

## Visibility Reduction due to Jet-Exhaust Carbon Particles<sup>1</sup>

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### ABSTRACT

Pyrolysis of hydrocarbon fuels leads to emission of free carbon in the exhaust of aircraft turbojet engines, visible as a faint dark trail. Carbon formation rises markedly when water injection is employed to augment thrust by 20-30 per cent in takeoffs under heavy loads or at high temperatures, the enhanced pyrolysis resulting from the lower combustion efficiency on such "wet takeoffs." During the takeoff run, while the aircraft is still moving at low speeds but with maximum thrust, dense dark smoke fills the exhaust wake, reducing visual ranges to as little as a few hundred feet. To cross-check recent measurements indicating carbon particulate emissions of the order of 15 lb per ton of fuel consumed in wet takeoff, Mie extinction coefficients have been computed for carbon particles of the size known to form as a sequel to pyrolytic freeing of carbon. These are used to make theoretical estimates of the *maximum* visual range to be expected if the carbon loading measurements were correct. A discrepancy exists in the sense that the predicted visual ranges are found to be some five times larger than the observed. It is concluded that a large fraction of total carbon emission leaves the tailpipe still unaggregated into large soot particles, whence the reported carbon loadings may be too low by a factor of as much as five. Consequently the aircraft operational hazards as well as the air pollution problems implicit in rising volumes of jet traffic at certain terminals may become rather more serious than has been predicted.

### 1. Introduction

Advent of turbojet aircraft has created a new source of visibility reduction at airfields, a reduction caused by the very aircraft that depend so heavily on good terminal visibility. Incomplete combustion of jet fuels, chiefly during the extreme engine operating conditions characteristic of takeoffs, leads to emission of dark smoke (carbon particles) from jet exhausts. Accumulation of such emissions under adverse conditions can aggravate terminal visibility problems, interfere with certain types of military aircraft operations, and with rising levels of commercial jet traffic could become more than a negligible source of air pollution (Appleman, 1956; George and Burlin, 1960).

The chief purpose of the following discussion will be to summarize some Mie-theory computations of absorption and scattering cross-sections of carbon particles and to use them in making certain estimates of opacity of jet-exhaust smokes. Discussion of those topics will be preceded by a brief summary of the physical chemistry of carbon-particle formation and comments on the greatly enhanced rates of carbon emission when water-injection is used to augment thrust during jet aircraft takeoffs, because these topics have strong bearing on the present problem.

### 2. Carbon-particle formation

Although the exact mechanism of formation of free carbon particles during combustion of hydrocarbons is not known, extensive study initiated because of the importance of the problem in many industrial processes (Parker and Wolfhard, 1950; Schalla, *et al.*, 1954; Tesner, 1961) has established that carbon formation is always associated with some form of *incomplete combustion*. When hydrocarbons are heated to a temperature high enough to begin pyrolytic breakdown of the hydrocarbon's molecular structure, soot formation will ensue if insufficient oxygen is provided to oxidize the molecular fragments all of the way to water and carbon dioxide. Rapid *quenching* before oxidation goes to completion is a frequent cause of carbon formation, a process turned to practical advantage in commercial production of carbon black. Even in combustion processes where the *over-all* air-to-fuel ratio is far above the stoichiometric ratio, as is true under typical aircraft turbojet combustion conditions, *local regions of inadequate oxidation* may develop. If quenching occurs in such cases before sufficient oxygen invades the pyrolyzed parcel of gases, carbon will be freed and soot formation is assured.

The mode of carbon aggregation following pyrolysis remains obscure, but it probably resembles a nucleation process. It is not yet known whether the aggregating entities are molecules of  $C_2$  or  $C_3$  or free radicals con-

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taining residual hydrogens that subsequently oxidize. But what is important for the jet-exhaust analysis that follows is the well-established point that the carbon nuclei proceed to grow by accretion of other carbon skeletons or radicals until they form crystallites with surprisingly uniform diameters of about 20 Å (Angstrom units), as revealed by electron microscopy (Watson, 1959) and confirmed by surface-area determinations using adsorption techniques.

Formation of high concentrations of the above type of crystallites is followed (probably because of quenching due to rapid motion through and beyond the turbojet flame zones) by rapid cessation of crystallite growth near the 20 Å size under typical conditions. Further aggregation next proceeds via coagulation of the crystallites into what Schalla *et al.* (1954) aptly term "popcorn balls." These popcorn-ball-like spheres are what will here be designated as "carbon particles," for they represent the smallest carbon aggregates leaving the jet engine as smoke. Their diameters are, like those of their constituent crystallites, remarkably uniform in most combustion processes; but, in turbojet combustion, diameters appear to range downward from an upper limit of about 500 Å to less than 100 Å.

