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History of Atmospheric Chemistry in Switzerland

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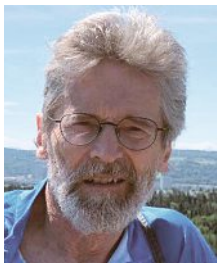
Abstract: This paper presents an overview on atmospheric chemistry, beginning with international aspects since the Roman Empire, and then focusing on the developments in Switzerland. Finally, the institutions dealing with atmospheric chemistry along with relevant scientists are briefly described.

Keywords: Aerosol · Air Pollution · Atmospheric chemistry · Ozone



Urs Baltensperger got his PhD from the University of Zurich in 1985, on chemical and morphological characterization of aerosols. Since then, his research interest focused on aerosol 'from cradle to grave', covering investigations of key processes determining the aerosol composition in the troposphere, the identification of their sources and sinks, new particle formation, secondary organic aerosol formation, source apportionment, aerosol-cloud interactions, and impact on climate and human health. He was head of the Laboratory of Chemistry at the Paul Scherrer Institute from 2000 until 2021, when he retired. He has also been Adjunct Professor at the Department of Environmental Systems Science at EHZ Zürich since 2006.

1. Introduction

While the term 'atmospheric chemistry' is relatively young, the history of the measurement of chemical species in the atmosphere in Switzerland is quite long. In the early years, such measurements were typically done by physicists and were included in the term 'meteorology' as part of atmospheric observations. Here, we include all measurements of chemical components in the atmosphere in the term 'atmospheric chemistry'. However, coverage of all types of atmospheric observations is beyond the scope of this paper. A comprehensive overview of the full range of atmospheric observations is found in the book edited by Willemse and Furger "*From weather observations to atmospheric and climate sciences in Switzerland*",[1] put together in the context of celebrating 100 years of the Swiss Society for Meteorology in 2016. Other reviews are listed in their specific contexts below. The interested reader is referred to those documents, as this article has neither the capacity nor the intention to repeat that wealth of information here. Another disclaimer refers to the climate change issue. This topic, while of highest relevance for our society, and to a great extent triggered by chemical species (such as CO_2, CH_4 and N_2O) emitted by human activities, is not treated here, as it cannot be given justice within the available space due to its complexity; rather, it would deserve an article of its own.

2. History of Air Pollution in and Beyond Switzerland

2.1 *Air Quality*

Our world has a long history of air pollution. An excellent book on air pollution was written by Peter Brimblecombe "*The Big Smoke, A history of air pollution in London since medieval times*".[2] Already Seneca wrote in a letter to Lucilius (Epistolae Morales CIV) about the year AD 65: "*Ut primum gravitatem urbis excessi et illum odorem culinarum fumantium quae motae quidquid pestiferi vaporis sorbuerunt cum pulvere effundunt, protinus mutatam valitudinem sensi*",[3] which reads in English: *As soon as I escaped from the oppressive atmosphere of the city, and from that awful odor of reeking kitchens which, when in use, pour forth all accumulated poisoned vapor and soot, I noted that my health was improving*. Brimblecombe reports that since early times, combustion played a key role in the generation of air pollution, giving numerous examples from the medieval times. Despite early attempts to control it, the air pollution in London continued to be severe, as evident from measurements of 'smoke' published by Russell in 1885 (see p. 153 of Brimblecombe's book).[2] Major smog events occurred from 1873 to 1982, with peaks in smoke and sulfur dioxide correlating with increased deaths. The worst one in the 20th century was the Great Smog of 1952, leading to some 4000 excess deaths. This resulted in a loud call for political action, and after some time, the government enacted the *Clean Air Act* of 5th July 1956.[2] The London fog of 1952 became a landmark in air pollution epidemiology because of its scale of the disaster. However, the problem was not restricted to England; in fact, an even earlier landmark of air pollution affected the Meuse Valley in Belgium in 1930.[4] The Ruhr valley in Germany experienced a similar situation, with the bad air quality reaching its climax during a severe smog episode in December 1962.[5]

Around 1945, a different type of air pollution emerged in the Los Angeles area. This triggered enormous efforts in research by Universities, governmental agencies and industries.[6] In 1952, Haagen-Smit characterized air pollution in Los Angeles by a decrease in visibility, crop damage, eye irritation, objectionable odor, and rubber deterioration.[7] He related it to large emissions of hydrocarbons and nitrogen oxides, *e.g.* through incomplete combustion of fuel in motor vehicles. Photochemical reactions then led to formation of ozone and aerosols. Different to the London smog, the Los Angeles (L.A.) smog requires strong photochemical action with high solar irradiation, and due to its secondary formation mechanism (where species were formed in the atmosphere rather than emitted) the situation was much more complex. The mechanisms involved in the formation of this type of smog rapidly became a research focus. In 1969, Goldsmith presented a comprehensive overview on the history, sources, mechanisms, and impacts of the L.A. smog,[8] summarizing that Los Angeles smog differs from the dominant type of air pollution in the United Kingdom by occurring more in hot than in cold periods, being oxidizing rather than reducing, occurring more frequently during dry rather than wet weather, being related to petrol rather than

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coal combustion and in being largely derived from motor vehicle exhaust rather than from household heating. Also in 1969, Friedlander and Seinfeld presented a dynamic model of photochemical smog.[9] As a result of the growing evidence of the importance of vehicle exhaust, the first generation of motor vehicle exhaust controls were applied to new cars.[8] In the mid 1970's, initially in California, vehicles were fitted with catalytic converters to reduce emissions of NO_X and VOCs, resulting in a gradual improvement of the situation, which however took several decades.

