THE INVENTION OF AIR CHEMISTRY - CHRISTIAN JUNGE (1912 -1996)

Ruprecht Jaenicke
Institute for Atmospheric Physics
University Mainz
55099 Mainz Germany

ABSTRACT

In 1963 the book “Air Chemistry and Radioactivity” by Christian Junge was published. The book was very well received and served for many years as a reference for trace substances and tracer studies in the atmosphere. Actually it has coined the term “Air Chemistry” or “Atmospheric Chemistry” as the name for a completely standalone science. His study was concentrated on those trace substances of the atmosphere, which many scientists of those days regarded as unavoidable dirt. However, substances like atmospheric aerosols and trace gases like methane, N_2O, and the chlorofluoromethanes (CFMs) might directly and indirectly influence weather, climate, and the environment. The Max Planck Society appointed Junge as director at the Max Planck Institute for Chemistry after a long search. Junge drove forward this research field so today it is a prosperous science.

INTRODUCTION

In 1995 the Nobel Prize for Chemistry was given to Paul Crutzen (1933 -), Mario Molina (1943 -), and Frank Sherwood Rowland (1927 - 2012) for their research on the chemistry of the atmosphere, in particular concerning the production and destruction of ozone. It was not only given to those extraordinary scientists, it also acknowledges a very important field of research, atmospheric or air chemistry. The term "Air Chemistry" was coined by Christian Junge with his book "Air Chemistry and Radioactivity" (Junge 1963).

In 2001, the then retired director at the Max Planck Institute for Chemistry in Mainz, Paul Crutzen, stated in a private video record[1]: "Well, I'm Chris Junge's successor as director here and I know his outstanding work not only on the properties of aerosols, but also he was one of the pioneers to look into the budgets of atmospheric gases - greenhouse gases - which nowadays are, of course, in the forefront of interest. For this reason I proposed Chris Junge in the early 80s for Nobel Prize of Chemistry". As early as the mid 1970s Junge's contribution to Atmospheric Chemistry was well accepted in the scientific community and many colleagues regretted that Junge had not been honored with that prize himself.

While Christian Junge contributed to several fields in air chemistry and meteorology, the work on atmospheric aerosols most probably is connected prominently with his name. But he also made contribution to atmospheric trace gases and the "Old Atmosphere".

EDUCATION AND SECOND WORLD WAR

Born in Elmshorn / Germany on 2 July 1912, Junge was formed by growing up in post WWI Germany. After his "Abitur" he wanted to study chemistry, but he was warned off by

[1] Cited from a film clip taken at the Max-Planck-Institute for Chemistry, Mainz Germany

unemployed men on the streets wearing posters “Chemiker sucht Arbeit”[2]. However he noticed the Zeitgeist and the increasing interest in flying and air traffic. He wanted to participate in that. So he decided on meteorology at the university of Graz (Austria), selecting Graz to begin with, because meteorologist Alfred Wegener (1880 - 1930) there “hatte die Lehrkanzel für Meteorologie und Geophysik inne”. Wegeners understanding of meteorology was that of atmospheric physics. Sadly, Wegener was lost in the ice of Greenland in 1930 on his last expedition. So Junge never had a chance of learning with Wegener.

After the first term, Junge continued meteorology in Hamburg and Frankfurt (1931-35). He finished his education with the PhD and a work about atmospheric condensation nuclei (Junge, 1935).

In Frankfurt he spent two more years as a university research assistant working on condensation nuclei and then he joined the “Reichswetterdienst” (German Meteorological Service) at the Office for Instruments in Hamburg. Automatically Junge was involved in the military connection of the Weather Service to the “Luftwaffe” with the onset of WWII. His first assignment in WWII was propaganda. It was thought that meteorological techniques best fit that task. So Junge had to launch propaganda pamphlets attached to weather balloons from eastern France in such a way, that they would fly across the frontline to the west. But easterly winds are rather rare in Western Europe and the success of this activity was rather limited. Quite impressive is his confession in his family diary that he deposited unused propaganda material in Basilique du Sacré Coeur in Paris. Junge spent the whole of WWII as a meteorologist on duty in many parts of the war theatre: North Africa (Derna, Libya), Crete, Italy, and France. That duty has shaped his view on meteorology and the atmosphere. A meteorologist at that time had to rely on the sparse meteorological observations coming in. He was responsible for the safe return of aircraft

[2] Chemist looking for work

comrades. So a lot of professional knowledge and profound imagination was required. Junge carried the full burden of the war and also served as a PW for two years, finally falling severely ill. Later he returned to the “Deutscher Wetterdienst” in Hamburg.

