (124), three-quarters of material from automobiles in less than 0.5 μm (47), combustion products in chain-like agglomerates (80), and liquid droplets from photochemical origin (80). Aitken particles are mostly sulfur (130), and jet engine exhaust lacy agglomerations of mass median diameter of 0.13 μm (170). Soot and fly ash are small compared to dust (68). Automobile particles have most mass in diameter range of 0.05 to 0.2 μm, small particles are more oxygenated (139), sulfate (141, 169), chloride, nitrate and ammonium (111). Small particles are important in visibility loss (169), benzo(a)pyrene (100, 113), polycylic aromatic hydrocarbons (1). From coal-fired plant fly ash the following elements showed consistent increases in concentration with decreasing particle size: Cr, Mn, Be, with greater effects for Pb, Tl, Sb, Cd, Se, As, Ni and S (131). The following elements are reported on the small sizes: S(36), Cl (82), V (82, 110), Mn (36, 92), Ni (141), Cu (92, 141), Zn (36, 92, 141, 185), As (118, 153, 185), Se (118, 185), Br (36, 47, 50, 52, 89, 92, 118, 153), Cd (172), In (52, 185), Sn (50), Sb (50, 118, 153, 185), and Pb (47, 48, 50, 89, 92, 110, 118, 141, 165, 172, 175). The amount in smaller particles increases with time of day (48, 130, 138).

Reported measurements found the following enhanced on <u>large</u> particles of a size distribution: salt aerosol (143), sandstorm (169), wind blown soils Fresno, California with K, Fe, Ti and Mn (47), dust or spray at Tallahassee, Florida with C, Cu, Ti, Cr, Mn, Fe (89), above coastal waters with 40% of the mass less than 3.5 μm (67), NO associated with particles larger than 2 μm (13), phosphate (111), polynuclear hydrocarbons (172), K bimodal in some components (89, 143). The chemical combination of S and N vary with particle size and time of day (137). Narrow size distribution was found for Zn and Cu (110). Particle sizes increased with plume aging (15). The following elements were on large particles: Na (48, but also small in 30), Mg (48, 185), Al (30, 36, 48, 50, 68, 82, 142, 153, 185), Si (30, 36,

153), S (48), Cl (48, also small 92), K (48), Ca (36, 47, 142, 153, 166, also small 48), Sc (118), Ti (36, 47, 48, 153), V (48), Cr (48, 89, 185), Mn (47, 48, 89), Fe (13, 36, 47, 50, 89, 118, 142, 153, 172, 185, and also small 48), Ni (48), Cu (89, 185), Zn (43, 48), Sn (48), Ba (48), Ce (118), Sm (118), and Th (118).

Time Patterns

Many measurements have found variations in total or individual component amounts of aerosols (26, 110, 152), not only seasonal (104) and in air mass origin (193) and sources (26), but some diurnal affects (26, 129, 130, 155, 171, 177) which may be quite rapid (37, 38, 64, 192), which of course depend on the location and geography. Holzworth (77) discusses a start-of-day increase, mid-day increase or decrease, early evening increase and a later night decrease. There is also the effect of relative humidity (104, 129 and section on Relative Humidity).

Maxima in early morning and evening have been observed for several places (77, 105, 199), especially with Pb and Br (36, 176), or in the desert with a maximum in the evening and a minimum in the morning (30). Zn in Fresno, California has a maximum in the early morning and evening (from automobile exhaust and tire dust?) (47). In Pomona, California, Zn, Pb, Br in small particles and Fe, Ca and Ti have the same diurnal effect for all three sizes (47). At the Los Angeles Harbor Freeway Pb peaks with heavy morning traffic and Fe and Ca on larger particles have the same diurnal patterns (47).

Elements have different diurnal cycles at Livermore, California (122), (1) a Na and Cl in phase (maritime) with each other, with a late night maximum for both, (2) all other elements (continental) except Br showing a mid-afternoon maximum and (3) a Br which had morning and evening peaks suggestive of automotive sources. In Los Angeles (176) a common source in the morning hours was indicated for Pb, Zn, Hg, Sg, As, In, Ba, Ag, Se, but Na, Cl, and Al had different diurnal patterns. Smog secondary conversion may account for 50% increase in optical thickness (179). Gas-to-particle conversion is significant (158). In Denver marked variations with time were found for Al, Si, Ca, S, Pb, and Cl (152).

There is significant variation of particle size with time (13, 81, 177) with submicron size having greater variation than larger sizes. Diurnal changes are also noted in carbon containing particles (129, 130). In Los Angeles non-polar organics dominate in morning, polar in afternoon (59). There is a variation with Santa Ana winds (from the desert to the east) (139). Diffusion profiles of SO_2, Cl_2 and organics (144) below temperature inversion differ vertically. The marine Arctic air mass has least material, frontal zones more, with Ti, Zn, Sn an order of magnitude more and Na, K, Ca 1.5 - 2 times more (193).