Due to the recognized importance of aerosols to *e.g.* visibility in the L.A. smog, researchers started early with studies on the chemical composition and source apportionment of aerosols, with a first study on aerosol chemical composition in Los Angeles by Mader in 1952.[10] In 1973, Friedlander performed a detailed source apportionment study for the Pasadena aerosol, where he determined the primary sources sea salt, soil, auto exhaust, fuel oil, fly ash, and cement dust, and also discriminated between primary and secondary aerosol components (directly emitted as particles and formed in the atmosphere from gaseous precursors, respectively).[11] This research was intensified with the growing evidence of the importance of aerosols (or particulate matter, PM) for human health, with the six-city study by Dockery *et al.* among the most highly cited papers on the association of PM with mortality.[12]

Public awareness on the detrimental effects of air pollution and political willingness to remedy are typically related to the perception of a highly severe situation. Since the situation concerning both the winter smog and summer smog in Switzerland was much less severe than in the areas mentioned above, little research was performed on these topics in Switzerland until the 1960's. One motivation behind the measurements that began then was to start early enough in order to avoid similarly critical situations as experienced in other countries.[13] Initially, these measurements focused on SO_2 and dustfall, mostly performed with Bergerhoff units[14] as *e.g.* described by Werner Jutzi in 1968.[15] Arnold Deuber completed a thesis at ETH Zürich on dustfall in 1969.[13] NO and NO_2 were also recognized as important indicators of air pollution, and first test measurements were performed *e.g.* in the city of Zürich in the 1960's;[16] a summary of early NO_X measurements is given by Sommer.[17]

A topic of air pollution that received high attention in Switzerland was the issue of acid rain. As described in an excellent review by Grennfelt *et al.*,[18] Robert Angus Smith had coined the term acid rain already in 1852. However, his pioneering work was largely ignored for nearly one century until the Swedish scientist Svante Odén described acid rain as a new and threatening environmental problem in the Swedish newspaper Dagens Nyheter in October 1967 and in a report in 1968[19] (cited in ref. [18]). This formed an opening for scientific and political collaboration, resulting in a treaty under the United Nations' Economic Commission for Europe UNECE), the Convention on Long-range Transboundary Air Pollution (CLRTAP) signed in 1979. Until 1999, eight protocols were signed under this Air Convention, committing parties to take far-reaching actions, not only with respect to acid rain but also with respect to several other air pollution problems (*e.g.* NO_X, NH_3, and volatile organic compounds (VOC), later also for particulate matter (PM)). Educational journals, such as Acid News (existing until today, see https://www.airclim.org/acidnews) were created, to report on problems of acidification and air pollution, and measures that are being taken to counteract them, including also cartoons (see Fig. 1).

The increasing awareness of an increasing variety of air pollution components along with the developing capabilities of measurement technologies resulted in increasing measurement intensity. Regular SO_2 measurements were performed in London already in the 1950's and began in the Ruhr area in the 1960's, showing very high SO_2 concentrations.[20] In Switzerland, SO_2 and

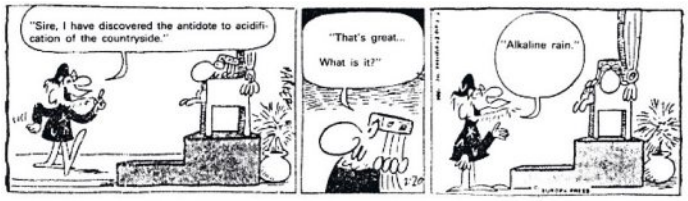

Fig. 1. Cartoon showing a not fully seriously meant antidote against acid rain from *Acid News* in **1984** (https://www.airclim.org/sites/default/files/acidnews_pdf/AN3-1984.pdf). Reproduced with permission. ©John Hart Studios Inc. All rights reserved.

sulfate measurements at the Jungfraujoch and in Payerne were launched by Empa in the context of the NABEL network in 1973 (see below). Fig. 2 presents the corresponding data from the Jungfraujoch, while the data for Payerne are found in the paper by Hueglin *et al*. in this issue.[21] Both datasets show the highest concentrations of SO_2 and sulfate concentrations around the mid 1970's, and a strong decrease afterwards, in line with the decrease in the maximum allowed sulfur content in diesel and fuel oil in Switzerland.[21] It is interesting to note that the SO_2 concentrations decreased even more than the sulfate concentrations. This is due to the shorter residence time of SO_2 compared to sulfate, such that sulfate is affected by emissions from a greater area, where SO_2 emissions may have been reduced at a later stage. Due to missing data in earlier years the rise of SO_2 with a clear peak is not seen, in contrast to SO_2 emission data showing a distinct peak in the years 1975 to 1980.[22] With time, Empa extended their activities at the Jungfraujoch (and the other stations of the NABEL network) to a highly comprehensive set of measurements.

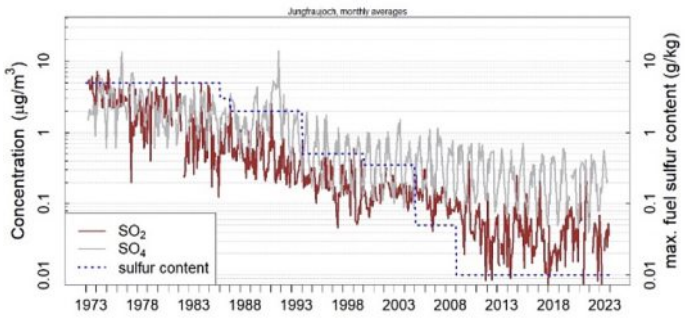

Fig. 2. Time series of monthly mean sulfur dioxide (SO_2) and particulate sulfate (SO_4) concentrations at Jungfraujoch since 1973.[23] The blue dashed line illustrates the maximum allowed sulfur content in diesel and fuel oil in Switzerland. Note the logarithmic presentation.

A related issue, the 'Waldsterben' (forest death, forest decline) arose in Germany in the early 1980s.[20,24] High concentrations of SO_2 are known to cause damage to most plants including trees, and severe and large-scale damage to forests was reported from Poland and particularly Czechoslovakia in areas with high SO_2 concentrations from lignite combustion.[25] Among the multiple initiatives started in Switzerland, the Swiss National project Sanasilva deserves special mention, as it had a big impact and resulted in a series of reports, with the last one published in 2015.[26] In contrast to the very high SO_2 and acid concentrations in Eastern europe regions, the concentrations in Central Europe were substantially lower and therefore direct effects of gaseous SO_2 and acid rain on tree leaves were not thought to be the primary cause of forest decline, but rather the indirect effects of wet and dry acidifying deposition over soil, thereby destroying the soil buffering system[25] and leading to critical ratios of base cations

to aluminum in the soil solution of tree rooting zones.[27-29] Acid fog has also been discussed as a possible reason for the decline, especially in areas with frequent fog occurrence.[25] The possible causes for the 'Waldsterben' were quite controversially disputed among scientists,[25,30] mentioning *e.g.* the observation that events of forest disease had also been observed in earlier times, and other causes (such as ozone, pathogens, insects such as bark beetles, or climate change)[25] may also have contributed to the observed forest declines.

