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# Stratospheric ozone measurements at Arosa (Switzerland): History and scientific relevance

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**Abstract.** In 1926 the stratospheric ozone measurements of the Light Climatic Observatory (LKO) of Arosa (Switzerland) started, marking the start of the world's longest total (or column) ozone measurements. These measurements were driven by the recognition of the importance of atmospheric ozone for human health as well as by scientific curiosity in this by then not well characterized atmospheric trace gas. Since the mid-1970s ground-based measurements of stratospheric ozone have also been justified to society by the need to document the effects of anthropogenic Ozone Depleting Substances (ODSs), which cause stratospheric ozone depletion. Levels of ODSs peaked around the mid-1990s as a result of a global environmental policy to protect the ozone layer implemented by the 1987 Montreal Protocol and its subsequent amendments and adjustments. Consequently, chemical ozone depletion caused by ODSs stopped worsening around the mid-1990s. This renders justification for continued ozone measurements more difficult, and is likely to do so even more in future, when stratospheric ozone recovery is expected. Tendencies of increased cost savings in ozone measurements seem perceptible worldwide, ~~also in~~ Arosa. However, the large natural variability in ozone on diurnal, seasonal and interannual scales complicates ~~to demonstrate~~ the success of the Montreal Protocol. Moreover, chemistry-climate models predict a "super-recovery" of the ozone layer in the second half of this century, i.e. an increase of ozone concentrations beyond pre-1970 levels, as a consequence of ongoing climate change. This paper presents the evolution of the ozone layer and the history of international ozone research and discusses the justification of these measurements for past, present and future.

## 1. Introduction

The world's longest time series of total (or column) ozone observations is from Arosa in the Swiss Alps, made at the "Light Climatic Observatory" (Lichtklimatisches Observatorium, LKO). The long total ozone dataset is valuable for long-term trend analyses of stratospheric ozone. In addition, other important ozone measurements, such as Umkehr and surface ozone measurements are being performed in Arosa. Since the 1970s, when anthropogenic stratospheric ozone depletion became a subject of public concern, the measurements at LKO became more and more important (Staehelin et al., 2016). A comprehensive report on the history of the LKO is presently in preparation (Staehelin and Viatte, in prep.). Here we focus on the justification to society for these measurements throughout the long history of the LKO in connection to the development of international stratospheric ozone research. This paper is based on the extensive correspondence by F. W. Paul Götz - ozone pioneer and founder of the LKO - which is treasured in the LKO archives located in Payerne, Switzerland, on the annual reports of the "Kur- und Verkehrsverein Arosa" (KVV Arosa, "Health Resort Authority of Arosa", see below), and on other research. Staehelin and Viatte (in prep.) divided the history of LKO into five distinct periods (see Sections 2-6 below). Section 7 includes some remarks on the future of measurements at the LKO, and a summary and conclusions are presented in Section 8.

## 2. Period 1921-1953: Friedrich Wilhelm Paul Götz

### 2.1. Therapy for tuberculosis prior to the availability of antibiotics

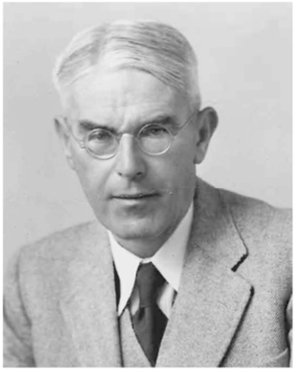
The first ozone measurements at Arosa were a part of medical research focused on the treatment of tuberculosis (TB). Before modern antibiotics became available (a few years after World War II), TB was a serious sickness with high mortality. The best available therapy for lung TB at the time was believed to be the “rest cure therapy” (as proposed, e.g. by Karl Turban, one of the leading medical doctor in Davos, see e.g. Virchow, 2004). At the end of the 19th century and the beginning of the 20th century, many sanatoria and hotels were constructed in Alpine villages such as Davos and Arosa. During “rest cure therapy”, which was fully developed in the first decades of the 20th century, the patients stayed outside on balconies during the day under strict hygienic conditions, usually for several months at a time. Recovery mainly occurred simply by resting. From a modern medical perspective, such rest under strict hygienic control (in order to prevent reinfection) in special lung clinics was probably the most helpful type of therapy before treatment by antibiotics became possible.

In 1905, Turban proposed opening an institute for scientific study of the “rest cure therapy” of lung TB (SFI, 1997). However, because of a lack of consensus among medical doctors, such an institute was founded only in 1922. On 26 March 1922, the municipality of Davos (“Landsgemeinde”) decided to create a foundation for an institute for high mountain physiology and tuberculosis research (“Institut für Hochgebirgsphysiologie und Tuberkuloseforschung”, today the “Schweizerisches Forschungsinstitut für Hochgebirgsklima und Medizin, SFI” in Davos). The resources for operating the institute mainly originated from a small fee that was paid by all guests of the town, who needed register when staying in Davos (a form of “tourist tax”).

The medical doctors of Davos and Arosa were convinced that the high altitude climate was an important factor for optimal recovery from lung TB and in order to study this further, the potentially relevant environmental factors needed to be investigated. At this point, Carl Dorno played an important role. He was a rich industrialist from Königsberg (Germany), who came to Davos because his daughter suffered from lung TB. She unfortunately passed away a few years after arriving in Davos, but Dorno remained and founded an institute to study the environmental factors important for treating TB using his own funds in 1907 (SFI, 1997). During the first World War and in the subsequent period of inflation, Dorno lost most of his financial resources. On 18 February 1923, the municipality of Davos decided to support the “Prof. Dorno Institute”, the nucleus of the world famous Physical Meteorological Observatory Davos (PMOD), which serves since 1971 also as World Radiation Center (WRC) of the World Meteorological Organization (WMO). When Dorno retired as director in 1926, the institute was integrated as an independent department into the institute for high mountain physiology and tuberculosis research in Davos and was financed by the Davos community, similar to the other institutes. Despite several studies, it was not possible to demonstrate the superiority of the Alpine climate for recovery from (lung) TB (Schürer, 2017).

## 2.2. F.W.P. Götz and the foundation of the LKO (LKS)

Friedrich Wilhelm Paul Götz grew up in Southern Germany (Göppingen, close to Stuttgart) and went to Davos for the first time prior to the beginning of the First World War to recover from lung TB, when he was working on his PhD thesis in astronomy (see Fig. 1). He stayed twice in the "Deutsche Heilstätte" sanatorium (1914-1915) and he was then released as "fit for work". For the following years (1916-1919) he intermittently taught at the "Fridericianum" German school in Davos and later worked with Dorno for an unknown duration (1919-1920). See Staehelin and Viatte for more details (in prep.).