A plume changes in size distribution as it ages (15) with particle size increasing. Tetroons and fluorescent material have been used to determine air parcel trajectories (192).

Altitude Variations

From near ground Cl, S, K, Ca and Ti decreased rapidly to half or less at 1 km altitude and then remained about constant to 10 km, indicating ground soil particles. Low Cl concentration suggests low sea salt concentration at higher altitudes, soil component quite constant with respect to altitude for Ti/Si, K/Si, and Ca/Si mass ratios, log normal distribution of these ratios (49, 102) with higher altitude maximum in size shifts to larger sizes (14). Altitude measurements in urban atmosphere (79) and unpolluted air (85, 168, 193) have been made. Over oceans aerosol marine type changes with altitude to continental aerosol (96, 148). Altitude variation is related to atmospheric temperature stratification (47). Pollution has been found in distinct layers up to 18,000 ft over California (51).

Relative Humidity

The number of particles becomes less with a drop in relative humidity (RH) and an increase in temperature (2), down to a RH of 30% (157). There has been little humidity effect on maritime aerosols (162). With RH, NH_4, CO_3, SiO_4, NO_2, NO_3 vary, also with wind direction and particle size (13). Particle size increases with RH (106). Changes as low as around 50% RH can make a difference in size distribution (61). Marine and urban aerosol may contain significant free H_2O at RH 40 to 75% from microwave absorption measurements (69, 177). Measurements have been made on hygroscopic sea salts (119). The scattering coefficient changes with RH (22, 83, 99, 107, 109). There is appreciable water and organic volatile matter in particulates in marine air (149, 186).

COMPOSITION

Analysis Methods

The analysis of aerosols to determine primarily the elemental and some chemical constituents can be made by modern instrumental techniques. In particular a variety of methods are used to excite the atoms (54, 127) whose radiation is characteristic of the elements, or by the effect of the particle on a beam of radiation or particles. For summaries of other methods see 121, 127, 128. Sometimes different names are used for the same technique or variations in the method depending on the exciting mechanism, the character of the radiation or beam after particle interaction, and the analyzing instrument.

One of the most widely used methods is neutron activation analyses (NAA). High energy or thermal neutrons are used to activate an element and the emitted gamma and x-radiation is characteristic of the element. A large number of elements can be determined simultaneously. Elements of atomic number less than sodium are not measured. The wide application is indicated by the number of research groups who have applied this method (5, 27, 33, 53, 54, 65, 70, 73, 74, 85, 93, 116, 127, 142, 153, 154, 155, 163, 176, 180, 190, 195, 196, 197).

Protons can be used as an activating particle for a large number of elements (1, 87, 88, 89, 92, 135).

High energy alpha particles can be used to excite the atoms for x-ray emission spectroscopy (54, 67) or to transmute Pb into Po and measure the alpha particles emitted (60).

Activation with gamma-ray photons producing radioactive nuclear isomers of the elements radiated and resulting in gamma radiation is useful for about 20 elements of atomic number greater than 31 (39). The high energy photons are useful for activation (7, 25, 142, 160, 180). The gamma-ray spectrograph is one of the instruments employed (82). The instrumental photon activation analysis (IPAA) is intermediate in sensitivity between neutron activation and atomic absorption (194).

The analysis of x-rays emitted by activated atoms can analyze up to about 81 elements. Various terms are used such as: x-ray fluorescence (11, 47, 54, 67, 76, 94, 125, 127, 131, 160, 161, 163, 166, 176, 187, 194), x-ray photoelectron spectroscopy (54, 131, 136), and x-ray energy spectroscopy (XES) (151, 152).

The scattering of protons (127, 133, 174) or alpha particles (95) is another approach.

Atomic absorption can analyze up to about 65 elements (31, 32, 33, 52, 73, 75, 127, 140, 141, 154, 156, 166, 172, 180, 195, 196).

Electron microscopy (10, 102, 107, 121, 123) is one of the oldest techniques for analysis. The scanning electron microscope (SEM) basically only examines the surface but information on particle size, shape, texture and topography of the surface is obtained. The electron spectroscopy for chemical analysis (ESCA) uses ultraviolet or x-ray photons to excite outer shells of atoms leading to ejection of electrons whose kinetic energy is reduced by the chemical binding energy and can therefore be used to identify elements by their chemical binding energy (121). ESCA is useful for N, Cl, Ca, S, Si, Pb, Hg, Br, Al, K, Cd, Fe, Mg, Na, C, N, B, P. It differentiates well between SO_4^{--}, SO_3^{--} and S (53, 127, 134, 135, 136, 137).

The emission spectrograph has long been a standard method (27, 44, 73, 85, 103, 110, 111, 127, 130, 131, 147, 197).

There are many other techniques including ion probe (13), flame photometry (147), infrared spectra (13), isotope ratios for C, O, S (57), optical microscopy (121, 123, 127), and x-ray analysis (121, 165).

