Aerosol Science and Technology: History and Reviews

Edited by David S. Ensor

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About the Cover

The cover depicts an important episode in aerosol history—the Pasadena experiment and ACHEX. It includes a photograph of three of the key organizers and an illustration of a major concept of atmospheric aerosol particle size distribution. The photograph is from Chapter 8, Figure 1. The front row shows Kenneth Whitby, George Hidy, Sheldon Friedlander, and Peter Mueller; the back row shows Dale Lundgren and Josef Pich. The background figure is from Chapter 9, Figure 13, illustrating the trimodal atmospheric aerosol volume size distribution. This concept has been the basis of atmospheric aerosol research and regulation since the late 1970s.

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CHAPTER 11

Aerosol Wars

A Short History of Defensive and Offensive Military Applications, Advances, and Challenges

Arthur K. Stuempfle

Introduction

The generation of particulate and liquid aerosols is an inevitable result of military operations during both peacetime and wartime. Matter can become airborne through the inadvertent creation of particulates by re-aerosolizing dust and dirt particles such as that caused by a helicopter downwash, the residue of propellant smokes or explosive bursts from artillery munitions, or any number of other means. Even more ominous is the deliberate or unintentional dispersion in aerosol form of liquid or solid chemical compounds, biological agents, or radioactive materials.

Military aerosols of all types are integral elements in every aspect of chemical warfare defense and can pose a serious threat to Soldiers and civilians during military operations. Aerosolized materials must be taken into account when planning and preparing an effective defense against chemical and biological materials. Chemical warfare defense and research efforts require an understanding of a wide range of interrelated topics where aerosols are involved. These topics include

- chemical weaponry,
- detection technology,
- respiratory and body protection,
- decontamination processes,
- prophylaxis and treatment,
- restoration and recovery,
- atmospheric transport modeling and prediction,
- indoor air quality, and
- operations research.

Other areas falling within the general arena of chemical warfare defense include the development and use of obscurants, defoliants, and riot control agents.

Entire chapters could be written on the history of each of these elements as they pertain to the corresponding aerosol science. However, this chapter presents only brief overviews of some important advances made in aerosol science as viewed from the military perspective on chemical warfare defense and offense. The primary elements discussed here are obscurants, chemical weaponry, respiratory protection, and atmospheric transport modeling and prediction, as well as how pre-1982 military sponsorship of aerosol science conferences contributed to the formation of the American Association for Aerosol Research (AAAR).

Discussion

Obscurants

Smokes and obscurants are considered force multipliers on the battlefield. An informative talk by Chris Noble at the 2006 International Aerosol Conference outlined the development of military smokes and obscurants and described how smokes have been used to obscure the battlefield and prevent target acquisition (Noble, 2011). An example of a successful use of smoke in a military operation is one conducted by Charles XII (1682–1718), the King of Sweden from 1697 to 1718 and a highly capable military general. In 1701, he used smoke aerosols generated by burning wet hay and straw to aid a river-bridge crossing (Figure 1).

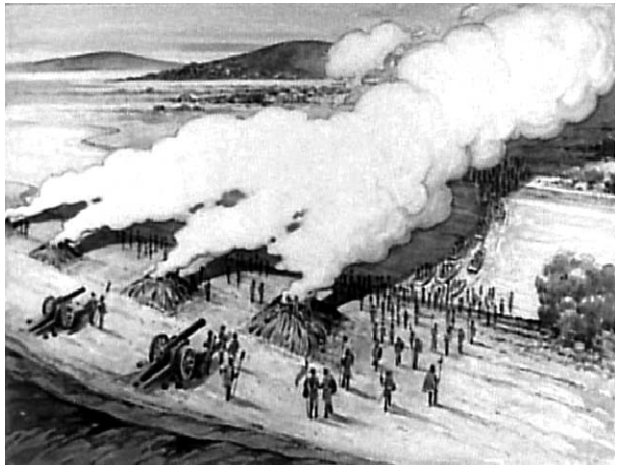


Figure 1. Obscuring smoke river crossing—1701.

Photo: Courtesy of US Army Chemical Biological Defense Command Historical Research and Response Team, Aberdeen Proving Ground, MD.

