

PLAINTIFF'S
EXHIBIT
FD-210

PARTICULATE AIR POLLUTION
GENERAL

8000 0614

SCF-FA-0050

PRODUCED BY FORD

PARTICULATE EMISSIONS

by W.R. Pierson

I. Aerosols - General

Suspended in the atmosphere are small particles of diameter up to, say, 0.01 cm (that is, 100 microns or 100 μ). Some of these particles are solid, some are liquid (usually containing dissolved solids), and some contain both a solid phase and a liquid phase. These particles are collectively called "particulates" or "aerosols."

The size distribution of particulates in continental air is usually found to be as in Fig. 1, the right-hand part of which has a slope approximating - 3 on a log-log plot of $dn/d \log r$ vs. r .

This is what is called the Junge distribution. A plot of $r^3 dn/d \log r$ vs. r for an aerosol following the Junge distribution looks like Fig. 2. Since $r^3 dn/d \log r = dV/d \log r$, what Fig. 2 says is that in a continental aerosol there are generally about equal amounts of particle volume (hence, mass) in each $\log r$ interval; for example, the mass between 0.1 and 1 μ is approximately equal to the mass between 1 and 10 μ . This holds true in polluted atmospheres sufficiently far from a single source but is not true in clean marine atmospheres, where large sea-salt particles predominate.

Particulates are involved in several atmospheric phenomena; as shown in Fig. 3, a given effect will generally be dominated by particles of certain sizes rather than by all the particulates in the air sample. For example, visibility reduction has to do mostly with particles in the size range of 0.1 to 1.0 μ .

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Gravitational settling velocities depend on particle size as shown in Fig. 4. It is gravitational settling that limits the upper end of the particulate size spectrum (Fig. 1). However, gravitational settling is unimportant for particles smaller than $1\ \mu$. The atmospheric residence time of airborne particulate matter is on the order of days or weeks. Mechanisms for removal of particles are, besides gravitational settling,

- 1) Inertial impaction onto surface obstacles (trees, buildings, and so forth) (mostly large particles)
- 2) Growth by coagulation with other particles (mostly small particles)
- 3) Growth by accretion of material from the vapor phase (including condensation of water vapor to form raindrops) (mostly large particles) (2 and 3 can then be followed by removal by gravitational settling and inertial impaction)
- 4) Washout by raindrops falling from the clouds above (mostly large particles).

There is, additionally, one major mechanism for particle shrinkage, though not usually complete disappearance; namely, evaporation.

Nucleation of particulates is of two types: 1) heterogeneous nucleation, i.e., buildup onto a tiny already-existing particle, or 2) homogeneous nucleation from the gas phase without benefit of nucleating centers. There are usually plenty of nucleating centers present to allow process 1), so that process 2), which requires a many-fold supersaturation of condensable material in the vapor phase, seldom occurs.

Processes of formation of aerosols include combustion; chemical reactions of gases in the atmosphere (e.g., photochemical smog); condensation from the vapor phase; injection by biological processes (e.g., spores, pollen); spray from the sea surface; dust stirred up by the wind or kicked up by men, animals, and machines; comminution (grinding up material by natural or man-made operations); and many other processes. The first two dominate in considerations of air pollution.

Representative particulate loadings for various situations on the earth's surface are shown below.

Table I. Examples of particulate loadings

<u>Place</u>	<u>Number/cm³</u>	<u>µg/m³</u>
Remote ocean	200	10
Remote rural	10000	35
Settled rural	35000	65
Industrial areas	200000	150
Auto exhaust pipe	10 ⁹	40000

II. Air Pollution Aerosols

Emissions inventories have produced the following numbers for the amounts of various materials emitted to the atmosphere. (Ref. 5 has most of these numbers.)

Table II. Estimated annual emissions, tons

(Particulate emissions refer to material emitted as particulates, and not particulates formed photochemically, etc.)

