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ARIE JAN HAAGEN-SMIT

1900—1977

A Biographical Memoir by
JAMES BONNER

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Biographical Memoir

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ARIE JAN HAAGEN-SMIT

December 22, 1900–March 18, 1977

BY JAMES BONNER

ARIE JAN HAAGEN-SMIT was born December 22, 1900, in Utrecht, a city in west central Holland. His father was the chief chemist of the Netherlands Royal Mint. The mint made gold and silver coins, and Arie's first chemistry lesson consisted of playing hide-and-seek with his sisters among piles of gold and silver bricks at the mint. He also had the opportunity to watch his father dissolve gold and silver coins to analyze them for their gold, silver, and copper content. But Arie's interest in chemistry was not aroused; he found the chemistry of gold and silver quite dull.

In high school, Arie became enthusiastic about mathematics. He taught himself calculus and found physics fascinating. He also became intrigued with languages, which he learned easily and found rewarding. In addition to English, he studied French, German, and Latin. His only poor grade in high school was in the Dutch language, and his wife, Zus, tells us that Arie was always a poor speller in Dutch.

During his high school days he also developed athletic skills. He became a rower and would begin rowing as soon as the canal ice melted in the spring. He also sailed on the lakes of Holland and was a champion boxer. Between rowing and boxing, he developed the largest biceps of any faculty member in the California Institute of Technology Division of Biology up to about 1960. After 1960, when he gained worldwide recognition for his outstanding work on air pollution, he always wore a coat with sleeves and it was no longer possible to check on the status of his biceps.

In 1918, Arie entered the University of Utrecht and chose chemistry as his major. His wife believes that he might have become a mathematician or a physicist were it not for the fact that he was counseled by university officials that no positions were available in these fields in Holland. He studied chemistry as an undergraduate with a minor in mathematics. (As my mother used to tell me, a little chemistry can do no harm, and I sympathized with Haagy. I also had a chemistry undergraduate degree with a minor in mathematics.)

When the time came for graduate school, Arie again chose Utrecht and organic chemistry, considering inorganic chemistry a dull "assembly of facts." His organic chemistry professor at Utrecht at that time was P. van Romburgh, a natural products chemist who soon had Arie isolating a dermatitis-inducing agent from the outer layers (arils) of the fruit of the cashew nut. The cashews, imported from Java, were exotic and made Arie feel that he was studying the world. The agent from the arils, which became the subject of Arie's first published paper, turned out to be a substance closely related to the dermatitis-inducing agents of poison oak and poison ivy, not surprising in that poison oak, poison ivy, and the cashew nut are all species of the same family and therefore closely related.

Van Romburgh retired in 1928 and was succeeded by the young Leopold Ruzicka, fresh from Zürich. Ruzicka, then the young giant of European organic chemistry, was totally immersed in the study of the isoprenoids, in particular, the isolation, structure, and synthesis of the sesquiterpenes. Arie's work with Ruzicka resulted in his thesis, *Investigations in the Field of Sesquiterpenes.* This work awakened in Arie a lifelong interest in the chemistry of the terpenes, from the lowliest isoprene to polyterpenes such as rubber.

Arie received his Ph.D. from the University of Utrecht in 1929 and stayed on as chief assistant in organic chemistry. In this position, which has no exact correlative in American chemistry departments, he was able to do his own research on natural products but was also obliged to supervise undergraduate laboratory courses. It was an enviable position, but poorly paid.

In 1930, Leopold Ruzicka was called back to Zürich to become professor of organic chemistry at the Swiss Federal Institute of Technology. He was immediately succeeded at Utrecht by Fritz Kögel, a German who brought with him his assistant, Hanni Erxleben. Arie stayed on as chief assistant.

Utrecht at that time was the world center for the study of plant hormones. Caltech, which eventually numbered three Utrecht graduates among its chemistry faculty, was basically a substation of Utrecht for plant growth hormone studies and the only center for plant hormone study in the United States.