The carbon particles (popcorn balls) may finally aggregate into larger, less regular structures, of which filaments usually represent the next stage, followed by matting of many filaments into flocculent lint-like masses whose sizes range into many tens of microns in some instances. These flocculent aggregates will here be termed "soot particles," following Schalla *et al.*, to distinguish them from the nearly spherical "carbon particles" with diameters of the order of a few hundred Angstroms. All of the above-outlined sequence of events,

beginning with pyrolysis and terminating with formation of a mixture of carbon particles and soot particles, takes place in a total transit time from turbojet combustor to tailpipe of only about 10 milliseconds.

### 3. Turbojet water injection process

Carbon emissions are readily noticeable from jet exhausts under all conditions of operation as soon as one sights along, or almost along, the flight path. However, here our chief concern will be with the unusually large quantities of carbon, posing visibility and air pollution problems, that are emitted when turbojet-powered aircraft employ *water injection* during takeoffs. Such takeoffs are referred to by pilots as "wet takeoffs"; but from the point of view of the casual observer they might well be called "black takeoffs," considering the intense dark clouds of carbon smokes emitted (Fig. 1).

Because turbojet thrust depends upon the mass as well as velocity of the exhausted combustion products, atmospheric density exerts strong influence on thrust. Variation of runway air temperature from winter to summer values can vary thrust from, say, 12,000 lb to only 8000 lb, with corresponding variation either in required length of takeoff run or in payload limit for a fixed run. To obviate this operational difficulty, effort has been devoted to finding ways of augmenting thrust during the takeoff period. Auxiliary jet devices (JATO) proved unwieldy, but by injecting water into the forward sections of the jet engines it becomes possible to attain substantial rise in thrust (Driggs and Lancaster, 1955; Casamassa and Bent, 1957). The thrust increase is due chiefly to the higher mass flux resulting from increased density produced by evaporative cooling of

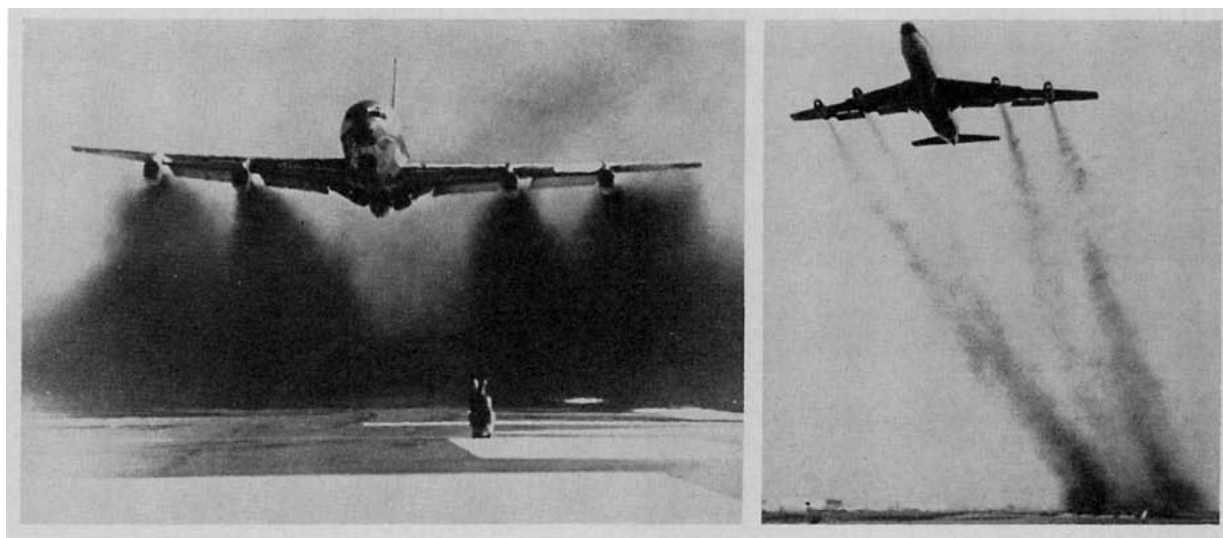


FIG. 1. Jet-exhaust carbon particles reducing local visibility during water-injection takeoffs of multi-jet transports. (Photographs by Los Angeles Air Pollution Control District.)

the water; but other favorable factors add to the thrust-increase. Alcohol is often added to prevent freezing of the injection mixture, but aside from its own slight fuel value, it acts essentially as does the water. Thrust increases of 25–30 per cent are readily obtained by use of water injection, but it is very important to note, in understanding carbon production in wet takeoffs, that fuel consumption rates rise far more than that percentage: *wet takeoffs actually involve very low engine efficiency.*

Surprisingly large volumes of water are employed in multi-jet wet takeoffs. The Boeing 707 and Douglas DC-8 each carry about 700 gallons of water for injection use in their Pratt and Whitney JT3 turbojet engines. This injection-water volume is about equal to the entire *fuel* capacity of the long-used DC3 transport! Operating rules call for use of water injection whenever runway temperature is over 40F, assuming standard loads, which means that wet takeoffs are almost the norm in many parts of the world throughout most of the year. The writer's observations of wet takeoffs by B-47 medium bombers indicate that water injection in that aircraft is ordinarily completed about one minute after beginning of the takeoff run, at which time the aircraft is usually about 1000 ft above the ground. Moment of termination of water injection is apparent even to the ground observer, since the darkness of the exhaust trails drops abruptly as injection ceases.