After a few years, the situation improved, and the perception of the importance of the forest decline issue diminished. Was it therefore wrong to warn the general public of a potentially similarly detrimental situation for the forests of Central Europe as it was clearly seen in Eastern Europe? Certainly not! It is the scientists' duty to express concerns on potential threats in the future, in the same way as it has been done successfully in the case of stratospheric ozone (see Section 2.4). Of course this must be done without exaggeration, but if then – as a result of the scientists' warnings – measures are taken and the disastrous scenarios are avoided we should certainly not call the scientists' warnings a mistake in the retrospective. It appears obvious that the rapid measures to reduce air pollution (especially the emissions of SO_2 by decreasing the maximum sulfur content in fuels and the corresponding decrease in wet deposition acidity) were important in avoiding the dark scenarios of the European forest dying to the same extent as it had indeed occurred in Eastern Europe. Without doubt, the health of the European forest has improved in many areas since the 1980's, although there are still challenges today, with eutrophication ('critical load') and climate change probably being the most important ones (for a comprehensive review the reader is referred to Seidl *et al.*)[31]

Besides large research activities, the acid rain situation in Northern Europe (to some extent also seen in Swiss Alpine lakes) and the threat of forest decline triggered also political measures and new definitions of air quality standards. This included the Swiss Federal Environmental Protection Act (*Umweltschutzgesetz* https://www.fedlex.admin.ch/eli/cc/1984/1122_1122_1122/de) introduced in 1983 as well as the *Luftreinhalte-Verordnung* (Ordinance on Air Pollution Control, published in 1985 and in force since 1986),[32] with air quality standards defined both for gaseous and particulate air pollutants. Hans Urs Wanner (PhD from ETH Zürich in 1965) was one of the early pioneers in air quality research; he established the environmental hygiene field at ETH Zürich. He was a member of the Federal Commission for Air Hygiene (Eidgenössische Kommission für Lufthygiene, EKL) since 1981 and presided over it from 1986 to 1996.[33] Wanner was a leading person in the development of the Swiss Ordinance on Air Pollution Control, with their air quality standards.

In addition, continuous measurements beyond those in Payerne and at the Jungfraujoch were intensified in the 1980's, *e.g.* within the NABEL network (Nationales Beobachtungsnetz für Luftfremdstoffe, see Section on Empa below) since 1981.[34] Concerning dustfall, heavy metals were determined besides the weight, such as lead and cadmium. As an example, the city of Zürich measured these heavy metals in dustfall since 1984, showing a distinct decrease after 1987 (in the case of lead to a great extent related to the introduction of unleaded gasoline in 1985, which was required for catalyst-equipped vehicles). Also within NABEL, deposition of heavy metals was measured on a regular basis since 1988. Other air pollutants followed, *e.g.* total suspended particulate matter (TSP) since 1991 and PM_{10} since 1997, or ozone since 1988.[34] Chemical constituents in aerosols received less attention, except for the sulfate content in aerosols measured at the Jungfraujoch and in Payerne since 1973, as described above.[21] Other exceptions are neutron activation analysis results by Dams and De Jonge from aerosols sampled on the Jungfraujoch from 1973 to 1975,[35] PIXE (proton induced X-ray emission) measurements of particulate matter by Wanner and colleagues in Zürich in 1978 and 1979,[36] or the chemical and morphological characterization of aerosols in the PhD thesis of Urs Baltensperger, which included a first aerosol source apportionment as shown in Fig. 3.[37,38] Note that the filters that were used for this analysis were still Total Suspended Particulate matter (TSP) samples, *i.e.* without an upper size cut off as used today with PM_{10} or $PM_{2.5}$ samples (particulate matter with an aerodynamic diameter smaller than 10 or 2.5 μm). A consequence of this is a much larger fraction of coarse mode particles than is found today.

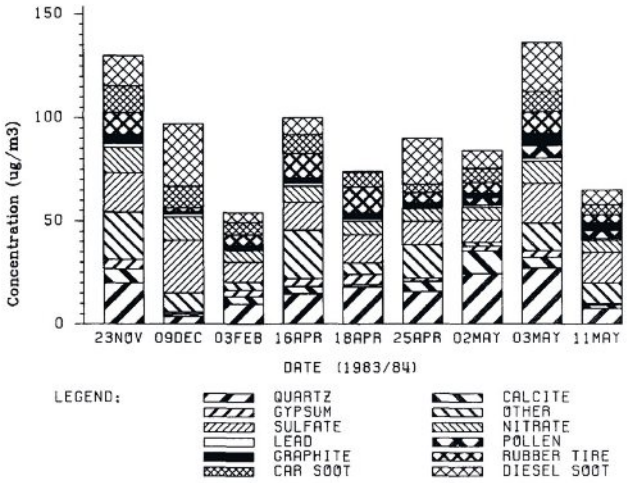

Fig. 3. Concentrations (μg/m³) of the main components of total suspended particulate matter (TSP) samples (*i.e.* sampled without a defined upper size cut-off) from a street with high traffic frequency in Zurich (Stampfenbachstrasse). Identified components are from top: Diesel soot (Dieselruss), Gasoline soot (Autoruss), Tyreware (Pneuabrieb), Graphite (Graphit), Pollen (Pollen), Lead (Blei), Nitrate (Nitrat), Sulfate (Sulfat), Other (Andere), Gypsum (Gips), Calcite (Calcit), and Quartz (Quarz). From the PhD thesis of Urs Baltensperger.[37,38]

2.2 *Surface Ozone*

The Swiss Ordinance on Air Pollution Control, in force since 1986, included also two standards for surface ozone, including a maximum 1-hour value of 120 μg/m³ to be exceeded only once an hour.[32] Already the first measurements showed that this value was exceeded frequently in the summer months.[34] This triggered a number of research campaigns, such as the Pollumet campaign (pollution and meteorology), which started in 1989.[39,40] Many studies followed, often combining extensive measurements and modeling, like the first study above. Obviously, the situation is substantially more complex for a secondary pollutant like ozone than for primary pollutants. As an example, NO first reduces ozone by titration in areas with high traffic, but increases ozone on longer time scales, through photochemical processes. While the ozone standard is still largely exceeded in Central Europe and Switzerland, it is clear that, while horizontal transport plays an important role, reduced anthropogenic emissions are the main reason for the reduced ozone peaks in hot summers.[41]

