The concentration and size distribution of airborne ferromagnetic particles

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ABSTRACT

A description of a device to remove ferromagnetic particles from an airflow is given. The instrument utilizes several sets of permanent magnets for the creation of a magnetic field of high inhomogeneity. A geometrically similar device without magnets serves as a reference precipitator. The particles were deposited onto electron microscope grids in the precipitators by magnetic and/or other forces. In comparing the size and number of particles on the grids of the precipitators, a "difference" distribution could be constructed. This is regarded as representing the spatial size distribution of airborne ferromagnetic particles. The significance and possible applications of the results are discussed.

Introduction

The total concentrations of particulate matter in polluted air range roughly from 100 $\mu g/m^3$ to 4 000 $\mu g/m^3$, the latter indicating heavy smog conditions in industrial areas. Average values run from 200 to 800 $\mu g/m^3$ for large, and from 100 to 200 $\mu g/m^3$ for small cities. (Junge, 1963). These values are one or two orders of magnitude higher than those characteristic for the average background concentrations.

The determination of the various components of this aerosol, both natural and artificial, is of known importance in a number of environmental sciences. Among them there are two fields—cloud physics and health sciences—where the size (or mass) distribution and concentration of particles of certain elements (or compounds) can play a leading role in processes like condensation, precipitation formation, visibility reduction and pulmonary penetration and retention.

It is also known that iron is one of the most important elements in the atmospheric aerosol for at least two reasons. First, in urban air this constituent is more abundant than lead or magnesium by factors of two to fifteen, and can be found in concentrations of one or two orders of magnitude higher than copper, zinc, manganese and others. (Magill et al., 1956; Lee et al., 1968). Further, an important fact is that the emission of iron seems to be steadily increasing. For this reason, there is a need to asses the possibly increasing degree of hazards connected with this emission.

From the point of view of cloud physics, on the other hand, the abundance of iron particles raises two interesting questions. One of them is connected with the role of these particles as potential ice forming nuclei, in the initiation and worldwide distribution of precipitation, while the other is concerning this role in the formation of noctilucent clouds.

Both question are related to what extent the meteoric influx, in the forms of prominent meteor showers and sporadic meteors, and producing an inhomogeneous, but continuous source of secondary particles, will play part, if any, in the phenomena mentioned above.

The problem of the ability of iron particles

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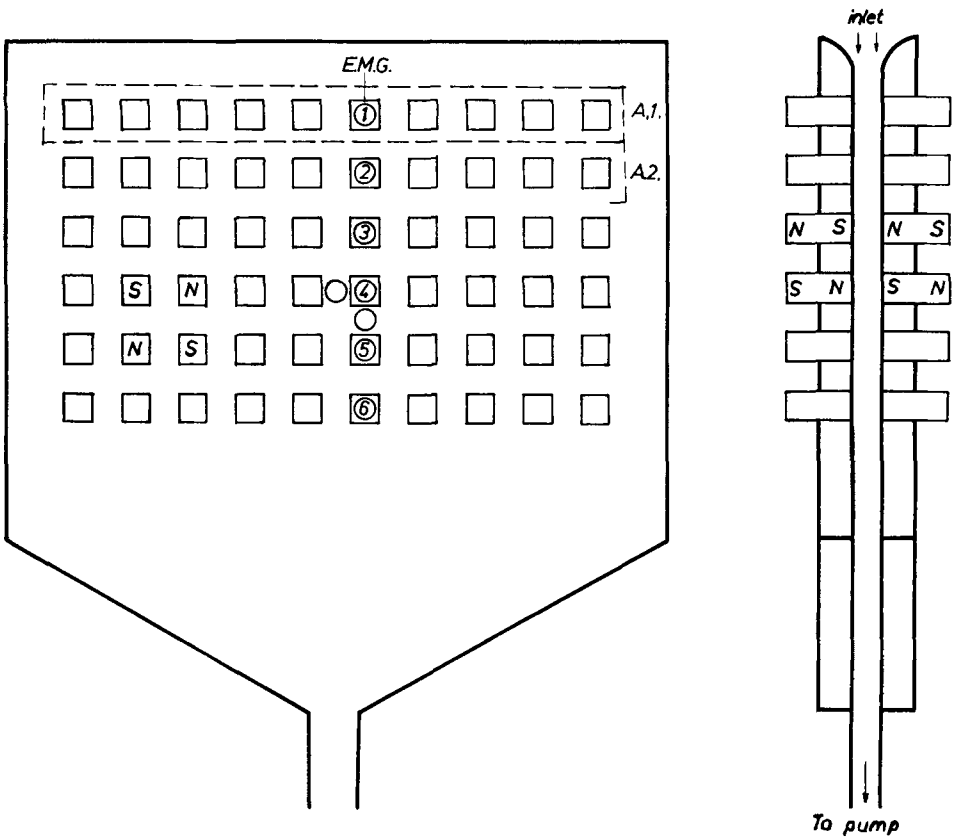