For organic compounds gas chromatography (73, 97, 115, 127, 157, and thin layer 140) and mass spectrometry (6, 127, 131, 145, 166) have been found to be very useful.

A list of some of the elements determined by the principal techniques is shown in Table 1 arranged by atomic number and technique. The table shows elements specifically mentioned in the references. This does not mean that only these elements can be determined by the listed methods, nor have all references been listed for each element. The considerable number of elements for each technique, though incomplete, should be an indication of the power of the technique. It would be desirable to have the "best" method given for each element but "best" depends on many factors such as sensitivity, sample preparation, interfering substances, the equipment available, the number of elements that can be measured simultaneously, and cost.

Geographical Variation

The elemental and chemical composition of aerosols is extremely variable with location, as well as time. On this topic there is also material under the sections following. Aerosol particles are complex mixtures of soluble and insoluble components and agglomerates (164) which vary with location, origin and history. Analysis of continental aerosols in West Germany found that about 60% of the total mass consists of water soluble material, 25-30% of organic matter (mostly water soluble), 25% of the mass is volatile at temperatures up to 150°C, and 35% of insoluble, mainly minerals (186).

Over remote oceans the sea salt is 95% $(NH_4)_2SO_4$ and over the North Atlantic 75% (123). At 200 m above the ocean off Southern California, 11% sea salt, 20% soil dust (silicates), 25% SO_4^{--}, NO_3^-, or NH_4^- (from gaseous reactions), and 20% or more volatile (partly water) with Cl/Na ratio 2.4 compared to 1.8 in sea salt (67). Over San Nicolas Island compared to Los Angeles smog showed size increases due to the anthropogenic source in Los Angeles and Pasadena contributions in Mn, Zn, Ca, Br, Fe, Pb, SO_4^{--}, NO_3^- and non-carbonate carbon (67) with decreases in Cu, Cl and no marked change in Na and K. Mixtures of marine and continental aerosols interact along the coasts (56). Noontime smog in Los Angeles may be 80% liquid (80), with organic material on the surface of the aerosol particles (162). In Pasadena smog nitrogen is usually found as amino type or pyridene rather than NO_3^- or NH_4^- (134). Small particles in smog are liquid, the larger ones solid (169).

In urban aerosols Ni, Cr, and Zn are more on the surface than in the particle interior (132), Pb, Zn, As, Ca, Pb, Hg, Se, and S may be 100 to 1000% enriched over crustal abundance (132). Small particles may stick to

large ones (158). In Chicago 12 elements (non-marine) were coherent but different from each other, but not coherent with Na, Cl, and Br (185). In U. S. cities Fe, Pb, Zn, Cu, Ni, Mn, V, Cd, Sn, Mo, Cu and Bi vary with the city and the time (43, 132).

In Japan Pb, SO_4^{--}, NO_3^-, Cl^-, and NH_4^- were measured (154). Water soluble material was 30-45%, and higher in June with high relative humidity (13). In Kyoto, Japan analyses for a year of K, Zn, Na, Br, W, La, Cd, Ag, Cs and Co were measured and season changes of Br, Na, and Zn in polluted and unpolluted areas were compared and an air mass dependency found (146).

In Belgium analyses made for SO_4^{--}, NO_3^-, NH_4^-, benzopyrene, rock elements Si, Al, Ti, highly enriched probably industrial As, Sb, Se, and Na, and Cl from the sea, with some Cl maybe from industry (153, 154). Industrial sources produce 30% of aerosol components in the northern hemisphere (45).

In the USSR chemical analyses for 10 elements in five varied locations found wide variations with location and time for Mg, Mn, Pb, Fe, Ni, Al, Ca, Cu, Ag and Zn (85). Similarly for different sites and altitudes in the world ranges of values for Na, Si, Cl, S, K, Ca and Ti have been determined (49).

Water soluble fraction of an aerosol increases as the size of the particle gets smaller (107, 124), 25% of aerosol in 0.16 to 0.4 μm particles, less with larger particles, at higher altitudes 4000-5000 m, 1.5 - 2 times smaller than at 100 to 1500 m (107). The composition of the water soluble part varies with the size (85).

In Texas, x-ray fluorescence has been used to measure Ca, Ti, V, Cr, Mn, Co, Ni, Cu, Zn, Hg, Pb, As, Br, Sr, Mo, and Zr (161). In the Blue Ridge Mountains of Virginia terpenes are not found in urban aerosol (184). Auto exhausts have benzoic acid and phenylacetic acid which are not found in urban air particulates and can therefore be used as a tracer (184). Only a small part of the observed material of Zn, Sb, Cu, Se and As can be accounted for in Maryland (194). Since silicates and chlorides are non-anthropogenic they can be subtracted from the total material to get the anthropogenic contribution (112). Wind generated aerosols have 20 times more organic carbon than the parent soil (29). Aitken nuclei, radii less than 0.2 μm, are mostly sulfur (187). In Hawaii (149) with heating the marine aerosol has a 50% reduction in light scattering, 65-75% in air with a definite island trajectory, and 20% in subsiding upper tropospheric air. In marine air the total mass loss is due to the volatilization of water and organics. The marine aerosol at Kumukahi is 50% of daytime Hilo aerosol when the city is exposed to trade winds.

