Reply to Referee 1

We would like to thank Referee 1 for his/her very competent and careful review and the valuable suggestions. We first show the comments of the reviewers, following our replies.

This is the first review of the paper by Staehelin et al. titled "Stratospheric ozone measurements at Arosa (Switzerland): History and scientific relevance". The manuscript depicts the history of Arosa's longest ozone record and describes different periods in the record. It is very important to understand the circumstances that brought public interest and financial support for the ozone research at Arosa. From the very beginning, the need for the continuous surface and total column ozone measurements was publicized to benefit public health (i.e. studies of the tuberculosis, UV-exposure related cancer, pollution, etc.). The importance of continuous measurements has been recognized due to short term (meteorological) and inter-annual (Dobson-Brewer circulation) variability that impacts ozone measurements. The laboratory studies of the CFCs in 1970-1980s and their link to the destruction of stratospheric ozone required verification by observations. It was possible through analyses of very long and stable record that provided information on ozone depletion detected in the 1990s. Researchers used Arosa record for ozone trend analyses and attribution of changes to anthropogenic activities, volcanic aerosols and climate drivers. The manuscript also outlines management approach that allowed Arosa research to continue with data collection during multiple potential interruptions for funding sources. This is very important lesson to current observational programs struggling with financial support. There are lessons learned on how to create successful project through collaboration between different organizations, including perseverance of scientists reaching out to the state government programs and private sector, and using scientific publications to emphasize the importance of the continued ozone record. Acknowledgement of the importance of ozone observations by international bodies of the World Meteorological organization and the Parties of the Montreal Protocol has been indeed very important tool used by observatories to seek continuous observation funding. There is also a concern about future move of Dobson instruments to Davos. This is not the first time when the merger of the Arosa and Davos ozone observing programs was proposed. In the past, it was possible to argue about importance of the continuous record and impacts of the local meteorological and anthropogenic influences on the record that can interfere with monitoring of ozone recovery. The studies of comparability of the records between Brewer measurements at two locations has been published that argue that both locations show little difference. However, it is still important to validate the move of the historical Dobson instruments to Davos. It is very important to provide information of the move to scientific community and engage scientists in the research of the impacts of location on continuous longest ozone record in the World.

Here are my suggestion for the paper.

1) *It will be nice to create the graph that would show the time line that combines information about the subject of the research at Arosa with funding information, and also includes important research publications dates in the time line. It may be possible to add this information in Figure 4 (Figure needs to be enlarged for final publication – too small font and hard to read).*

Our reply: We followed the idea of the referee and produced an overview Figure, Figure 9 in the revised manuscript. We took the data of Fig. 6 instead of Fig. 4 (originally submitted manuscript) as proposed by the referee.

2) *In Conclusion section, it would be nice to have "Lessons Learned" for managing of the station operations: few bullets about actions that helped to organize data gathering and research.*

Our reply: We followed the advice of the referee summarizing the most important points to run the station in bullets points (see Section 9 (Summary and Conclusions), 4. paragraph in the revised manuscript).

Detailed comments:

p. 5 line 143-144 "After the first conjectures that the amount of biologically active UV-radiation was determined by stratospheric ozone levels" – is there a reference for the paper? Is it Götz, 1926.

Our reply: The title of the paper Götz 1926 shows that Götz was aware that ozone is responsible for the transparency of the atmosphere for solar UV-B radiation. This relationship was first discussed by Hartley (1871). However, we wanted to emphasize that Götz who was interested to understand UV radiation at the Earth's surface started to study stratospheric ozone because he was not satisfied by measurement of UV-climatology. In a presentation to the Royal Institute of Public Health in May 1929 he explains his scientific approach (which was obviously different to the view of Dorno from Davos): "From the beginning the measurements of the UV light in Arosa are not only designed on a statistical basis, but also driven by the desire to understand the variability. And the observed higher UV-levels in the autumn with respect to the spring observed in the series 1921 to 1924 can only be explained by the weaker ozone layer in the autumn. The variations of the ozone layer which has first been observed quantitatively by Fabry and Buisson are decisive for the comprehension of the behavior of the UV-Radiation which is essential for the life" (for more detail see Staehelin and Viatte, in prep.). We modified the manuscript, see Section 2.3, 2. paragraph.

p.10, line 297, please provide the year when Payerne joined the GRUAN program.