Junges significance for atmospheric aerosol research rests on three subjects, Aerosol size distribution, Aerosol chemistry, and the Stratospheric aerosol layer.

RESEARCH

His professional position after returning to work was that of an experienced weather forecaster and a trained atmospheric physicist with an interest in chemistry. That laid the ground for his scientific career. Academic research suited Junge much better, so he returned to Frankfurt for habilitation (teaching permit for universities) and became “Privatdozent” in 1953. These years resulted in remarkable progress on atmospheric aerosols (Junge, 1952b).

The understanding of the atmospheric aerosol at that time was that of a “zoo” of rather independent particle populations, unconnected and different in particle size: Aitken particles, small ions, small medium ions, large ions, Langevin ions, ultra large ions, haze particles and so on (Israël et al 1932). That seemed to be a consistent picture, because particle physics offered a zoo as well. But the atmospheric particle zoo (line spectra) was created because of convenience. Israel et al (1932) explained the two-fold differentiation in the evaluation of the data. If the data are connected by straight lines instead of a curve, distinct concentrations of populations are created. Junge added his own measurements about the optically active atmospheric particles. His training as a meteorologist, being often forced to develop synoptic charts out of sparse data, encouraged him to draw a line through all the aerosol concentrations points obtained so far. He wrote: “For simplicity, the line spectra of the Aitken particles were converted to continuous

distributions“ (Junge 1952b). This way, the equation (concentration density proportional to r^{-3} with r=radius) for the continuous aerosol size distribution was created: Its simplicity was so convincing that colleagues raised the equation into the rank of a “law”. Most probably that was initiated by an remark of Siedentopf (1906 - 1963) to an oral presentation of Junge at that time: “A r^{-3}-law of radii distribution is not present in atmospheric dust only, but also in interstellar material.“

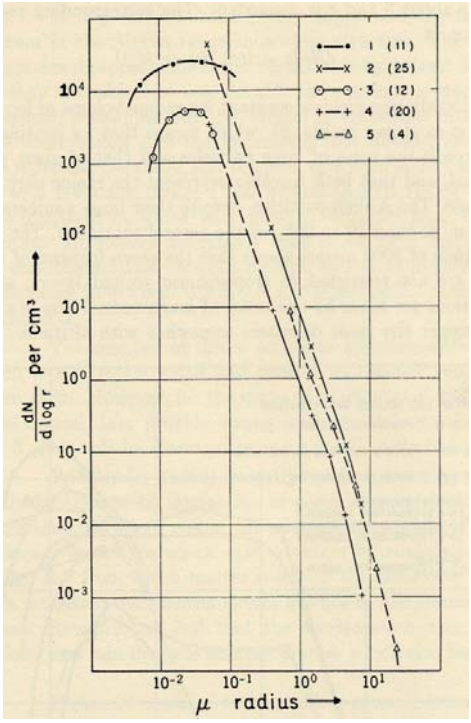

Fig. 1: Atmospheric aerosol size distribution: The dashed line is filling the gap (Junge 1953, 1955)

That straightforward picture has dominated the scientific view for many years. I remember well a discussion with Kenneth Whitby (1925 -- 1983) in Minneapolis in 1971, when he was questioning that double logarithmic straight line by Junge and planned to deviate from it. Later Whitby introduced his view (Whitby 1973) of the multimodal nature of the aerosol and Jaenicke et al (1976) added the mathematical formalism used today.

The employment and scientific future was dim in Germany in the 1950s and so Junge followed an invitation by Dr. Helmut Landsberg, than Director of the Geophysics Directorate, USAF Cambridge Research Center, Bedford, Mass and moved to the US.