Sea Salt

Marine aerosols are formed by the evaporation of sea water which goes into the atmosphere by spray or bubble bursting. The mechanism of aerosol production is complex and the relationships between the variables not well understood (23, 63, 188). The proportions of the chemical constituents in the marine aerosol produced from the sea are not as in sea water, and vary from element to element (23), and ocean (34). There is, for example, a decrease in the amount of chlorine relative to sodium (67, 161) and the amount of organic material is greatly increased (73). Further, the importance of the marine aerosol decreases with altitude until at about 2000-3000 m it has been displaced by the continental aerosol so that it is essentially continental in nature (14, 96, 148). The Cl/Na ratio at 200 m above the sea is about 2.4 compared to 1.8 in sea water (67); at this altitude the aerosol was 11 percent sea salt. The Cl/Na ratio has been measured up to a value of 3 to 6 over the North Pacific Ocean (173). Tiny particles do not have the same composition as sea salt which varies above the sea (56, 80), but sulfuric acid and sulfates are appreciable (43, 123). The organic material in a particle may be in the range of 1-19% of the total (in Bermuda, 73), or as much as 35% (in the Virgin Islands, 162) with a protective coating on the particles (150). The volatile material may be 40% (80).

The elements reported from marine aerosols include: Na (21, 74, 185, 191), which can also be crustal in origin; Cl (21, 89, 92, 143, 185, 191); Mg (151); K (21, 33); Sr (33); Ca (33); I (119); Br (21, 119); and F (180). Fluorine is suggested as an excellent indicator of marine aerosols (180). Even marine aerosols are not of pure sea salt constituents (56, 96), and material from continents is important (24). Measurements over the subtropical North Atlantic found Fe, Al, Mn, La, Sc, Cr and Co apparently from continental origin, and Pb, Cu, Sb, Se, As and V from some non-crustal source (74). Na, Cl, SO_4 and K measured a decrease inland 6 km from the sea with large particles settling out in Israel (189). Surface maritime air changes as it goes inland from the ocean (14). One-fifth to one-quarter of nuclei instrumental in cloud formation are salt nuclei from the sea (46). Cl, Br, I, also Ca, Mg, Na have ratios in marine aerosol different from sea water and which vary with size range (4). Along coasts there is a mixture of marine and continental aerosols (14, 56).

Soil Constituents

When material from the earth's crust such as soil is blown into the air to form aerosols it reflects the composition elements of the source and is in the larger particles.

In Fresno, California, K, Fe, Ti, and Mn from wind blown soil was found in large particles, and also Ca from soil and a cement plant (47). In San Francisco, Fe, Al, Na, and Cl (93) from soil, and since Al, Mn, Fe, Sc, Ba, Cr, La, and Sm have the same wind dependence they must also be from the soil

(31, 93). Si, Na, Fe (112) are rock elements (154); Sc, Mn, La, and Cr are crustal (74); Fe, Al, Mn, La, Sc, Cr, Co are continental (94); and from dust or sea spray Cl, Ca, Ti, Cr, Mn, Fe (89). Al, Co, La, Eu, Th have the same levels as the earth's crust (25). The bulk of particles is in Si, Fe, Al, Ca, and Mg (105, 199), and small amounts of Ni, Cr, Mn, and Cu in the desert (105). Crustal elements Sc, La, Ce, Sm, Mn, Fe, and Mg correlate with Cd and Hg (90), Si from quartz and mica, Al from mica, Ca and Mg from limestone (32). One set of measurements in Denver found 80% of mass quartz, limestone, micas, and less than 1% industrial (32). Some other measurements by x-ray and optical microscopes on particles in Denver found 5% soil, 57% auto emission, quartz 9%, fly ash 8%, 17% miscellaneous minerals, and 4% miscellaneous (165).

From the Karakoram Desert, Fe, Ca, Zn and especially Si were found (30). For soil detection Al, Mn and possibly V/Na ratio have been recommended (119, 145). The natural aerosols from ground found Na on particles larger than 0.5 μm, Si, and Al larger than 1.0 μm (48). An analysis of 70 world sites' minerals found all high in silica (40). Silicates comprise 20% of aerosol over the oceans (67). In Eastern Washington state appreciable Si (71) was found. Silicates and carbonates are found in desert dust (76). At the South Pole (196) trace elements have been measured. Analysis of soluble aerosol inorganic origin for 13 compounds in USSR found $CaSO_4$ was about one-fourth of the material, a larger percent in particles larger in r = 1 μm with $Ca(HCO_3)_2$, $(CaCO_3)$, K_2SO_4, NaCl, NH_4NO_3, NH_4OH, but in r less than 1 μm $MgCO_3$, Na_2SO_4, $(NH_4)_2SO_4$, NH_4Cl, HCl, HNO_3 and H_2O (85). Major inorganic constituents are silicates, oxides of Al, Fe, Mn, Mg, $CaCO_3$ and sea water, in urban environment 40-60% sulfates (21). Fe and Ti have been reported from soil (92). Fe, Al and Si have been noted as suitable for crustal component normalization (40, 52, 53), and aluminum is particularly suitable (52, 125).