While still a Utrecht graduate student, Frits Went developed a biological assay for the plant-growth substance. Kögel, with Arie's assistance, set out to isolate the active principle, the plant-growth substance.

In 1954, Arie isolated this active material—then called heteroauxin, now auxin. The work, published that same year, established indole-3-acetic acid as a plant-growth material with auxin activity.

The isolation of this material laid the cornerstone of our knowledge of plant-growth regulation. In 1935, Kenneth V. Thimann at Caltech independently isolated indole-3-acetic acid from a different source. Where Arie had used human urine, Thimann used culture medium from the fungus *Rhizopus*—but the substance was the same. Indole-3-acetic acid was not isolated from higher plants and shown to be a hormone until 1946, but the effects of Arie Jan Haagen-Smit's findings about the chemical nature of the plant-growth regulator spread wide long before the compound was established as a natural plant component.

Arie never claimed special credit for this great discovery. Neither did he claim credit for an even more important find, made in the summer of 1935. Frits Went, then a faculty member at Caltech, spent that summer in Utrecht working with Arie. They discovered that substances never found in nature but chemically similar to indole-3-acetic acid, such as alpha-naphthalene-acetic acid, can mimic completely the action of indoleacetic acid in the control of plant growth. From this discovery—not patented by the discoverers—grew the whole field of chemical control of plant growth, the invention of 2,4-D as a weed killer, the idea of selective herbicides, the whole field of agricultural chemicals, and a multibillion-dollar business worldwide. This 1935 finding was monumental, its importance documentable only many years after the fact.

Meanwhile, work in Utrecht on the plant-growth hormone took a curious twist. The Went bioassay for auxin activity is highly specific for the natural hormone or closely related derivatives. In a series of papers published in Hoppe-Seyler's *Zeitschrift für physicalische Chemie* in 1933 and 1934, Kögel, Haagen-Smit, and Erxleben described the isolation in pure form and the structure determination of two active plant hormones, auxin a and b.[1] Auxin was revealed to be a

[1] These investigations took place from 1931 to 1936. From 1934 to 1935, I was a postdoctoral fellow in the Department of Chemistry at the University of Utrecht. I got to know Haagen-Smit, Kögel, and Erxleben very well. Erxleben was responsible for chemical degradations and structure determination, Haagen-Smit for isolation and biological assays, and Kögel for overall master planning, writing, and publication. So far as I know, Haagen-Smit had nothing, or next to nothing, to do with the chemistry or structure determination of auxin a and b.

trihydroxymonocarboxylic acid of eighteen carbon atoms. Auxin b contained one less carbon atom and one hydroxyl group, as well as one carbonyl group. Auxin a was isolated from human urine; auxin a and b were isolated from corn oil. (Only a single sample, from Hungary, contained the two hormones. All subsequent corn oil samples were free of both auxin a and b.)

Though it was later possible to obtain degradation products and determine their structure, the isolation of auxins a and b could never be repeated and turned out to be a scam perpetrated by Hanni Erxleben. Erxleben left detailed notebooks in which samples were properly recorded. After the end of World War II, J. A. Vliegenthart, the new professor of organic chemistry, reinvestigated these samples by mass spectrometry.[2]

"Authentic" auxin a turned out to be cholic acid; similar findings were made with respect to the degradation products. It is now believed that auxins a and b never existed. Haagen-Smit writes, "It is possible that the initial mistake was to advertise the purity of auxin a prematurely. Professor Kögel's eagerness to publish and his dictatorial behavior possibly made it very difficult for Miss Erxleben to retract her error, although this could have been done quite readily in the early period. It was Erxleben's persistence in covering up which led to the unwitting involvement of many associates."[3]

In any event, Haagen-Smit did not worry greatly about auxin a. Initially, he believed it existed, and after his arrival at Caltech he established a factory to produce it. When this factory produced only indole-3-acetic acid with no sign of

[2] J. A. and J. F. G. Vliegenthart, "Reinvestigation of authentic samples of auxins a- and b-related products by mass spectrometry," Proceedings of the *Recueils des Travaux Chimique des Pays-Pas,* 85(1966):1266–72.