Although the smoky exhaust of a wet takeoff is quite noticeable even after the aircraft has left the runway, the really intense visibility reduction occurs along those early portions of the takeoff run in which the aircraft is still moving slowly but with maximum thrust maintained to achieve high acceleration. Under the latter conditions, the amount of carbon emitted per *unit length of flight path* is extremely high, so a wet takeoff leaves behind, all along the runway, a low dark layer of carbon smoke whose opacity tends to increase backwards towards the starting position. It is not uncommon, in takeoffs of six-jet B-47 aircraft, to have horizontal visual ranges reduced to as little as 200–300 ft within the smoke, a condition rendering hazardous the multiple takeoffs of the type occasionally used in military operations. In commercial jet operations, where only single takeoffs are involved, an operational hazard arises mainly from persistence of the smoke layer under conditions of stability and low winds such that subsequent landing operations may encounter lowered visibility.

In a detailed study of jet exhaust carried out under difficult and often hazardous conditions by the Los Angeles Air Pollution Control District, George and Burlin (1960) concluded that "the practice of water injection results in emissions of smoke in excess of legally allowable opacity limits but the duration of the emission is well within the allowable three-minute time limit." Hence no direct action has been taken in Los Angeles on this new pollution problem.

The writer's observations of wet-takeoff emissions have been made chiefly with respect to B-47 operations at Davis-Monthan AFB, Tucson, Ariz. Under typical mid-morning conditions in Tucson, a highly repetitive pattern unfolds: The existence of a shallow unstable layer, topped at about 400–500 ft above terrain by the remaining upper portions of the almost invariably present nocturnal inversion, leads to fairly rapid (order of a minute) clearing of the runway itself as the heated exhaust gases and contained carbon particles quickly rise convectively to the inversion base. The result of that first phase is the appearance, at the altitude of the inversion base, of a long dark smoke-patch of width and length comparable with those of the runway itself, but densest near the starting position. A further microclimatological feature typically present in the morning hours at Tucson, namely slow down-valley drift of air in the subinversion layer, then carries the dark patch northwestward across the city, eventually adding it to the typical morning pall of pollution observable down the Santa Cruz River Valley to the northwest of Tucson. A series of these dark patches are often arrayed in bead-like fashion from the air base to the northwest part of the city when a series of successive B-47 takeoffs have occurred during the morning hours. The writer had observed these peculiar patches over Tucson for several years before he established their origin in the above process by observing a number of them throughout their entire history, the typical drift-time being of the order of an hour before they become lost in the morning pall to the northwest of the city. Observations of these patches of water-injection smoke from light aircraft revealed that they are often extremely thin by the time they have drifted only a few miles from their source, having vertical thicknesses of only a few tens of feet in some cases.

#### 4. Mie cross-sections for carbon particles

In order to gain further insight into certain quantitative aspects of jet-exhaust opacity and to obtain cross-checks of an optical nature on the particulate sampling studies of George and Burlin (1960) of carbon emission rates for turbojet engines, this section will examine the absorbing and scattering properties of small carbon spheres, making use of the Mie theory.

Aggregation of numerous crystallites into nearly perfect spheres of diameter of the order of a number of hundreds of Angstroms was noted above in Section 2. Because of the characteristically low hydrogen-carbon ratio of turbojet soots, the difficulty cited by Millikan (1961) should not be serious here. Coagulation of the popcorn-ball carbon particles into filaments and flocculent masses will alter the effective Mie properties of the constituent particles in the sense that, for a given mass-loading of carbon in the exhaust, the attenuating effects must tend to *decrease* as such flocculation proceeds.

Hence, the following will tend to give an *upper limit* to the theoretical opacity for jet-exhaust emissions, a point to be borne in mind in comparing these results with direct observations (Section 6).

Ruedy (1941) undertook a limited number of Mie calculations some years ago, but lacked the advantage of modern computing techniques and consequently was forced to employ only rather poor approximations to the appropriate equations from the Mie theory (van de Hulst, 1957). Availability of IBM 650 Mie programs, developed by the present writer's colleagues for use in prediction of the radar scattering and absorbing properties of water drops and ice spheres, made possible accurate computation of the Mie cross-sections of carbon particles.

The optical data were taken from the work of Senftleben and Benedict (1918), who determined the complex index of refraction  $m^*$  of carbon for six wavelengths in the visible spectrum, where  $m^* = m(1 - ik)$ ,  $m$  being the ordinary refractive index and  $k$  the absorption term. Because there is only slight variation in either the real or the imaginary part of the index for carbon over the full spectrum, the writer used only the two extrema from their data, namely  $m^* = 1.90(1 - 0.36i)$  for wavelength  $0.436 \mu$ , and  $m^* = 2.00(1 - 0.33i)$  for  $0.623 \mu$ .