Fig. 1. The magnetic precipitator, consisting of two sets of permanent magnets alternating the polarity of the individual magnets. *E.M.G.*, electron microscope grids (circles with and without numbers). The distance between the walls is 3 mm.

to initiate an ice cloud will be discussed in the paper which follows (Prodi & Wirth, 1972). In the work reported here, the concentration and particle size dsitribution of ferromagnetic material (probably mostly iron) was measured at the Osservatorio Scientifico over Verona, Italy (elevation: 284 m a.s.l.), during a four-day period (14.9–17.9 1971). The Observatory is located on a hill facing the wide plane of the Po Valley (about 85 000 km^2), more than 200 m above it and above the town of Verona. The only remarkable pollution source in the area is a small foundry on the plane, close to the town.

Method of measurements

To our knowledge, the ferromagnetic fraction of atmospheric aerosol in the range between about $5 \times 10^{-6} < r < 10^{-4}$ cm has not been investigated, as far as particle methods are concerned. However, simple devices can be designed to meet different requirements of both pollution and cloud physics studies.

The force acting on ferromagnetic particles having magnetic moment **M** has the form:

$$\mathbf{F} = \mathbf{M} \cdot \nabla \mathbf{H} \qquad (1)$$

where $\nabla \mathbf{H}$ is the gradient of the inhomogeneous field. According to this equation, any magnetic precipitator should be designed in a way to ensure a high inhomogeneity of the magnetic field.

Two sets of permanent magnets have been arranged, as shown in Fig. 1, in lines and columns, alternating the polarity of the individual magnets. These sets were assembled facing the opposite polarities at close distance (3 mm). The air was sampled between them at a flow

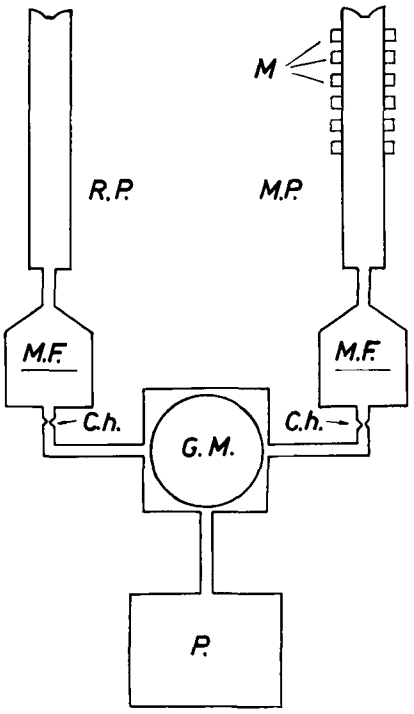

Fig. 2. Sampling arrangement. *M*, permanent magnets; *M.P.*, magnetic precipitator; *R.P.*, reference precipitator; *M.F.*, membrane filter holders; *C.h.*, calibrated holes; *G.M.*, gas meter; *P.*, pump.