Junge’s interest in the chemistry of atmospheric aerosols started with his research into the growth behavior of Aitken particles (Junge 1952a). The growth of particles in a water vapor enriched atmosphere is of great importance for cloud physics. Junge found a growth factor for particles, still widely referenced today. At that time, the idea of the mixed nature of atmospheric aerosol particles evolved. Well mixed particles seemingly made it easy for followers to study the

chemical composition, because the meteorology seemed to be of minor influence. Investigations could be done without carefully follow the history of a specific air parcel. Chemical studies of the atmospheric aerosol were separated from meteorology! Today that connection is remembered again.

In the US, Junge interest moved again to the chemistry of atmospheric aerosols. What was the state of the art at that time? Atmospheric aerosol particles serve as cloud condensation nuclei. Certain chemicals should foster this process. Electron microscope diffraction diagrams of individual particles showed ammonium sulfate being present (Jacobi et al 1952). Those and other results showed roughly the quality of aerosol particles. Quantitative data were completely absent. 50 miles outside of Boston (Round Hill) Junge made measurements with impactors and micro techniques, primarily with color reactions (Junge, 1954). Impactors allow the division of aerosol into fractions, large nuclei of .08 to .8 µm and giant nuclei of .8 to 8 µm in radius. Those measurements were later complemented by measurements in Florida (Fig. 2). The pronounced chemical difference between particles larger and smaller than .8 µm radius was established. It probably influenced Whitby later in his idea about the multimodal structure (and different production mechanism) of the atmospheric aerosol.

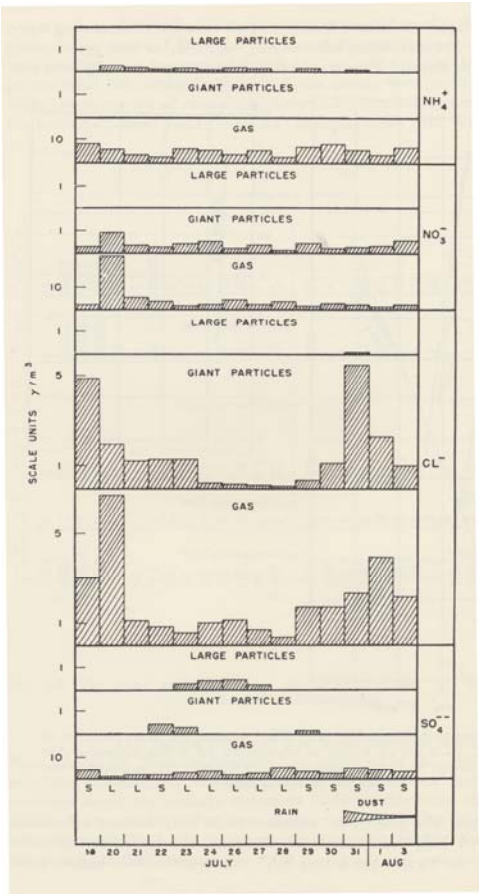

Fig. 2: The chemical composition in Florida, Junge (1956). γ/m^3 are $\mu g/m^3$.

It is interesting to note the dust event in August (Fig. 2). As a meteorologist trained in active duty Junge easily identified Saharan dust. Despite

that, other scientists (Delany et al, 1967) later selected the “pristine” air of Barbados for studying micrometeorites, because they could not imagine Saharan dust crossing the Atlantic Ocean. The benefit from this ill fated initiative now is the impressive time series of Saharan dust being transported to Barbados (Prospero 1999). In connection with the chemistry of atmospheric aerosols it is interesting to note, that Junge was the first in the US to establish a precipitation chemistry network (Junge et al 1956), following the Scandinavian original (Egnér et al 1955).

October 4, 1957 was a turning point in US-American research strategies. The USSR had launched successfully the first satellite, much earlier than expected. All research efforts in US research installations were moved to space. The public pressure was huge. What to do about an “Atmospheric Chemist” in an Air Force research establishment?