For each of these two wavelengths, values were computed for the Mie normalized cross-sections  $\sigma_t$ ,  $\sigma_a$ , and  $\sigma_s$ , where  $\sigma_t = \sigma_a + \sigma_s$  and the respective subscripts denote total, absorption, and scattering. They were computed for selected values of the Mie parameter  $\alpha = 2\pi a/\lambda$  up to 8, where  $a$  is the radius of the particle and  $\lambda$  is the wavelength in question. For visibility analyses, one is chiefly interested in the *total* attenuation cross-section  $\sigma_t$ , defined as the quotient of the effective total attenuation cross-section divided by the actual or geometric cross-section  $\pi a^2$ . Table 1 summarizes the computer re-

TABLE 1. Mie total attenuation cross-sections  $\sigma_t$  for carbon particles of radius  $a$  at two wavelengths  $\lambda$ .

$\lambda(\mu)$	$\alpha = 2\pi a/\lambda$								
	0.2	0.4	0.6	0.8	1.0	1.5	2.0	4.0	8.0
0.436	0.20	0.46	0.86	1.45	2.09	2.82	3.00	2.68	2.46
0.623	0.18	0.42	0.82	1.44	2.17	2.94	3.09	2.68	2.46

sults for  $\sigma_t$ . It will be noted that  $\sigma_t$  exhibits only very slight spectral variation from the deep violet ( $\lambda = 0.436\mu$ ) out to the orange red ( $\lambda = 0.623\mu$ ), justifying use of only the extrema of the Senftleben and Benedict (1918) indices.

To display the nature of particle-size variations in all three of the Mie cross-sections, as well as to suggest the relative contributions of Mie absorption and Mie scattering at small  $\alpha$ , Fig. 2 has been plotted from the calculated values for just the wavelength  $\lambda = 0.623 \mu$ . Note especially that, for very small  $\alpha$ ,  $\sigma_t$  is due almost wholly

to *absorption*, but for large  $\alpha$  scattering is about equal in importance with absorption in producing total beam attenuation in a cloud of carbon particles.

By use of  $\sigma_t$  data for both wavelengths, and interpolation to infer the small spectral variation at intermediate wavelengths, the curves of Fig. 3 were constructed to reveal both the wavelength and particle-size dependence of the total attenuation cross-section for carbon spheres. Most carbon particles emitted by

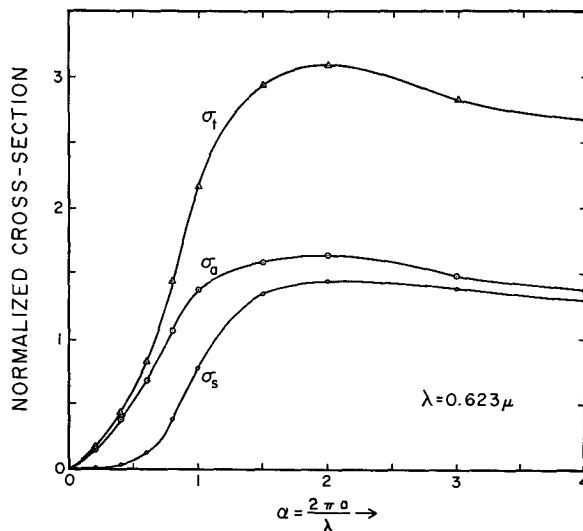


FIG. 2. Mie cross-sections for carbon spheres at a wavelength of 0.623 microns.

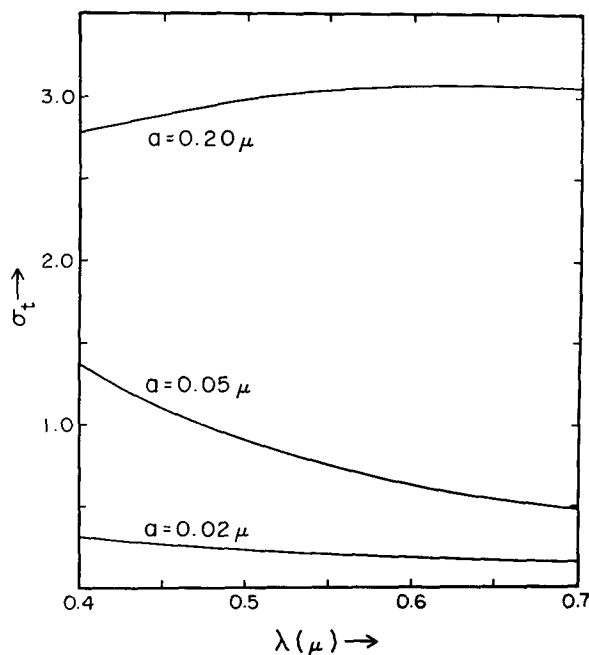


FIG. 3. Visual-region wavelength dependence of the Mie total attenuation cross-section for carbon spheres of three radii  $a$ .