velocity of 2.5 cm sec^{-1}. To detect the particles collected by the magnetic precipitator (M.P.), several electron microscope grids were placed on each magnet along the line in the direction of the flow. For comparison, a few grids were also located at intermediate sites between some of the magnets. Other mechanisms than the capture of ferromagnetic particles by the magnetic forces can be effective in removing particles from an airflow between two walls close to each other. To evaluate their importance in our case, a set of similarly prepared grids was located in another geometrically identical sampler without magnets (reference precipitator, R.P.). The arrangement is shown in Fig. 2. The two airflows were euqalized by two calibrated holes downstream of the membrane filter (M.F.) holders. The difference between the numbers of particles found on the two sets of grids were considered as the collected ferromagnetic fraction of the atmospheric aerosol. The comparison with the grids in R.P. was also useful to determine which combination of parameters (distance between the walls, length of the inhomogeneous field, velocity of the flow carrying the aerosol) was giving the highest rate of deposition of the particles. At a fixed geometry of the precipitator, the flow rate was adjusted to the highest value which was giving statistically the same distribution (and concentration) of particles on the last row both in the M.P. and in the R.P.

As for any precipitator, the problem arises how to evaluate the capture efficiency of the instrument. In principle, this could be done both by theory and experiment.

Theoretical calculations cannot be made because there exist wide variations in aerosol structure, density, chemical composition and shape of particles.

By experiment the problem can be solved by testing the precipitator with an artificially produced ferromagnetic aerosol. Besides the difficulties connected with the production and control of the real chemical and physical properties of this aerosol, one cannot be sure if the particles would be representative of the natural ones.

In our case it was the instrument itself which suggested the possibility to consider the rows of magnets, located perpendicularly to the airflow, as independent precipitators. If N_1 and N_2 are the numbers of particles of radii r_i collected on the grids over the first and second rows respectively, while the actual concentration of them is $f(t)$ in the air, then (Junge, 1953):

$$N_1 = E(r)\, f(r)\, V \tag{2}$$

$$N_2 = E(r)\,[f(r) - E(r)\, f(r)]\, V \tag{3}$$

and the collection efficiency of the first row, E, is

$$E = \frac{N_1 - N_2}{N_1} \tag{4}$$

The result of such a computation will be shown in Fig. 3 for the different size classes of ferromagnetic particles. It is seen that the above considerations about the size dependence of captured particles are at least statistically valid, independently of the unknown composition, origin and shape of the material. At the given experimental conditions the collection efficiency for particles of 0.3 μm is 50 % and drops

very quickly to 20 % for particles of 0.16 μm in diameter.

The investigation of particles deposited on the sets of carefully selected, carbon coated electron microscope grids in both parts of the instrument was made by means of a Siemens Elmiskop II electron microscope at a magnification of 8 000. Ten "squares" of each grid were inspected representing a total area of 6.4×10^4 μm^2 and the number and shape of particles found were recorded in 20 size classes from 6.2×10^{-6} cm to 3.0×10^{-4} cm. The classification was made by direct visual observations while a few electron microscope pictures were taken for comparison with the results of other investigations and for demonstration.

Results and discussion

The particle shapes identified on the grids of both the magnetic and reference precipitators were spherical, spheroidal, irregular and fluffy, similar to those found by Hemenway & Soberman (1962), Bigg et al. (1971) and others from rocket soundings between heights of about 80–160 km.