**Friedrich Wilhelm Paul Götz**

| | |
|---|---|
| 1891 | Born on 20 May in Heilbronn (Germany) |
| 1891-1910 | Childhood in Göppingen (near Stuttgart, Germany) |
| 1910 | Start of Studies in mathematics, physics and astronomy in Heilbronn (Germany) |
| 1914-1915 | Davos: recovery from tuberculosis at «Deutsche Heilstätte» |
| 1916-1919 | Intermittently high school teacher at the «Fridericianum» (German School) in Davos, Switzerland |
| 1919 | Dissertation, University of Heidelberg (Germany), thesis on the photometry of the moon surface |
| 1919-1920 | Part-time coworker of Dorno in Davos |
| 1921 | Habilitation and lecturer at the University of Zürich, Switzerland |
| 1932 | Marries Margarete Karoline Beverstorff (27. Dec.) |
| 1940 | Promotion to «Titular-Professor» at University of Zürich, responsible for teaching courses in meteorology |
| 1950-1954 | Physical and mental illness (arteriosclerosis) |
| 1954 | Died on 29 Aug. in Chur (Switzerland) |

**Figure 1.** Biography of F.W. Paul Götz, founder of the Light Climatic Observatory in Arosa.

It appears that Götz was the main driver behind the initiative to make atmospheric measurements at Arosa. He likely first contacted the Arosa medical doctors and together they subsequently made a request to the managing committee of the KVV Arosa in March 1921 to hire Götz in order to make climate studies relevant for health. The KVV Arosa (Kur- und Verkehrsverein Arosa, "Health Resort Authority of Arosa") was an organization that had a fairly large budget, mainly supported through the "tourist" tax (i.e. a fee to be paid by foreigners/guests staying in Arosa), which was also used to cover the costs of various other activities that currently fall under the responsibility of the municipality. This request was supported by the General Assembly of the KVV Arosa that took place on 20 August 1921 and Götz was asked to found the "Light Climatic Station" (LKS), which later became known as the "Light Climatic Observatory (LKO)". The measurements taken at the LKS were to complement the meteorological observations made at Arosa since 1884 by the Swiss national weather service (now "MeteoSwiss"). These atmospheric measurements were thought to be relevant for studying recovery from TB. Arosa was the first municipality to finance an institute with the task of studying environmental factors favorable to curing (lung) TB. The support Götz obtained from the KVV Arosa was rather modest and later he secured additional regular funding from both the Chur-Arosa railway company and the Arosa municipality (for more detail see Staehelin and Viatte, in prep.). The LKS measurements were made on the roof of the Inner-Arosa Sanatorium, where the "Grand Hotel Tschuggen" is located at present (see Fig. 2).

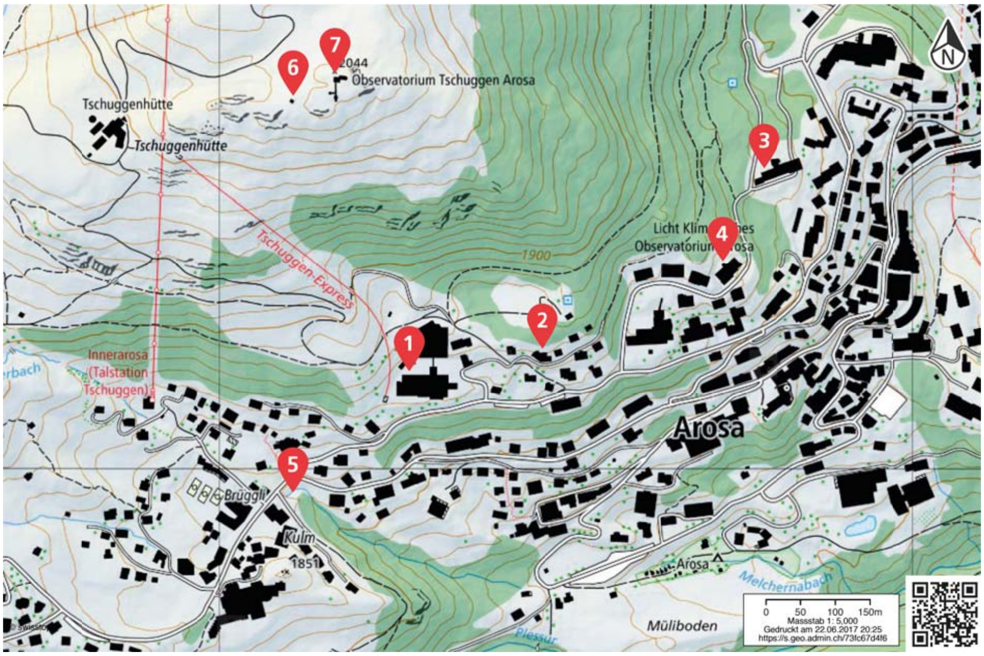


**Figure 2.** Map of important locations relevant to the Arosa Light Climatic Observatory (LKO). LKO measurement sites: 1) Sanatorium Inner-Arosa; 2) Villa Firnelicht; 3) Florentinum; 4) Haus zum Steinbruch. Other sites: 5) Götzbrunnen (fountain in honor of Götz); 6) Hut where Götz made his nighttime measurements in Tschuggen; 7) Astrophysical observatory at Tschuggen. With permission of swisstopo (Swiss digital maps, geo.admin.ch).

For the first few years Götz was able to borrow an instrument from Dorno (who was based in Davos, see 2.1) to measure "biologically active ultraviolet (UV) radiation". This instrument had been adapted and used by Dorno and consisted of a photoelectric cell with a cadmium (Cd) cathode (Levy, 1932). Götz published several papers using measurements covering the period November 1921-May 1923 (Götz 1925, 1926a and b). He found the first indication of the seasonal variability of stratospheric ozone in the northern mid-latitudes, with a minimum in autumn and maximum in spring, a very important result, which would later help to understand the global issue of stratospheric circulation. This result was in fact published earlier than the well-known publication of Dobson and Harrison (1926). Dorno did not agree with Götz's Cd-cell results, and this led to an open dispute published in the literature (Dorno, 1927). It seems likely that there were also some personal difficulties between Dorno, who was 26 years older, and Götz, which became more evident with time. It also appears there were issues between the medical doctors from Davos and Arosa, with the latter suggesting that the scientific studies made in Arosa should be coordinated with those from Davos. They also asked that the institute for high mountain physiology and tuberculosis research in Davos (Institut für Hochgebirgsphysiologie und Tuberkuloseforschung in Davos) be renamed to include Arosa, but it seems that these efforts failed since members of the Davos community wanted a higher financial contribution from Arosa to the institute (based on the principle of equal duties, equal rights ("gleiche Rechte, gleiche Pflichten")). The KVV Arosa was, however, not willing to pay the requested amount.