Junge proposed studying micrometeorites in the stratosphere as a possible threat to satellites. The stratosphere had been of scientific interest anyhow, because of the nuclear test explosions and the possibility of tracing the transport in the stratosphere. In addition, Penndorf (1954 for the troposphere) and Bigg (1956) had speculated from twilight measurements about the vertical

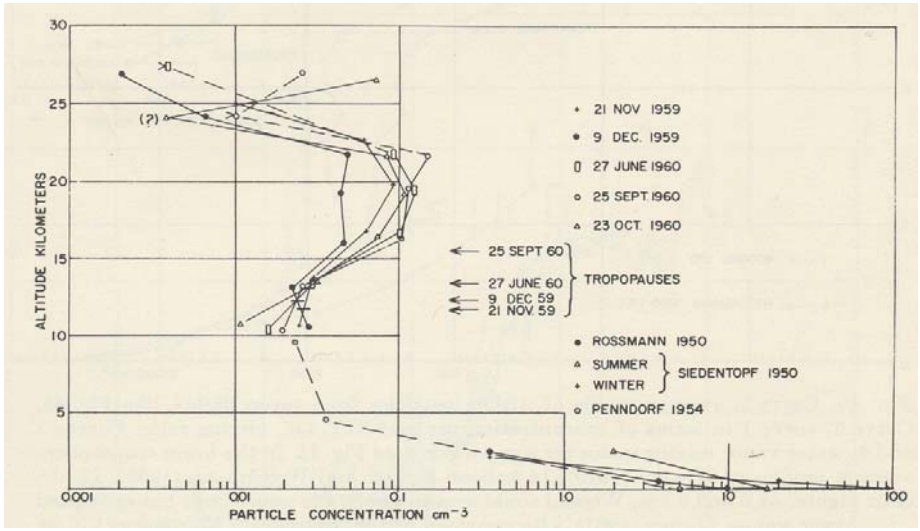

Fig. 3: Five stratospheric profiles of large particles obtained over Sioux Falls, South Dakota (Chagnon et al 1961)

distribution of dust and a stratospheric dust layer. However, proof though measurements on location was missing.

Junge's proposal was well received and he was given plentiful resources for conducting that research. So he launched balloons from Sioux Falls and Hyderabad, India. Additionally impactor probes were secretly[3] carried on U2-reconnaissance planes, at that time unknown to the public. The result surpassed all expectations: The Stratospheric Aerosol Layer (Fig. 3) was "detected" and confirmed (Junge et al 1961). The aerosol surface of this layer continues to play a key role in the explanation of the ozone hole and climatic research today.

However, the success story came to a sudden end: With his chemical interest, Junge was looking into the chemical composition of the particles collected (Junge et al 1961). He found an abundance of $SO_4^=$, a sulfate layer and a clear indication at that time of the source earth and not space. Therefore, up to the present day, that layer carries the name Junge-layer or sulfate layer.

As the research was no longer connected to space, Junges position at the USAF Cambridge Research Center was abruptly terminated and he turned to Germany in 1962 and became Professor for Meteorology at the University of Mainz.

UNIVERSITY OF MAINZ

In Mainz, Junges personal income was secured, but the professional situation was very dim. There were only a few students left at the department. The financial budget of the institute was barely 10% of that of a single director at the Max Planck Institute. There was only a limited number of rooms for scientists to work. All his attempts to change that, failed. He even organized a research shack with support from the Volkswagen-Stiftung and the University had objections. Hiring non-meteorologists at a meteorological institute was almost impossible, because of the

[3] Secretly means, the carrier was not revealed to Junge, because even as US-citizen he was from Germany. The probes carried only the information of height and exposure time, but no location