jet engines will be of size corresponding to the lower two curves. The upper one, for  $a = 0.2 \mu$ , is included to show the maximal attenuation that occurs for particles with radius of about a few tenths of a micron ( $\alpha$  near 2-3 in the visible). One must, of course, keep well in mind the fact that particles of the latter size have very much greater masses than those of the lower curves. It would, for example, require densely-packed coagulation of some  $10^3$  of the particles corresponding to the lowest curve to yield one particle of the size corresponding to the upper curve. The ratio of the geometric cross-sections of those two sizes is 100, and the ratio of their  $\sigma_t$  happens to average about 15-fold across the full spectrum. Hence, for a given *total* mass of carbon, attenuation is about 1.5 to 2 times more intense if one has 1000 particles of  $a = 0.02 \mu$  densely packed into a single sphere of radius  $0.2 \mu$ .

### 5. Analysis of jet-exhaust opacity

In this section fuel consumption data will be combined with the George and Burlin (1960) exhaust-sampling measurements of carbon loading and with the above Mie attenuation data to estimate runway opacities. Comparison of the latter with observations of visibility reduction will serve to cross-check the reported carbon loadings. For these purposes the operating characteristics of the J-57 engine (identified in commercial version as the Pratt and Whitney JT3) will be assumed. This engine powers the B-47 and B-52 bombers, the KC-135 tanker, and a number of smaller Air Force aircraft such as the F-100, F-101, F-102, as well as the Navy's F4D, A3D, and F8U-1. In addition, the JT3 version of the same engine is used in the DC-8 and certain of the 707 air transports.

Data (American Airlines, 1958) on this engine indicate that its nominal thrust of 10,000 lb can be increased by use of water injection to about 13,000 lb with a corresponding specific fuel consumption of 0.95 lb per hr per lb thrust, whence it consumes fuel at a rate of about 12,300 lb per hr = 3.4 lb per sec in wet takeoff. Four-jet aircraft such as the two cited transports are thus burning fuel at a rate of 13.6 lb per sec. By contrast, in cruise at 85 per cent of maximum continuous power and about 8000 lb thrust per engine and specific fuel consumption of 0.77 lb per hr per lb, the four-jet aircraft would use fuel at a rate of only about 6.9 lb per sec. Thus wet takeoff entails roughly *twice* the fuel consumption rate of cruising, yet delivers only about 30 per cent more thrust, an indication of the reduced efficiency that is the price paid for the short period of augmented thrust and that thereby leads to excessive carbon emission over the runway.

In the Los Angeles Air Pollution Control District's measurements, George and Burlin (1960) arrived at a value of 27 lb of *total* particulates per ton of fuel for an engine using water injection at a pressure ratio of 2.74,

which corresponds closely to wet takeoff conditions. (By contrast, minimum particulate emission occurred with engine settings corresponding to landing-approach conditions, the rate then being only 15 lb per ton.) The particulates were collected in a train consisting of three modified Greenberg-Smith impingers followed by a Whatman filter thimble. The first two impingers were operated with the orifices under water, the third was dry. In the collections made under simulated wet takeoff, 49 per cent of the measured particulates were trapped in the impinger waterbaths, the remaining 51 per cent was caught in the thimbles. Of the first of those two components, the fraction that was neither water soluble nor soluble in methyl chloroform constituted 10 per cent, referred to total particulates, and this may be regarded as comprising the carbon aggregates large enough to be trapped in the impingement process. No differentiation on solubility grounds is reported for the 51 per cent trapped in the thimbles; but it was completely combustible, so the possibility that it was all carbon cannot be rejected here. Hence a *maximum* of 61 per cent of all *collected* particulates may have been carbonaceous.

To obtain an *upper* limit to carbon opacity, we may proceed to assume that 61 per cent of the 27 lb per ton of fuel was carbon, whence a four-jet wet takeoff might involve carbon emission at as large a rate as 0.11 lb per sec. Since final takeoff speed for aircraft of the type here considered is usually in excess of 150 mph (e.g., 185 mph for the Boeing 707), we will be considering conditions representative of about the *middle* of the takeoff run if we assume a speed of 100 mph. For an aircraft of 130 ft wing-span (Boeing 707) we may reasonably assume the carbon smoke to be spread through a wake region whose section normal to the takeoff path is of the order of 150 ft wide, and observations suggest an initial smoke-filled height of perhaps 30 ft. Hence each second's carbon output,  $0.11 \text{ lb} = 50 \text{ gm}$ , is dispersed throughout a slab of volume of some  $2 \times 10^{10} \text{ cm}^3$ , whence the carbon loading in the exhaust prism is about  $2 \times 10^{-9} \text{ gm cm}^{-3}$  near mid-runway, if we accept George and Burlin's results.