Although Carl von Clausewitz (1780–1831) never specifically used the words "fog of war" (circa 1827), the concept of how weather and smoke clouds on the battlefield can deceive the eye and result in general confusion is indicative of the role that military aerosols play in defensive and offensive operations (von Clausewitz, 1976).

Smokes have been used to screen ongoing military activities and operations and can be employed as smoke screens or smoke blankets. Properly delivered, visual smoke curtains can obscure large areas and distances such as the New York City skyline (Figure 2A). Placing multiple numbers of fog oil smoke generators along a line can blanket a large area, resulting in reduced visibility (Figure 2B). Colored smokes can also be used for signaling purposes as demonstrated at Fort McClellan, Alabama, in 1963 (Figure 3A).

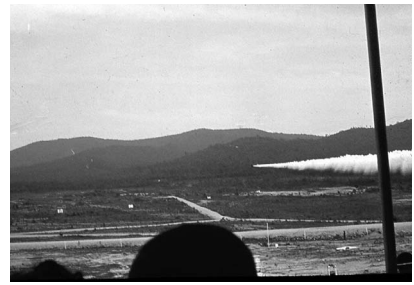
(A) Aircraft-generated smoke curtain—New York City, 1924

(B) Fog oil smoke generator

Figure 2. Smoke generation methods.

Photos: Courtesy of US Army Chemical and Biological Defense Command Historical Research and Response Team, Aberdeen Proving Ground, MD.

(A) Signaling smokes—1963



(B) Smoke curtain—1963

Figure 3. Uses of smoke.
Photos: Courtesy of the author.

In some cases, smokes can produce collateral effects as well. During a smoke training exercise near the Black Hills National Forest area in 1998, the 300th Chemical Company provided smoke coverage in support of a medium girder bridge company. The smoke operation was conducted on an open, private ranch. About 1½ hours into the 3½-hour operation, approximately 100 head of cattle appeared and contently grazed inside the smoke cloud. Apparently, the smoke either provided pleasing shade and a cooling effect, or shielded the animals from harassing flies.

Some of the earlier effective smoke screens used FS (sulfur trioxide dissolved in chlorosulfonic acid), which was developed between 1929 and 1930. FS produces an intense white cloud when aerosolized. Unfortunately, when FS reacts with atmospheric moisture, sulfuric acid and hydrochloric acid drops are formed, which can be quite detrimental to materials such as nylon. In another example of collateral effects during a live fire demonstration, FS smoke was released from an aircraft spray tank to establish a smoke curtain, similar to that shown in Figure 3B. Suddenly, the winds shifted, sending the acidic smoke over the viewing stands where the VIPs were seated. The ladies of the audience did not appreciate the holes that were caused in their stockings.

More recently, as a consequence of laser technology, it was determined that infrared (IR) screening smokes were needed. The development of such obscurants to counter IR guidance and target acquisition systems required intense research into the fundamental properties of aerosols and electromagnetic wave propagation processes. Smoke screens played a significant role on the mobile battlefield during the 1967 Yom Kippur War between Israel and several Arab countries. Those experiences contributed to

an initiative in the early 1970s by the US Army Materiel Command (AMC) to bring together a team of scientists and engineers to form a Smoke Task Force. The objectives of the Smoke Task Force were twofold: to devise methods to quickly deploy a smoke screen around armored vehicles, and to develop advanced smokes that could counter laser-guided munitions. The efforts were headed by personnel at the Chemical Systems Laboratory located in Edgewood, Maryland. Theoretical studies at Edgewood by the Obscuration Sciences Branch of the Physics Division led to the successful demonstration of an effective IR countermeasure (Embury, 2002a, 2002b, 2004; Embury et al., 1993, 1994a, 1994b). This countermeasure came about through a change in the geometry of the obscurant material and by making it a conductive substrate rather than a dielectric. However, the desire to produce a one-way obscuring smoke or make an object totally invisible may be beyond the reach of practicability even with the future development of metamaterials (Boyle, 2006).

Chemical Weaponry

The weaponization of chemicals to achieve military objectives dates back to as early as 1000 B.C. when arsenical smokes were used by the Chinese. The history of chemical warfare and an excellent overview of weapons development from the decades following World War I (WWI) to the present have been documented by Jeff Smart, Command Historian of the US Army Research, Development and Engineering Command (RDECOM), Aberdeen Proving Ground, Maryland (Smart, 1997).