A detailed chemical analysis will enable to determine the fraction of ferromagnetic material originating from outer space. Authors of several studies of the so-called "black spherules" found in deep sea sediments, polar ice, volcanic dust, snow crystals and in the atmosphere (for instance: Crozier, 1960; Wright et al., 1963; Hodge & Wright, 1964; Kumai, 1966; Vittori, 1968, respectively) tend to agree about their meteoric origin. Not much is known, however, about the man-made production of these spherules and, in addition, the very recent rocket and satellite flight's results of Fechtig et al. (1968), Farlow et al. (1970), Fechtig & Fenerstein (1970) have failed to give conclusive evidence of both the dust belt around the Earth at heights of 80–120 km and the previously reported particle fluxes. No assumption was therefore made about the origin of ferromagnetic particles, and we treated this particle population simply as a contamination characteristic of moderately polluted area. Therefore two different curves were drawn (Fig. 4) of the "surface concentration" vs. size of the particles sampled simultaneously in the M.P. (curve *b*) and in the R.P. (curve *a*); in both cases the average grids area scanned was 0.064 mm². What is of interest in this case is the concentration of particles in the sampled air. This total number of particles deposited was inferred from the concentration on the grid in the first row, that was uniform, as it was checked by inspecting a grid (open circle in Fig. 1) between two polar pieces. The concentration was computed with the aid of the capture efficiency previously mentioned (Fig. 3) knowing the total volume of air sampled (20 m³) and the total useful area of the precipitator (161 cm²).

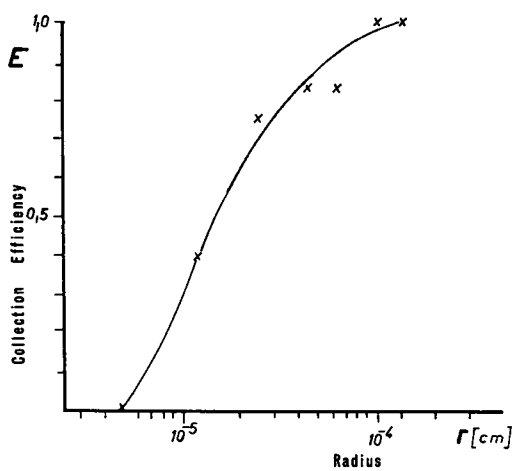

Fig. 3. Collection efficiency of the magnetic precipitator for the different size classes of ferromagnetic particles. The original 20 size classes have been reduced to seven, between the fixed limits, for smoothing the curve.

In Fig. 5 the particle number concentration vs. size is shown as deduced from the R.P. (curve *A*), and the M.P. (curve *B*) and the difference, i.e. the ferro-magnetic particle concentration distribution (curve *D*). For sake of comparison, an average distribution of continental aerosols, given by Junge (1963) is also shown (curve *C*).

The regular decrease in the concentration of ferromagnetic aerosol for $r \geq 2 \times 10^{-5}$ cm is very similar to that established for various particles and can be represented by the usual equation

$$dN/d(\lg r) = cr^{-\beta}$$

where dN is the number of particles in the $d(\lg r)$ range, c is a constant and β is, in our case, equal to 2.1. The total concentration of

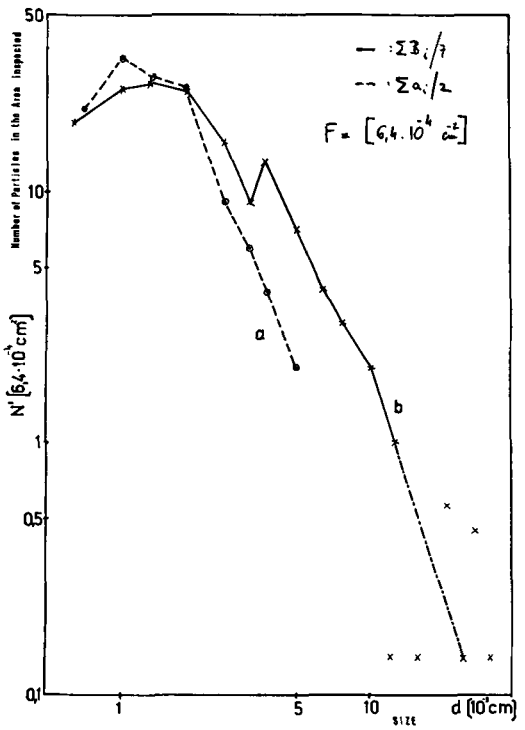

Fig. 4. Average number of particles in the inspected area vs. size of the particles sampled simultaneously in the M.P. (curve *b*) and R.P. (curve *a*). The average grid area scanned was 0.064 mm²; seven grids have been inspected in the M.P. and two in the R.P.

this fraction is 1.8 particle per cc, the 94 % of which falls below 0.6 μm radius.