From this last estimate plus the Mie coefficients for total attenuation from Section 4, we may now examine the visual range in the exhaust wake. Relevant data are presented in Table 2. We continue to assume that we deal only with compact carbon spheres, and take their density as about  $2 \text{ gm cm}^{-3}$ . For three particle radii ranging from rather smaller than the largest reported jet-exhaust carbon particles ( $a \approx 0.02 \mu$ ) up to radii about as large as are produced under controlled conditions in carbon black manufacture ( $a \approx 0.20 \mu$ ), the corresponding number densities  $n$  are shown for the above-described exhaust region for a carbon loading of  $2 \times 10^{-9} \text{ gm cm}^{-3}$ . Next is shown the corresponding aggregate *geometric* cross-section  $n\pi a^2$  per unit volume on the simple assumption of a monodispersed smoke

of the given particle size. Such a total cross-section is least, of course, for the largest particles with fixed loading. If all three particle sizes had Mie normalized attenuation cross-sections of unity the  $n\pi a^2$  values would represent the effective attenuation coefficient in the exhaust prism. But  $\sigma_t$  varies about 15-fold (Fig. 3) in the size range in question, so we introduce values of  $\sigma_t$ , taking for estimation purposes a wavelength of  $0.5 \mu$  in Fig. 3, and compute the actual attenuation (scattering plus absorption) total cross-sections (i.e., extinction coefficients)  $b \equiv n\pi a^2 \sigma_t$ , shown in the next-to-last column.

The in-smoke visibilities  $V$  corresponding to these values of  $b$  can be estimated from the Koschmieder visual range formula

$$V = b^{-1} \ln(\epsilon^{-1}),$$

where  $\epsilon$  is the visual brightness-contrast threshold. For midday conditions an appropriate value of  $\epsilon$  is about 0.003, whence  $V = 6/b$ , approximately. In the last column of Table 2,  $V$  has been computed using the latter

TABLE 2. Exhaust-region visual range estimates.

$a$ ( $\mu$ )	$n$ ( $\text{cm}^{-3}$ )	$n\pi a^2$ ( $\text{cm}^{-1}$ )	$\sigma_t$ ( $\lambda = 0.5 \mu$ )	$b$ ( $\text{cm}^{-1}$ )	$V$ (ft)
0.02	$4 \times 10^7$	$5 \times 10^{-4}$	0.25	$1 \times 10^{-4}$	$2 \times 10^3$
0.05	$2 \times 10^6$	$2 \times 10^{-4}$	0.90	$2 \times 10^{-4}$	$1 \times 10^3$
0.20	$4 \times 10^4$	$5 \times 10^{-6}$	3.0	$2 \times 10^{-4}$	$1 \times 10^3$

relation, the results being expressed in feet. It will be seen that the visual ranges vary only by a factor of two despite the ten-fold range of carbon particle size, and that even the lowest of these is considerably larger than the writer's estimate of visual ranges of 200–300 ft in exhaust smokes for six-jet B-47's. Adjusting the latter to match a four-jet commercial transport, one might expect visibilities of about 300–400 ft.

## 6. Discussion

Although the discrepancy between the lowest estimated visibility of Table 2 ( $10^3$  ft) and observational estimates of about 300–400 ft is small enough that one may say that the marked opacity of jet exhausts on wet takeoff is tolerably well explained by the combination of the particulate counts and the Mie extinction values, it is desirable to ask where in the present analysis one might suspect error.

First it should be pointed out that the writer's estimates of visual range of the order of 300–400 ft for four-jet transports are not likely to be too low. In multiple-B-47 takeoffs, pilots are required to carry out their takeoff runs with only about 150 ft between successive aircraft and pilots report that it is ordinarily not possible to see the aircraft immediately ahead because of obscuration by the dark exhaust. Although the fuel

consumption and power data used in Section 5 pertain to commercial air transport practice, the type of engines and the approximate takeoff speeds are closely similar to those of the B-47. Hence, with allowance for number of engines, a visual range of 400 ft is probably an upper limit here, whereas assumptions made in the preceding section tend to give a *lower* theoretical limit to  $V$ .

Although some variations in the visual contrast threshold might raise our estimates of  $V$ , the writer's observations refer to midday conditions where the frequently used "standard threshold" of 0.02 is definitely too large. (However, even if one did use  $\epsilon = 0.02$ , the logarithmic dependence on  $\epsilon$  is such that  $V$  would only be reduced by about one-third, i.e., to no less than 600–700 ft.) Nor would any adjustment for the role of absorption (as contrasted with scattering) help to account for the present discrepancy between predicted and observed visual ranges. In the case of carbon-particle extinction for small particle sizes, absorption exceeds scattering, yet any allowance for this will only tend to *exaggerate* the present discrepancy, inasmuch as absorption tends, with visual targets of non-zero intrinsic brightness, to *increase*, not *decrease* contrast. Thus, although a precise analysis would deal separately with absorption and scattering, the results would not alter the sense of the present conclusions.