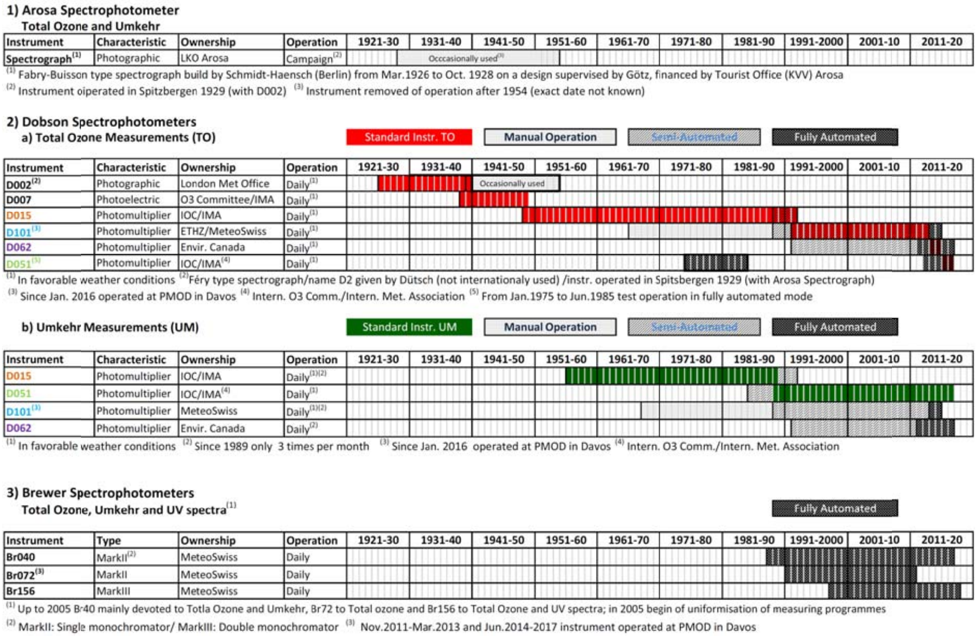
### 2.3. LKO under Götz

1926 was an important year for Götz. After the debate regarding cooperation between the Arosa and Davos medical doctors took place (for more details see Staehelin and Viatte, 2018) Götz moved into the "Villa Firnelicht" (see Fig. 3), which is very close to the Inner-Arosa Sanatorium, where measurements had previously been performed (see Fig. 2). Evidence suggests that Götz used family resources to build the large house, probably the inheritance from his father, Paul Götz, who owned a ironmongery ("Eisenwarenhandlung") in Göppingen (Trenkel, 1954) and died in 1926. "Villa Firnelicht" offered space for atmospheric observations on the roof and a balcony. It hosted three apartments and was therefore too large for just Götz and his wife. When Götz moved into "Villa Firnelicht" the institute was renamed to "Light Climatic Observatory" (Lichtklimatisches Obervatorium (LKO)). Götz invited colleagues to come to the LKO for sabbatical-type collaborations and to make atmospheric observations.

After the first conjectures that the amount of biologically active UV-radiation was determined by stratospheric ozone levels, Götz devoted a large part of his time to stratospheric ozone research. He realized that studying stratospheric ozone required suitable instrumentation and using resources from the KVV Arosa he mandated the Schmidt-Haensch company based in Berlin (Germany) to construct a Buisson-Fabry type of a sun spectrophotometer, with a design supervised by him. The instrument was delivered and used by Götz in hisexpedition in Spitzbergen (see below), but it is not known to us why it subsequently was only very rarely used. In 1926 Götz started a very fruitful collaboration with Gordon Dobson, a British physicist and meteorologist at the University of Oxford, who had just developed his first spectrophotometer (Walshaw, 1989). Götz began continuous total ozone measurements at Arosa using an instrument called a Fery spectrograph, which was developed by Dobson (Staehelin et al., 1998a). Later, Götz used improved sun spectrophotometers also constructed by Dobson (abbreviated as Dx, where x is the fabrication number; see Fig. 4). Dobson was very interested in the favorable climate and good weather and working conditions at the LKO. Thus, he arranged that the instruments were formally made available to the LKO through the International Association of Meteorology and Atmospheric Sciences (IAMAS, an association of the International Union of Geodesy and Geophysics (IUGG)). This allowed Götz to make total ozone observations at Arosa for many years, while it would have been very difficult for him to buy such spectrophotometers. After 1948 these instruments were formally borrowed through the International Ozone Commission (IO3C) of the IAMAS. Götz became one of the leading ozone researchers. He developed the "Umkehr method", which provided the first reliable information about the vertical ozone profile. This method is based on the "Umkehr effect", which Götz discovered during his expedition to Spitzbergen in 1929 (Götz, 1931). The first series of Umkehr measurements (besides a limited number of observations made in Oxford in 1931) was performed together with Dobson and his coworker Meetham on the roof of the "Villa Firnelicht" in 1932/33 (Götz et al., 1934).

**Figure 3.** "Villa Firnelicht", Götz's house in which the LKO, Götz's observatory was hosted (see text).

Götz was active in the international research community, as a member of the International Radiation Commission from 1932-1936 (Int. Rad. Com., 2008) and as a member of the IO3C from 1948, when it was formally established at the Seventh IUGG Assembly, until 1954 (Bojkov, 2012). Götz's research interests were broad, concerning many aspects of weather and climate, leading also to the publication of two books focusing to the statistical analysis of meteorological observations made at Arosa (Götz, 1926b; 1954).



**1) Arosa Spectrophotometer**
**Total Ozone and Umkehr**

| Instrument | Characteristic | Ownership | Operation |
|---|---|---|---|
| Spectrograph(1) | Photographic | LKO Arosa | Campaign(2) |

(1) Fabry-Buisson type spectrograph build by Schmidt-Haensch (Berlin) from Mar.1926 to Oct. 1928 on a design supervised by Götz, financed by Tourist Office (KVV) Arosa
(2) Instrument oiperated in Spitzbergen 1929 (with D002) (3) Instrument removed of operation after 1954 (exact date not known)

**2) Dobson Spectrophotometers**
**a) Total Ozone Measurements (TO)**

| Instrument | Characteristic | Ownership | Operation |
|---|---|---|---|
| D002(2) | Photographic | London Met Office | Daily(1) |
| D007 | Photoelectric | O3 Committee/IMA | Daily(1) |
| D015 | Photomultiplier | IOC/IMA | Daily(1) |
| D101(3) | Photomultiplier | ETHZ/MeteoSwiss | Daily(1) |
| D062 | Photomultiplier | Envir. Canada | Daily(1) |
| D051(5) | Photomultiplier | IOC/IMA(4) | Daily(1) |

(1) In favorable weather conditions (2) Féry type spectrograph/name D2 given by Dütsch (not internationaly used) /instr. operated in Spitsbergen 1929 (with Arosa Spectrograph)
(3) Since Jan. 2016 operated at PMOD in Davos (4) Intern. O3 Comm./Intern. Met. Association (5) From Jan.1975 to Jun.1985 test operation in fully automated mode

**b) Umkehr Measurements (UM)**

| Instrument | Characteristic | Ownership | Operation |
|---|---|---|---|
| D015 | Photomultiplier | IOC/IMA | Daily(1)(2) |
| D051 | Photomultiplier | IOC/IMA(4) | Daily(1) |
| D101(3) | Photomultiplier | MeteoSwiss | Daily(1)(2) |
| D062 | Photomultiplier | Envir. Canada | Daily(2) |

(1) In favorable weather conditions (2) Since 1989 only 3 times per month (3) Since Jan. 2016 operated at PMOD in Davos (4) Intern. O3 Comm./Intern. Met. Association

**3) Brewer Spectrophotometers**
**Total Ozone, Umkehr and UV spectra(1)**

| Instrument | Type | Ownership | Operation |
|---|---|---|---|
| Br040 | MarkII(2) | MeteoSwiss | Daily |
| Br072(3) | MarkII | MeteoSwiss | Daily |
| Br156 | MarkIII | MeteoSwiss | Daily |

