Some Physical Relationships of Fine Particle Smoke

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THE smoke rising from a grass, brush or forest fire is primarily formed by the condensation of moisture and other vapors produced through pyrolysis and combustion. This smoke formation depends on the rate at which the surrounding air moves into the fire to form particles from the rich source of gaseous vapor which the fire generates.

Measurement of the size of such particles and the other physical, chemical and electrical properties which they possess during their existence as airborne particles, is an extremely complicated problem. Field observations show that a vigorously burning fire produces an extremely high concentration of very fine particles. Most of them are smaller than a half micron in diameter, quite a few are invisible. The visible smoke often appears blue by scattered light and yellow by transmission.

When a fire occurs in an area having rich fuel supplies but not enough air for complete combustion, it produces surges of black carbonaceous smoke. Such smoke has a wide variety of particle sizes, ranging from the submicroscopic to ash particles which are millimeters or even centimeters in cross section. On the other hand, when green leaves, needles and small branches burn slowly, the dense smoke which develops has a yellow or white appearance. Since

this type of fire evaporates a considerable amount of water, this type of smoke is made up of the largest liquid particles formed during the pyrolytic or incomplete combustion process characterizing such fires. The size of such watery particles depends on the humidity of the air, the nature of the fuel and the amount of water condensed, thus their size is variable and unstable.

The formation of smoke particles from the condensation of hot gases produced from low vapor pressure substances involves a growth process in which the primary particles are rarely larger than one micron.

EARLY EXPERIENCE WITH FIRE PARTICLE SMOKES

During the early phases of World War II, I developed a smoke generator for screening personnel and facilities from enemy surveillance in Europe, Africa and the South Pacific. This artificial fog generator[1] produced extremely uniform fine particles having a diameter of 0.6 micron.

To produce such particles, I passed diesel lubricating oil through a flash boiler, subjecting it to a temperature in excess of 450°C. The oil vaporized to produce a boiler pressure of 10 pounds per square inch. This hot oil vapor was then permitted to escape at sonic velocity through a small orifice into the ambient air. This high speed jet of vapor entrained great quantities of the surrounding colder air, which then quenched the hot supersaturated gas and diluted the particles thus formed within a few milliseconds.

This device produced a nearly monodispersed liquid droplet fog with a diameter of 0.6 micron. Our first generator[2] vaporized 10 gallons of oil per hour (11 grams per sec.), thus producing 1×10^{14} particles per second. This formed a highly opaque white fog. Later units utilized 50 to 100 gallons an hour and produced a fantastic amount of smoke which looked like a natural fog. During these experimental studies more than 30 years ago, we tried to make larger particles by controlling the condensation rate of the vapor. Although we had ideal conditions for growing such particles, we were not able to make them much larger than one micron in

diameter despite many attempts.[3] Our theoretical as well as field studies showed that the particle size which was the most effective for screening purposes was 0.6 micron (the middle of the visible spectrum).

Under the optimum operating conditions of the generator, we produced a particle concentration greater than 1×10^{10} particles per cc at the completion of the condensation process. Fortunately the rate of dilution was so great that the concentration of the screening smoke particles was reduced to less than 100,000 per cc within a few meters of the jet orifice.

Somewhat similar relationships probably exist in a forest fire except that the entrainment velocities, amount of supersaturation and vapor concentrations are considerably lower. The gas and particle formation reaction is, however, much more complex.

THE PRESENT MEASUREMENT OF SMOKE FROM FOREST FIRES

In reviewing the current literature[4] concerning the various aspects of the smoke from grass, brush and forest fires, I have noted that most of the quantitative data is based on sampling with an E.P.A.-type high volume sampler.

In view of the observable features of such fires, these measurements could be misleading if used without information about the concentration of the small particles. The high volume sampler is a device which depends on measuring the weight of the particles of an aerosol. Thus it reflects the presence of ash from intense burning and other large particles resulting from incomplete combustion. The high volume sampler gives virtually no information on the *number* of particles that are smaller than one micron. Thus it is essential that the relative concentration and size spectrum of the fine particles be obtained by other methods of the smoke particle size spectrum produced in the various types of fires utilized by fire ecologists. It is also of critical importance to establish similar particle number and size spectrums in wildfires of both the fast moving as well as the giant convective column types. Until this is done, it will

not be meaningful to evaluate the relative air pollution caused by various prescribed burning techniques as compared to the effluent of wildfires.