Member since 2008, (fully) certified in 2015 (see revised manuscript, Section 4.2, 1. paragraph)

p.10, lines 297-298, "To apply the statistical Langley plot method a large number of ozone observations with different solar angles is required and .." – Was Langley plot method applied to other Dobson instruments outside of Arosa? What is the limitation of the Langley method and why it was not successful at other stations?

Our reply: The statistical Langely plot method was used in the Halley Bay total ozone record of the British Antarctic survey, which was used to discover the ozone hole over Antarctica (Farman et al., 1985, see Section 4.2, second paragraph in the revised manuscript). The application of the statistical Langley plot method requires a large number of individual measurements covering an extended range of my-values of a large number of days and the application of the method is time consuming. I think the requirement of a large number of measurements and the rather time consuming data analysis are the reason why the method is not commonly used as well as the regular intercomparisons organized under the auspices of WMO.

p. 15, line 438-440. Please provide information about

Our reply: For technical information concerning new electronics and use of Brewer spectrophotometers at Arosa we refer to the report of Staehelin and Viatte, which is in prep. whereas the use of Arosa data to study temperature sensitivity is discussed in the first paragraph in 6.2.4 in the revised manuscript.

p. 16, lines 452-469. This section needs to be expanded to provide more information on how Arosa data contributed to the AO, NAO and ENSO studies. It is mentioned very briefly now, but it is very important contribution.

Our reply: We extended the discussion in the revised manuscript.

p. 16, line 484, since this paper discusses the value of the long-term observations, it would be good to state that the observations provides reference for validation and improvement of the models.

Our reply: We added Eyring et al., 2013 as reference

p. 18, Summary and Conclusion: It would be nice to organize this section to more clearly emphasize benefits of Arosa record to the ozone science. You already have statements in regards to validation of satellite stability, consistency in the merged satellite records. I wonder if it can be separated into individual paragraphs that can read as an expanded bullets.

Our reply: We summarized our experience from the last decades in form of bullet points (see above)

p. 18, lines 549-553. I suggest adding the years of Arosa record to each of the justifications. It will show reader how sometimes focus of the society comes back in later years when more theoretical information and observations become available to renew the subject of the early studies.

Our reply: We modified the manuscript accordingly (Section 8, second paragraph).

p.18, lines 553-555 – this needs to be a separate paragraph concentrating on the need for future ozone measurements. The validation of future satellite stability, resolving differences between satellite

records, and help to bridge possible breaks in the satellite records can be mentioned here. As well as continuous validation of the CCM, CTM and regional model, can be added to this paragraph.
Our reply: The points related to the future including validation of numerical simulation are discussed in the second paragraph of Section 8 in the revised manuscript.
Mentioning of the Copernicus project is also important. Help with validation of the new ozone measuring instruments is one of the Dobson network objectives. Providing training for future scientists, including training of operators from developing countries must be mentioned in this section.
Our reply: These points are addressed in the last paragraph of Section 8.
p. 18 lines 555-559 – this is very important section on the management of the funding for station observations, including personal communications of scientists with funding agencies. This needs to be emphasized in the "lessons learned" .
Our reply: This discussion is covered by 5. paragraph of Section 8.
p. 18, line 565 – remove second "cost".
Our reply: Done
p18, lines 560-566, p. 19 lines 567-578. It is important to mention guiding role of the WMO ozone SAG committee and Ozone research managers' report that set goals for the WMO Dobson network. It seems that this venue should be used more often than not to support the funding needs for individual countries. You should also emphasize that Arosa record on its own would not be able to succeed or become widely used without collaborations with the WMO Dobson network and NDACC. The recent efforts in the Brewer COST action project are aimed at creating the organized collaborative network of ozone measurements in Europe and homogenization of the near-real time processing of ozone data from multiple Brewer instruments. The effort to retain all individual station data on the same scale was the most important step in creating the homogenized operations for the Dobson network in 1980s.
Our reply: We tried to cover these points in the last paragraph of Section 8.
We also improved several formulation in the manuscript.