An appreciation of chemical and biological defense research must begin with an understanding of the threat that such compounds present to soldiers and must take into account today's potential terrorism threats to unprotected civilians and first responders.

The possibilities of using toxic materials on the battlefield were considered well before the 16th century when Leonardo da Vinci (1452–1519) suggested an offensive use of toxic materials against war ships (McCurdy, 1977). Da Vinci proposed the following, "Throw poison in the form of powder upon galleys. Chalk, fine sulfide of arsenic, and powdered verdegris may be thrown among enemy ships by means of small mangonels. And all those who, as they breathe, inhale the said powder with their breath will become asphyxiated."

However, the modern use of chemicals on the battlefield began in earnest as a result of the substantial advances in the manufacture and use of industrial

chemicals. Germany's chemical industry during the early 1900s enabled the first large-scale use of toxic chemicals to influence the battlefield. In April 1915, the Germans released chlorine from 1,600 large and 4,130 small cylinders (a total of 168 tons of chlorine) to create a vapor cloud that resulted in significant French and Algerian force casualties (Figure 4A). Over a 2-day period, estimates were that 5,000 French and Algerian Soldiers were killed, and at least 10,000 more were disabled. The era of chemical warfare had begun. In a retaliation effort, the French countered in kind in September 1915 (Figure 4B). The necessity of using protective masks became urgent (Figure 5).

(A) First German chlorine attack—April 1915

(B) French gas attack—September 1915

Figure 4. World War I chlorine gas attacks.

Photos: Courtesy of US Army Chemical and Biological Defense Command Historical Research and Response Team, Aberdeen Proving Ground, MD.

(A) Protective masks—April 1915

(B) Chlorine cylinders, Battle of Loos—September 1915

Figure 5. Allied response to German use of chemical weapons.

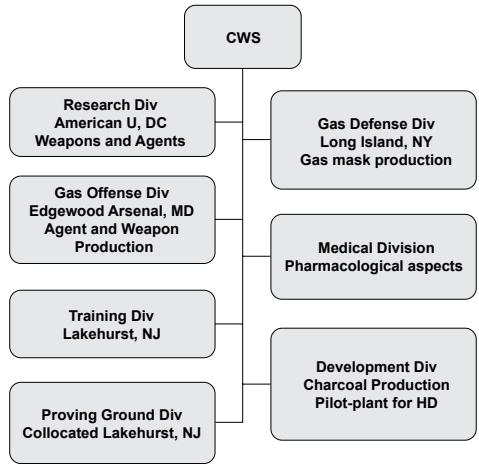
Photos: Courtesy of US Army Chemical and Biological Defense Command Historical Research and Response Team, Aberdeen Proving Ground, MD.

During WWI, more than 3,000 different chemicals were studied and considered for use as possible chemical warfare agents; 45 were actually employed in combat operations. The range of materials first included gases but quickly shifted to liquids and solids dispersed as aerosols (Prentiss, 1937).

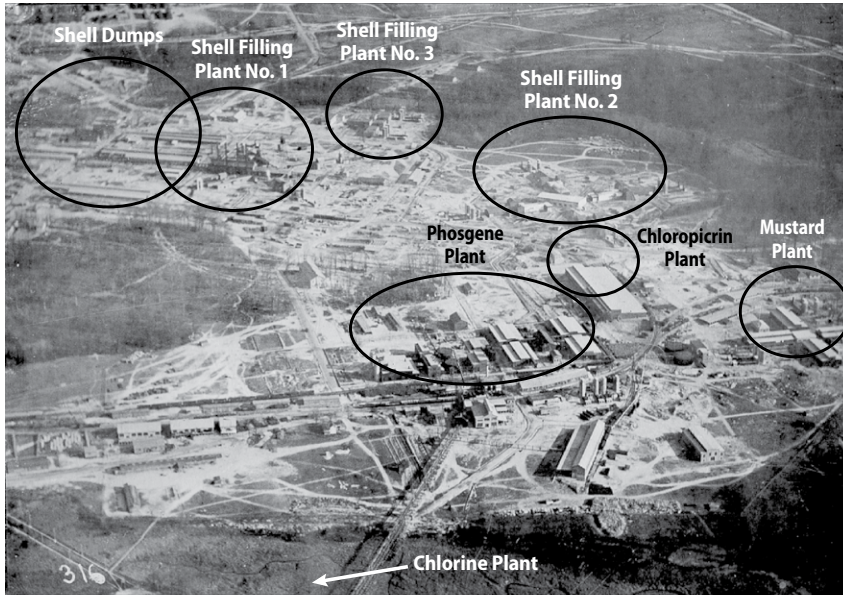
The United States entered WWI in 1917 unprepared for chemical warfare. The responsibilities for chemical warfare defense and offense were divided among several government agencies. The Bureau of Mines undertook protective mask production because of its experience in developing mine gas and rescue apparatus; research on chemical agents and weapons was performed at American University in Washington, DC; pharmacological aspects of chemical warfare defense fell under the Medical Department; and chemical agent production and munition-filling plants were the responsibility of the Ordnance Department with production facilities instituted at the Edgewood Arsenal in Maryland.