Non-Soil Constituents

The relative concentration of materials greater than that found in crustal materials is taken as evidence of origin from a non-crustal source. Soil constituents are high in Na, Fe (112), silicates, chlorides (112), Si, and Al on particles larger than 1 μm (30), and Na larger than 0.5 μm (30).

In marine aerosols over sub-tropical North Atlantic Ocean, Sb, As, Se, V, Pb, and Cu are not from crustal material (74). Identifications made as to the enhancement of non-crustal materials, particularly in urban environments at various locations and conditions have found the following:

Mn, Fe, Hg (ratio to Al) for an industrial area (153)

As, Sb, Se, may be industrial (154)

Pb, Cl, S, due to human activities in Denver (151)

Cd, Ag, Sn, Sb, Hg, Co, and Br higher concentration than in crust and sea (90), also V, Zn, Cu, Cd, Pb, and Se (33)

Cl, Br, V, and As enhancement from Washington, D. C. into Virginia forest (52)

Puget Sound, S anthropogenic (71)

Zn, Se, Br, Hg, Sb in San Francisco (93)

Zn, As, Se, Sb, Hg, and Pb, several orders more (25)

In Los Angeles it has been estimated that 50% of aerosols are due to secondary reactions between gases, or gases and particles (175). It has also been found that 30% of mass on clear days has benzene soluble organic, and 50% on polluted days (181)

In North Atlantic aerosols 10 to 1000x crustal concentration (33) for Pb, V, Sb, Se and different depending whether winds were from Africa or North America.

In the Antarctic Zn, Cu, Sb, Se and Br are higher than expected from volcanoes and marine activity (195, 196)

Terpenes are in high concentration in the Blue Ridge Mountains of Virginia (184) but not when the air comes from urban regions.

In agricultural material released by plants in the USSR, pollen, terpenes, etc. was nearly $9\mu g/m^3$ (85).

Hydrocarbons

There is a long list of hydrocarbon compounds which have been identified in urban pollution from man's activity (20, 166). In addition there are compounds from natural sources, land and sea. Background measurements have been made (157). The terpenes are produced from natural sources in millions of tons (58).

Over remote oceans there is $2\text{-}4\ \mu g/m^3$ with compounds of 14 to 32 carbon atoms (34). In Bermuda the total organic carbon on aerosols has been measured as 1-19% of the aerosol with the percent decreasing with increasing salt content. The organic carbon mass loading was 0.15 to $0.47\ \mu g/m^3$ (73). In the Virgin Islands the aerosols were 35% organic material with a protective coating on the aerosols such that there is little humidity effect (162). The fatty acids above the Ligurian Sea were $0.050\ \mu g/m^3$ and over

the Pacific Ocean 0.08 to 4 $\mu g/m^3$. Coastal fog coated by evaporation retarding monolayer remains as 0.3 to 2.6 μm stabilized droplets (80).

Organic material (ether soluble) at Mainz, Germany was 18%, Denselbach (rural) 9%, Irish West Coast 8%, Jungfrau Joch 3%, Cape Verde Islands 1.5%, with a total of 55 organic compounds identified (101).

In the USSR 25 $\mu g/m^3$ of organic origin was found in the cities (85). Auto exhaust contributes to hydrocarbon content. Traffic correlations with hydrocarbons and nitrogen oxides by type (gas chromatography) have been found (115). Airports also contribute (52). At Tucson, Arizona the benzene soluble organic fraction was 5-8%, consistently less than 15%, and the water soluble fraction 2-5%.

Many measurements have been made on hydrocarbons in the Los Angeles Basin. Of the volume 60-80% of the total particulates, and of the mass 25-50% was organic (138). Carbon accounts for 2/3 of the total organic aerosol in Los Angeles (143). In Los Angeles organic matter is mostly in small particles, in a cascade filter 30% of the particles with 12% of the mass is collected on the backup filter (33, 139). Condensed organic vapors peak in a volume at a size of 0.35 μm (65). In Pasadena only the particles larger than 0.1 μm had a volatile component (65). The carbonate carbon is low compared to non-carbonate carbon, especially smaller than 0.6 μm (129, 130). Most carbonate carbon is in particles larger than 2 μm (130).