(1) Up to 2005 Br40 mainly devoted to Totla Ozone and Umkehr, Br72 to Total ozone and Br156 to Total Ozone and UV spectra; in 2005 begin of uniformisation of measuring programmes
(2) MarkII: Single monochromator/ MarkIII: Double monochromator (3) Nov.2011-Mar.2013 and Jun.2014-2017 instrument operated at PMOD in Davos

**Figure 4.** Sun photometers operated at Arosa from 1921-present (for more details see Staehelin and Viatte in prep.).

During World War II, the KVV Arosa's financial support of the LKO was substantially decreased and Götz considered leaving Switzerland. Karl Wilhelm Franz Linke, professor and director of the Institute for Meteorology and Geophysics of the Goethe University of Frankfurt am Main (Germany) made him two offers to move to Frankfurt. At the same time Heinrich von Ficker, professor at the University of Vienna and director of the Central Institute for Meteorology and Geodynamics, asked Götz to become professor in Vienna (Austria). However, Götz decided to stay in Arosa in the Swiss Alps. If Götz had moved to Frankfurt or Vienna in World War II, the column ozone measurements made at LKO would likely have come to an end after just about one decade of measurements.

A few years after World War II, when modern antibiotics become available, the reasons for atmospheric studies related to tuberculosis therapy at LKO gradually became obsolete (Schürer, 2017). Moreover, many of the rich clients, who had been important to some of the sanatoria, no longer could afford to travel to Switzerland because of the 1930s economic depression. However, starting in the 1930s, Arosa was progressively promoted as a winter sport resort area. In November 1943, Götz provided a new justification for the measurements at LKO, proposing that the excellent air quality in Arosa was a "natural resource" and that such resort areas should quantify their air quality to obtain an objective grading (Götz, 1954). This proposal was part of a project for the "medical enhancement" of Switzerland's resort areas ("Medizinischer Ausbau der Kurorte"), which was termed "climate action" ("Klimaaktion") and funded by the Swiss Federal Office for Transport (Schweizerisches Bundesamt für Verkehr). Through this project, Götz obtained support to study air pollution by making surface ozone measurements. He was convinced that high ozone concentrations were an indication of healthy alpine air, since at the time polluted urban air had low ozone concentrations (caused by the high city-center NOx emissions). After World War II, Götz significantly increased efforts to obtain additional support for research at LKO by applying for a wide range of grants, which allowed him to hire collaborators who assisted him with measurements and scientific work.

Götz suffered from physical as well as mental (arteriosclerosis) health problems in the last years of his life (Trenkel, 1954) and he died at the age of 63 in 1954. Dr. Gertrud Perl was his main assistant from 1948 onwards, She continued making measurements even after Götz's death, but on the roof the Florentinum Sanatorium (see Fig. 2), because of difficulties with Götz's wife, who owned "Villa Firnelicht". Unfortunately, the Dobson instrument was damaged during transport to the Florentinum, so that there are a few months of data missing from the Arosa total ozone time series.

## 3. Period 1954-1962: First intermediate period

After Götz's death, it was uncertain for several years whether the measurements at LKO would at all continue. Jean Lugeon, the director of MeteoSwiss (Meteorologische Zentralanstalt at the time), supported the ozone measurements at Arosa during this critical period. He knew Götz personally, since they had taught together at the University of Zürich, and he was aware of the scientific value of the measurements. He was also the coordinator of the Swiss contribution to the International Geophysical Year (IGY) coming up 1958, in which total ozone measurements of Arosa were recognized as geophysically significant data set. For a few years, the Swiss National Science Foundation (SNSF) contributed to the salary of Perl in addition to the support received from the KVV Arosa and the Arosa municipality. From 1957 onwards, the Arosa total ozone measurements were additionally supported by MeteoSwiss. Hans-Ulrich Dütsch, a former graduate student of Götz (see Sect. 4.1), also played an important role for the continuation of ozone measurements at Arosa. He wrote a letter to the councilor of the Swiss Federal government in Bern responsible for the Federal Department of Home Affairs. In his response, the councilor indicated that MeteoSwiss could be mandated to assume the responsibility for the Arosa ozone measurements based on several resolutions of the World Meteorological Organization (WMO), which advised the national meteorological services to undertake ozone measurements. It was suggested that the Federal Meteorological Commission ("Eidgenössische Meteorologische Kommission"), the committee responsible for overseeing MeteoSwiss, should consider this in a comprehensive way, also looking at additional options, such as moving the LKO measurements to nearby Davos. Dütsch disagreed with the move to Davos, since he feared that this might lead to a serious discontinuity in the ongoing Umkehr measurements, because of larger aerosol contamination in Davos. In the end, the LKO stayed independent and was not integrated into MeteoSwiss, but MeteoSwiss and KVV Arosa provided financial support and measurements were continued at Arosa.

## 4. Period 1962-1985: Hans-Ulrich Dütsch

### 4.1. Dütsch and international ozone science

After Dütsch completed his PhD thesis in 1946 (title: “Photochemische Theorie des atmosphärischen Ozons unter Berücksichtigung von Nichtgleichgewichtszuständen und Luftbewegungen", Photochemical theory of atmospheric ozone under consideration of non-equilibrium states and air movements), he first worked as a physics teacher at a high school (Gymnasium) in Zürich. However, he remained interested in ozone research and eventually decided to pursue a career in science (see Fig. 5). From 1962-1965 he lived with his family in Boulder (Colorado, USA) while working as a researcher at the newly founded National Center for Atmospheric Research (NCAR). Together with Carl Mateer, Dütsch was the first to use modern computers to retrieve vertical ozone profiles with the Umkehr method.

In 1965 Dütsch was appointed as full professor at the ETH Zürich (ETHZ) where he served as director of the Laboratory of Atmospheric Physics (LAP, later to merge with the Institute of Climate Sciences to become the Institute for Atmospheric and Climate Science (IAC)). Dütsch’s research continued to focus on ozone, and thus he extended Swiss ozone measurements (see Section 4.2).

**Hans-Ulrich Dütsch**

| | |
|---|---|
| 1917 | Born on 26 Oct. in Winterthur (Switzerland)<br>Childhood in Winterthur |
| 1940 | Diploma in theoretical physics with a minor in meteorology, University of Zürich |
| 1943-1946 | Graduate student of Getz |
| 1947-1962 | High school (Gymnasium) teacher in physics in Zurich, continuing ozone research |
| 1950 | Visitor at Mass. Inst. Technol., MIT, USA |
| 1962-1964 | Researcher at the High Altitude Observatory in Boulder (CO, USA)<br>Head of the Ozone Research Program at the newly founded NCAR (CO, USA) |
| 1965-1985 | Professor at ETH-Zurich |
| 2004 | Died on 27 Dec. in Zürich (Switzerland) |

**Figure 5.** Biography of Hans-Ulrich Dütsch.

During Dütsch’s first years at ETHZ the main motivation for atmospheric ozone measurements at Arosa and Payerne was improving the understanding of the “high atmosphere” circulation patterns. This was with the aim of providing improved weather forecasts; the relationship between the vertical distribution of ozone and synoptic meteorological conditions was a major research topic in the 1960s and the early 1970s (see Breiland, 1964). Publications using measurements from the nearby Hohenpeissenberg Observatory (located in Bavaria, Southern Germany) revealed links between ozone levels and synoptic weather types (Hartmannsgruber, 1973; Attmannspacher and Hartmannsgruber, 1973, 1975).