In June 1918, the War Department centralized all gas warfare functions and established the Chemical Warfare Service (CWS) (Figure 6). In 1920, all chemical warfare functions, including chemical training, research, and gas mask production, were centralized at the Edgewood Arsenal in Maryland (Figure 7).

Chemical weaponization is the process of effectively disseminating a liquid or solid payload in the appropriate particle size range depending on the intended respiratory or dermal effects. Research into chemical weaponry continued at Edgewood on a peacetime basis until the United States entered World War II (WWII) in 1941. In 1942, the Edgewood Arsenal was renamed the Army Chemical Center. Preparations for engaging in chemical warfare resumed in earnest and notable advances were made in understanding the behavior and effects of aerosols on the battlefield. Prior to the ban on open-air testing of chemical munitions in 1969, research continued on how to rapidly and effectively disseminate liquids and solids. One system in use during the 1960s for assessing the munition effectiveness for aerosolizing its payload was the ultra-high-speed camera.



(A) CWS made permanent July 1, 1920; centralized at Edgewood Arsenal.



(B) Edgewood Arsenal

Figure 6. Chemical Warfare Service (CWS)—June 28, 1918.

Note: (A) Courtesy of author; (B) Photo: Courtesy of US Army Chemical and Biological Defense Command Historical Research and Response Team, Aberdeen Proving Ground, MD.

Figure 7. Chemical warfare school.

Photos: Courtesy of US Army Chemical and Biological Defense Command Historical Research and Response Team, Aberdeen Proving Ground, MD.

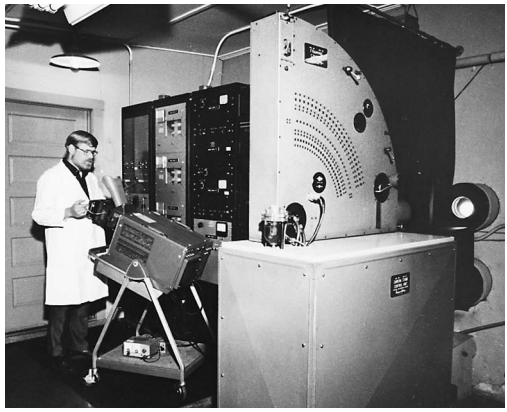
High-speed photography at 1.3 million frames per second enabled the initial breakup of a munition to be captured on film (Figure 8A). The bursting of a liquid-filled plastic sphere device is shown in Figure 8B. The cloud front expands at an average rate of 1,875 feet per second.

However, determination of the particle size distribution of the initial cloud could not be discerned due to the limited focal length and depth of field of the photographs.