[3] From W. P. Jacobs, *Plant Hormones and Plant Development* (London: Cambridge Univ. Press, 1979), p. 57.

auxin a, he isolated indole-3-acetic acid from plants and from urine and let it go at that.

Although Utrecht was the original center of plant hormone work, Caltech grew in importance with the appointment of Herman Dolk and Kenneth Thimann to the faculty in 1930. The plant hormone group was further enlarged by the appointment of Frits Went and Johannes van Overbeek. In 1935, Thimann left Caltech to establish a competing plant hormone center at Harvard, and he persuaded Haagen-Smit to come to Harvard for the 1936–1937 academic year. At Harvard, however, the chemistry faculty could not decide whether or not there was such a thing as biochemistry, although Thimann, with a Ph.D. in that subject, certainly tried to convince them. In 1937, with Harvard in doubt about the wisdom of appointing more biochemists, it was relatively easy for Frits Went and Thomas Hunt Morgan, the chairman of the Division of Biology at Caltech, to persuade Haagen-Smit to return to Caltech. Arie and his wife Marie (known to all as Zus), rapidly took root in Pasadena, where they raised their children—Maria, Margaret, and Johanna (today Maria Van Pelt, Margaret Daniel, and Johanna Demers), and a son, Jan, from Arie's first marriage.

CALIFORNIA INSTITUTE OF TECHNOLOGY

At Caltech Haagen-Smit undertook a variety of tasks to determine, by trial and error, which would interest him the most. With David Bonner he found that adenine was a leaf-growth factor for radish leaves. With this author and James English, he discovered that the wound hormone active on bean plants was 1-decene-1, dodecanoic acid. With Joseph Koepfli and Gorden Alles, he isolated the active principle of *Cannabis sativa* (marijuana). With Edward Tatum, he identified the chemical nature of the precursor of the *Drosophila* eye pigment, and with several of his students he attempted the reisolation of auxins from plant material—attempts that always resulted in the isolation of indole-3-acetic acid.

Terpenes, however, remained his greatest love. He investigated the leaf oil of the California bay tree, *Umbellularia californica,* whose principal component is umbellulone. He separated the terpenes of guayule leaf oil, *Parthenium argentatum,* and, with Nicholas Mirov of the U.S. Forest Genetic Station in Placerville, determined the composition of the turpentines of a wide variety of pine trees. Separating the components of the pine turpentines by fractional distillation, he remarked in later years how simple it all would have been if he had waited until gas chromatography had been invented. He wrote what became a classic chapter on the chemistry, origin, and function of essential oils in plants for Gunther's 1948 *The Essential Oils.*

From the late 1940s onward, Haagen-Smit undertook a massive program to determine the flavor components of the pineapple. Reports of his studies with Arthur Prater, Clara Deasy, and Justus Kirchner were published in a series of articles beginning in 1945. This work led in turn to investigations of the flavor components of wines, onions, and garlic. Haagen-Smit passed air over plants enclosed in translucent plastic chambers, collecting in a cold trap the volatile flavoring materials evaporated from the plants. Investigating the chemical nature of these volatiles—that is, the substances distilled off plants exposed to the heat of sunlight—he found that there were many, particularly terpenes, that were distilled out of leaves and wasted. In some cases, the amount of terpenes wasted through distillation by sunlight amounted to one-quarter or more of the total photosynthate of the plant.

Haagy had many graduate students, including two of my brothers, Walter and David. He was good with students, suggesting interesting projects for them to work on, giving helpful suggestions, and teaching a fascinating advanced class on the chemistry of natural products. This class was always well attended. One of Haagy's graduate students told me several years after his departure from Caltech that his notes on the course "Chemistry of Natural Products" contained more meat and information than his notes from any other class he had taken as a graduate student at the Institute.

In addition to sharing the academic burdens of teaching and supervising graduate students, Arie served as the Division of Biology's first executive officer, a position he held for six years. I remember there was so much work to do as executive officer at the time that each evening, instead of a briefcase, Arie took home a suitcase full of papers to work on. In the morning, he brought back the suitcase full of resolved work.