Referee 2: I found the paper very interesting, as I'm not a specialist in stratospheric ozone so historical details of how some of the researched developed was new to me. Overall the paper is well written and I only saw a couple of minor grammatical errors. It gives a detailed description of how total column ozone measurements were initiated at Arosa with the aim of studying the impact of mountain air on recovery from tuberculosis and eventually contributed to our understanding of the stratospheric ozone layer and the damage being caused to it by our emissions. The story it tells of pressures from competing scientists and institutes or the difficulties in maintaining funding, illustrate the ways in which science develops in the face of many challenges. It highlights the importance of maintaining high-quality and long-term measurement facilities as the information they give can evolve with time as we become aware of new processes the consequences of human activity. The site is of continued importance for studying the impact of climate change on ozone as well as other factors. My only suggestion would be that the abstract could possibly put a greater emphasis on the value of the historical measurements and continuing them into the future, in the light of climate change. Grammatical notes are in the attached pdf.

Please also note the supplement to this comment: https://www.atmos-chem-phys-discuss.net/acp-2017-1079/acp-2017-1079-RC2supplement.pdf: Could you add a couple of sentences to outline the basic operation of these types of instrument and why the Umkehr was such a significant improvement? Not all readers may be familiar with the optical methods used to measure total column ozone.

Johannes Staehelin, 15.032018

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

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Abstract. In 1926 the stratospheric ozone measurements were started atof the Light Climatic Observatory (LKO) of Arosa (Switzerland) started, marking the beginningstart of the world's longest series of total (or column) ozone measurements. Theyse measurements were driven by the recognition of the importance of atmospheric ozone is important for human health, as well as by scientific curiosity about what was, at the time, an illin this by then not well characterized atmospheric trace gas. From around the mid-1950s to the beginning of the 1970s studies of high atmosphere circulation patterns that could improve weather forecasting was justification for studying stratospheric ozone. 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. In the mid-1970s, a paradigm shift occurred when it became clear that the damaging effects of anthropogenic Ozone Depleting Substances (ODSs), such as long-lived chlorofluorocarbons, needed to be documented. This justified continuing the ground-based measurements of stratospheric ozone. Levels of ODSs peaked around the mid-1990s as a result of a global environmental policy to protect the ozone layer, implemented throughby the 1987 Montreal Protocol and its subsequent amendments and adjustments. Consequently, chemical destruction of stratospheric ozone started to slow ozone depletion caused by ODSs stopped worsening around the mid-1990s. To some extent, this raises the question as to whether continued ozone observation are indeed necessary. In the last decade there has been a 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. Ttendency to reduce the costs associated withies of increased cost savings in making ozone measurements globally seem perceptible worldwide, also including at Arosa. However, the large natural variability in ozone on diurnal, seasonal, and interannual scales complicates the ability to demonstrate the success of the Montreal Protocol. AndMoreover, chemistry-climate models predict a "super-recovery" of the ozone layer at mid-latitudes 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. These factors, and identifying potentially unexpected stratospheric responses to climate change, support the continued need to document stratospheric ozone changes. This is particularly valuable at the Arosa site, due to the unique length of the observational record. This paper presents the evolution of the ozone layer, and the history of international ozone research, and discusses the justification for of thesese measurements in the for past, present and into future.

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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). This~~e~~ long total ozone dataset is extremely valuable for long-term trend analyses of stratospheric ozone. In addition, other important ozone measurements, such as Umkehr and surface ozone measurements were also made at~~re being performed in~~ Arosa. Since the 1970s, when anthropogenic stratospheric ozone depletion became a subject of public concern, the measurements at LKO grew in importance ~~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 societal justification~~ to society~~ for these measurements over~~throughout~~ the long history of the LKO, particularly highlighting the link~~ 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 stored~~treasured~~ in the LKO archives located at MeteoSwiss 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. Following Staehelin and Viatte (in prep.) we divide~~d~~ the history of LKO into five distinct periods (see Sections 2-6 below). Section 7 looks at the potential pathways into ~~includes some remarks on~~ the future of measurements at the LKO. Finally~~, 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