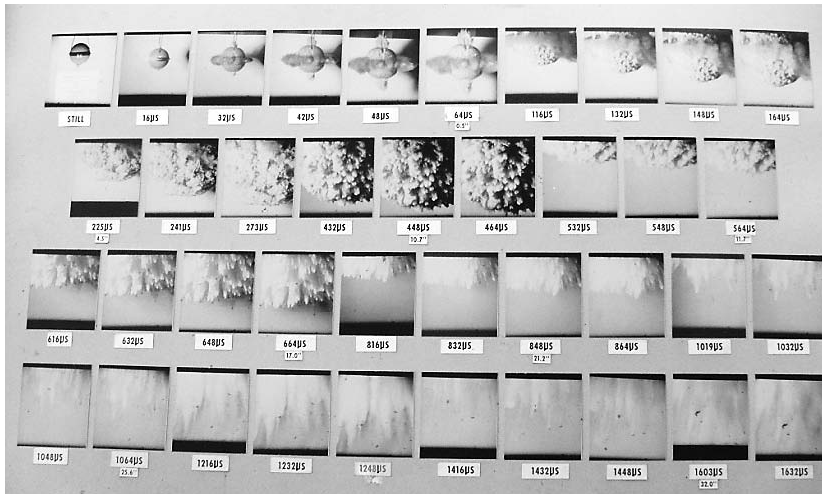
In 1947, Dennis Gabor (1900–1979), a British/Hungarian scientist, developed the theory of holography; however, it was not until the invention of the laser in 1960 that major advances in this field became possible. The laser provided the coherent light source necessary for making holograms. The military quickly recognized that holograms had the potential to greatly extend the depth of field and to "see" into clouds. As early as 1963, the Air Force pursued holographic technology for particle size determination, and the Army Chemical Center sponsored work with Technical Operations Research in Burlington, Massachusetts, to develop a laser-hologram system to capture images of exploding munitions in test chambers (Zinky, 1965). The proof of principle of the Hologram Recording System, demonstrated in 1965, enabled the measurement of particles during their dynamic formation in the size range of 3–30 micrometers at a concentration up to 100,000 particles per cubic centimeter (Figure 8C).

The need to determine the aerosol size generated by various explosive or spray systems and to track cloud travel in the environment led to the development of novel methods for collecting samples of particulate aerosols, especially biological aerosols under field conditions.

Among these sampling devices is the now familiar Andersen Aerosol Sampler (Andersen, 1958) (Figure 9A). This product of military research advanced the methodology to more clearly define the particle size distribution of aerosols. Another example is the Rotorod Sampler (Metronics, 1969) (Figure 9B). This device captured atmospheric particles in the size range of 1 to 10 micrometers at an effective sampling rate of 52 liters per minute, and with a U-shaped collector rod, particulates from 10 to 100 micrometers at 118 liters per minute. Edgewood researchers extended this device into a moving filament rotating sampler from which the time history and concentration of an aerosol cloud could be obtained.



(A) High-speed camera—1.3 million frames per second



(B) Liquid agent dissemination sequence

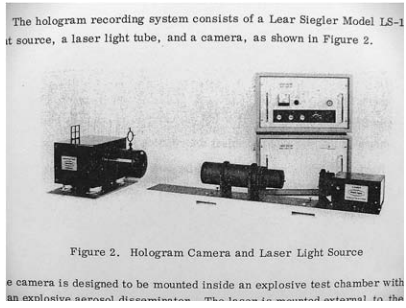

The hologram recording system consists of a Lear Siegler Model LS-1
t source, a laser light tube, and a camera, as shown in Figure 2.

Figure 2. Hologram Camera and Laser Light Source

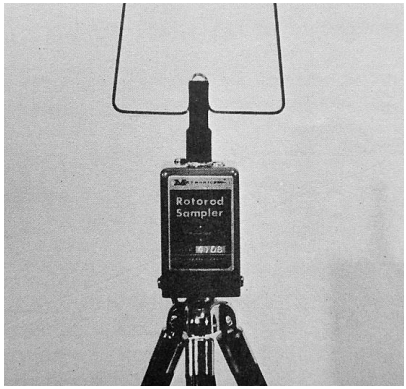
e camera is designed to be mounted inside an explosive test chamber with

(C) Hologram recording system—1965

Figure 8. Munition dissemination assessment methods.

Photos: (A) Source: Courtesy of US Army Chemical and Biological Defense Command Historical Research and Response Team, Aberdeen Proving Ground, MD; (B) and (C) Courtesy of the author.

(A) Andersen Aerosol Sampler—1958



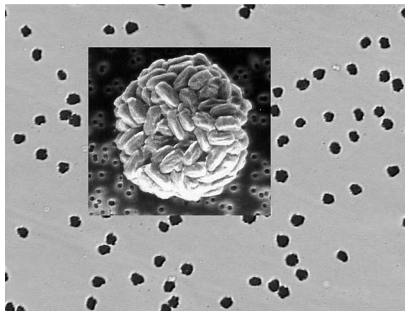
(B) Rotorod Sampler—1962

Figure 9. Aerosol sampling devices.
Photos: Courtesy of the author.