THE VARIATIONS IN SMOKE PRODUCED BY GRASS, BRUSH AND FOREST FIRES

Komarek has correctly pointed out[5] that the major problem posed by forest fire smoke is the reduction in visibility. Since, as mentioned earlier, the most effective particle size for reducing daytime visibility is a 0.6 micron spherical particle, it seems obvious that the most effective smoke pollution control strategy for such fires should utilize techniques that produce particles either smaller than 0.3 micron or larger than 1 micron. In view of the present techniques that seem to be feasible in conducting annual or 2-year interval burns in areas free of underbrush, it is desirable, if at all possible, to make particles larger than one micron since gravity would then accelerate their removal from the atmosphere. This would thus utilize the coldest, slowest, wettest fire that it is possible to burn.

On the other hand, when the fuel loads are high, such as in saw grass, chapparal, heavy undergrowth and the burning of lumbering slash, it would probably be best to plan fires so they burned rapidly, efficiently, hot and with adequate air entrainment so that the smoke is not black and the particles are considerably smaller than 0.3 micron. Such smoke would appear bluish and at times would be invisible except for the heat waves produced.

Since most of such fires are burned in areas where the ambient air is not polluted, the atmosphere can probably accommodate a fairly high concentration of these invisible or blue haze particles without reaching concentrations where coagulation would lead to a larger particle size. While such particles are not eliminated from the atmosphere by gravity, they are effectively removed by cloud droplets and precipitation. The larger particles serve as cloud condensation nuclei while the smaller ones diffuse to the cloud droplets due to their high Brownian motion.

More work should be directed toward evaluating the properties of black smoke, both in relation to visibility reduction, health effects

and the possibility that such particles may be good scavengers for gaseous constituents in the atmosphere. Little is known about their structure and other properties, except that they are very different than liquid droplet particulates.

These relationships of physical and chemical reactions of smokes emphasize the importance of making quantitative surveys of the submicron particle concentrations produced by various types of fires. This cannot be done with the high volume sampler, although measurements with it are useful, especially with fires that generate large quantities of ash.

It is worth noting that a single 100 micron particle (0.01 cm) is equivalent in weight to 5 million 0.6 micron particles, or a billion having a diameter of 0.1 micron!

DEVICES FOR MEASURING FINE PARTICLES

The measurement of fine particles is extremely easy to accomplish. The Gardner Small Particle Detector* or the Environment/One Fine Particle Detector** are highly portable, instant sampling instruments which quantitatively measure the concentration of fine particles.

About 5 years ago we conducted 10 low level transcontinental survey flights with such instruments and presently are operating a global surface network of fine particle monitoring stations utilizing them.

OPERATING PRINCIPLE OF FINE PARTICLE DETECTOR

These devices operate on the principle of the Wilson Cloud Chamber. The air to be measured is drawn into a humidified chamber and a vacuum of 20 inches of mercury is produced in an adjoining chamber. The moistened air sample containing the particles is then suddenly expanded into the vacuum. The high degree of supersaturation with respect to water, which is produced by the sudden

* Gardner Associates, 3941 Carman Rd., Schenectady, N.Y.

** Environment/One Corporation, 2773 Balltown Rd., Schenectady, N.Y.

adiabatic expansion into the vacuum, causes all airborne particles larger than 0.003 micron to accept water vapor to form a cloud. The larger the number of particles in the sample air, the denser the fog that forms.

The number of particles in the sampled air is determined by measuring the amount of light transmitted through the fog which forms, using a stabilized light source and a photocell. A logarithmic scale on an instrument dial provides values ranging from 65 to more than a million particles per cubic centimeter. Such a measurement can be initiated and completed in less than 15 seconds. The instrument is calibrated based on a device which in turn was calibrated with an Aitken counter.

THE PATTERN OF CONCENTRATIONS OF FINE PARTICLES IN GLOBAL AIR

With such an instrument we have made thousands of measurements[6] throughout the world. Austin Hogan, of our Research Center, has developed[7] the concept of an "aerosol climatology." Our surveys have enabled us to assign various concentrations of fine particles to the air pollution classification shown in Table 1. Some of my personal observations are summarized in Figure 1.