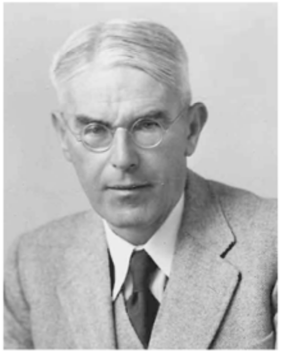
The first ozone measurements at Arosa were a part of medical research focused on the treatment of pulmonary tuberculosis (TB). Before modern antibiotics became available (a few years after World War II), TB was considered as a serious ill~~sick~~ness with high mortality rates. The best available therapy for treating~~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 doctors in Davos at the time, 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 more 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 indeed the most helpful type of therapy before treatment by antibiotics became possible.

The medical doctors of Davos and Arosa were convinced that the high altitude climate was an important factor for optimal recovery from TB. To study this further, the potentially relevant environmental factors needed to be investigated. Already i~~I~~n 1905, Turban proposed opening an institute aimed to study the scientific effectiveness of the "rest cure therapy" of pulmonary~~lung~~ TB (SFI, 1997). However, because of a lack of consensus among medical doctors, this ~~such an~~ institute was founded only 17 years later 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 staying in the town, who needed to 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 pulmonary~~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 Observatory Dorno~~"Prof. Dorno Institute"~~, the nucleus of the renoned~~world famous~~ Physical Meteorological Observatory Davos (PMOD), which~~ serves~~ since 1971 also serves as the World Radiation Center (WRC) of the World Meteorological Organization (WMO), a center for international calibration of meteorological radiation standards within the global network. When Dorno retired as director in 1926, the institute was integrated as an independent department into the Swiss research institute for high mountain physiology and tuberculosis research in Davos and was financed by the Davos community, similar to the other institutes. Despite numerous~~several~~ studies, however, it was never shown that~~not possible to demonstrate the superiority of~~ the Alpine climate was a superior environment for recovery from pulmonary~~(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 pulmonary~~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) after which~~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 some months during the ~~an unknown duration (~~1919-1920 period~~)~~. See Staehelin and Viatte (in prep.) 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	Founding of Lightclimatic Observatory (LKO) at Arosa
193~~2~~1	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	I~~Physical and mental i~~llness (including 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~~ initiate~~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. It was~~, mainly~~ supported mainly 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 nowadays are subject of communal responsibility~~currently fall under the responsibility of the municipality~~. Götz's~~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 objectives of~~measurements taken at~~ the LKS were to complement the meteorological observations made at Arosa since 1884 by the Swiss national weather service (now "MeteoSwiss") by~~. These atmospheric~~ measurements which were thought to be relevant for studying the recovery from pulmonary TB. Thus in 1921 Arosa was the first municipality to finance an institute with the task of studying environmental factors favorable to curing (pulmonary~~lung~~) TB. The support Götz obtained from the KVV Arosa was rather modest and he later ~~he~~ secured additional regular funding from, ~~both~~ the Chur-Arosa railway company,~~ and~~ the Arosa municipality and the canton of Grisons (for more detail see Staehelin and Viatte, in prep.). The LKS measurements were made on the roof of the Inner-Arosa Sanatorium, where nowadays the "Grand Hotel Tschuggen" is located ~~at present~~ (see Fig. 2).