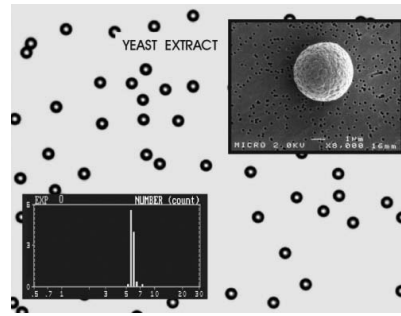
Providing aerosol test standards for the calibration and quantification of the performance of aerosol sampling devices sometimes makes use of liquid or particulate atomizing systems. The concept of an aerosol sprayer originated in 1790 when self-pressurized carbonated beverages were first introduced in France. This technology was advanced prior to and during WWII by Department of Agriculture researchers, Lyle Goodhue and William Sullivan, who developed the first portable aerosol spray can (U.S. Patent No. 2,321,023, 1941). Their invention was patented in 1941, and during WWII, the small cans were filled with insecticide and pressurized by a liquefied fluorocarbon gas for use against malaria-carrying insects. This invention led the way for the many practical applications of aerosol cans common today.

Today, ink-jet technology is available to produce custom aerosols of known composition and size at practically any desired rate (Figure 10). The Ink Jet Aerosol Generator (IJAG) invention (U.S. Patent No. 5,918,254, 1999) at Edgewood is used to test the sensitivity of bioaerosol detection candidates at very low threat levels of a few particles per liter of air. The IJAG is also used for creating uniformly sized aerosols for the calibration of sampling devices at rates up to 2,000 particles per second.

(A) The IJAG provides a source of well-characterized biological aerosol particles at low concentrations to test BW detection equipment. An ink jet cartridge, loaded with water and dissolved or suspended material, produces 50 micrometer droplets which are quickly dried in a vertical oven, producing the desired residue particles.



(B) Clusters of BG spores



(C) Spherical particles of yeast extract

Figure 10. Ink jet aerosol generator (IJAG).

Photos: Courtesy of US Army Chemical and Biological Defense Command Historical Research and Response Team, Aberdeen Proving Ground, MD.

Respiratory and Body Protection

In addition to his weaponry ideas, Leonardo da Vinci also had ideas for respiratory protection in the 16th century. His advice for protection against toxic powder was to "have your nose and mouth covered over with a fine cloth dipped in water so that the powder may not enter" (McCurdy, 1977). A more refined respirator for firefighters and mine workers was developed in 1850 by John Stenhouse of Glasgow, Scotland. It consisted of a wood charcoal filter element and velvet-lined facepiece with an elastic headband to provide

a tight fit (Smart, 1999). However, WWI found the allied armies unprepared for gas attacks even though respirators were used in civilian matters. Efforts were immediately undertaken in 1915, primarily by the British, to develop respirators to protect the wearer from noxious vapors. However, researchers were aware that aerosols of lesser volatile compounds could be expected and therefore also included particulate filtration in the protective masks.

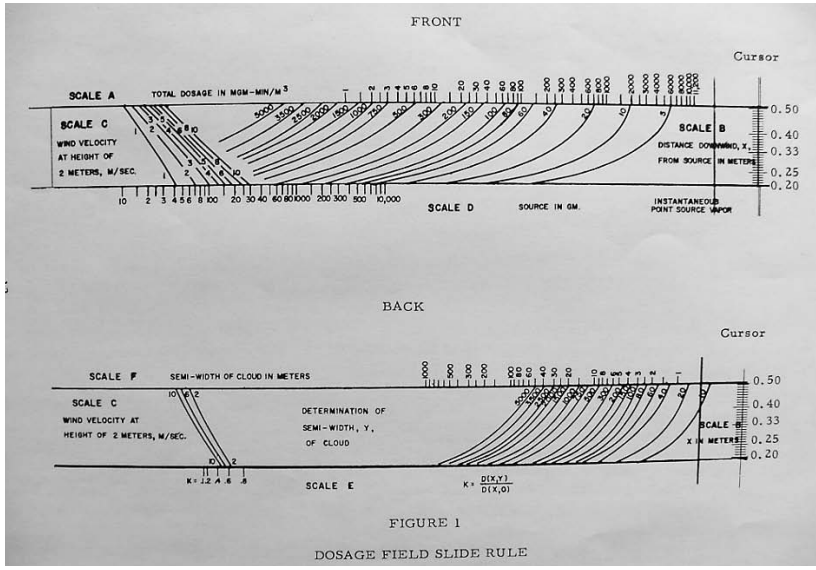
A discussion of the United Kingdom's development of particulate filters was presented by J. M. (Don) Clark at the Second Symposium on the History of Aerosol Science (Clark, 2005).