rigid structure of a university. The chemists at the university had their doubts, because Junge's scientific subjects (trace gases of the atmosphere) had been regarded as "unavoidable dirt" or contamination by them, not worth a scientific study. In addition, Junge became a victim of a scientific storyteller (a Münchhausen). As part of his seed money he purchased a very expensive instrument, the Goetz centrifuge for aerosol particle collection. That instrument was a complete failure because of overstated advertisements[4] of that colleague at the highly praised California Institute of Technology. But Junge tried to pursue his ideas with persistence and soft money. Together with Kurt Bullrich (1920 – 2010) and Hans-Walter Georgii (1924 -), he was among the first to start a "Collaborative Research Centre" of the German Research Foundation (DFG). He started investigations of trace gases (CO, Hg, N_2O) and aerosols (size distribution, development of an ion counter of high sensitivity, acidity of aerosols). His idea was to document those trace substance concentrations worldwide and reveal their sources, budgets, and effects on the environment. Only a meteorologist with his knowledge, experience and imagination on transport in the atmosphere could initiate that. He planned participation in ocean cruises with the new German research vessel "Meteor". Instruments for those tiny concentrations were not available commercially, so they had to be developed. All instruments had to be calibrated. He definitely was not an off-the-shelf scientist and taught that to students. His ability to develop a complete picture out of sparse observations proved to be helpful.

MAX PLANCK INSTITUTE FOR CHEMISTRY

While Junge was still in the US, on the other side of the ocean, Professor Paneth (1887 - 1958), then director at the Max Planck Institute for Chemistry in Mainz, died suddenly. The Max Planck Society (MPG) swiftly formed a successor committee to fill the vacancy. That turned out to be an endless undertaking. It changed its name from successor committee to

[4] In the oral history interview with Lee A. DuBridge search for Goetz in http://resolver.caltech.edu/CaltechOH:OH_DuBridge_1, 4 June 2012

appointment committee (because another academic member of the institute had reached retirement age) to future commission (the closure of the institute was proposed by prominent scientists). It carried on for almost a decade. Many famous scientists (Hans E. Suess (1909 – 1993), Gösta Rudstam, Rudolf Ludwig Mößbauer (1929 - 2011), Hermann Hartmann (1914 - 1984)) were appointed and they rejected for different reasons. Many new fields were explored and discarded. Internal solutions were discussed. Junge came into play only, when a joint appointment (Johannes Geiss (1926 -)) with the university was discussed and Junge opposed such an idea[5]. In an exploratory conversation with Professor Wolfgang Gentner (1906 - 1980), then Vice-President of the MPG, Junge was reluctant to accept an appointment of the MPG, because he never had a formal education as chemist. But Gentner convinced Junge, that he could hire chemists instead. And Junge suddenly realized the huge opportunities he would have in such a position[6].

In 1968 he was appointed director at the Max Planck Institute for Chemistry and formed the Department of Atmospheric Chemistry.

That department became a success story. Junge offered young scientists an environment to follow their own career. He continued to teach “High Atmosphere” at the university, fulfilling a great desire of the MPG. Young scientists joined Junges group. He could attract many scientists from different fields. So the research gained in depth and in breadth. He formed four groups on atmospheric aerosols, trace gases, reaction kinetics, and the “old atmosphere”. The work deepened the understanding of the physics and chemistry of the atmosphere.

[5] MPG-Archives
[6] Personal communication from Junges family history

Fig. 4: 1972 at the Max Planck Institute in Mainz: Liebl, Warneck, Neuling, Schmidt, Hahn, Schidlowski, Greese, Seiler, Kuhning, Winkler, Schmidt, Respondek, Schmidt, Eichmann, Beck, Slemr, Ketseridis, Schütz, and Jaenicke among others.

The Old Atmosphere started to explore the accumulation of free oxygen in the atmosphere during the evolution of the earth using geological means. Only free oxygen makes life, as we know it today possible. This whole research shaped thinking in terms of budgets in addition to concentrations. Later that research drifted to the carbon cycle and the evolution of life (Schidlowski 2009).

Junge was a pioneer in climate issues. Already in 1971 he participated in a compilation about "Inadvertent Climate Modification" SMIC-Report (1971). In a presentation[7] at the General Assembly of the MPG in 1975 he emphasized the climatic importance of atmospheric trace

[7] „Die Entstehung der Erdatmosphäre und ihre Beeinflussung durch den Menschen", Hauptversammlung der MPG 1975 in Hamburg

substances, including CO_2 and chlorofluoromethanes (CFM). The founding of the Max Planck Institute for Meteorology Hamburg in 1975 rests on Junge's initiative.

Reaction chemistry help in forming trace gas budgets. That included the active participation of other fields, like micro-biology and soil science.