SMOG AND MICROCHEMISTRY

Until well into World War II, the gasoline produced in southern California was produced by the straight fractional distillation of crude oil and principally contained saturated hydrocarbons. In the summer of 1943 a butadiene plant for the manufacture of one monomer for a synthetic rubber opened in Los Angeles. It quickly became surrounded by a fog of beautiful, eye-irritating, orange vapor: Smog had been born. The catalytic cracking of petroleum, which began on a large scale at this time, led to the production of a vast array of unsaturated hydrocarbons, and soon the aerosol we now know as smog was not only generated abundantly in industrial Los Angeles but also drifted from Los Angeles inland to the San Gabriel Mountains. It travelled east as far as Riverside and even New Jersey! The aerosol, contained under the inversion layer characteristic of summer and fall days in southern California, could not rise up and be diluted.

In the late 1940s, no one knew the chemical nature of the smoggy aerosol, although it was widely suspected that it had something to do with emissions of petroleum products. The Western Oil and Gas Association, the industrial association of petroleum companies, engaged the services of the southern branch of Stanford Research Institute to determine the chemical nature of smog. They found that smog was caused by sulfur dioxide emissions. Stringent laws were immediately passed in Los Angeles County, putting a lid on SO_2 emissions, and soon Los Angeles had the cleanest air—from the standpoint of SO_2 pollution—of any major city of the United States.

But smog continued to get worse, and at this point Haagen-Smit intervened. His training and experience in microchemistry—that is, the determination of the chemical nature of substances available only in very small quantities—stood him in good stead. He and his constant colleague, Charles E. Bradley,[4] determined that the aerosol was composed of polymerized oxidation products of unsaturated hydrocarbons. They further showed that these unsaturated hydrocarbons were released from gasoline storage tanks, from the gasoline tanks of automobiles, and were also present in the exhaust of automobiles. Further study showed that the formation of smog was even more complicated because it was not due to unsaturated hydrocarbons alone, but to their oxidation by ozone.

Early in the course of these investigations (work done in collaboration with Milton Zaitlin, Herbert Hall, and W. Noble) it was also found that smog injured plants. Sensitive plants such as spinach and alfalfa were used for sometime to determine smog severity at smog-measuring stations throughout Los Angeles County. Haagen-Smit and Charles Bradley also worked out a simple quantitative method for

[4] Charles E. Bradley was the retired head of chemical research for the United States Rubber Company and the first professional chemist ever employed in the rubber industry in the United States.

determining ozone concentration in the air: Put a piece of bent, and therefore stressed, rubber tubing into an air sample and determine how long it takes for the stressed rubber to crack—a simple, elegant, and quick test for ozone concentration in air.

The single, localized smog source of 1943 was quickly controlled by the reduction of leaks from the butadiene plant. In the years after 1943, however, smog in Los Angeles grew ever more intense and pervasive. In 1947 the Los Angeles County Air Pollution Control District was formed by an act of the California legislature to study the problem. It was also provided with the legal tools to enforce measures necessary for improving the situation. Haagen-Smit was instrumental in persuading city, county, and state officials to establish this organization and institute these important measures. By the late 1940s, Haagen-Smit not only knew the nature of smog, he realized the magnitude of the problem of dealing with it and the need for action on a wide front. More research was needed, for example, to find out in detail how the high oxidant levels in Los Angeles air were generated.

From 1950 to 1959, Haagen-Smit took a year's leave of absence from his academic post to lead the research efforts of the Los Angeles County Air Pollution Control District. These further studies confirmed the details of the photochemical cycle by which primary pollutants were transformed into eye irritants and polymeric aerosols. The primary agents in this process were the oxides of nitrogen that originated in all high-temperature combustion in air (the combination of oxygen and nitrogen at high pressure and temperature and the rapid quenching of the reaction). These conclusions were not at all readily accepted by the automobile industry, however, and it was not until 1954 that general agreement as to the chemical nature of smog and the photochemical nature of its genesis was achieved.