During the beginning of WWII, the filter papers from captured German gas mask canisters were sent to Edgewood for the purpose of reverse engineering. Large quantities of the German-designed filter paper that contained 14% of asbestos fibers were manufactured in the United States for protective masks, but even better smoke filters were needed. The assistance of university and industrial scientists was solicited, and major advances in the theory and technology of aerosol filtration resulted from the work of Irving Langmuir (1942) and Victor LaMer (1951). The requirement for high–airflow rate air purifiers to serve in collective protection shelters gave rise to the development of high-performance filters. The Edgewood Arsenal thereby became the sole supplier of filters for the Manhattan Project, which used them to confine airborne radioactive particles in the exhaust ventilation systems of experimental reactors. These became known as absolute, super-interception, and super-efficiency filters. In 1961, Humphrey Gilbert authored a report called *High-Efficiency Particulate Air Filter Units, Inspection, Handling, Installation* (Gilbert, 1961), and the term "HEPA filters" stuck.

Atmospheric Transport Modeling and Prediction

The transport of aerosols from munition functioning has required research into the complex arena of micrometeorology. In the case of biological aerosols, which can be infective at very low inhaled dose levels, the downwind transport is long range, in some cases covering hundreds of miles downwind depending on the release point. For chemical aerosols, the downwind transport of aerosols and vapors is of shorter range because atmospheric dilution and diffusion lessen the potential hazard. The first means of predicting transport distances made use of hand calculations and slide rules. The first straight logarithmic slide rule was invented in 1632 by William Oughtred (1574–1660), an English clergyman and writer on mathematics.

During the 1950s and 1960s, mechanical calculators with a constant multiplier function, such as the Marchant calculator, made numerical estimations by hand much faster. A slide rule for estimating downwind hazard levels based on O. G. Sutton's mathematical models (Sutton, 1947) was then invented (Stuempfle et al., 1962), and other slide rules were developed for quickly obtaining aerosol property values (TSI Incorporated, 1981) (Figure 11).