Global particulates from natural sources	2,400 X 10 ⁶
Global particulates from pollution	100 X 10 ⁶
Total U. S. air pollution (1968)	214 X 10 ⁶
Total U. S. particulates from air pollution (1968)	28 X 10 ⁶
Total U. S. gaseous emissions from gasoline-driven vehicles	81 X 10 ⁶
Total U. S. gaseous emissions from diesel vehicles	1.3 X 10 ⁶
Total U. S. particulate emissions from gasoline-driven vehicles	0.5 X 10 ⁶
Total U. S. particulate emissions from diesel vehicles	0.3 X 10 ⁶

(The last figure does not include railroads and other non-highway diesel fuel consumption, which is about equal to the consumption by road diesels.) Particulates formed in the atmosphere from gaseous pollutants (that is, sulfates from SO₂, nitrates from NO and NO₂, particulates formed by atmospheric photochemical reactions of hydrocarbons, etc.) amount to some 200 X 10⁶ tons worldwide annually, or twice as much as the emissions of particulates qua particulates - so-called "primary" particulates - given in Table II.

III. Air Pollution by Aerosols from Vehicle Engines

In discussing particulates from motor vehicles we will consider only the primary particulates and ignore the secondary particulates (e.g., photochemical smog) even though the latter are very important. As seen in Table II, the total mass of primary particulates from motor vehicles is a small part of the pollution from vehicles or of the aggregate pollution picture nationwide and world wide. But within a restricted locality, the particulate contribution of the automobile can be significant as shown in Table III (from Refs. 4 and 7).

Table III. Vehicle particulate emissions and total particulate pollutant emissions in selected urban areas (includes gasoline- and diesel-powered vehicles).

<u>Urban area</u>	<u>Tons of particulate annually</u>	
	<u>Total</u>	<u>Motor vehicles</u>
Washington, D. C.	35000	5700
New York and North N. J.	231000	33800
Kansas City	60000	5000
Jacksonville, Fla.	14000	600
St. Louis	147000	4700
Los Angeles County	43000	16400

In the L. A. area, vehicles contribute an estimated 38% of the total particulates.

An idea of the rate of emissions from an individual vehicle can be obtained from the following (See Refs. 6, 7, 23):

Total particulate from automobile exhaust

≈ 0.35 g/mile or ~ 5 g/gallon of fuel consumed

Total particulate from diesel exhaust ~ 50 g/gallon of
fuel consumed

The level of emissions depends on fuel composition, engine design, operating conditions, and other variables.

Gasoline-engine exhaust particulate contains the following (if leaded fuel is used):

Pb (mostly in compounds) 20 to 40%

Carbon particles and various organic compounds, including raw or partially burned lubricating oil - variable $\approx 40\%$

Compounds of Ca and Mg $\sim 2\%$

Cl and Br (in Pb compounds mostly) - 15 to 30%

Sulfur (largely as sulfate) - up to a few %

Fe and Fe oxides - a few %

Zn - a few tenths %

Al, Si, Cu - traces

The Pb, Cl, and Br are present by virtue of the tetra-ethyl lead together with the ethylene chloride and bromide added to scavenge the lead. Lead compounds that have been reported include PbClBr , $\text{NH}_4\text{Cl}(\text{PbClBr})_2$, $(\text{NH}_4\text{Cl})_2 \text{PbClBr}$, $[\text{Pb}_3(\text{PO}_4)_2]_3\text{PbClBr}$ if phosphorus additives are present, PbSO_4 , and $\text{PbO} \cdot \text{PbSO}_4$ (see Refs. 4, 25, 30, 31, 32, 36). The lead halides form by condensation from the vapor phase as the cooling exhaust gas reaches the 350°C range. The Mg, Ca, and Zn are primarily from the lubricating oil, the ferrous material from wear

and rusting, the sulfur from the small amount (ca. 0.035%) present in gasoline. Solid hydrocarbons arise from the same sources as the gaseous ones in general. The carbon particles constitute a combustion aerosol, as would be formed in any flame.

Most of the particles in gasoline-engine exhaust are very small, as shown in Fig. 5. Particles as small as $0.01\ \mu$ are present in large numbers. Composition varies with size; for example, Fe is found mostly in the larger particles, while Pb concentrates in the small ones, as would be expected from their respective modes of formation.

Diesel engine exhaust particulate contains carbon particles (~50%), paraffin hydrocarbons (including unburned fuel) (~50%), paraffin acids (~1%), H_2SO_4 (~1%), Si (~0.01%), Fe (~0.005%), Zn (~0.005%), and traces of Al, Ca, Ni (see Refs. 41, 44); there will be compounds of barium or other elements if smoke suppressants are used. (Refs. 6, 7, 40, 41) The particles tend to be larger than those from gasoline-engine exhaust (see Ref. 6).