Stratospheric ozone depletion resulting from anthropogenic emissions was first publicized in the 1970s. Molina and Rowland (1974) and Stolarski and Cicerone (1974) independently discovered that chlorine radicals destroy

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stratospheric ozone in a chain reaction. Furthermore, Molina and Rowland postulated that chlorofluorocarbons were a possible source gas for stratospheric chlorine. The chemical industry, with market leader DuPont, strongly objected to the view of Molina and Rowland. DuPont went so far as to launch an advertisement in the New York Times in 1975 stating that "Should reputable evidence show that some fluorocarbons cause a health hazard through depletion of the ozone layer, we are prepared to stop production of the offending compounds". This provided a new justification for making high quality total ozone measurements, namely as a basis for reliable long-term trend analysis. This was a new challenge for making ground-based total ozone measurements since stratospheric ozone (in extratropics) can vary by as much as ± 20 % from day to day, whereas anthropogenic stratospheric ozone changes were (and still are) on the order of only a few percent per decade.

Dütsch was one of the few scientists making important contributions to ozone research both before and after the debate on anthropogenic ozone depletion had started. Prior to this, Dütsch was largely curiosity-driven and had been interested in better understanding stratospheric ozone climatologies. For example, Dütsch (1974) provided basic science that served later to validate numerical simulations of anthropogenic ozone depletion. He also contributed to the IO3C, serving as member from 1957-1961, as secretary for 15 years (1961-1975), before being elected as president (1975-80), and being named an honorary member in 1984. He was also the main organizer of two important ozone symposia (the Quadrennial Ozone Symposia, organized for the IO3C) that took place in Arosa in 1961 and 1972. For more information on Dütsch's research, see also Staehelin et al. (2016.)

### 4.2 Ozone measurements at LKO under Dütsch

In 1956, Dütsch was able to find resources to put the Umkehr ozone measurements in Arosa on a regular, operational basis. When Gertrud Perl had to leave Arosa in 1962 because of health problems, Dütsch took the responsibility and scientific leadership of the LKO, although he was at that time still living in Boulder (CO, USA). A large part of the observations, particularly the Umkehr measurements, were performed by students, under the tutelage of Perl and others, until Kurt Aeschbacher became responsible for the LKO measurements in 1964, remaining so until November 2001. When Dütsch became professor at ETHZ in 1965, financial support of measurements at LKO (total ozone and Umkehr) continued as before (i.e., via KVV Arosa and Arosa municipality). In addition to the spectrophotometric measurements, Dütsch initiated ozone sonde measurements, which allowed obtaining detailed information on the ozone vertical profile. In 1966/67, these balloon measurements were operating from Kilchberg (close to Zürich), and were taken over in August 1968 by MeteoSwiss and made from Payerne, 140 km Southwest of Zürich, on the Swiss plateau (Jeannet et al., 2007). Since then, Payerne has become a member of "The Global Climate Observing System (GCOS) Reference Upper-Air Network" (GRUAN), which is an international observing network - under the auspices of WMO - of sites measuring essential climate variables above Earth's surface.

When Dütsch was responsible for the LKO, total ozone and Umkehr measurements were routinely performed using two Dobson spectrophotometers (see Fig. 4). To obtain the total ozone, only direct sun observations were performed. Dütsch applied the statistical Langley plot method to update the instrumental constants of the Dobson instruments every year (Dütsch, 1984). To apply the statistical Langley plot method a large number of ozone observations with different solar angles is required and therefore the observers need to choose suitable meteorological conditions, e.g. cloud free conditions lasting for at least several minutes. Each year Dütsch went to Arosa for several days to check all the total ozone measurements for reliability and to apply the statistical Langley plot method. This led to small corrections being made to the total ozone measurements for the previous year and some small changes to the instrumental constants for the following year. Students, who usually stayed in Arosa for several months at a time, made the Umkehr measurements, which need to be started prior to sunrise every morning.

In 1973, the LKO measurements were moved from the “Florentinum” to “Haus Steinbruch” (see Fig. 2), at a distance of a few hundred meters. The working conditions at the LKO were much better at “Haus Steinbruch” than at the “Florentinum”, however the running costs were more expensive (for more detail see Staehelin and Viatte, in prep.). In 1978, the first international intercomparison of Dobson spectrophotometers took place in Arosa. This was organized by Dütsch under the auspices of WMO. The results of this first intercomparison exercise at Arosa were not satisfying, e.g. as “differences between (standard) instruments led to a debate as to which should be used as the standard for the intercomparison” (see Staehelin et al., 1998a). However, this debate only deepened the insight how necessary such comparisons are, fostering the reputation of Swiss ozone research. Dütsch continued to apply the statistical Langely plot method to update the instrumental constants.

## 5. Period 1985-1988: Second intermediate period

### 5.1. International development and the importance of the Arosa total ozone time series

In the early 1980s, as new information about reaction rate constants in ozone chemistry became available, it seemed that chemical ozone depletion by ODSs was considerably less than had been predicted in the late 1970s (Benedick, 1991). However, in 1985 the Antarctic ozone hole was discovered (Farman et al., 1985), and the international ozone research community was able to demonstrate that the ozone hole was caused by the chlorine and bromine in halocarbons, which were largely of anthropogenic origin. New insight came through the discovery (Solomon et al., 1986) that the chlorine and bromine species are very efficiently converted into ozone destroying forms on the surface of polar stratospheric cloud particles, acting as efficient catalysts in the cold polar stratospheric vortex (for reviews see Rowland, 1991; Peter, 1997; Solomon, 1999).

In the mid-latitudes, the first analysis based on the by then still relatively short record of measurements by the Total Ozone Mapping Spectrometer (TOMS) instrument onboard the Nimbus 7 satellite also showed rapid ozone decline (Heath, 1988). However, ground-based total ozone measurements such as those made using Dobson instruments did not confirm the large downward trends suggested by the satellite data. (Data from most ground stations are deposited in the international data archive (presently World Ozone and Ultraviolet data center (WOUDC), presently operated by Environment and Climate Change Canada). This discrepancy led to the 1988 publication of the International Ozone Trend Panel report (IOTP, 1988). The report demonstrated that TOMS data available at the time were not reliable enough for trend analysis because of inappropriate treatment of the degradation of the diffuser plate. Later these data were reanalyzed more extensively using additional wavelengths in the retrieval algorithms and results were significantly improved (Stolarski et al., 1991). It turned out that also some of the data from the ground-based instruments were not of high enough quality to carry out reliable long-term trend analyses. This was attributed to calibration issues with the Dobson instruments, which showed frequent sudden changes when compared to TOMS overpass data (IOTP, 1988). Rumen Bojkov, Secretary of the IO3C (1984-2000), used TOMS data to provide "provisionally revised" ground based measurements, which, however, had weaknesses such as not correcting for sulfur dioxide ($SO_2$) interferences leading to potential errors in ozone trends based on Dobson series (e.g., De Muer and De Backer, 1992).