The question of particle *form* demands further comment. The Mie data pertain only to spheres, which make them directly applicable to the "carbon particles" as here defined, but the studies of Schalla *et al.* (1954) indicate that agglomeration of the carbon particles into filaments and matting of the latter into flocculent soot particles occurs in turbojet exhausts as in other carbon-releasing processes. They give no information as to how close to the exhaust ports of the turbojet engine their electron photomicrography samples were taken, which is an important point inasmuch as collection well aft of the exhaust would afford increased time for flocculation. Since George and Burlin (1960) collected within an inch or so aft of the noise-suppressor tailpipes, minimal flocculation time was available in their case. The latter fact would tend to reduce over-all collection efficiency as will be elaborated below. Here, it need only be noted that *if* the bulk of the carbon flocculates by a few plane-lengths aft of the engine then the effective attenuation coefficient  $b$  for George and Burlin's (1960) carbon loading would be considerably *lower* than the values of order  $10^{-4} \text{ cm}^{-1}$  appearing in Table 2. This follows incontrovertibly from the general rule that once aggregation has proceeded past particle sizes such that their Mie parameter  $\alpha$  is larger than that corresponding to the peak of the  $\sigma_t$ -curve in question, continued aggregation, with fixed mass loading, can only *reduce* the over-all attenuation. Thus to suggest that *soot particles* predominate over *carbon particles* in the darkest portions of jet exhausts would only exaggerate the discrepancy between pres-

ent optical estimates of opacity and the carbon loadings found by George and Burlin. (It may be noted also that the same trend would result from admitting that some appreciable fraction of the particulates trapped in the filter thimbles downstream of the impingers might have been combustible material other than carbon.)

Thus the theoretical visual ranges inferred in Table 2 are likely to be *lower* than, but are surely not higher than, the visual ranges that would occur with an overall carbon loading of  $2 \times 10^{-9}$  gm cm<sup>-3</sup> as found in the Los Angeles work. Hence one must ask whether George and Burlin's measurements might have led to *underestimates* of the carbon emissions of the turbojet engines they studied. Reasons for regarding this as likely will now be given.

The Greenberg-Smith impingers used by George and Burlin had modified slits in the first two impingers of each train. The standard 0.082-inch circular orifice was replaced by a stainless steel 0.045-inch by 0.6-inch slit-style orifice, and was operated at the somewhat higher-than-normal pressure-drop of 3.5 inches Hg. From this information one may calculate, using the equation for subcritical flow through an orifice (Prandtl, 1952), that the impingement speed was about 150 m/sec. Impaction efficiency factors due to Ranz and Wong (see Green and Lane, 1957) may be used to find the particle-size cutoff for impingement in these modified devices. The writer's computations indicate zero collection for particles of diameter of  $0.3 \mu$  or less, 25 per cent at a diameter of  $0.4 \mu$ , and 70 per cent at  $0.5 \mu$ . Clearly, the deposits found in the Greenberg-Smith impingers could not be due to what are here termed carbon particles (diameters below  $0.05 \mu$ ). Rather, such dark deposits must have represented collections of those *soot particles* that had already agglomerated from the carbon particles in the short time required for the combustion products to pass from combustor to tailpipe.

Materials trapped in the Whatman filter thimbles that followed the above impingers accounted for about half of the total particulates caught. It was noted that an uncommon degree of penetration of these thimbles was occurring, as revealed by a black deposit on the thimble holders themselves. A collection cutoff near a diameter of 0.1 micron was thought to be achieved with these thimbles [communication from A. P. Fudurich, Los Angeles APCD]. However, this is also above the diameter limit of  $0.05 \mu$  for the carbon particles here considered, so the latter could, apparently, have passed in quantity through the collection train employed by George and Burlin (1960).

The crucial question then becomes the following: Could a fairly large fraction of the carbon emitted in wet takeoff still be, at moment of passing out the tailpipe (George and Burlin's sampling point), only in the early stages of aggregating from carbon particles to soot particles? In the usual coagulation equation