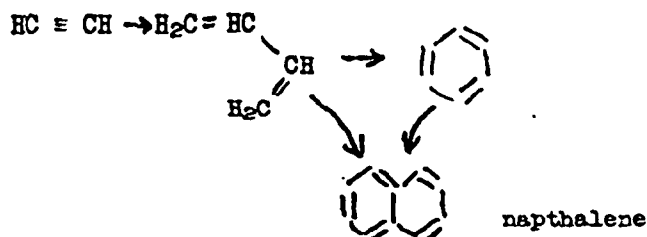
A given particle from a vehicle exhaust is unlikely to be simple in composition but rather will generally be a hodge-podge of inorganic and organic material. Because of their vapor pressures, the organic species can generally be expected to condense from the gas phase, normally onto some already-existing site. The amount of particulate matter experimentally found thus depends on experimental conditions. If the sample is collected hot, organic material will be in the vapor phase and will be missed. If the sample is cooled below ~50°C, water droplets will be collected unless the stream is diluted. Some organic constituents will evaporate after collection.

The unburned fuel comprising much of the diesel particulate has a room-

temperature vapor pressure of the order of 10^{-3} mm Hg (e.g., cetane, $C_{26}H_{54}$) and hence could not exist alone in a condensed phase unless the surrounding atmosphere were about 1 ppm diesel fuel by volume (10 ppm by weight); this illustrates the proposition that complex processes must be at work in order that certain species be observed at all.

Common to both gasoline exhausts and diesel exhausts, and indeed to combustion of any fossil fuel, are compounds called polycyclic hydrocarbons or polynuclear aromatic hydrocarbons (PNA). A few of them are shown in Fig. 6. There are a number of related compounds which, though not strictly hydrocarbons, should be considered along with the PNA's; examples are shown in Fig. 7.

PNA's are produced in greatest amount when the engine is run rich. They are present in gasoline to the extent of a few ppm, of which some 0.1 to 0.2% survives the combustion process and is emitted in the exhaust (see Ref. 24). A primary source of PNA's in exhaust, however, is thought to be a buildup from molecular fragments formed by pyrolysis of fuel or lubricating oil in the combustion chamber (see Ref. 8 Ch. 20, Refs. 39, 45):



and so forth. About 30 PNA's have been found in automobile exhaust. They amount to some 0.1% of the particulate from a gasoline engine and a similar (Reference 46) percentage of the particulate from a diesel engine.

The vehicle has been estimated to contribute 2 to 40% of the compounds of this class found in urban atmospheres, depending on the city. (The main source of PNA in city air is the burning of coal, particularly in antiquated residential heating units; the strongest doses to individuals come from smoking -- cigarette smoke is full of PNA's.) The reason for concern about PNA's is that many of them, and their methyl derivatives, are carcinogens.

Factors affecting PNA emissions are the following: High aromaticity in the fuel gives high PNA emissions. High consumption of lubricating oil gives high PNA emissions. The presence or absence of lead anti-knock fluid seems to have little or no effect on PNA emissions if the rest of the gasoline composition is held constant. Avoiding a rich fuel mixture strongly reduces PNA emissions but little is gained by continuing to increase the air-fuel ratio beyond the stoichiometric value and into the lean region. Emissions of PNA's are at their highest during acceleration and deceleration. As a rule, emission controls for PNA's are the same as those for hydrocarbons in general and for CO: air injection thermal reactors on the manifold, catalytic converters, etc. Particulate traps reduce PNA emissions but not to an impressive extent. Much of the PNA produced in the automobile ends up in the crankcase; presumably, then, the control of blowby suppresses PNA emissions.

Fuel factors affecting the mass of emitted gross particulate matter are the following: The presence of Pb gives higher emissions (though Habibi and Jacobs of DuPont claim the opposite), mostly due to an increase in the amount of inorganic material (primarily Pb salts). However, there is some evidence that leaded fuel reduces the emission of carbon and/or carbon compounds during cold start and warmup. An increase in the amount of scavenger (ethylene dichloride, ethylene dibromide) in the anti-knock fluid causes emission of more particulate matter. Fuel additives such as upper cylinder lubricant seem to increase

particulate emissions; detergents may do the same, though this is disputed. Some investigators believe that anything that decreases engine deposits (more scavenger, presence of detergent additive) will increase particulate emissions; this seems possibly true since some 15% of the Pb consumed in running on leaded fuel is retained as deposits in the engine, exhaust system, etc. Another 10% of the Pb consumed seems to be retained in the lubricating oil. Replacement of tetra-ethyl lead with tetramethyl lead has no discernible effect on particulate emissions. High fuel aromaticity gives higher particulate emissions.