To judge the significance of the distribution represented by curve *D* in Fig. 5, we have converted our results into cumulative mass (concentration) distribution. In this calculation we have used a density of 3 g cm^{-3} for the particles in the previously selected seven size classes, the centers of which were choosen to form "equivalent" radii. These, being 10–20 % smaller than the formers (where the percentage was arbitrarily selected) were used as the starting point for the calculations. The calculated masses then were expressed in micrograms/m^3, and plotted (Fig. 6) together with values deduced from measurements made by means of a six stages cascade impactor and analyzed by atomic absorption spectrophotometric analysis (Lee et al., 1968). This comparison demonstrates an agreement between the results of two different methods of measurement which seems to be good for particles below about 2.5 μm This agreement is, however, not so good comparing the total iron contents, as our result is about one order of magnitude smaller than that of Lee et al. At any case both distributions seem to represent the iron content of aerosol in a not heavily polluted air (Lee's measurements were made at Fairfax, a suburb of Cin-

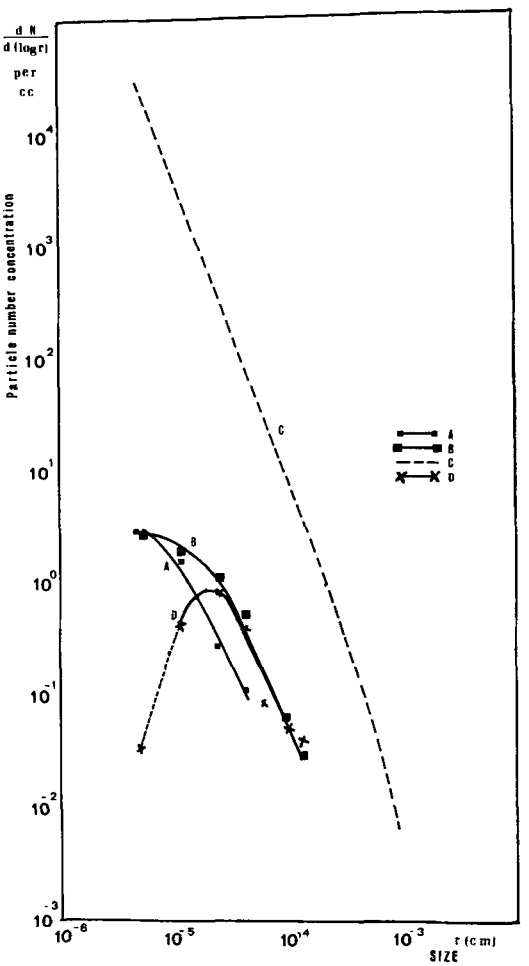

Fig. 5. Particle number concentration vs. size constructed by the particles found in the R.P. (curve *A*), and in the M.P. (curve *B*); the difference of the two gives the ferromagnetic particle concentration distribution (curve *D*). For sake of comparison, an average distribution of continental aerosol, given by Junge (1963), is also shown (curve *C*).

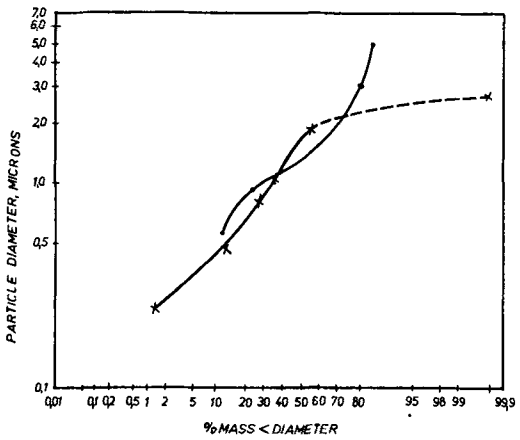

Fig. 6. Cumulative mass (concentration) distribution as calculated by our measurements (line with crosses) and derived from the measurements of Lee et al. (1968) at Fairfax, Cincinnati (line with dots).