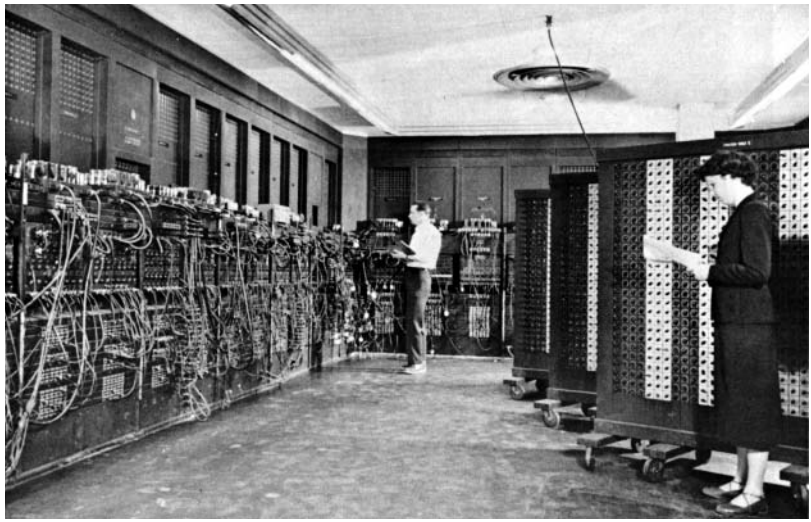


(A) Cloud Travel Slide Rule—1962

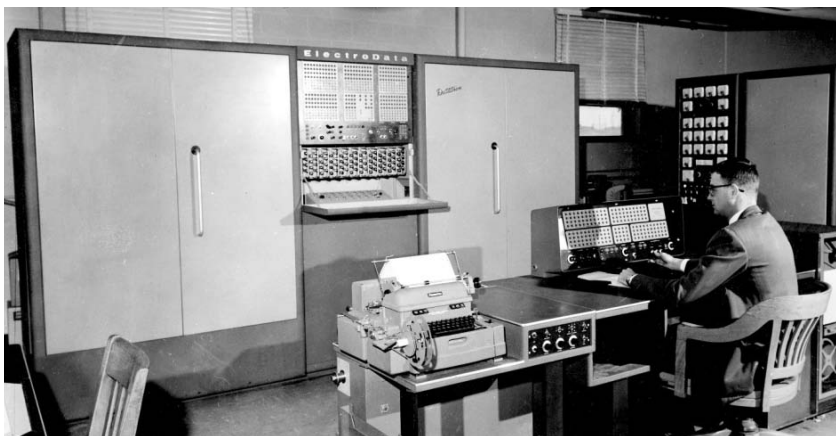
(B) Aerosol Properties Slide Rule—1981

Figure 11. Slide rules.
Photos: Courtesy of the author.

The advent of the electronic computer provided the capability for advanced atmospheric transport modeling and prediction. The first electronic computer supported by Department of Defense research funds was the Electronic Numerical Integrator and Calculator (ENIAC) (Figure 12A).



(A) Electronic Numerical Integrator and Calculator (ENIAC)—1946
Ballistics Research Laboratory, Bldg 328



(B) Datatron 205 Electrodata Computer—1960

Figure 12. Early electronic computers.

Photos: (A) Courtesy of US Army Chemical and Biological Defense Command Historical Research and Response Team, Aberdeen Proving Ground, MD; (B) Courtesy of the author.

Development of ENIAC began in 1943 but was not completed until 1946. It was used at the Ballistic Research Laboratory at Aberdeen Proving Ground, Maryland, to compute ballistic firing tables and was capable of reducing the time of calculation for a 60 second shell trajectory from 20 hours of hand calculations to 30 seconds. It consisted of 18,000 vacuum tubes and 70,000 resistors. A smaller-sized digital computer acquired by the Edgewood Arsenal for rapid and more accurate estimates of downwind cloud and aerosol transport was the Datatron 205 Electrodata Computer (Stuempfle, 1960) (Figure 12B).

An additional benefit of operating such a power-consuming and heat-generating device, with its 1,500 vacuum tubes that generated 56,000 BTU per hour, was that the room containing it needed to be air-conditioned whereas the rest of the government building was not. In 1960, this electronic data processing system cost $150,000. The discovery of transistors in 1958 eventually led to the development of second generation computers that were smaller, faster, and less power-consuming. Over the past 50 years, major advances in computer technology have occurred, and a typical programmable hand calculator purchased for $20 can now rapidly perform all the tasks that required massive equipment and intensive programming labor in 1960.

Aerosol Research Conferences

In the 1970s, one of the early technical objectives of the Edgewood Arsenal was to assemble a team of scientists to develop improved obscurants, especially against laser systems. This required knowledge of aerosol physics and chemistry. Starting in June 1978, Ed Stuebing brought together, on an annual basis, a number of recognized experts in a variety of technical specialties to share research advances in aerosol physics. The first conference topics focused on the optical and physical properties of aerosols and aerosol characterization methods (Stuebing, 1978). Interest in the conference grew as its proceedings became known. The number of attendees increased and soon included researchers from the government, academia, and industry, as well as international visitors. Some of the attendees at the CSL Aerosol Science Conference in 1981 are shown in Figure 13.

Figure 13. CSL Aerosol Science Conference—1981.
Photo: Courtesy of the author.

These conferences brought together researchers with a variety of specialties who contributed to the understanding of aerosol behavior and thus provided a stimulus for creating a national organization for aerosol researchers. In collaboration with key personnel from the scientific community, these series of information exchanges served as the forerunner to AAAR, which held its first conference in Santa Monica, California, in February 1982 (Ensor, 2000). Subsequently, the First International Aerosol Conference was held in Minneapolis, Minnesota, in 1984 (Liu et al., 1984). Through these conferences, the advances gained from aerosol science research are now exchanged on an international basis.

Conclusion

This chapter touched very briefly on some historical developments regarding obscurants, chemical aerosol generation and characterization, holography, particulate filter development, aerosol sampling techniques and calibration devices, spray can technology, atmospheric transport, and computer technology. The military and government sponsorship of research efforts has resulted in the advancement of knowledge and the technologies of aerosol science.

Acknowledgments

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