Even so, the story of the study of smog is in large part the story of Arie Jan Haagen-Smit single-handedly fighting the resistance of the American automobile industry. The industry gradually gave in, bit by bit. One day, for example, I saw Haagen-Smit chuckling in the hall and asked him what was so funny. He said, "Today I had three vice-presidents from the Ford Motor Company in my office. Last year I would have had to go to Detroit to see them, if I could have seen them at all."

Automobiles, however, were not the only producers of hydrocarbons. The enormous oil-fueled electric power plants in southern California were also major producers of air pollutants, and in 1957 Haagen-Smit took a further leave of absence to study the control of air pollution in the plants of the Southern California Edison Company. Through these investigations he was able to bring about major reductions not only in hydrocarbon emissions but also in the emission of nitrogen oxides. And in 1960, when the state of California established a motor vehicle pollution control board, Haagen-Smit chaired its criteria committee. He also lobbied for, and was instrumental in the formation of, California's Air Resources Board. In 1968 he became its chairman.

In this position, he was responsible for enforcing the Board's regulations concerning maximum allowable pollutants in automobile exhausts. Each car manufacturer was required to certify at the beginning of each model year that its models conformed to California emission requirements. If a company's cars did not conform, it was not permitted to sell cars in California. One year (I think it was in 1969) the Volkswagen Company failed to register its compliance with the Air Resources Board regulations. Haagen-Smit promptly posted a ban on the sale of Volkswagens in the state. He took enormous flak from the entire automobile industry but the ban stood, and within ten days Volkswagen produced its certificate of compliance. As Haagen-Smit said at the time, "I'm sorry it wasn't General Motors, but, in any case, that's real power!"

Haagen-Smit had the courage and conviction to give up his more academically respectable studies to attack the urgent social problem of smog. Unlike most scientists, he did not stop at identifying the problem but went on to implement its cure. The polluting effects of the automobile would have remained unchecked but for his influence on legislation at the county, state, and national level.[5] Rigorous of thought, humorous, and persuasive, he was instrumental in establishing the Los Angeles County Air Pollution Control District. His recommendation that California establish a statewide Air Resources Board was promptly adopted by the state legislature, with Haagen-Smit appointed as the Board's chairman. He was successful at every step of his quest for more stringent air pollution control legislation and was directly responsible for the catalytic converters now mandatory on automobiles to remove the last vestiges of unburned hydrocarbons from their exhausts.

Haagen-Smit retired from Caltech in 1971 but retained his position as chairman of California's Air Resources Board until 1973. He continued to work on smog abatement and had the pleasure of attending the dedication of the new Arie Jan Haagen-Smit Laboratories of Air Pollution Research of the Air Resources Board. He was also a member of the President's Task Force on Air Pollution (1969–1970) and a member of the Environmental Protection Agency's National Air Quality Advisory Committee from 1971 to 1976.

Haagen-Smit contributed his creativity and energy to the

[5] Haagen-Smit was widely regarded—internationally as well as nationally—as the authority on smog and air pollution and on methods for diminishing such pollution. So universal was his fame that he was known to all local residents of southern California as "Dr. Haagen-Smog."

problem of smog control in every way he could manage. In a thoughtful mood he once said to me: "I have studied the chemical nature of smog, and I've studied where it comes from. I've studied how to control the sources and the political measures necessary for regulation of emission of pollutants. I've done it on a local basis and I've taken it to the state basis and I've done quite a bit on a national basis. There's only one facet of air pollution control that I think I could have done more for and that's city planning." But this is characteristic Haagen-Smit modesty. So far as smog and its elucidation and control are concerned, Haagen-Smit did it all.

He passed away March 18, 1977.