The rate of Pb emissions depends strongly on the immediate history of the engine and on engine speed. At low speed with a clean engine, the Pb emitted is only a small fraction (say, 20%) of the amount consumed, and deposits build up. With sustained light-duty service, Pb emissions eventually approach 60% of the Pb consumed. If, after a period of such low-speed driving, the car is suddenly accelerated, the Pb emissions can be as high as 2000% of the amount consumed, owing to the ejection of deposit material, much of it in large (up to 5000 μ diameter) flakes. With protracted running at high speeds the loosening of deposited material will finally subside and the rate of Pb emissions will be just a little less than the rate of Pb consumption in the fuel. If now low-speed driving is resumed, Pb emissions will be very low (say, 10% of that consumed) and will climb slowly to the low-speed value. The emission rate is very erratic, however, since large-particle emissions can be affected by thermal or mechanical shock, changes in gas velocity, etc. There may be another maximum associated with deceleration.

In general, severe engine operating conditions mean that a higher percentage of the Pb consumed is emitted and the particles are bigger. Over the life of

the car, some 75% of the Pb consumed is emitted as particulate. Emissions tend to increase with age.

Emissions of gross particulate matter are highest when the engine is cold. Particles can be expected to be larger at low exhaust gas velocity owing to enhanced opportunity for coagulation and wall loss, and indeed it appears (Ref. 36) that nitrate and sulfate particles are larger at lower road speed. It is presumably true that oil burners are smoke producers. Diesel particulate emissions rise very steeply when the engine load approaches its maximum rating.

Various means of control for gross particulates have been tried. A system of fluted exhaust pipes (for cooling) followed by a mesh-filled box and a cyclone collector (DuPont, Ref. 28) reduced Pb emissions 10-fold. Other systems (Ref. 42) include thermal precipitation in a packed bed or acoustic coagulation in a fluidized bed, or (Ref. 32) a simple trap muffler. The control of blowby gets rid of much of the problem, since the particulate emissions in blowby are in quantities approaching the tailpipe emissions.

Catalysts for the control of gaseous pollutants seem to be very effective in destroying organic particulates. With lead-free fuel, nearly all of the exhaust particulate matter is organic. It is therefore anticipated that the use of catalysts, especially since such use apparently must be accompanied by the use of lead-free fuel, will eliminate the problem of exhaust particulates. Particulate emissions from attrition of the catalyst itself, by chemical attack, exfoliation, pellets wearing against each other, etc., should perhaps be considered, especially in view of the fact that such material is, after all, a catalyst.