$$dn/dt = -Kn^2,$$

where  $n$  is particle concentration and  $K$  is the coagulation constant, the value of  $K$  appropriate to the high temperature of the jet exhaust (725K) is about  $2 \times 10^{-9}$  cm<sup>3</sup>/sec for  $a = 0.02 \mu$  (near the maximum for carbon particles as found by Schalla *et al.*). From Table 2 we note that  $n = 4 \times 10^7$  cm<sup>-3</sup> for  $a = 0.02 \mu$  if we assume the George-Burlin carbon loading figure; whence  $-dn/dt$  is about  $0.3 \times 10^7$  cm<sup>-3</sup>sec<sup>-1</sup>. Thus something like 6 sec is the half-life against coagulation for such particles present in that concentration. This is so long compared to the transit-time (order of milliseconds) for passage of the combustion products from combustor to tailpipe that predominantly *uncoagulated* particles must be expected. Of course, we actually need here a carbon particle concentration of about six times that just assumed, since Table 2 shows a visual range for  $a = 0.02 \mu$  some six times the 300-ft visual estimate cited earlier. Use of a six-fold higher  $n$  lowers the half-life by a factor of 36, i.e., to about a *sixth of a second*, but even this is long compared to in-engine transit times with exhaust speeds of order 3000 ft sec<sup>-1</sup> in wet takeoff; so we must conclude that *a large fraction of all emitted carbon leaves a turbojet tailpipe still in the form of carbon particles of only a few hundred Angstrom units diameter*. That is, soot formation takes place principally in the wake, aft of the engine, and collections of particulates made *at* the tailpipe will tend to underestimate total carbon unless collection efficiency is high down to a few hundred Angstroms, a difficult goal to achieve.

One might object that the above coagulation rate, based on an equation applicable only to coagulation by Brownian motion, may underestimate actual coagulation under the intense stirring within the exhaust of a jet engine. The Smoluchowski equation for the contribution to coagulation made by turbulence (Green and Lane, 1957) is

$$-dn/dt = (32/3)a^3n^2S,$$

where  $S$  is the velocity shear generating the turbulence field. With jet velocities of order 3000 ft sec<sup>-1</sup> and jet diameters of order 1 ft,  $S \doteq 3 \times 10^3$  sec<sup>-1</sup>, so for carbon particles of radius  $a = 0.02 \mu$ , the coagulation constant which multiplies  $n^2$  on the right side of the Smoluchowski equation becomes about  $4 \times 10^{-12}$  cm<sup>3</sup> sec<sup>-1</sup>, which is about *two orders of magnitude smaller* than  $K$  in Brownian coagulation. The physical reason for the slight effect of turbulence here is the small particle radius  $a$ . If  $a$  were, say,  $0.2 \mu$ , turbulent coagulation would proceed about ten times faster than Brownian coagulation in presence of shears of magnitude typical of jet exhausts; but for the carbon particles of concern here, turbulent stirring is negligible compared to Brownian effects.

## 7. Conclusion

Emission of free carbon particles with diameters of the order of a few hundred Angstrom units accounts for

the dense black smoke emitted in the exhaust of turbojet engines during takeoffs in which water injection is employed to augment thrust. Measurements made by the Los Angeles Air Pollution Control District, when combined with operating data for multijet wet takeoffs, imply total carbon loading in the wake of about  $2 \times 10^{-9}$  gm cm<sup>-3</sup>. Using extinction coefficients calculated for carbon particles using Mie theory, and assuming (in order to maximize *computed* opacity) all of the carbon to be in form of spheres of radius  $0.02 \mu$ , that value of carbon loading implies a visual range not less than about 2000 ft, whereas direct observation plus pilot reports indicates in-smoke visual ranges no greater than about 400 ft near mid-runway for the case of four jet transports powered by engines employing water-injection to augment takeoff thrust.

From consideration of the collection techniques employed by George and Burlin (1960) in the Los Angeles studies, it seems very likely that most of the carbon particles with diameters under  $0.05 \mu$  must have escaped collection, tending to make their estimates of carbon loading too low. Considerations of coagulation rates plus physics and chemistry of carbon-particle formation make it, in fact, plausible to assume that the bulk of the free carbon leaving a turbojet engine is found in particles of diameter below  $0.05 \mu$ .

If, as these arguments seem to indicate, the discrepancy between observed visual ranges and visual ranges computed from Mie theory and the George-Burlin carbon loading figure results from the latter being unduly low, we must conclude that actual carbon emissions may run as much as five times higher than indicated by the Los Angeles studies. That is, in wet takeoff, carbon emission may approach 80 to 90 lb per ton of fuel consumed. This would still leave engine fuel-use efficiency at better than 95 per cent level, so is by no means ruled out on that ground. Air pollution implications of such an emission would be rather more serious than has been assumed in the past, so it appears to be important, for aviation operational reasons as well as for urban pollution reasons, to secure further experimental data and observational data on the form and amounts of carbon particulates released by turbojet engines in wet takeoff. Although certain new engine types (e.g., Pratt-Whitney J-75 turbofan) will not require water injection to achieve *currently* required takeoff thrusts, large numbers of operating aircraft will continue to contribute to this problem. And, since emission of carbon is not inconsequential even in absence of water injection, rising jet traffic may render this a serious problem at busy terminals in the near future. If, as has been concluded

here, the total carbon emission rates for wet takeoff were substantially underestimated in the sampling techniques employed in the Los Angeles study, then so also were the emissions under other engine operating conditions. Even under landing-approach conditions particulate emission as measured by George and Burlin (1960) ran 15 lb per ton of fuel; so the correction factor of about five inferred here would raise total carbon emissions at an active air terminal to levels that deserve more serious consideration from the viewpoint of air pollution and air safety.

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