HONORS AND DISTINCTIONS

BOARDS AND COMMITTEES

1943–1946	Chairman, Netherlands-American Society
1945	Chairman, Southern California Section, Society for Experimental Biology and Medicine
1950–1960	Member Advisory Committee, American Cancer Society
1962	Southern California Section, American Chemical Society, held various offices from 1950 on; Chairman, Board of Trustees
1950–1972	Los Angeles County Air Pollution Control District, Senior Consultant
1954	Member, State Committee of Technical Experts on Air Pollution (Beckman Committee)
1955–1960	Member, Environmental Health and Air Sanitation Advisory Committee, State of California
1957–1958	Consultant, Southern California Edison Co.
1960	Member, Los Angeles County Medical Assoc. Committee on Environmental Pollution
1960	Member, Clean Air Committee for Los Angeles area, L.A. Chamber of Commerce
1960–1962	Member, Motor Vehicle Pollution Control Board, State of California
1967–1970	Trustee, Arboretum Foundation of Los Angeles County; President (Honorary Trustee 1971–1977)
1963	Member, Presidential Advisory Panel on Environmental Pollution, U.S. Office of Science and Technology
1964–1973	Member, Advisory Committee for Biology and Medicine, Atomic Energy Commission
1965	Member, Committee on Pollution, Subpanel on Air, National Academy of Sciences
1967	Member, Technical Advisory Panel to the Assembly Committee on Transportation and Commerce, California State Legislature
1967–1968	Chairman, Technical and Scientific Advisory Committee, State of California Air Resources Board
1968–1973	Chairman, State of California Air Resources Board

1968–1971 Member, Environmental Health Sciences Advisory Committee, U.S. Department of Health, Education & Welfare

1969–1972 Member, Governor's Environmental Quality Study Council

1969–1970 Chairman, President Nixon's Task Force on Air Pollution

1970–1971 Member, Advisory Committee on Application of Aerospace Technology, for Mayor of Los Angeles

1971 Member, Sea Grant Advisory Panel, University of Southern California

1971 Member, Environmental Control Seminar Programs in Rotterdam, Warsaw, Prague, and Bucharest, for U.S. Department of Commerce

1971 Member, Conference of Interdisciplinary Communications Program, Smithsonian Institution

1971 Member, Committee for Motor Vehicle Emissions, National Academy of Sciences

1971 Member, Editorial Board, Excerpta Medica (The Netherlands)

1971–1973 Chairman, National Air Quality Criteria Advisory Committee, Environmental Protection Agency

1972 Reviewer, Extramural Research Projects and Grants, Environmental Protection Agency

1973 Consultant, Division of Biomedical and Environmental Research, Atomic Energy Commission

1974–1976 Member, Science Advisory Board, Environmental Protection Agency

HONORS AND AWARDS

1947 Knight of the Order of Orange Nassau, The Netherlands

1950 Fritzsche Award, American Chemical Society

1955 Certificate, Pure Air Committee

1957 Special Clean Air Award, Los Angeles County

1958 Frank A. Chambers Award, Air Pollution Control Association

1959 Medal of Merit, Daughters of the American Revolution

1964 Richard C. Tolman Award, Southern California Section, American Chemical Society

1969 Hodgkins Medal, Smithsonian Institution
1969 Fellow, The Franklin Institute
1970 B. Y. Morrison Memorial Lectureship Award, Agricultural Research Service, U.S. Department of Agriculture
1972 Honor Scroll, American Institute of Chemists
1972 Frederick Gardner Cottrell Award, National Academy of Sciences
1972 Monsanto Award for Air Pollution Control, American Chemical Society
1973 National Medal of Science
1974 Rheinland Preis (Germany)
1974 Tyler Ecology Award
1974 Eliot Cresson Gold Medal, The Franklin Institute

PROFESSIONAL SOCIETIES

Air Pollution Control Association
American Chemical Society
American Association for the Advancement of Science
Botanical Society of America
Botanical Society of The Netherlands
Dutch Chemical Society
Institute for Food Technology
New York Academy of Sciences
Royal Academy of Science, The Netherlands
Sigma Xi
Society for Experimental Biology and Medicine
Society of Plant Physiologists
Society for the Study of Development and Growth
Swiss Chemical Society
Alpha Chi Sigma
National Academy of Sciences

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1934

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