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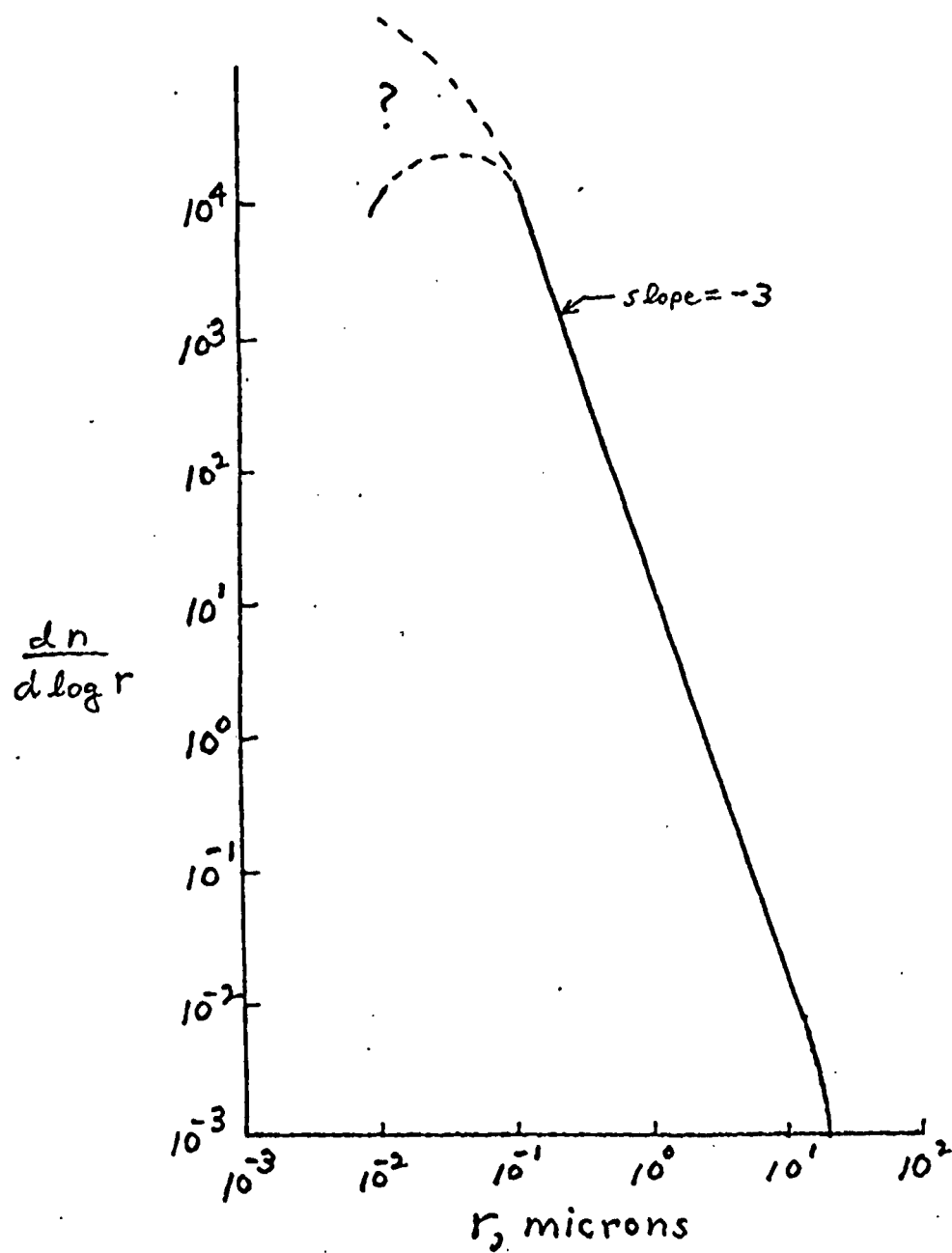


Fig. 1— The Junge distribution.

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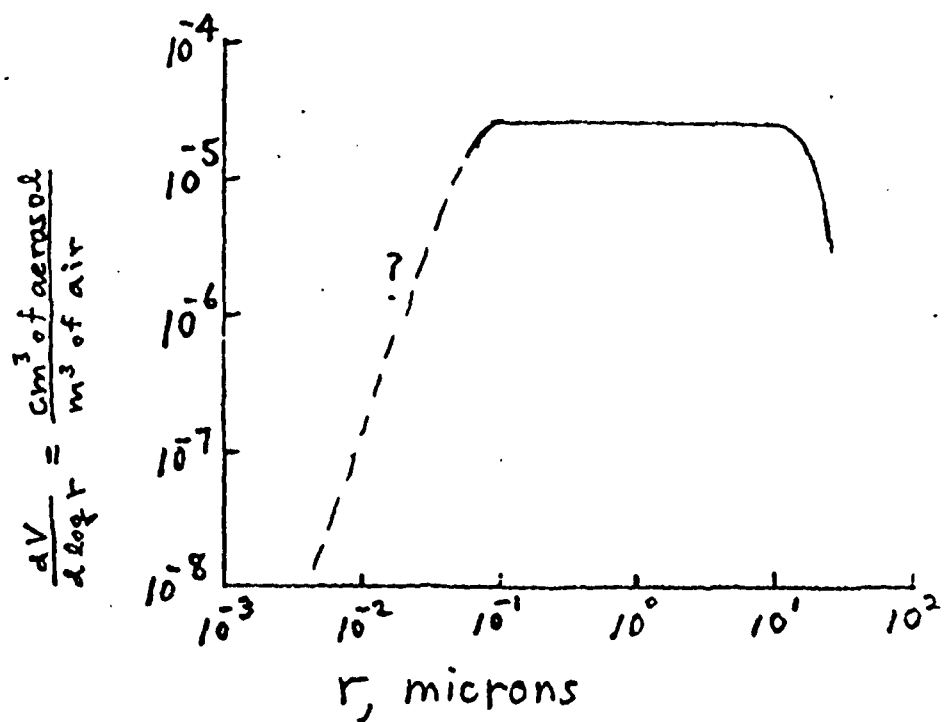