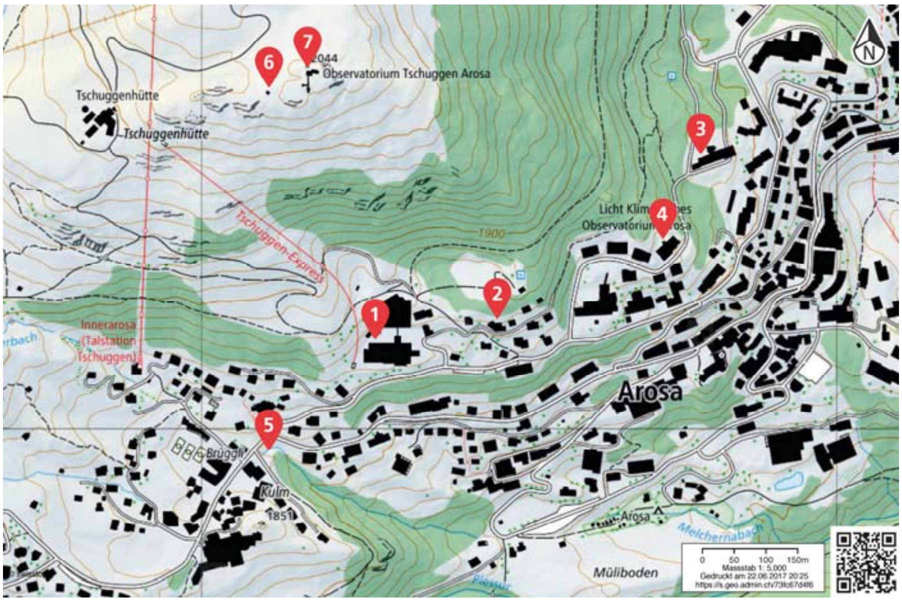


Figure 2. Map of important locations relevant to the Arosa Light Climatic Observatory (LKO). LKO measurement sites: (1) Sanatorium Inner-Arosa; (2) Villa Firnelicht; (3) Florentinum; (4) Haus zum Steinbruch. Other sites: (5) Götzbrunnen (fountain in honor of Götz); (6) hHut where Götz made his nighttime measurements in Tschuggen; 7) aAstrophysical 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 Section 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. This turned out to be, a very important result, which would later contributing help to develop a better understanding the global issue of stratospheric circulation patterns. This seasonal cycle represents one pillar on which the modern understanding of the Brewer-Dobson circulation rests. In fact, Götz published tThis 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 surfacedbecame more evident with time. It also appears there were issues between the physiciansmedical 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. T, but it seems that these efforts failed probably since members of the Davos community wanted a largerhigher financial contribution from Arosa forto 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.

Figure 3. “Villa Firnelicht”, Götz’s house in which the LKO, Götz’s observatory was hosted (see text).

2.3. LKO under Götz

1926 was an important year for Götz. After the sobering debate regarding cooperation between the Arosa and Davos medical doctors ~~took place~~ (for more details see Staehelin and Viatte, in prep. ~~2018~~) Götz moved into the “Villa Firnelicht” (see Fig. 3), which is very close to the Inner-Arosa Sanatorium, where measurements had ~~previously~~ been previously 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 an 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 the "Light Climatic Observatory" (Lichtklimatisches Obervatorium (LKO)). Götz invited colleagues to come to the LKO for sabbatical-type collaborations and to make atmospheric observations.