The most important application of the long-term measurements from Arosa (see Fig. 6) was probably their use in the 1988 IOTP report. The Arosa time series was the only Dobson dataset that required no correction and was much longer than any of the other ground-based measurement records. Results from Neil Harris's PhD thesis were published in the IOTP and showed, for the first time, significant decreases in stratospheric ozone in the northern mid-latitude winter season (Harris, 1989). He used two different approaches, namely (1) dividing the individual records into two periods of similar length using measurements going back to 1957 and (2) developing a novel multiple linear regression model taking into account trends for different months. In this model the downward trend started in 1970, and the analyses also showed that the negative trend was not sensitive to the start year. At present, standard Dobson measurements are based on observations of two (AD) wavelength pairs, which allow to minimize the interference by aerosols, a technique introduced during the International Geophysical Year (IGY) in 1957-58. To further support his main conclusion, Harris (1989) also used single other wavelength pair (C) data from Arosa, which are available as representative (homogenized) measurements since 1931. Again, he found similar negative total ozone trends at most other sites in the northern mid-latitudes (IOTP, 1988).

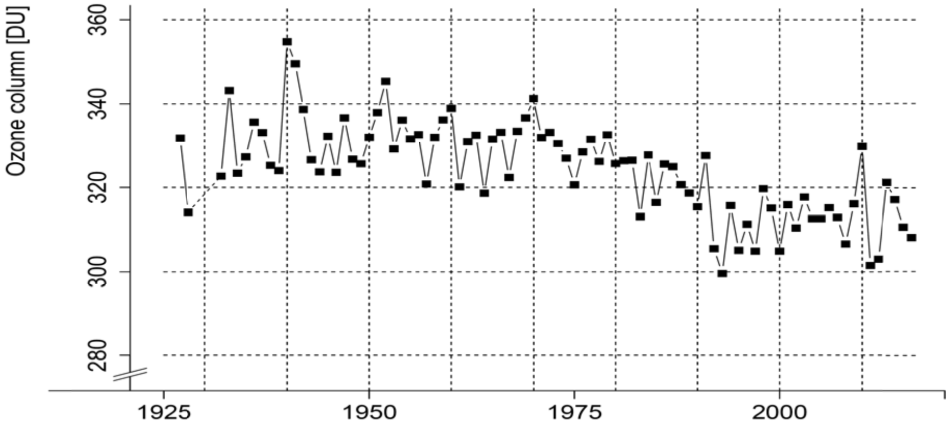

**Figure 6.** Annual mean total column ozone values measured at the world's longest continuous spectrophotometer site in Arosa, Switzerland, from 1926-present. The ozone column in Dobson units, where 100 DU correspond to a 1-mm thick slab of pure ozone gas at standard conditions (273.15 K, 1000 hPa).

### 5.2. Continuation of measurements at the LKO

After Dütsch's retirement in 1985, the continuation of Swiss long-term ozone measurements again became uncertain. The professor succeeding Dütsch focused on another research topic, and consequently the ETH Zürich argued that the continuation of operational ozone measurements did not fall under the responsibility of a university. Conversely, MeteoSwiss, which already was responsible for the ozonesonde measurements since 1968, argued that such long-term measurements needed scientific analysis by a well-qualified scientist, which MeteoSwiss was not able to support (a hiring freeze for permanent positions existed at the federal level at the time). Dütsch again wrote a letter to the responsible minister of the Federal government to point out the importance of the Arosa ozone measurements. Representatives from the Swiss Federal Office for the Environment (the "Swiss EPA") argued that ozone research in Switzerland needed to be continued since expert ozone researchers served a vital role to provide advice to policy makers regarding both stratospheric (in terms of the Vienna Convention and Montreal Protocol) and tropospheric ozone. Subsequently, a commission of the Swiss academy of Natural Sciences was tasked to analyze the situation. Government representatives as well as Swiss ozone researchers were invited to their meeting. Again, it was considered whether it made sense to move the LKO measurements to Davos (PMOD), but no decision was made in this regard. Nevertheless, MeteoSwiss and the ETH Zürich (i.e. IAC, Institute for Atmospheric and Climate Science, Laboratory of Atmospheric Physics (LAPETH) prior to 2001) agreed to continue the measurements, with the former officially accepting to take responsibility for the continuation of the ozone measurements at Arosa (total ozone and Umkehr) as well as the ozonesondes launched from Payerne, and the IAC at ETH Zürich consenting to continue ozone research. The agreement - implying that the person responsible for the LKO operations was moved to a MeteoSwiss position, whereas the IAC filled a scientific position with a major focus on ozone research became effective at the beginning of 1988.

## 6. Period 1988-2014: Ozone measurements and research at MeteoSwiss and IAC (ETHZ)

### 6.1. International Development: The Montreal Protocol

Since 1988, the most important justification for ozone measurements at LKO Arosa (total ozone und Umkehr) and ozone sonde launches in Payerne has been the documentation of the effect of ODSs on the stratospheric ozone layer and the effectiveness of the Montreal Protocol. Chemical ozone depletion by ODSs is expected to evolve very similar to the evolution of Equivalent Effective Stratospheric Chlorine (EESC). EESC provides an estimate of the total amount of halogens in the stratosphere, calculated from emission of chlorofluorocarbon and related halogenated compounds into the troposphere (lower atmosphere) and their efficiency in contributing to stratospheric ozone depletion (hence "effective"), and by taking the higher ozone destructiveness of bromine appropriately into account (hence "equivalent"). EESC peaked in the second half of the 1990s and subsequently showed a slow decrease, which is attributable to the Montreal Protocol, but in its slowness dictated by the long lifetimes of the emitted substances (see Fig. 7a). Total ozone measurements at Arosa are broadly consistent with long-term evolution of EESC (Staehelin et al., 2016) showing record low values in the early 1990s (Fig. 7b). The recovery of the ozone layer is a slow process and the signal of any sort of turnaround in the Arosa total ozone time series is still indistinct. Figure 7b shows the large interannual variability of the annual means, which is normal for a single measurement station and renders an attribution of the change in the downward trend difficult. While model results suggest that the Montreal Protocol and its amendments and adjustments have helped to avoid millions of additional skin cancer cases, Fig. 7b indicates that the global network of ozone station measurements needs to remain strong to in order to achieve a clear detection of the trend reversal and a proper attribution of the reasons.