Fig. 2 - The Junge distribution

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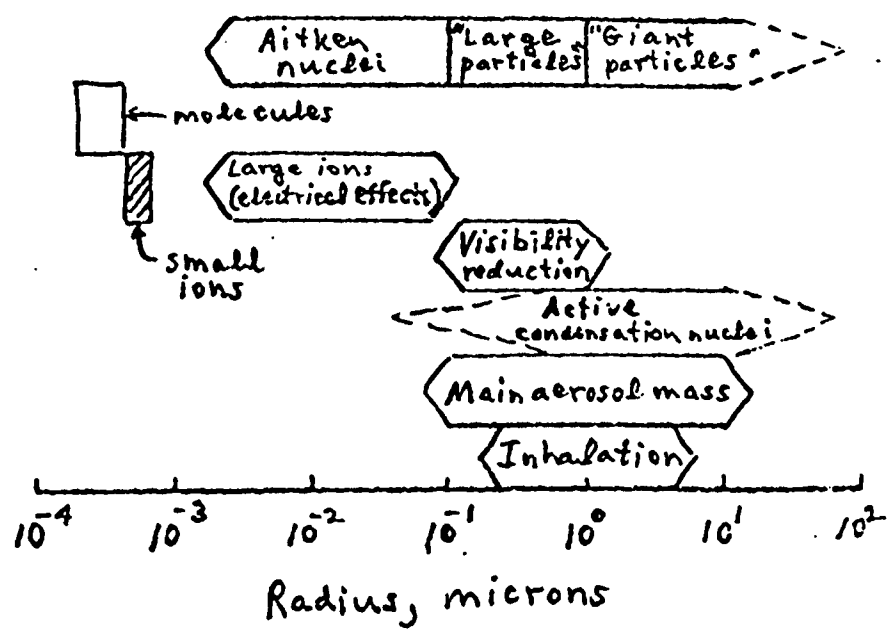