2.3 *Air Quality Today*

Comparing today's air quality with the past one can truly speak of a success story. Similar to SO_2 and sulfate at the Jungfraujoch (shown in Fig. 2), most measured air quality variables show a remarkable decline since the installation of the Ordinance on Air Pollution Control in 1986. The improvement is best seen in the annual NABEL reports of the Swiss Federal Office for the Environment (FOEN, in German: Bundesamt für Umwelt, BAFU), available at https://www.bafu.admin.ch/bafu/de/home/themen/

luft/publikationen-studien/publikationen/nabel-luftqualitaet.html. While *e.g.* 20 years ago most air pollutants were above the limits, most of them are today within the limits. Notable exceptions are ozone as described above, and $PM_{2.5}$, where the exceedances occur mostly in the Southern part of Switzerland.[42] These improvements in air quality allowed for epidemiological studies on air quality – health relationships at much lower concentrations than 20 years ago. These studies revealed that there are significant health effects also at lower concentrations than the current standards allow. Therefore, the World Health Organization developed in 2021 new air quality guidelines for particulate matter ($PM_{2.5}$ and PM_{10}), ozone, nitrogen dioxide, sulfur dioxide, and carbon monoxide, defining the lowest values, where health effects are clearly substantiated through the new studies.[43] As a consequence, the EKL, under the lead of Nino Künzli (Swiss Tropical and Public Health Institute, TPH, Basel), reviewed the currently valid standards in Switzerland. Following the Swiss Federal Environmental Protection Act, which requires the protection also of vulnerable people from adverse effects of air pollution, the EKL prepared a report with recommendations for new standards,[44] which are in most cases in line with the new air quality guidelines of WHO. These new standards, if adopted by the Swiss government, would again be exceeded in most cases in Switzerland, which points to the need for a continued strive for improvement in air quality, despite the success story so far.

An important question remains: In all investigations on the links between air pollutants and health, the best correlation was always found between $PM_{2.5}$ and mortality, as was already the case in the landmark paper by Dockery *et al*.[12] However, there are obviously different aerosol components with quite different toxicity, which tend to average out in a typical aerosol mix. This raises the question if a better link to mortality can be found through a specific aerosol component. Black carbon as well as the concentration of ultrafine particles (particles smaller than 100 nm in diameter) have been discussed, without a conclusive answer (see ref. [44] for a summary of the discussion). Recently, the oxidative potential of PM has emerged as such a possible link, however, there are a variety of tests for the oxidative potential, which provide to some extent different results (as expected). More research is needed to identify the best possible marker (or combination of markers) for the oxidative potential. Additional efforts are also needed to control emissions of nitrogen-containing air pollutants, mainly ammonia emissions from agriculture,[45] since nitrogen deposition in large areas of Switzerland still substantially exceeds critical loads of nitrogen set by the UNECE Convention on Long-range Transboundary Air Pollution to protect semi-natural ecosystems.[27,46]

2.4 *A Century of Ozone Measurements in the Swiss Alps*

In contrast to the rather late initiation of ozone measurements related to smog, Switzerland was one of the forerunners in ozone measurements in Alpine regions. A comprehensive review is presented by Johannes Staehelin and Pierre Viatte in their book on '*The Light Climatic Observatory Arosa*', telling the story of the world's longest atmospheric ozone measurements.[47] Ozone was already discovered in 1839 as a chemical substance by Christian Friedrich Schönbein in Basel.[47] Due to its early recognized importance for life on earth it was one of the first chemical species in the atmosphere that obtained high interest.[48] F. W. Paul Götz founded the Light Climatic Observatory (LKO) in 1921, and started regular and continuous total ozone observations as early as 1926,[48] initially in an attempt to determine the potentially relevant environmental factors in the treatment of tuberculosis, as described in the historic overview by Staehelin *et al.*[49] Besides ozone, Götz was also interested in Saharan dust events[50] (see also Annex in Oeschger *et al*. by Haeberli[51] for additional information). Ozone was also measured by other groups; as an example, Daniel Chalonge from France measured ozone on the Jungfraujoch also in 1926,[52] and the groups of Chalonge and Götz measured ozone simultaneously in Lauterbrunnen and at the Jungfraujoch in August 1933, finding indications for increasing ozone concentrations with altitude.[53] The following is a short summary of the comprehensive book by Staehelin and Viatte.[48]

In addition to the ozone concentration measurements, first ozone profiles were obtained in Arosa already in 1932/1933, where measurements were then performed on a routine basis from 1956 on and were to become the world's longest total ozone series. After the death of Götz in 1954, the ozone measurements in Arosa were continued by Gertrud Perl for a short time period. After some uncertainties, Hans Ulrich Dütsch, who became professor at ETH Zürich in 1964, accepted the responsibility for the measurements, with support by MeteoSwiss. The reliable total ozone series of Arosa played an important role in the International Ozone Trend Panel report,[54] in which for the first time significant negative winter trends in total ozone in the northern midlatitudes were documented, probably an important ingredient for political approval of the Montreal Protocol. It is thus an excellent example for the importance of such long-term measurements. Despite its importance, the continuation of the measurements was highly uncertain again with the retirement of Dütsch in 1985, until the LKO became part of MeteoSwiss and a joint leadership with the ETH Zürich was created in 1988, with Johannes Staehelin at ETHZ being data quality advisor for the ozone measurements of MeteoSwiss.

The experience of these ozone measurements exemplify the difficulties that long-term measurements have to cope with (and we will see more examples further below): they are often initiated and sustained by the persistence of individual scientists, and their value is at least initially often not widely recognized by the general audience or the funding agencies. Moreover, financial constraints often threaten the continuation even of highly successful time series, in this case the documentation of anthropogenic stratospheric ozone depletion and later on its recovery as a result of the 1987 Montreal Protocol. Another important issue of long-term series is the need for homogenization. Instruments (or parts of instruments) need to be replaced, procedures need to be adapted (such as optical alignment), and any of these instances require a great deal of homogenization efforts. This is especially true for such a long series as the Arosa ozone data, where a careful analysis showed that for total ozone only data from 1930 can be used (the reader is referred to the already mentioned book[48] reporting on the tremendous efforts by Staehelin and others to reach homogeneous ozone data series).