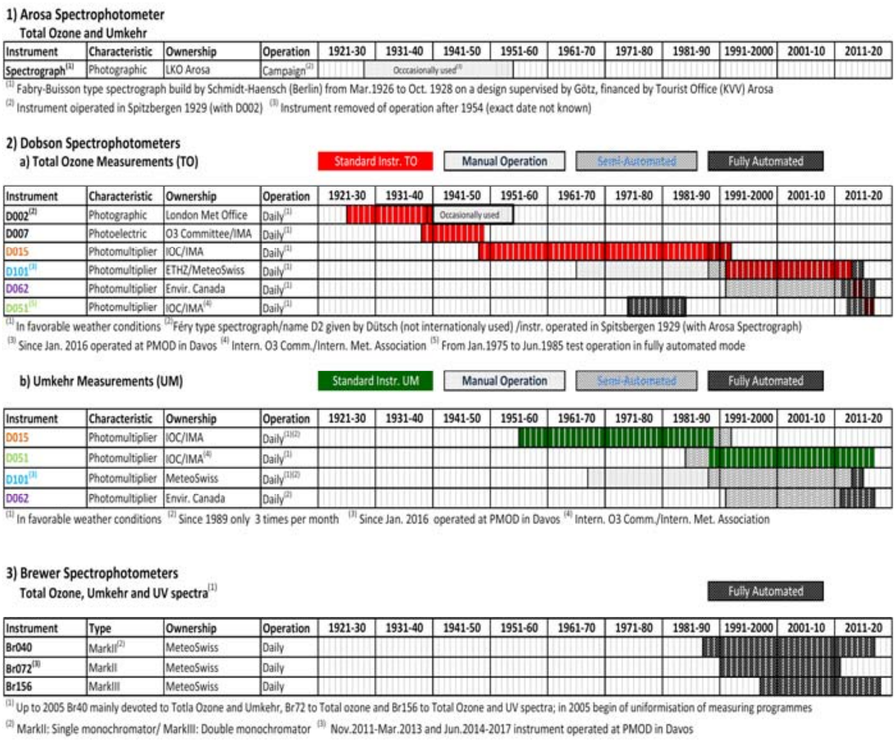


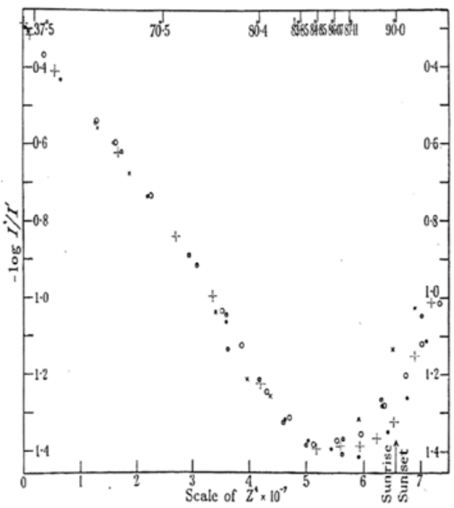
Figure 4. Sun photometers operated at Arosa from 1921-present (for more details see Staehelin and Viatte in prep.).

Hartley (1881) was the first to postulate that atmospheric ozone is responsible for absorbing solar light in the UV-B spectrum. Because the amount of biologically active UV-radiation is~~was~~ determined by stratospheric ozone levels, Götz devoted a large part of his time to stratospheric ozone research (see Staehelin and Viatte, in prep.). 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 his expedition to~~in~~ Spitzbergen (see below), but it is un~~not~~ known to us why it was 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, since~~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. The sun photometers constructed by Dobson measure the intensity of solar radiation at wavelength pairs in the range of 300-340 nm at the Earth's surface. Three different types of instruments were constructed by Dobson (Dobson, 1968) which are shortly characterized in Fig. 5. In order to minimize the falsifying effects of atmospheric aerosols on total ozone measurements the two wavelengths pairs method was introduced during the International Geophysical Year (1958).

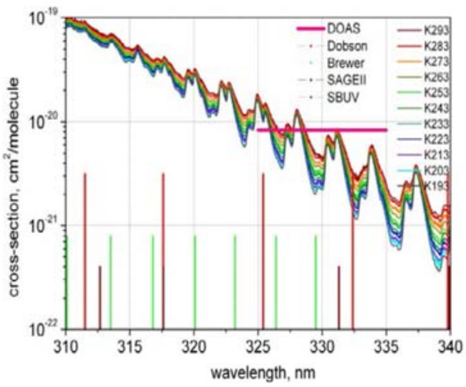
Götz became one of the leading ozone researchers. In the second half of the 1920s and the first half of the 1930s a key research question was how ozone is distributed in the vertical. Surface measurements e,g, from Arosa indicated low tropospheric ozone concentrations and rather unprecise measurements suggested ozone maxima in the mid-latitudes (in partial pressure) at altitudes of around 40-50 km (see Dobson, 1968). The Umkehr method developed by Götz et al., 1934 (see Fig. 5), however, showed maximum concentrations rather at 20-22 km. This was considered a scientific breakthrough providing 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).

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 International Ozone commission (IO3C) created in~~from~~ 1948, when it was formally established at the Seventh IUGG Assembly, until 1954 (see Bojkov, 2012). Götz's research interests were broad, concerning many aspects of weather and climate, and le~~a~~d him~~ing also~~ to publish ~~the publication of~~ two books on focusing to the statistical analysis of radiation measurement and meteorological observations made at Arosa (Götz, 1926b; 1954).



Ozone profile by the **Umkehr method**: Zenith sky measurements (wavelengths pair C) as function of time including sunrise or sunset (time is represented by the solar zenith angle written at the top of the Figure at the left, from Götz et al., 1934). Zenith solar sky radiation at surface is determined by ozone absorption and scattering. Zenith sky radiation at surface is progressively diminished by atmospheric ozone absorption near sunset; when the sun reaches the lowest elevation angles the scattering at higher altitudes becomes predominant which causes the reversal (Umkehr). Umkehr curves contain information on ozone profile which can be determined by a retrieval algorithm.