Fig.3 — Particle size ranges associated with various effects

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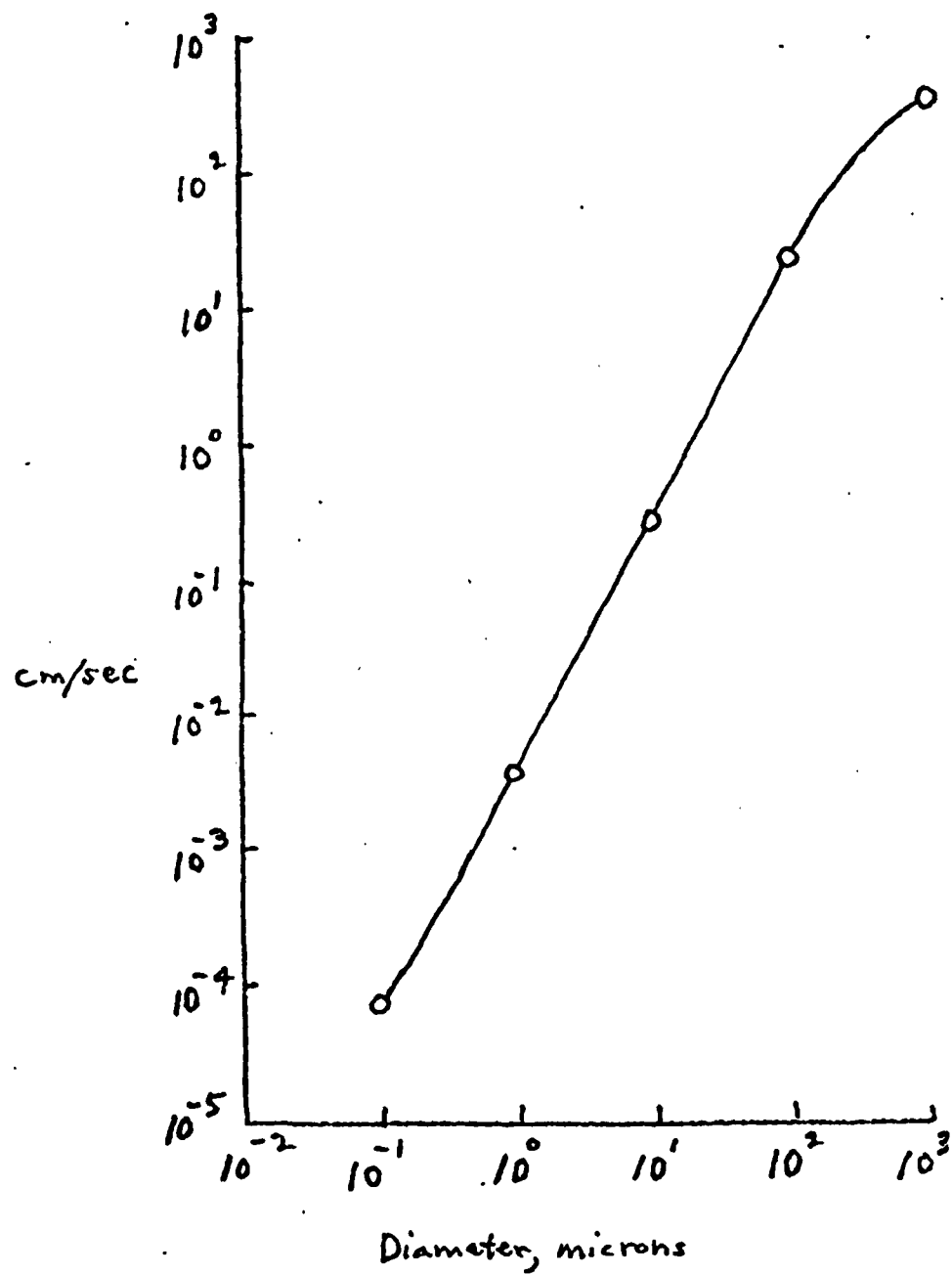
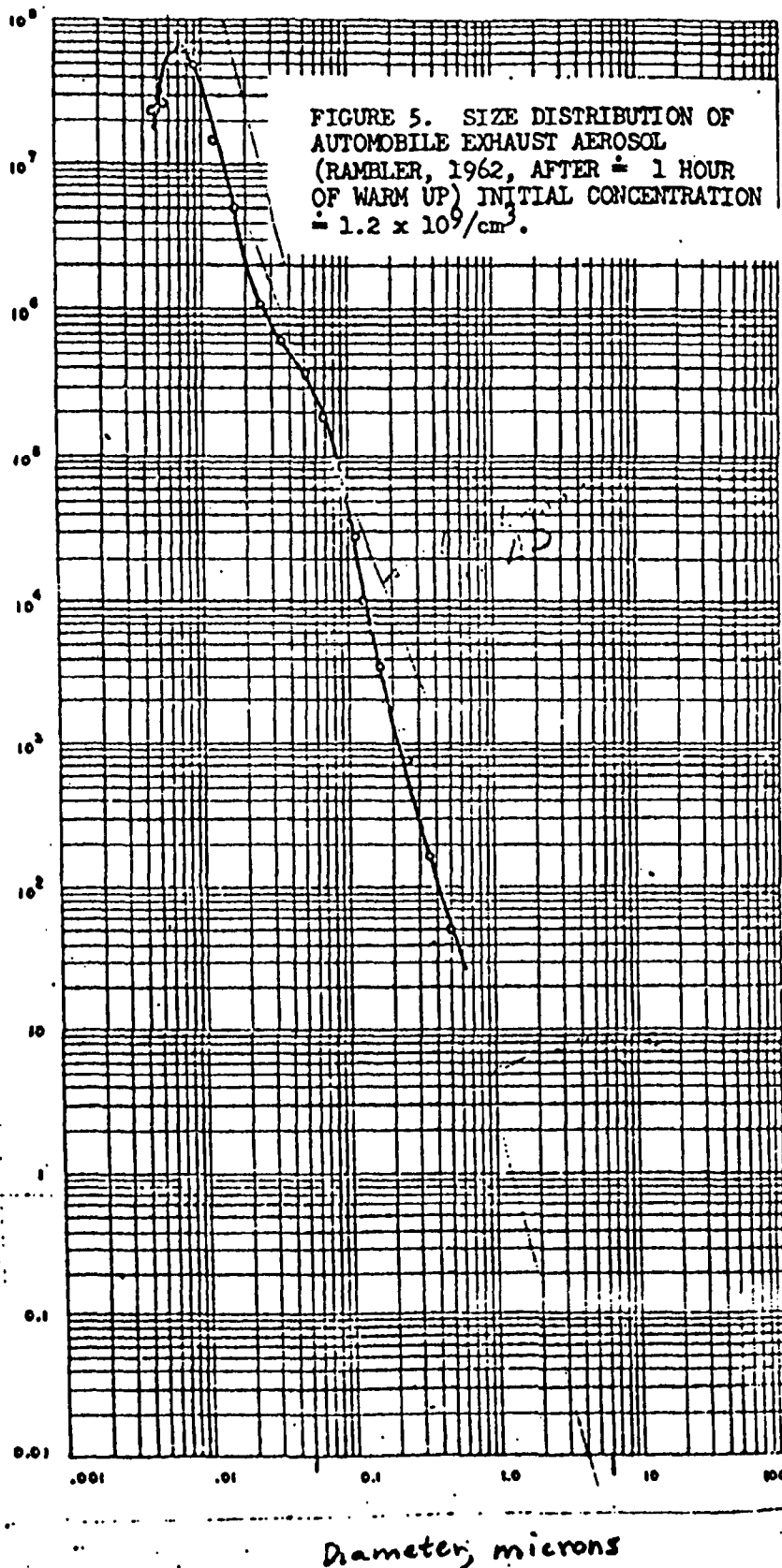