2.5 *The Jungfraujoch as a Research Infrastructure for Atmospheric Observations*

In the early years, most of the measurements were performed within campaigns, with the notable exceptions of *e.g.* the NABEL network or the ozone measurements in Arosa (see above). Successful long-term measurements, however, require solid infrastructures. An excellent example of such an infrastructure is the High-Altitude Research Station Jungfraujoch, which, due to its unique location in an unspoiled high alpine environment at 3,500 m above sea level, the year-round accessibility *via* the Jungfrau Railways, and the excellent infrastructure, provides unique conditions for successful research in various disciplines (see https://www.hfsjg.ch/en/jungfraujoch/history/, from which the following text is extracted). Research began as soon as the railway (initiated by Adolf Guyer-Zeller) was completed in 1912. Alfred de Quervain, famous meteorologist and Greenland explorer, was the driving force. On his initiative, the Jungfraujoch Commission of the Schweizerische Naturforschende Gesellschaft (now Swiss Academy of Sciences, SCNAT) was founded in 1922. Early examples of research at the Jungfraujoch comprise: astronomical observations (*e.g.* Emile

Schär, Geneva, from 1922); the altitude dependence of cosmic rays (Werner Kolhörster and Gubert von Salis, from 1923); or ozone measurements and stellar spectroscopy (Daniel Chalonge, from 1928, see also the section on ozone above). In 1931, the research station was inaugurated, and in 1937 the Sphinx observatory was completed (see the book by Leander Diener for a comprehensive historic overview on the early years at the Jungfraujoch),[55] with an English translation to follow. Since then, extensive research has been performed under the auspices of the International Foundation High Alpine Research Station Jungfraujoch (extended by the astronomical observatories at Gornergrat in 1967). This research covered a wide range of fields, such as meteorology (including atmospheric chemistry, as mentioned above), physiology, glaciology, radiation, astronomy, and cosmic rays. An overview on these early years is found in the chapter by Hans Balsiger and Erwin Flückiger in the book by Willemse and Furger.[1] Only developments concerning atmospheric chemistry can be mentioned in the following. Marcel V. Migeotte from the University of Liège, Belgium and colleagues started to analyse the solar radiation at the Jungfraujoch with high-resolution spectrometers. With time, the absorption of the solar radiation through the Earth's atmosphere by the increasing amount of climate-relevant trace gases became more and more important, and since 1986 the Belgian group has continuously monitored the development of the Kyoto-Protocol related greenhouse-gases CO_2, CH_4, N_2O and SF_6 as well as the ozone layer destroying chlorofluorocarbons (CFCs) (see Balsiger and Flückiger in ref. [1]).

In 1988, the Paul Scherrer Institute (PSI) started aerosol measurements at the Jungfraujoch. In 1994 the Swiss contribution to Global Atmosphere Watch (GAW) programme of the World Meteorological Organization (WMO), established in 1989, was launched through a decision of the Swiss Government and under the lead of MeteoSwiss. This GAW-CH programme allowed for a substantial enlargement of the aerosol measurements at the Jungfraujoch, along with support of a wide variety of other atmospheric observations, including aerosol optical depth, ozone, greenhouse gases and reactive gases complemented by strong activities in data quality assurance. For more information on the GAW-CH programme the reader is referred to the report by Gerhard Müller and Pierre Viatte.[56] Today, a variety of long-term and campaign-wise activities are ongoing at the Jungfraujoch. Among the long-term activities, ICOS (Integrated Carbon Observation System, see https:// https://www.icos-cp.eu/) and ACTRIS (Aerosol, Clouds and Trace Gases Research Infrastructure, https://www.actris.eu/), two European Research Infrastructure Consortium (ERIC) networks with a strong component at the Jungfraujoch are mentioned here. More information on ACTRIS is found in the paper by Mohr and Gysel-Beer in this special issue.[57] For a full overview on all on-going research activities at the Jungfraujoch the reader is referred to the annual activity reports of the International Foundation High Altitude Research Stations Jungfraujoch and Gornergrat (https://www.hfsjg.ch/en/publications/activity-reports/).

2.6 *Chambers for the Investigation of Atmospheric Chemistry Processes*

Atmospheric chemistry is not limited to atmospheric observations, but also includes the investigation of chemical reaction mechanisms. Suitable means for such investigations are atmospheric simulation chambers. Haagen-Smit used a fumigation room to investigate the impact of the Los Angeles smog on plants and its effects on eye irritation.[7] In Europe, Groth and colleagues built the 'Grosse Bonner Kugel' to study atmospheric processes.[58] In Europe, atmospheric simulation chambers experienced a boost with the start of the EUROCHAMP project (Integration of European Simulation Chambers for Investigating Atmospheric Processes), which started in 2004 and lasted over three funding periods until 2022 (from Switzerland, PSI participated in all three projects). The legacy of this long-term project includes, besides a wealth of results in original publications, a Practical Guide to Atmospheric Simulation Chambers, and the reader is referred to this openly available book for further information.[59] Today, these EUROCHAMP activities are part of the above mentioned ACTRIS research infrastructure.

At CERN, Geneva, Jasper Kirkby initiated the construction of an atmospheric simulation chamber to test Henrik Svensmark's thesis that ionization from galactic cosmic rays (GCRs) facilitates ion-induced nucleation and formation of new particles to such an extent that this would be the dominant cause for today's warming.[60] To simulate GCRs, he planned to use a pion beam available at CERN. After a long induction period of more than 10 years, the project finally took off in 2009 and rapidly emerged into a highly successful project with the participation of more than 20 groups worldwide. The Collaboration was able to show that the Svensmark hypothesis cannot explain today's warming,[61] but besides that provided new insight in a wide variety of new particle formation fields. Comprehensive overviews are found in the review by Kirkby[62] as well as in the article by Dada and colleagues in this special issue.[63]

2.7 *History of Atmospheric Chemistry from Archives*

In parallel with the developing importance of the history of air pollution and climate change the interest in learning about past atmospheric composition has been growing. However, obviously no atmospheric data were available for these early times. Therefore, ice cores and other archives were explored for their suitability to retrieve such missing information. Hans Oeschger from the University of Bern was one of the pioneers in ice core research. Already in 1965 he and colleagues reported on sampling of polar ice for radiocarbon dating.[64] While the research interest focused mainly on the ice shields of the poles initially, Oeschger and colleagues explored also the potential of alpine glaciers as archives of atmospheric chemistry and found Colle Gnifetti in the Monte Rosa region to be a suitable site.[51] This paper included two important Annexes: the first one by Heinz Gäggeler (University of Bern and PSI, which was at that time still the Swiss Federal Institute for Reactor Research (Eidgenössisches Institut für Reaktorforschung, EIR)), who reported on the first ice core dating using ^{210}Pb measurements (with a full independent publication in 1983),[65] and the second one by Wilfried Haeberli (University of Zurich), who presented a review on Saharan dust events in the Alps. In the following years, Oeschger developed a program on climate research, mainly through polar ice cores (see section on the University of Bern below for the further developments), while Gäggeler initiated an Alpine ice core program, which was later led by Margit Schwikowski (PSI). Soon, chemical information was also retrieved from such ice cores. Dietmar Wagenbach (University of Heidelberg) published the first sulfate record from Colle Gnifetti,[66] showing a distinct peak of sulfate in the 1970's, followed by the same decline as seen in atmospheric measurements described above (Fig. 2). Schwikowski and colleagues confirmed these results, also with an ice core from Colle Gnifetti, noting that the decline in SO_2 emissions occurred later in Eastern Europe than in Switzerland.[67] A later paper also shows the consistency of these records with various other ice cores from the Alps.[68]