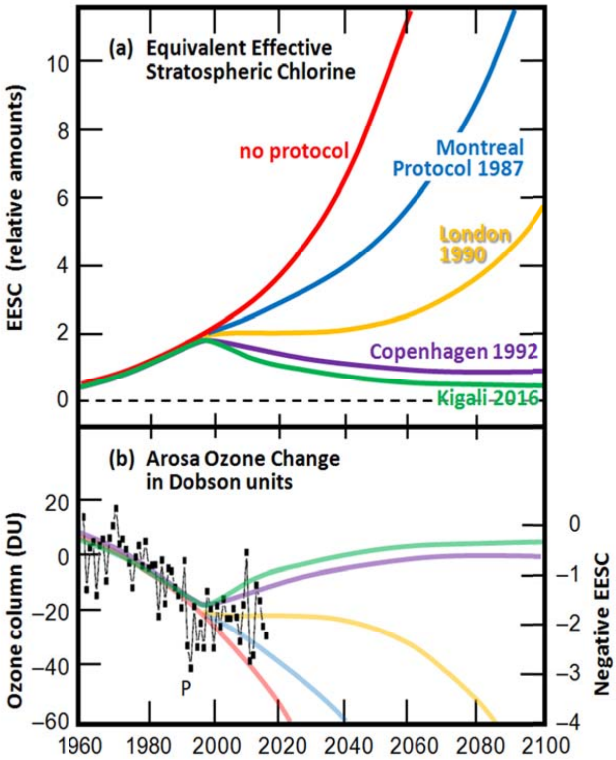

**Figure 7.** (a) Relative abundance of Ozone Depleting Substances (ODSs, i.e. volatile halocarbons) expressed as equivalent effective stratospheric chlorine (EESC) for the mid-latitude stratosphere, shown for various scenarios (demonstrating the impact of the Montreal Protocol and its subsequent Adjustments and Amendments). EESC can be viewed as a measure of chemical ozone depletion by ODSs and takes into account the temporal emission of the individual ODS species as well as their ozone depleting potential. (b) Arosa annual mean ozone columns (black symbols, as in Fig. 6) in comparison with the scenarios in (a). “P” marks the eruption of Mt. Pinatubo in 1991, which has aggravated the ozone loss.

## 6.2 LKO and related activities

### 6.2.1 Cooperation between MeteoSwiss and IAC (ETHZ)

The cooperation between MeteoSwiss and the IAC of ETH Zürich ensured that the different strengths of the two institutions were fully utilized. MeteoSwiss had the expertise and resources to renew the infrastructure at the Arosa station and was also able to guarantee reliable long-term operation through permanent contracts for technicians and scientists. On the other hand, IAC (ETHZ) had the possibility to lead scientific research, for example, with PhD theses that produced results published in the scientific literature. The use of ozone measurements as basis for scientific research requires high quality data and the results from the ETH studies thus provided both, a feedback mechanism in terms of data quality and enhanced visibility of the ozone measurements.

### 6.2.2 Renewal of the LKO infrastructure

When Meteoswiss become responsible for the LKO ozone measurements in 1988, the instrument infrastructure required renewal and extension. This was completed under the leadership of Bruno Hoegger and included constructing a spectrodome to house the two Dobson spectrophotometers as well as semi-automation of the Dobson total ozone measurements and full automation of the Dobson Umkehr measurements (Hoegger et al., 1992). Three Brewer instruments were also purchased between 1988 and 1998, thus allowing increased reliability of the Arosa total ozone series by complementing the Dobson Umkehr measurements and by providing instrumental redundancy (see Fig. 4). Furthermore, additional UVB measurements were added. Stübi et al. (2017a) demonstrated the excellent stability of the Arosa Brewer triad over the past 15 years.

### 6.2.3 Homogenization of the Arosa total ozone and Umkehr timeseries

The Dobson instrument D15 was the main instrument used to measure total ozone in Arosa from 1949 to 1992 (see Fig. 4). Archie Asbridge (formerly of Atmospheric Environment Canada) inspected this instrument after it was taken out of service in 1992, and it turned out that it had been operated in optical misalignment. Using the overlap between total ozone measurements of the D15 and D101 instruments, the latter of which was calibrated against the world standard instrument in 1986 and again in 1990, the Arosa column ozone time series was adjusted to the scale of the world primary Dobson instrument (for more detail see Staehelin et al., 1998a and Scarnato et al., 2010). The Arosa Umkehr timeseries also required homogenization (Zanis et al., 2006).

### 6.2.4 Foci of scientific studies since the 1990s

The comparison of the unique Arosa total ozone timeseries from Dobson and Brewer instruments has allowed studies of the differences between the two instrument types (Staehelin et al., 1998a; Scarnato et al., 2009, 2010) as well as their long-term behavior since they are calibrated in different networks. In the 1990s, quantification of the downward ozone trends was the main reason for making long-term stratospheric measurements (comp. Section 5.1, and Staehelin et al., 1998b, 2001). These trends were seen as a consequence of increasing ODS concentrations. Subsequent studies were also devoted to understanding the potential contribution of other processes enhancing the observed downward trends, including long-term climate variability, e.g. in connection with tropopause altitude (Steinbrecht et al., 1998) and the North Atlantic Oscillation (NAO) or Arctic Oscillation (AO) (Appenzeller et al., 2000; Steinbrecht et al., 2001; Weiss et al., 2001). The unique length and high quality of the Arosa total ozone and Umkehr measurements also meant they were important for the EU project CANDIDOZ (Chemical and Dynamical Influences on Decadal Ozone Change; Zanis et al., 2006; Brunner et al., 2006; Harris et al., 2008). Later, as the ODS concentrations have decreased, documentation of the "turn around" in stratospheric ozone trends became more and more important (e.g. Mäder et al., 2010). The Arosa time series was also used to introduce the concept of extreme value theory in ozone science (Rieder et al., 2010a, b). This allowed to attribute extreme ozone values to events of various origins, such as dynamical factors as ENSO or NAO, or chemical factors, such as cold Arctic vortex ozone losses, or major volcanic eruptions of the 20th century, e.g. Mt. Pinatubo.

### 6.2.5 Tropospheric ozone

The surface ozone measurements from Arosa are unique and very valuable for tropospheric chemistry studies. Surface ozone was measured already in the 1930s by Götz to quantify the contribution of tropospheric ozone to the total column, and later continued by the careful and representative surface ozone measurements made in the 1950s (Götz and Volz, 1951; Perl, 1961). Thanks to these measurements it was possible to show that surface ozone concentrations increased by more than a factor of two from the 1950s to 1990 (Staehelin et al., 1994). This has commonly been attributed to the large increase in ozone precursor emissions (nitrogen oxides, volatile hydrocarbons, and carbon monoxide) resulting from the strong economic growth in industrialized countries following World War II. The surface ozone measurements made at Arosa and Jungfraujoch were pillars in the studies of Parrish et al., (2012, 2013), which contributed to an important report by the Task Force of the Hemispheric Transport of Air Pollution (HTAP). HTAP was organized in 2005 under the auspices of the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (LRTAP Convention) to study intercontinental transport of ozone in northern mid-latitudes. Based on these data, Parrish et al. (2014) compared three state-of-the-art chemistry climate models (CCMs) to show that simulated surface (baseline) ozone trends over Europe were about a factor two smaller than those seen in the available observations.