Motor Vehicles

One of the most important sources of atmospheric pollution is the automobile. In addition to emission particulates a significant part of the gaseous products of combustion in time become converted to aerosols, or are absorbed on aerosols. As noted in the section on Time Patterns the aerosol levels in many places have correlated with traffic patterns. The most important elemental measurements have shown that from antiknock compounds in gasoline Pb and Br or their ratio correlates with traffic, and is in small particles as expected from high temperature combustion (15, 35, 36, 87, 112, 125, 155, 184, 195). Chlorine is also an ingredient in leaded gasolines as has been noted (30, 52, 53, 142), but since there are other sources, as well as the sea, it is not as specific for motor vehicles.

The elements Zn, Ba, Co (43, 52, 53, 142) have also been found on aerosols from motor vehicles, especially in tunnels, also Ni and Cd (53). The barium may come from diesel smoke suppressant (52), Al, Ca and Fe from crustal material near highways (142). The auto emission particles have been measured at 57% of the total (165). There is a correlation of traffic with C_2-C_5, and C_4-C_8 aliphatic hydrocarbons, and C_6-C_{10} aromatic carbon compounds measured by gas chromatography (115). The auto exhaust aerosols have benzoic acid and phenyl acetic acid not found in other urban aerosols (184).

Elements

The results of some of the measurements of the elemental composition of aerosols are summarized here. It should be kept in mind that although an aerosol particle in some cases may be of uniform composition, in general it is not the case, especially in urban atmosphere which is polluted. But even in natural source aerosol particles the composition is complex. The elements are easier to determine; however ordinarily the elements are not in pure form, but combined in chemical compounds whose nature is not as easy to determine in the free air. Some of the results are referred to in other sections.

Nitrogen. The chemical form depends on size and time of day (137).

Sodium. Comes from the sea or sea salt (48, 112, 125, 176, 185, 194), from mostly large particles (48), with no size effects (118), from natural and ground sources in ocean and crust (162), with Cl/Na ratio for sea salt (119).

Magnesium. Is mineral in origin (199), and on large particles (48, 185).

Aluminum. Is mineral in origin (199), from soils (119, 125, 176), wind blown (48), on large particles (50, 176, 185), more than 1.0 μm (48), from natural aerosols and ground (30), its use as ratio element to determine if aerosols are of crustal origin (52, 119).

Silicon. Is mineral in origin (199), soil (112), natural aerosols from ground larger than 1.0 μm particles (30), in Eastern Washington state particles are mostly (71).

Sulfur. Has no major fluctuations with lack of a discrete source (36). There is an increase in amount on smaller particles with the time of day in Los Angeles (138); found on very small particles (48) with submicron particles 10 to 21% variation in concentration (139). In Pasadena, California SO_4^{--} predominates by night and SO_3^{--} by day (134). It is due to human activity in Denver (151). In Puget Sound, Washington, many particles contain anthropogenic origin material (71). It may be appreciable in filter collected material (66). The chemical composition depends on the size and time of day (137). There may be a difference in the isotope ratio depending on source (57).

Chlorine. Is produced by sea salt (112), from automobile and sea, replaced by Br on aerosols with loss of Cl from marine aerosols mostly on smaller particles (120), small particles (82), sea salt Cl/Na ratio (119), on large particles (89), on large and small particles (48), from automobile exhaust (112, 125), with Pb and Br from automobile exhaust on particles less than 0.5 μm (175), non-anthropogenic (112), narrow size distribution in U. S. cities (110), due to human activity in Denver (151).

Potassium. Comes from background oceanic terrestrial aerosols and anthropogenic contributions associated with dispersion of rock (125).

Calcium. Is mineral in origin (199), from soil and cement in Fresno, California (47), sedimentary in origin (47), large particles (47, 48, 89, 142, 185), diurnal pattern in Pomona, California (47), same diurnal pattern as Fe on Harbor Freeway, Los Angeles (47).

Titanium. Comes from the soil (92).

Vanadium. Is found on small particles from combustion processes (48, 50, 176), identified with, and therefore a good tracer for residual oils (52, 61, 80, 110, 130, 194, 198). Over the North Atlantic Ocean it has 13 times the crustal concentration (33), possibly from dust (119). The V-bearing particles may be scavenged out of the air by large particles associated with vehicles (142). It is on small particles (82).

Iron. Is mineral in origin (47, 92, 130), from soil (90, 112), on large particles (13, 48, 50, 89, 142, 172, 185), in U. S. cities has the greatest concentration on large particles 0.6 to 1.8 $\mu g/m^3$ (110), around 3.5 μm in size (141), from residual combustion (61). In Pomona, California it has the same diurnal pattern as Ca and Pb (47), and at the Harbor Freeway the same as Ca (47).

Nickel. Is produced by combustion processes (48), oil combustion plants (52), residual oil combustion (61), and motor vehicles (53).

Copper. In Chicago was found on large particles (185), from residual fuel oil combustion (61). Care must be exercised in its measurement (53).