3. Developments at Individual Institutions in Switzerland

3.1 *ETH Zürich and EAWAG*

Albert Waldvogel became Professor of Atmospheric Physics at ETHZ in 1985. His research focus was on radar meteorology and cloud physics rather than ozone as Dütsch before. He led the interdisciplinary project WaBoLu (Wasser, Boden, Luft, *i.e.*

water, soil and air) which investigated the interdependence of the cycles of water, soil and air. Within this topic, the project investigated winter precipitation at Mount Rigi, *e.g.* the impact of riming on precipitation chemistry.[69]

Johannes Staehelin (see above) had earned his PhD in 1983 at EAWAG, advised by Jürg Hoigné and Werner Stumm. Jürg Hoigné was professor at EAWAG and ETH Zürich, performing research on photo- and ozone chemistry, until his retirement in 1997. Although Hoigné's research was focused on processes in the aqueous phase, it became gradually clearer that such studies are also important for the aquatic phase in the atmosphere (*i.e.* aqueous droplet chemistry). Likewise, Laura Sigg studied mostly processes in water samples, but included also SO_2 oxidation studies in atmospheric water.[70] In an effort to strengthen atmospheric chemistry at EAWAG, J. Alistair Kerr, an expert in kinetic measurements of gas-phase reaction rates, was hired in 1987, and became professor at ETH Zürich in 1994. With his retirement in 1999 the activities in atmospheric chemistry at EAWAG were ended.

Also in 1999, Thomas (Tom) Peter became professor for atmospheric chemistry in the Institute for Atmospheric and Climate Science at ETH Zürich (https://usys.ethz.ch/en/people/profile.thomas-peter.html). He investigated the fundamentals of aerosols and clouds, their physical processes and chemical reactions, and their influence on the composition of the air.[71] He worked both experimentally with laboratory experiments to study the thermodynamics and kinetics of individual trapped aerosol particles or ensembles of particles under atmospheric conditions, and theoretically, using physicochemical models to study microscale aerosol processes and their impact on atmospheric chemistry up to the global scale. With Peter's presence at ETHZ, stratospheric chemistry experienced a boost where he contributed both as co-author on several international scientific reports on the evolution of the stratospheric ozone layer, and by his leadership in the World Climate Research Program SPARC (Stratospheric Processes and their Role in Climate), which he co-chaired from 2007 to 2012. Peter was also one of the driving forces behind an interdisciplinary Sinergia project funded by the Swiss National Science foundation (Infectivity of influenza viruses in expiratory aerosols under ambient temperatures and humidities). An interesting outcome of this ongoing project is the influence of pH on the inactivation time of viruses.[72] Peter retired in 2023. As his successor, Colette Heald from MIT became professor and head of the atmospheric chemistry group at ETHZ in 2024. She investigates the composition and chemistry of the global atmosphere and how these impact and interact with air pollution, climate change, and ecosystem health (see https://iac.ethz.ch/group/atmospheric-chemistry.html).

In 2004, ETH Zurich opened another professorship in atmospheric science, and Ulrike Lohmann started her position as professor for Experimental Atmospheric Physics in the Institute for Atmospheric and Climate Science (see https://iac.ethz.ch/group/atmospheric-physics.html). Her research focuses on the role of aerosol particles and clouds in the climate system, with specific interests on cloud microphysical processes including the formation of cloud droplets and ice crystals and the influence of aerosol particles on the radiation balance and on the hydrological cycle in the present, past and future climate.[73] She combines laboratory work and field measurements (*e.g.* on the Jungfraujoch[74]) on cloud and aerosol microphysics with the representation of them in different numerical models.

In addition, Claudia Mohr became Professor for Aerosol Chemistry in 2023, at ETHZ and PSI. Her activities are described below in the section on PSI. Moreover, there are a large number of other groups at the Institute for Atmospheric and Climate Science at ETH (see https://iac.ethz.ch/). As their focus is on other topics rather than atmospheric chemistry they are not further described here.

3.2 *Empa*

As detailed in the paper by Hueglin *et al*. in this special issue,[21] concerns about the harmful effects of long-range air pollutant transport on natural ecosystems prompted the Organisation for Economic Cooperation and Development (OECD) to initiate an international research and measurement program in 1972. In 1979, the measurement activities were transferred to the European Monitoring and Evaluation Programme (EMEP) in support of the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP). An overview of the history of EMEP is found in the review by Torseth *et al.*[75] The Swiss Federal Office for the Environment (FOEN; at that time called the Federal Office for Environmental Protection) entrusted Empa with performing measurements at the two stations Payerne and Jungfraujoch, which began in 1973. In 1978, FOEN designated Empa to build and operate a new national network for the measurement of air pollutants, *i.e.* the National Air Pollution Monitoring Network (NABEL). By the end of 1979, the network consisted of eight stations measuring gaseous compounds and particulate matter (PM).[76] A full description of the further developments of the NABEL network, which currently comprises 16 stations, is found in the above mentioned paper by Hueglin *et al*.[21]

From 1979 to 1989, Robert Gehrig, whose career at Empa began in 1977, was responsible for the planning, installation, and operation of the NABEL network. In 1989, he was followed by Brigitte Buchmann, and since 2002 Christoph Hueglin has been the head of NABEL. The NABEL network is embedded in the Laboratory for Air Pollution/Environmental Technology at Empa, which was led by Werner Jutzi when NABEL was founded. Peter Hofer then became laboratory head in 1987, Brigitte Buchmann in 2002, and Lukas Emmenegger in 2012. In addition to NABEL, the lab comprises the World Calibration and Quality Assurance Centers for WMO (lead: Christoph Zellweger and Martin Steinbacher, (see chapter by Buchmann *et al.* in ref. [1]), and it includes research groups on Atmospheric Modelling and Remote Sensing (lead: Dominik Brunner[77]), Climate Gases (lead: Stefan Reimann[78]), Emissions and Isotopes (lead: Joachim Mohn[79]), and Laser Spectroscopy (lead: Béla Tuzson[80]) (see https://www.empa.ch/de/web/s503/research).