## 7. Future of ozone measurements at the LKO

### 7.1 International Demands

There is a general demand of proof of the effectiveness of the Montreal Protocol and a heads-up of how climate-related changes will affect the ozone layer, i.e. what the impacts are of the anticipated stratospheric cooling and the enhanced Brewer-Dobson circulation, and what this means for polar, mid-latitude and tropical ozone.

Recovery of the stratospheric ozone layer in response to the reduction of ODS concentrations controlled by the Montreal Protocol is slow (see Sect. 6.1) and requires continued long-term stratospheric ozone observations. ODSs most directly impact ozone in the upper stratosphere, where photolysis leads to the release of halogen radicals from these species. Extensive data analyses carried out under the auspices of the SI2N activity (commonly sponsored by SPARC (Stratosphere-troposphere Processes and their Role on Climate), IO3C, IGACO-O3/UV (Integrated Global Atmospheric Composition Changes), and NDACC) (Network for Detection of Atmospheric Composition Changes) highlighted issues related to the availability and uncertainty of measurements, such as of merged satellite datasets, and trend analysis techniques. (See the special journal issue jointly organized between Atmospheric Chemistry and Physics, Atmospheric Measurement Techniques, and Earth System Science Data: Changes in the vertical distribution of ozone – the SI2N report). Recently, Steinbrecht et al. (2017) presented the latest analysis of upper stratospheric ozone trends confirming the expected increase in upper stratospheric ozone in extratropics. It will be important to continue high quality stratospheric ozone measurements to be able to follow the slow recovery of the ozone layer in response to the changing burden of stratospheric ODSs, including nitrous oxide ($N_2O$), which is likely to become the dominant species for stratospheric ozone depletion in future (Ravishankara et al., 2009; Portmann et al., 2012).

Climate change will modify the distribution of stratospheric ozone in different ways (see e.g. Arblaster et al., 2014). Increasing greenhouse gases cause decreasing stratospheric temperatures modifying reaction rates leading to increasing extratropical stratospheric ozone concentrations. At the same time, however, the polar stratospheres are not expected to cool on average. Furthermore climate change is expected to enhance the Brewer Dobson Circulation which transports ozone from the main tropical source region to the extra-tropics (Butchart, 2014). Modification of the Brewer Dobson Circulation is expected to increase stratospheric ozone in the mid-latitudes above levels of recovery in response to the decrease in ODSs alone. This has been termed “super recovery”. ~~On~~ contrast, the enhanced transport out of the tropics is expected to result in a decrease in stratospheric ozone in these regions. The enhancement of the Brewer Dobson Circulation is, however, still under debate, with state-of-the-art CCMs projecting an increase but only controversial observational evidence being available. Importantly, the expected enhancement depends strongly on the climate change scenario investigated, thus it is essential that high quality measurements are continued. The unique length of the Arosa timeseries is particularly useful for documenting the effects of climate change on ozone since the dataset covers a period of almost 40 years when the stratosphere was relatively undisturbed by anthropogenic influence, about 25 years in which anthropogenic ODSs increased in (stratospheric) concentration, and the latest period with the slow decrease of EESC. The Arosa timeseries will therefore play a crucial role in the coming decades to further document ozone changes in the Northern mid-latitudes, including the predicted “super recovery” expected to become important around 2030 (e.g. Hegglin et al., 2015).

### 7.2 Continuation of measurements at the LKO

The MeteoSwiss board of directors decided in 2015 to explore the possibility of moving the Arosa measurements to the PMOD in Davos. Such a move could not only help to master financial restrictions, but might also offer the advantage of the excellent technical infrastructure, platforms and expertise that is available at PMOD in Davos. Within this program the Dobson instruments are currently completely automated (comp. Fg. 4). However, before such a move is to take place, an adequate period of overlapping measurements at both sites (Arosa and Davos) is essential. A break in the world's longest total ozone time series would be very unfortunate. However, the relocation is particularly challenging as stratospheric recovery from ODS is expected to be slow (see Sec. 6.1) leading to small ozone changes and requiring therefore measurements of very high quality (i.e. very high stability). At present simultaneous total ozone measurements of Brewer instruments of ~~Dvaos~~ and Arosa have been analyzed and presented (Stübi et al., 2017b)

## 8. Summary and Conclusions

Homogenous long-term records such as the total ozone record from Arosa are very valuable for trend analyses in climate science. Reliable long-term, ground-based total ozone measurements are also crucial for validation of ozone observations from space, particularly in terms of validating the long-term stability of merged satellite datasets (e.g. Labow et al., 2013). The extraordinary length of the Arosa record was particularly important for a wide range of studies, including the analysis of stratospheric ozone variability related to long-term climate variability such as NAO/AO (Appenzeller et al., 2000) and El Nino Southern Oscillation (Brönnimann et al., 2004) as well as the evaluation of the (early part of the) Twentieth Century Reanalysis Project (Compo et al., 2011; Brönnimann and Compo, 2012).

Justification for the LKO measurements changed from (1) study of environmental factors possibly important for the recovery from tuberculosis, to (2) study of air quality being an important natural resource in resort areas, to (3) enhancing understanding of atmospheric physics to improve weather forecasts, to (4) quantification of anthropogenic ozone destruction by ODSs, and finally to (5) document the effectiveness of the Montreal Protocol. In future, if stratospheric ozone gradually recovers as expected in response to the decreasing burden of ODSs, continued observations will be necessary to document the effects of climate change on stratospheric ozone, as predicted to by CCMs, i.e., through enhancement of the Brewer Dobson Circulation. The reasons for continuing the Arosa measurements have thus changed many times over the past decades. Initially it was never imagined that such a long record would have been made. A key element for this success was the motivation of the scientists and technicians involved: it appears that it was Götz's initiative that started field observations at Arosa, and twice the efforts of Dütsch were crucial in ensuring that measurements continued.

It is difficult to obtain funding for such continuous observations through normal science funding agencies such as the Swiss National Science Foundation (SNSF), since an additional few years of measurements usually do not result in novel scientific conclusions; this has been experienced by several other networks, for example, Network for Detection of Atmospheric Composition Changes (NDACC). The success of the Montreal Protocol probably contributed to the decrease in number of ozone measurements submitted to WOUDC that took place in the last years (Geir Braathen, personal communication). This might exacerbate in future as cost of monitoring cost are under pressure in many countries. However, we believe that such routine measurements are the responsibility of developed countries. Institutions like national meteorological services, although they also may experience financial shortfalls, are ideally suited to carry out these types of measurements since they are (in contrast to universities) capable of making long-term commitments and have the possibility to hire permanent staff, something which is becoming more and more difficult at modern universities. Universities have the advantage of being able to focus on particular issues (e.g. through PhD theses) for a limited time, resulting in articles in peer-reviewed journals. Here it is important to stress the relevance of scientific activities using long-term observations. Excellent collaboration has existed between MeteoSwiss and IAC (ETHZ) for the past three decades, however, this particular type of cooperation seems less and less feasible in future as the permanent scientific position required to maintain this is no longer supported by ETHZ. In other countries the required research is integrated in the same institution (e.g. the German Weather Service (DWD) in Germany or the "Centre National de la Recherche Scientifique (CNRS)" in France) - a problem that still waits for proper solution for the Swiss longterm ozone measurements..

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