Zinc. Is found on larger particles than Pb (28), on larger particles (43, 53, 143), in an increasing amount with particle size (13), Zn/Pb ratio from motor vehicles is too small to account for Zn levels in large U. S. cities, (125, 142), from motor vehicles (53), not from motor vehicles (142), related to activity of motor vehicles, from tires (?) (52), on small particles in Chicago (185), more in Antarctic than expected (195), in Fresno, California on small particles in morning and evening from tires (47), combustion (47), in Pomona, California more small particles with different diurnal pattern than Pb and Ca (47).

Arsenic. Is found primarily on small particles (118).

Bromine. Has been identified as being on small particles produced by leaded gasoline combustion and correlated with traffic patterns (5, 13, 32, 47, 50, 53, 80, 87, 89, 93, 103, 112, 118, 126, 130, 155, 161, 175, 176, 185), more than in sea and crustal materials (90). The Br/Pb ratio may be used (87, 103) to separate natural and pollution sources (50, 130). It is in sea salt (119). There is more in the Antarctic than expected (195).

Strontium. Has been identified as an anthropogenic source in San Jose, California (47).

Cadmium. Is on small particles (172), correlates with Sc, La, Ce, Sm, Mn, and Fe (90), and is produced by motor vehicles (53).

Lead. This important pollution element is found in small particles produced by leaded gasolines and correlates with traffic patterns (13, 35, 43, 47, 48, 50, 53, 87, 89, 125, 126, 141, 160, 161, 165, 172, 175, 194). The Pb/Br ratio is characteristic of leaded gasolines (87) and may be used to separate material and pollution sources (93, 185). Lead may be attached to larger particles near a freeway (28). Lead isotopes on the west coast of the U. S. are different than in the Los Angeles Basin (28). It is due to human activity in Denver (151). The concentration in U. S. cities is in the range 1.3 to 3.2 $\mu g/m^3$ (110). Over the North Atlantic Ocean there is 3000 times the crustal concentration (33).

Sources

In addition to the material in the preceding sections there are listed here some of the elements which have been identified with particular sources.

Residual fuel oils. Have been identified with V (197), Se, Sb (52, 130), Ni (43).

Automobiles. For Pb and Br see references under elements, also benzoic acid and phenylacetic acid (184).

Airports. V, Mn, Sb, Zn from jet fuels and tires (52), Ba from diesel vehicles (52), and As from tires (52).

Jet fuels. In Los Angeles, Al, Au, In, La, Ti, V, Ba, Dy, Te, and U above detectable limits, only Al, Ti, and Ba about 0.1 ppm (140), large agglomerates (170), mostly carbon (41, 130).

Air mass. Marine Arctic has the smallest number, two orders below mean frontal zones, Ti, Zn, Sn order of magnitude above average (at 300 m tower) (193) Na, K. Ca 1.5-2 times above mean (146), chemistry and air mass correlate (14, 18, 146).

Local sources. Give larger variations Cu, W, Cr, Zn, Sb, Ga, Br, Ag, Fe, and Ce, but same are Na, K, Ti, Al, Sm, Eu and correlations between Cr-Sb-Zn, Sc-Co-Th-Ce, and Ca-Mg (185).

Metallurgical plants. Cr, Zn (161), Pb, Zn, As, Hg, Cu, Fe oxide (61).

Coal-fired power plants. Elements and particle size (131).

Table 1. Techniques for elemental analysis.

Element	Neutron Activation	Proton Activation	X-ray Fluorescence	X-ray Fluorescence Radioactive Source	X-ray Photon Activation	Proton Scattering	Atomic Absorption	Emission Spectroscopy
1 H						133, 174		
2 He						133		
3 Li						133	156	
4 Be						133	57, 73 156	73
5 B						133		
6 C						133		
7 N						133		
8 O						133		
9 F	44				180	133		
10 Ne	44					133		
11 Na	118, 120 180, 185		11	160	7, 194	133	149, 156 180, 196	
12 Mg	62, 93 180, 185		11		7	133	149, 156 196	
13 Al	44, 62 119, 140 180, 185		11, 36 86	160	186	133	156	55

Table 1 (continued).

Element	Neutron Activation	Proton Activation	X-ray Fluorescence	X-ray Fluorescence Radioactive Source	X-ray Photon Activation	Proton Scattering	Atomic Absorption	Emission Spectroscopy
14 Si		89	11, 36, 86, 94		194	133	156	
15 P						133		
16 S	27, 47 62, 55	89, 133	11, 36 47, 86 94, 187			133		55
17 Cl	47, 112 185	89, 133	11, 36 47		7, 82 180, 194	133		
18 A								
19 K	5, 27 47, 62	89	11, 25 36, 47 187				149, 156 180, 196	
20 Ca	14, 62 155, 185	89	11, 36 47, 187	160, 161	7, 194		149, 156 196	
21 Si	27, 62 93, 118 180, 196							
22 Ti	44, 62 93, 180	89	47, 86	25, 161	7, 194		156	

Table 1 (continued)