cinnati, USA), which is the main constituent of elements in solid form in it. The existing differences could be explained by several reasons. First, we have to take into account that our results should have been affected by particles of a "mixed" nature from the point of view of ferromagnetism. It should also be mentioned that for the relatively small area of the grids investigated, the "particle method" could not be said to be safe in detecting the large ferromagnetic particles, which have a very small spatial concentration anyway. On the other hand, the magnetic precipitator is able to extend the limits of measurements to the smaller size range (to about 0.3 μm in diameter) where the cutoff appears. The cutoff itself is connected with the forces acting in the inhomogeneous field of the permanent magnets as it was previously described. Other reasons could be a real decrease in the concentration of the ferromagnetic particles or a certain limit for their exhibiting ferromagnetic behaviour.

As to the first possibility, recent studies of the Mössbauer isotopes in atmospheric air by Kopcewitz & Dzienis (1971) seem to confirm the higher iron content, but point out that it is existing in the form of ultrafine ($d = 10^{-6}$ cm) particles. Being in superparamagnetic state, the decrease in E of the magnetic particles of our precipitator could be explained.

The results of measurements available in the literature generally refer to the mass concentration (or distribution) of various elements deduced from microchemical analysis of the aerosol. The bulk methods, however, do not provide *detailed* informations about the possible hazards of given chemicals to human beings, for it is not possible to give more than a rough guess for the retainment by the lungs. Naturally, however, it is much more difficult to determine the chemical composition of individual particles, except if the size separation is accomplished while the particles are airborne. From this point of view, the above described simple method and its findings can be considered useful in hygiene, atmospheric physics and chemistry.

Appendix

Curve A in Fig. 5 (particles collected on the grids of reference precipitator) shows that a certain fraction of atmospheric aerosol had been removed from the flow passing between the walls. The removal of particles by the walls is due to several effects. (Being in the range of low enough Re numbers, the Stokes' law applies to the movement of the particles, if the particles can be considered spherical.) The resistance of the medium is increased close to the walls by a factor $(1 + 0.56\ (r/x))$ (Fuchs, 1964), where r is the radius of the particle and x is its distance from the wall. The deviation from the Stokes velocity, that is the presence of the wall, becomes appreciable when $x \leqslant 10r$.

By means of the values of x for different radii it is possible to compute the volumes V_i, from which the particles of corresponding sizes had been taken away by the walls. The ratio of this volume to the total volume sampled, V, should be equal to the ratio of the number of particles (N_i) found on the reference precipitator to that of the aerosol content of the volume sampled for the individual particle sizes. This last measurement has not been made; instead, an average size distribution of continental aerosol was used for computations. Having performed such a calculation for two particle sizes ($r = 5.6 \times 10^{-6}$ and $r = 2.0 \times 10^{-5}$ cm) a slightly lower concentration of particles than the one shown by curve A in Fig. 5 has been found. The difference can be explained on account of Brownian diffusion, electrostatic deposition and the deviation from the Stokes' flow due to the irregular shapes of particles.

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КОНЦЕНТРАЦИЯ И РАСПРЕДЕЛЕНИЕ ПО РАЗМЕРАМ АТМОСФЕРНЫХ ФЕРРОМАГНИТНЫХ ЧАСТИЦ

Написано инструмент, составленный для селекции ферромагнитных частиц из даннаго течения воздуха. Прибор использовает перманентные магниты для создания магнитного поля высокой неоднородности. Подобный по-внешнему инструмент без магнитов служил контрольному преципитатору. Частицы из воздуха были оседены на решетках электронного микроскопа в приборах из-за магнитных и других сил. Оценивая размеры и число частиц с помощю электронного микроскопа, было возможно конструировать разницы распределений, что учитывается распределением ферромагнитных частиц в воздухе. Объясняется значительность и возможная применяемость результатов.