The research on trace gases moved more into budget studies (N_2O, CO, ^{12}CO, ^{14}CO, microorganisms, seawater, OH radicals and CFMs). Such studies require interdisciplinary cooperation, a possibility Junge had implemented much better at the MPIC, than was the case at the university in those times.

Aerosol research expanded into organic compounds, desert aerosols, and undisturbed atmospheric aerosols. The idea was to document the natural state of the atmosphere as long as it still was possible.

He even speculated about the influence of mankind on the evolution of the Earth's atmosphere (Junge 1975), an idea that later has been widely popularized by the Nobel Prize-winning atmospheric chemist Paul Crutzen

When Junge retired from office in 1979, the ground was laid for the successors and resulted 1995 in a Nobel prize for Chemistry to Paul Crutzen. Many of his students and collaborators later had leading scientific positions in Germany. Today the Max-Planck-Institute for Chemistry works mainly on issues of air chemistry, a term once coined by Christian Junge.

THE PERSON

Working with Christian Junge was a demanding undertaking, a mind remodeling and mind stretching procedure. He was an inspiring teacher, always giving broad views rather than pure collections of facts. That means he had developed a coherent view of the atmosphere (world), sorting out and cataloguing the different published ideas. This way, the inexperienced

student (researcher) was helped to avoid false avenues of research. This is just in contrast to many teachers and text books today, offering the student all the conflicting ideas and observations and then leaving them alone. It also means that front and end fit together, making the picture complete and sound. This way of teaching is only possible, if the teacher has a sound knowledge, experience, and bright scientific imagination.

d 1979 in

And that also was the way, that Junge tackled research. He had visions, questions, and models about subjects that other people had never dreamed off, even if it was lying in front of their eyes. So he started studying those atmospheric traces, which chemists often had seen, but regarded as unavoidable disturbances (dirt) rather than as study objects. In naming such problems, one could draw it into attention and create new insights. Some people certainly don't like this kind of approach. They often study an object to the last comma, but fail in seeing the important issues.

Junge was a passionate experimenter and experimental data always convinced him. Such data came first and models came later. In tackling scientific issues, the questions came first and then the instrument needed to explore them. And if the instrument or procedure was not at hand, he developed it. Also, this approach is unlike that of many others, who have an instrument and are looking for applications. This way often the abilities of an instrument are exaggerated (see the problems with the Goetz' aerosol centrifuge (Goetz et al 1961)). To some of his colleagues those forward looking views not were always appreciated, stirring up settled life and complacent positions.

Junge had served the scientific community in many positions: President of the Commission on Atmospheric Chemistry and Global Pollution of IAMAP (1967-75), President of IAMAP 1975, President of GAeF (Gesellschaft für Aerosolforschung) 1977. On the other side, the scientific community honored his work with many awards. To name a few: Member of the Leopoldina Academy of Science 1965, Alfred-Wegener-Medal of the "Verband Deutscher Meteorologischer Gesellschaften" in 1968, Carl-Gustav-Rossby-Medal of the American Meteorological Society in 1973, Korrespondierendes Mitglied der Mainzer Akademie der Wissenschaften und Literatur 1976, Dr. phil. nat. h.c. Universität Frankfurt 1978, Das große Verdienstkreuz des Verdienstordens der Bundesrepublik Deutschland 1981

In 1999, the European Aerosol Association finally has established the Junge Memorial Award in honor of Christian Junge. The Junge award is intended to recognize the outstanding research contributions of an individual, who has shaped a completely new field of aerosol science and/or technology, as Junge did for Air Chemistry.

Junge retired almost abruptly from atmospheric science after reaching retirement age and once his successor was on duty. He moved close to one of his daughter's residence at Lake Constance. There he followed his leisure pursuit - history and anthropology. In 1991 he even wrote a draft "Die Unersättlichkeit des Menschen; Gedanken zur Evolution seiner Intelligenz"[8]. Christian Junge died 18 June 1996 in Überlingen at the Lake Constance surrounded by his family.