3.3 *Paul Scherrer Institute, PSI*

The Paul Scherrer Institute, PSI was formed in 1988 by fusion of the Swiss Federal Institute for reactor Research (Eidgenössisches Institut für Reaktorforschung, EIR) and the Swiss Federal Institute for Nuclear Research (Schweizerisches Institute für Nuklearforschung, SIN). In 1984, Heinz Gäggeler became head of the chemistry laboratory at the EIR and later professor of radiochemistry at the University of Bern and the EIR. Being also highly interested in glaciers as archives of past climates (see above) he aimed for a better knowledge on the transfer function from the atmosphere to the snow. This involved both the vertical transport of air constituents from the planetary boundary layer to the free troposphere and the scavenging of these constituents by snow. Therefore, he started with aerosol measurements in 1986, taking advantage of a newly developed instrument, the epiphaniometer,[81] at the Jungfraujoch and also at Colle Gnifetti (4,450 m asl Monte Rosa region, Fig. 4).[82]

In 1988, Gäggeler created an aerosol research group in his laboratory, which was headed by Urs Baltensperger. In 1995, the aerosol measurements at the Jungfraujoch were embedded in the Global Atmosphere Watch (GAW) programme of the World Meteorological Organization (WMO), overseen and supported by MeteoSwiss. Today, these aerosol measurements are among the most comprehensive aerosol programs worldwide and are part of the ACTRIS ERIC (The Aerosol, Clouds and Trace Gases Research Infrastructure, see Section 2.5). More information on the history of ACTRIS ERIC is found in the review by Laj *et al*.,[83]

Fig. 4. Left: Urs Baltensperger, Heinz Gäggeler, and Martin Emmenegger (from left to right) in front of the solar panel for the epiphaniometer on Colle Gnifetti, 4450 m asl. Right: The epiphaniometer box and battery installed in a snow pit on Colle Gnifetti.

and for ACTRIS-CH (Switzerland) in the paper by Mohr and Gysel-Beer.[57]

In 2000, the Laboratory of Atmospheric Chemistry was founded and was headed by Urs Baltensperger. In the first years, the laboratory consisted of the groups Gas Phase Chemistry (later renamed to Gas Phase and Aerosol chemistry, head: André Prévôt), Aerosol Chemistry (head: Ernest Weingartner), and Ecosystem Fluxes (head: Rolf Siegwolf). The Laboratory has developed into a highly successful lab, performing field and laboratory experiments on gas-phase and aerosol processes and their impacts on air quality, clouds, climate, and health, complemented with stable isotope measurements in the biosphere (until 2017, when with the retirement of Rolf Siegwolf these activities moved to the Eidg. Forschungsanstalt für Wald, Schnee und Landschaft, WSL). In 2011, Josef Dommen became head of the newly founded 'smog-chamber' group. After his retirement in 2018, Julia Schmale became leader of this group, followed by Imad El-Haddad after her move to EPFL in 2019. With the move of Ernest Weingartner to the Fachhochschule Nordwestschweiz in 2014 Martin Gysel became group leader of the Aerosol Physics and Optics group. Urs Baltensperger retired in 2021. Kaspar Daellenbach became leader of the newly founded Aerosol and Health group in 2022. In 2023, Claudia Mohr became head of the Laboratory of Atmospheric Chemistry and professor at ETH Zürich. In 2016, The Laboratory of Environmental Chemistry was founded as the second laboratory with an environmental portfolio in the Department for General Energy Research. It was headed by Margit Schwikowski and consisted of her own group (Analytical Chemistry, with a focus on ice core chemistry from high altitudes) plus the group of Markus Ammann (originally called Surface Chemistry, now Multiphase Chemistry). Schwikowski is a member of the board of the international foundation Ice Memory, with the mission to extract ice cores from selected glaciers worldwide and store them safely in Antarctica, as rapidly as possible. These ice cores shall then be used in the future for later 'histories of atmospheric chemistry'. With the retirement of Margit Schwikowski in November this year this high-altitude ice core activity will be discontinued at PSI and probably also in Switzerland, and the group of Markus Ammann has already been integrated into the laboratory of Claudia Mohr.

Research highlights of these groups include: the detection of oligomers in secondary organic aerosol in the new smog chamber;[84] the discovery of the importance of secondary aerosol in Beijing's air pollution in wintertime;[85] and numerous other papers on aerosol source apportionment where André Prévôt has gained a leading role worldwide; the characterization of new particle formation at the Jungfraujoch;[86] the link of the oxidative potential of aerosols to human health in the whole of Europe and its potentially higher importance than the mere aerosol mass;[87] an opinion paper on aerosols and health;[88] or work on heterogeneous photochemistry in the atmosphere.[89] More details on the current research activities within the Laboratory of Atmospheric Chemistry are found in the individual papers by Mohr and Gysel-Beer,[57] Ammann *et al*.,[90] and Dada *et al*.[63] of this special issue.

3.4 *EPFL*

At the École Polytechnique Fédérale de Lausanne (EPFL), early activities in atmospheric chemistry were mostly related to technological developments. Ludger Wöste joined EPFL in 1980, first with research on the photodynamic behavior of metal clusters, and from 1981 on laser applications. Together with Jean-Pierre Wolf, he built a mobile lidar (light detection and ranging) for the remote sensing of trace gases.[91] This technique was later also extended to the remote sensing of aerosol particles. In 1989, Wöste left EPFL to accept a professorship at Freie Universität Berlin. Wolf first joined Wöste in the move to Berlin, and after other intermittent locations became professor in physics at the University of Geneva, continuing with research on a wide variety of laser applications.[92]

Hubert van den Bergh, who had started at EPFL as a PostDoc in 1973, performed research on a wide variety of laser applications and became professor in 1990 (in the chemistry department while Wöste had been in the physics department). He used the DIAL (Differential absorption lidar) technique to determine the concentrations of trace gases in the atmosphere.[93] Van den Bergh retired in 2013.

Bertrand Calpini joined the group of van den Bergh in 1990. Since 1993 he led the lidar group, building *e.g.* a pump-and-probe lidar[94] or a lidar for the remote sensing of aerosol and water vapor at the Jungfraujoch.[95] Calpini then joined MeteoSwiss in Payerne in 2002.