TABLE 1. FINE PARTICLE CLASSIFICATIONS

Concentration	Cloud Cond. Nuclei (CCN) n/cc > 0.1 micron	Condensation Nuclei (CN) n/cc > 0.003 micron
Very Low	50– 119	50– 299
Low	120– 319	300– 999
Light	320– 679	1,000– 4,999
Moderate	680–1,999	5,000– 49,999
High	2,000–7,499	50,000–149,999
Very High	> 7,500	> 150,000

During the course of these field observations with the Gardner Small Particle Detector, I discovered that by using 2″ of mercury vacuum (1 division on the vacuum dial) instead of 20″ of vacuum as is used for measuring condensation (Aitken) nuclei, the concentra-

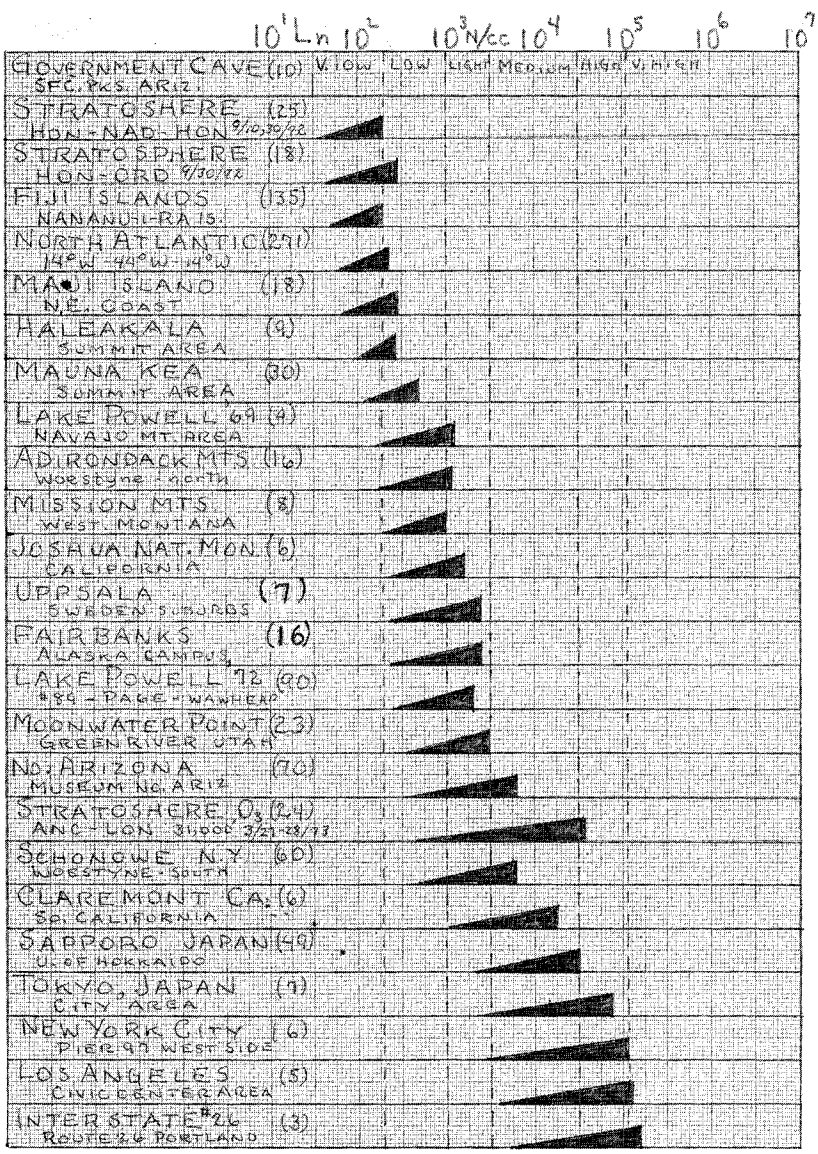

FIG. 1. Levels of large and small atmospheric particles in various locations in the world.

tion of particles per cubic centimeter indicated by the instrument provided a reasonably accurate indication of the number of "large" particles in the air sample. These are the particles which serve as nuclei for the formation of cloud droplets in the natural atmosphere. My observations of these large particles show a consistent relation to the "purity" of the air, ranging from less than 65/cc in extremely clean oceanic air, to concentrations ranging from 3000 to 8000/cc in the badly polluted air of cities.

In the bluish haze produced by a nearby forest fire causing a noticeable smell of wood smoke in the air, the concentration of fine particles is comparable to that found in air classified in the "moderate pollution" category such as is typical of much of the suburban air of the United States. Figure 2 indicates the levels of such "large" and "small" fine particles which I have measured during a number of occasions when I have encountered the smoke of forest and grass fires. For comparison, I have included values for the smoke particles produced in a campfire in the open woods and the smoke in a rock shelter of the sort probably used by our cavern dwelling ancestors. Other interesting sources are also included in the table.