REFERENCES

E.K. Bigg: The Detection of Atmospheric Dust and Temperature Inversions by Twilight Scattering. *J. Meteorology* **13** (1956), 262-268

[8] The Insatiability of Mankind; Thoughts About the Evolution of His Intellect. Junge was quite disappointed that this draft was not even read by highly praised academic members of the Max Planck Society. Quite disappointed he devoted it to "Meinen Freunden und Bekannten zur Unterhaltung und Anregung, Überlingen August 1991"

A.C. Delany, A.C. Delany, D.W. Parkin, J.J. Griffin, E.D. Goldberg, B.E.F. Reimann: Airborne Dust Collected at Barbados. *Geoch. Cosmoch. Acta* **31** (1967), 885-909
H. Egnér, E. Eriksson (1955): Current data on the chemical composition of air and precipitation. *Tellus* **7** (1955), 134-139
A. Goetz, O. Preining: Bestimmung der Größenverteilung eines Aerosols mittels des Goetz'schen Aerosolspektrometers. *Acta Physica Austriaca* **14** (1961), 292-304
H. Israël, L. Schulz: Über die Größenverteilung der atmosphärischen Ionen. *Meteorol. Z.* **49** (1932), 226-233
W. Jacobi, C. Junge, W. Lippert: Reihenuntersuchungen des natürliches Aerosols mittels Elektronenmikroskops. *Arch. Meteorol. Geophys. u. Bioklimatol.* **A5** (1952), 166-178
R. Jaenicke, C.N. Davies: The Mathematical Expression of the Size Distribution of Atmospheric Aerosols. *J. Aerosol Sci.* **7** (1976), 255-259
C. Junge: Übersättigungsmessungen an atmosphärischen Kondensationskernen. Gerlands Beiträge zur Geophysik **46** (1935), 108-129
C. Junge: Das Größenwachstum der Aitkenkerne. *Ber. Deut. Wetterdienst US-Zone* **38**, (1952a), 264-267
C. Junge: Gesetzmäßigkeiten in der Größenverteilung atmosphärischer Aerosole über dem Kontinent. *Berichte des deutschen Wetterdienstes in der US-Zone* **35** (1952b), 261-277
C. Junge: Die Rolle der Aerosole und der gasförmigen Beimengungen der Luft im Spurenstoffhaushalt der Troposphäre. *Tellus* **5** (1953), 1-26
C. Junge: The chemical composition of atmospheric aerosols. I. Measurements at Round Hill Field Station, June-July 1953. *J. Meteorology* **11** (1954), 323-333
C. Junge: The size distribution and aging of natural aerosols as determined from electrical and optical data on the atmosphere. *J. Meteorol.* **12** (1955), 13-25
C.E. Junge: Recent investigations in air chemistry. *Tellus* **8** (1956), 127-139
C.E. Junge: *Air Chemistry and Radioactivity*. New York and London 1963
C. Junge: Die Entstehung der Erdatmosphäre und ihre Beeinflussung durch den Menschen. *Max-Planck-Gesellschaft Jahrbuch* (1975), 309-317
C.E. Junge, C.W. Chagnon, J.E. Manson: Stratospheric aerosols. *J. Meteorology* **18** (1961), 81-108
C.E. Junge, J.E. Manson: Stratospheric aerosol studies. *J. Geophys. Research* **66** (1961), 2163-2182
C.E. Junge, P.E. Gustafson: Precipitation sampling for chemical analysis. *Bull. Am. Meteorol. Soc.* **37** (1956), 244-245
R. Penndorf: The vertical distribution of Mie particles in the troposphere. *Geophys. Research Papers USAF* **25** (1954), 1-12
J.M. Prospero: Long-term measurements of the transport of African mineral dust to the southeastern United States: Implications for regional air quality. *J. Geophys. Res.*, **104 (D13)** (1999), 15917-15927
M. Schidlowski: Frühe Evolution des Lebens auf der Erde: Geologische und biogeochemische Befunde. *Z. geol. Wiss.* **37** (2009), 237-260
SMIC: Inadvertent Climate Modification. Report of the Study of Man's Impact on Climate (SMIC). MIT-Press 1971
K.T. Whitby: On the Multimodal Nature of Atmospheric Aerosol Size Distribution. *Paper presented at the VIII International Conference on Nucleation Leningrad* 1973