Modelling of atmospheric chemistry was performed at EPFL through various activities. Alain Clappier performed research on numerical modelling and simulation of the atmosphere as well as air quality studies in different cities from 1990, for example to interpret field observations together with van den Bergh and Calpini. Clappier became Professor of climatology at CNRS and University of Strasbourg in France in 2008, while for some time still being linked to EPFL. Isabelle Bey did atmospheric chemistry modelling at EPFL from 2001 to 2008,[96] from where she moved to the Center for Climate Systems Modelling (C2SM) at ETH Zürich as Executive Director.

Michel Rossi joined EPFL in 1991 coming from Stanford Research Institute. He investigated the uptake of NO_2 and other gases to model aerosol substances such as soot,[97] sea salt and ices in laboratory studies. These activities were discontinued in 2008, and Rossi moved to the Paul Scherrer Institute (PSI).

Satoshi Takahama joined EPFL as Assistant Professor in 2012. His research was focused on FTIR spectroscopy for the chemical characterization of atmospheric aerosols.[98] In 2020, he became senior scientist in the group of Athanasios Nenes.

Athanasios Nenes became Professor at EPFL and head of the newly founded Laboratory of Atmospheric Processes and their Impacts in 2019. A central focus in his research program is atmospheric particulate matter (aerosols) and their impacts on atmospheric processes that impact clouds, climate, air quality and biogeochemical cycles https://www.epfl.ch/labs/lapi/our-research/). Both experimental and modelling work is included. Nenes has been instrumental in the development of the widely used thermodynamic equilibrium model ISORROPIA for gas-particle equilibrium partitioning calculations.[99] As an example of applications, Nenes *et al*. evaluated the links between ammonia and nitrate

availability, water content and pH in atmospheric aerosols.[100] The paper by Ibikunle *et al*. in this special issue gives an example on Nenes' research activities at EPFL.[101]

Julia Schmale became tenure track assistant professor and head of the newly founded Extreme Environments Research Laboratory at EPFL in Sion in 2019 (https://www.epfl.ch/labs/eerl/eerl-home-page/research/). Her research aims to provide deeper process understanding in the ongoing transformation of extreme environments, such as the polar and alpine regions, by putting an emphasis on *in situ* field measurements, *e.g.* with participation in large ship expeditions.[102] Together with other data products (*e.g.* from satellite observations, reanalyses products or model simulations) she links atmospheric processes to the cryosphere, ocean, land, and human activities.[103] Further information on her research is found in her paper in this special issue.[104]

3.5 *University of Bern*

Although not directly involved in atmospheric chemistry, the Division of Climate and Environmental Physics at the Physics Institute of the University of Bern has a long and highly successful history in climate-related ice core analysis including the elucidation of the history of atmospheric species in ice cores, mostly from the polar regions. Hans Oeschger had founded this institute in 1963 and served as its director until his retirement in 1992. He was followed by Thomas Stocker, who, together with his colleagues in the Division, continued the successful work on polar ice core research. Upon his retirement in 2024, Hubertus Fischer became the new head of the Division.

At the Department of Chemistry, Biochemistry and Pharmaceutical Sciences of the University of Bern, Heinz Gäggeler performed not only research on radiochemistry, but also on environmental chemistry, as described above, until his retirement in 2009. Originally within Gäggeler's group, Sönke Szidat (since 2017 Associate Professor) has developed methods for the determination of radiocarbon in atmospheric aerosols at the University of Bern since 2000. These results are important *e.g*. for the distinction between fossil and non-fossil carbon in the atmospheric aerosol, thus facilitating its source apportionment.[105]

3.6 *University of Basel*

Markus Kalberer started in 2016 as Professor in atmospheric science at the University of Basel. He performs research on the chemical composition of organic aerosols using a variety of mass spectrometric methods including ultrahigh-resolution mass spectrometry, and the reactivity of organic aerosols. He is also involved in the ICOS-Cities activities in Basel. Moreover, he investigates health-relevant components in the atmospheric aerosol, using *e.g.* model cells to assess the aerosol impact on health,[106] and he develops methods to determine the aerosol oxidative potential and the components that drive it.[107] More information on this rapidly developing topic is found in the separate article on oxidative properties of atmospheric particles and their biological effects, which Kalberer wrote together with Barbara Rothen-Rutishauser (University of Fribourg), Kaspar Daellenbach (PSI), and Jean-Jacques Sauvain (University of Lausanne).[108]

3.7 *Agroscope*

Agroscope, an institution affiliated with the Swiss Federal Office for Agriculture, is the Swiss federal centre of excellence for research in the agriculture and food sector. It was formed in 2014 from a variety of organizations such as the former Swiss Federal Research Station for Agricultural Chemistry and Environmental Hygiene, Liebefeld-Bern, or Reckenholz, Zürich. It performs research on agricultural topics on the one hand and provides links with practices (farmers, *etc*.) (https://www.agroscope.admin.ch/agroscope/en/home/about-us/organization.html). In the 1990's, Albrecht Neftel from Liebefeld was one of the leading scientists in the ozone research at that time. Today, Christof Ammann in the Climate and Agriculture group of Agroscope performs research on agricultural emissions such as ammonia (see the separate article in this issue).[45]

3.8 *MeteoSwiss*

MeteoSwiss has a long history in meteorology, of which a number of chapters in the book by Willemse and Furger[1] give testimony. Their strongest involvement in atmospheric chemistry measurements refers to ozone, on the one hand continuing the worldwide unique series of continuous ozone measurements in Arosa (column measurements) and on the other hand by profiling of the ozone concentration in Payerne (using radio-soundings and micro-wave measurements). In addition, MeteoSwiss plays a highly important role in the implementation of the Global Atmosphere Watch (GAW) and the Global Climate Observing System (GCOS) programmes, both aiming at securing long-term observations of the chemical composition of the atmosphere. In Switzerland, the activities of these two programmes are coordinated by the Swiss GAW/GCOS Office at MeteoSwiss, and many Swiss institutions receive funding through MeteoSwiss for these activities.

3.9 *Other Institutions*

Finally, there are a number of institutes active in atmospheric science, however, with a limited portfolio in atmospheric chemistry. Examples are the University of Applied Sciences North-western Switzerland, where Ernest Weingartner develops instruments for physical aerosol properties, but also for black carbon, or the Physikalisch-Meteorologische Observatorium Davos/World Radiation Center (PMOD/WRC), where a global network for aerosol optical depth within GAW is operated.

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