These are the particulate levels of "natural" aerosols to which man's physiological development has been accommodated over several millions of years.

The air pollution problem of most urban or industrialized areas is of a very different sort. Although the particle concentrations may be comparable, the make-up of the particulates is not similar. Such substances as lead, mercury, asbestos, coal and quartz dust, cadmium, DDT, the nitrogen oxides, sulfuric acid, unburned hydrocarbons and the "witches smoke" of synergised particulates produced by exotic chemicals, provide insults to our respiratory system to which we have not had the time for adaptation.

THE DETECTION OF OZONE AND NITROGEN DIOXIDE IN THE ATMOSPHERE

Several years ago I discovered[8] that the Gardner Counter could also be used as an extremely effective and highly sensitive ozone

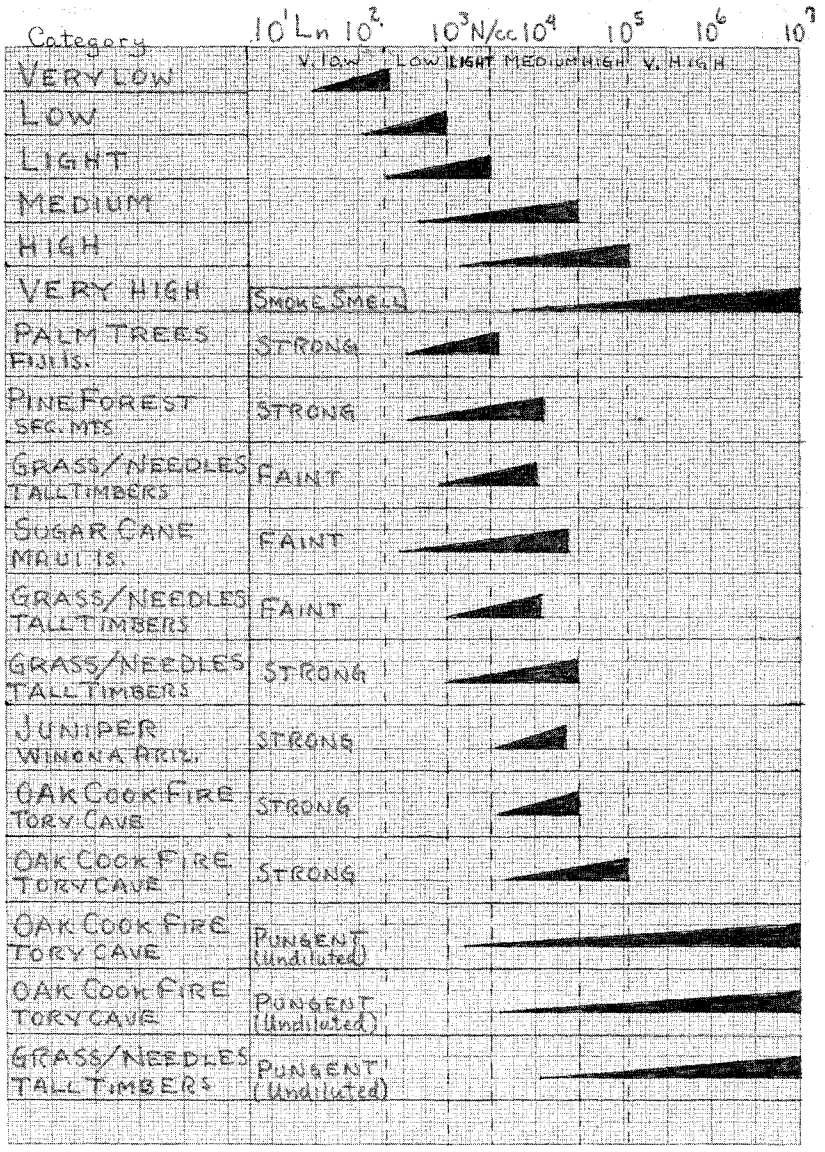

FIG. 2. Concentration of large and small atmospheric particles from various types of fire.

detector. At that time I had not established that the reaction with saturated iodine vapor or a cyclic olefin such as β-Pinene (Gum Turpentine is a good source) was specific for producing gas to particle reaction with ozone.

Since then I have established that when a sample of air containing ozone in the concentration of a few parts per billion is suddenly introduced into a 50 liter plastic bag containing several crystals of iodine or a few drops of turpentine, a tremendous number of particles are suddenly produced in the air containing the iodine or terpene vapor. A measurement of the concentration of particles thus formed can be interpreted in terms of the amount of ozone in the air. Figure 3 shows this relationship.