Fig. 4 - Settling velocity for unit-density spheres in air at 0°C and 760 mm Hg

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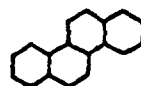
from Whitby et al., "Generation of Small Ions," July 1969

$\frac{dN}{d(\text{diam})}$ or $\frac{dn}{2\pi d \log r}$

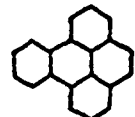


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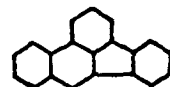
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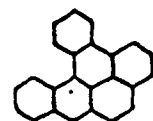
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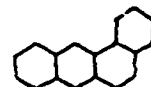
Benzo(e)pyrene
(1, 2-benzopyrene)



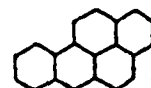
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(3, 4-benzofluoranthene)



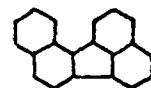
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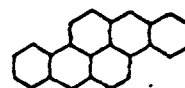
Benz(a)anthracene
(1, 2-benzanthracene)



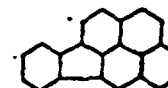
Benzo(a)pyrene
(3, 4-benzopyrene)



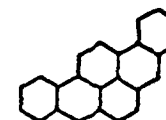
Benzo(j)fluoranthene
(10, 11-benzofluoranthene)



Dibenzo(a, h)pyrene
(3, 4, 8, 9-dibenzopyrene)



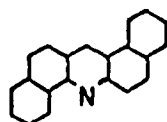
Indeno(1, 2, 3-cd)pyrene
(o-phenylenepyrene)



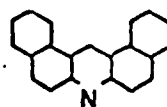
Dibenzo(a, i)pyrene
(3, 4, 9, 10-dibenzopyrene)

FIGURE 6

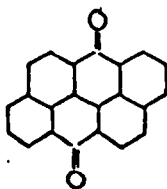
Carcinogenic Polynuclear Aromatic Hydrocarbons
Identified in Urban Air



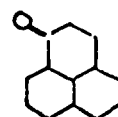
Dibenz(a, h)acridine
(1, 2, 5, 6-dibenzacridine)



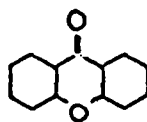
Dibenz(a, i)acridine
(1, 2, 7, 8-dibenzacridine)



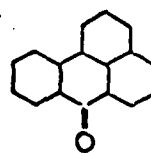
I
Anthanthrone



II
Phenalene-9-one



III
Xanthene-9-one



IV
7H-benz(de)anthracen-7-one

FIGURE 7

Aza-Heterocyclics and Polynuclear Carbonyl
Compounds Identified in Urban Air

8000 0638