Element	Neutron Activation	Proton Activation	X-ray Fluorescence	X-ray Fluorescence Radioactive Source	X-ray Photon Activation	Proton Scattering	Atomic Absorption	Emission Spectroscopy
23 V	27, 41 44, 62 180, 196	89	11, 36 47, 187	25, 161	82		156	55, 73
24 Cr	5, 44 62, 93 185, 196	89	36, 47 86	80, 160 161	7, 194		156	73
25 Mn	27, 44 62, 93 180, 196	89	36, 47	25, 160 161			141, 149 156, 160 180	55, 73
26 Fe	44, 62 180, 185 196	89	11, 13 36, 47 86	25, 160 161	7		141, 149 156, 160 180	55
27 Co	5, 27 44, 62 93, 180		11	161			156	
28 Ni	44, 47	89	11, 47 86	25, 161	7, 194		73, 141 149, 156	55, 73
29 Cu	25, 27 44, 47 62, 185	89	11, 47 86	25, 160 161	194		73, 141 149, 156	55, 73

Table 1 (continued).

Element	Neutron Activation	Proton Activation	X-ray Fluorescence	X-ray Fluorescence Radioactive Source	X-ray Photon Activation	Proton Scattering	Atomic Absorption	Emission Spectroscopy
30 Zn	5, 27 44, 62 185, 196	89	11, 13 36, 47	25, 160 161	7, 194		73, 141 149, 156 160	73
31 Ga	27, 47 62, 155		47		39			
32 Ge	44			25				
33 As	27, 44 62, 73 155, 185		36, 47	161	7, 194			
34 Se	27, 44 47, 50 118, 176		36, 47 187		7, 37 194			
35 Br	5, 27 44, 62 185, 196	89	36, 47 112, 187	25, 160	7, 39 194			
36 Kr								
37 Rb	5, 44 47, 155		47		7, 194		156	
38 Sr	44, 47		36, 47	25, 160	39		156	

Table 1 (continued).

Element	Neutron Activation	Proton Activation	X-ray Fluorescence	X-ray Fluorescence Radioactive Source	X-ray Photon Activation	Proton Scattering	Atomic Absorption	Emission Spectroscopy
39 Y					7, 39 194			
40 Zr			86	25, 160	7, 39 194			
41 Nb								
42 Mo	44			25, 160				
43 Tc								
44 Ru	44							
45 Rh					39			
46 Pd			86					
47 Ag	5, 27 62, 93 155				39			
48 Cd	5, 44		11, 86		39		52, 73 156	73
49 In	27, 46 62, 176 185				39			

Table 1 (continued)

Element	Neutron Activation	Proton Activation	X-ray Fluorescence	X-ray Fluorescence Radioactive Source	X-ray Photon Activation	Proton Scattering	Atomic Absorption	Emission Spectroscopy
50 Sn	44		86	160			127	73
51 Sb	5, 44 118, 180 185, 196		127		7, 194			
52 Te	41							
53 I	44, 93 119, 155				7, 194			
54 Xe								
55 Cs	5, 44 118				194		156	
56 Ba	5, 41 44, 93 155, 176		86, 127		39		127	73
57 La	5, 27 41, 44 62, 118							
58 Ce	27, 44 62, 93 180			160	7, 194			

Table 1 (continued)

Element	Neutron Activation	Proton Activation	X-ray Fluorescence	X-ray Fluorescence Radioactive Source	X-ray Photon Activation	Proton Scattering	Atomic Absorption	Emission Spectroscopy
59 Pr	44							
60 Nd	44							
61 Pm								
62 Sm	27, 44							
	62, 93							
	118, 155							
63 Eu	27, 62							
	93, 155							
	180, 196							
64 Gd	44							
65 Tb	44							
66 Dy	41							
67 Ho								
68 Er	44				39			
69 Tm				160				
70 Yb	44, 50				39			
	197							

Table 1 (continued)

Element	Neutron Activation	Proton Activation	X-ray Fluorescence	X-ray Fluorescence Radioactive Source	X-ray Photon Activation	Proton Scattering	Atomic Absorption	Emission Spectroscopy
71 Lu	44, 50 197							
72 Hf	44, 50 155, 197				39			
73 Ta	44, 50 155							
74 W	5, 27 44, 62 155			160	39			
75 Re	44							
76 Os	44				39			
77 Ir	44				39			
78 Pt	44				39			
79 Au	44, 93 155, 156				39			
80 Hg	27, 44 47, 62 93, 176		47	160, 161	39		73	

Table 1 (concluded)

Element	Neutron Activation	Proton Activation	X-ray Fluorescence	X-ray Fluorescence Radioactive Source	X-ray Photon Activation	Proton Scattering	Atomic Absorption	Emission Spectroscopy
81 Tl								
82 Pb		89	11, 13	25, 160	7, 39		52, 141	73
			36, 86		194		149, 156	
			94, 176				160, 196	
83 Bi							156	
84 Po								
85 At								
86 Rn	44							
87 Fr								
88 Ra								
89 Ac								
90 Th	27, 44							
	50, 93							
	118, 155							
91 Pa								
92 U	41, 44		160					

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