In my early experiments conducted in the extremely clean air of the North Atlantic, the deserts of the Southwest and the snow country of northern Japan, I found that identical values of the concentration of particles would be formed using either the iodine or turpentine reaction.

Subsequently in less clean air environments having larger numbers of nuclei as a background, a striking difference was observed between the iodine and turpentine reactions. Within the past year I have discovered that a very small amount of nitrogen dioxide (of the order of a part per billion) will greatly inhibit or entirely prevent the increase in particle production in air that contains ozone. Thus, by using this extremely simple portable, inexpensive and highly sensitive instrument, it is extremely easy to detect the presence of ozone as well as nitrogen dioxide in the parts per billion range.

Most of my field measurements up to the present time indicate that nitrogen dioxide occurs in air that has been modified by man's activities. The presence of nitrogen dioxide seems to relate to the effluent of automobiles and other such sources of hot flames that oxidize the nitrogen in the air.

Whether the forest fire is also a generator of this noxious gas is thus far uncertain based on my field studies. This is an area that I now intend to investigate in considerable detail.

The rise of industrial technology has been so rapid that our physiological adaptation to these new forms of atmospheric pollu-

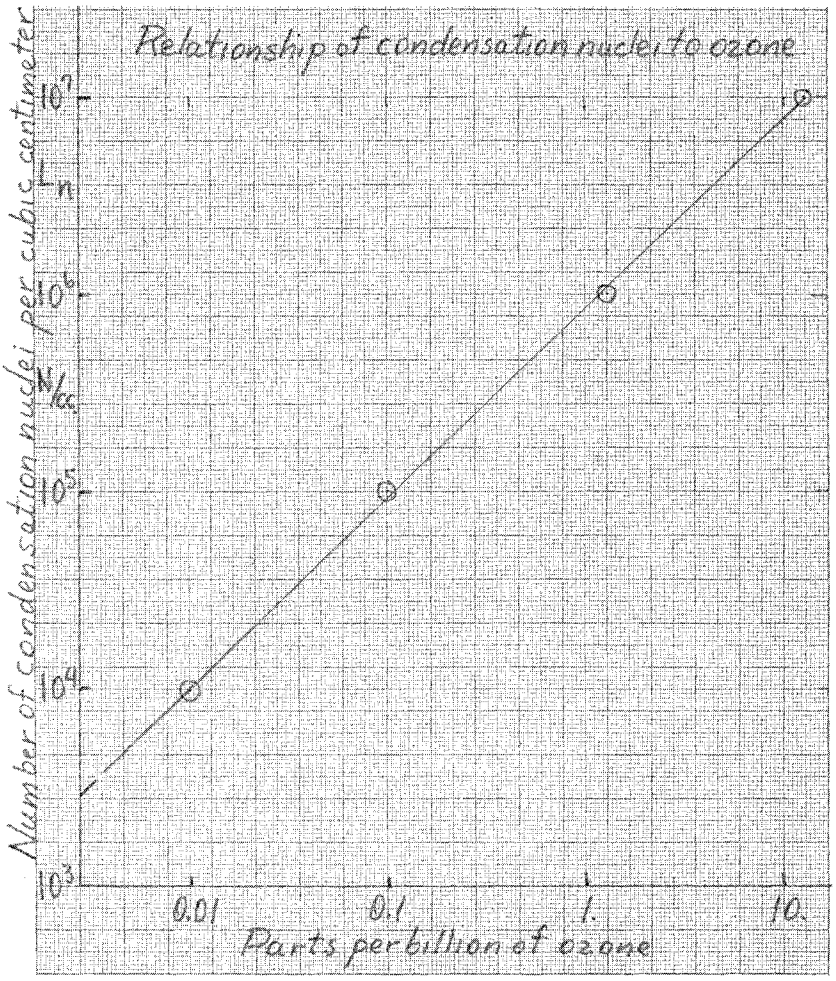

FIG. 3. Relationship of condensation nuclei to ozone.

tion has apparently not been possible. This is indicated by the rapid increase in respiratory ailments of people living in the vicinity of urban settlements. Even those living outside of the city are not free of these problems, since the air encountered along heavily traveled

highways contains some of the highest pollution levels that can be found in the free atmosphere.

The proper use of fire in the environment properly attuned to certain levels of tolerance, coupled with an adequate understanding of the physical, chemical and electrical reactions that might be utilized in adjusting practice to result, offers challenging avenues to the eco-pyroloist.

I hope they will rise to their opportunities.

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