Wavelengths used in total (column) ozone measurements (see text):
Féry spectrograph (photographic detection): wavelengths pairs: 306.2/326.4; 305.2/323.2; 302.2/326.4
Dobson instrument with photoelectric detection: 311.0/330.0
Dobson instrument with photomultipliers:
wavelengths pairs: A: 305.5/325.4; B: 308.8/329.1; C: 311.45/332.4; D: 317.6/339.8.
After International Geophysical Year (IGY (1958): AD wavelengths pairs used to minimize aerosol interference.



(World primary) Dobson instruments are calibrated by the Langley plot method.

Ozone absorption cross sections in the Huggins band at different temperatures and wavelengths used in different instruments (ACSO, 2015, Figure 3).

Figure 6. Ozone observations by instruments designed by Dobson.

During World War II, the KVV Arosa's financial support forof 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 heGötz had moved to Frankfurt or Vienna duringin World War II, the column ozone measurements made at LKO would likely have come to an end after just about one decade of measurements.

Already during the 1930s economic depression, rich clients, who had been important to some of the sanatoria, no longer could afford to travel to Switzerland. Moreover, a~~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 one characteristic ~~an indication~~ of healthy alpine air, since at that~~e~~ time the (heavily) polluted urban air had low ozone concentrations (caused by the high city-center NOx emissions titrating ozone). 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.

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In the last years of his life Götz suffered from ~~physical as well as mental (arteriosclerosis)~~ health problems (including arteriosclerosis) ~~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 and ~~, S~~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" the LKO had to move to the Florentinum Sanatorium (see Fig. 2) at the end of 1953. 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 during this period.

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) in~~coming up~~ 1958, in which the total ozone measurements at~~of~~ Arosa were recognized as a geophysically significant data set. For a few years, the Swiss National Science Foundation (SNSF) contributed to Perl's~~the~~ salary ~~of Perl~~ in addition to the support received from the KVV Arosa~~, and~~ the Arosa municipality and the canton Grisons. 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 head (minister) councilor of ~~the Swiss Federal government in Bern responsible for~~ the Swiss

Federal Department of Home Affairs in Bern. In his response~~,~~ we read ~~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 thate 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 that were started in 1956 by Dütsch (see Section 4.2), 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 airflow ~~movements~~), he first worked as a physics teacher (mainly) 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. 6~~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~~ merged in 2001 with the Institute of Climate Sciences to become today's ~~he~~ Institute for Atmospheric and Climate Science (IAC)). Dütsch's research continued to focus on ozone, and he ~~thus he~~ continued, pursued and extended the Swiss ozone measurements (see Section 4.2).

Hans-Ulrich Dütsch

1917	Born on 26 Oct. in Winterthur (Switzerland)
	Childhood in Winterthur
1940	Diploma in theoretical physics with a minor in meteorology, University of Zürich
1943-1946	Graduate student of Göetz
1947-1962	High school (Gymnasium) teacher in physics in Zurich, continuing ozone research
1950	Visiting scientist or at the Massachusetts. Institute of . Technology., MIT, USA
1962-1964	Researcher at the High Altitude Observatory in Boulder (CO, USA)
	Head of the Ozone Research Program at the newly founded NCAR (CO, USA)
1965-1985	Prof.essor at the Swiss Federal Institute of Technology Zürich (ETH Zürich) Zurich
2004	Died on 27 Dec. in Zürich (Switzerland)

Figure 65. 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) and the relationship between the vertical distribution of ozone and synoptic meteorological conditions become a important research topic in the 1960s and the early 1970s (see Breiland, 1964).

Stratospheric ozone depletion resulting from anthropogenic emissions was first publicized in the 1970s. Molina and Rowland (1974) as well asand Stolarski and Cicerone (1974) independently discovered that chlorine radicals destroy stratospheric ozone in a chain reaction. Furthermore, Molina and Rowland postulated that chlorofluorocarbons were a possible source gas for stratospheric chlorine. The chemical industry, particularlywith 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 the extra tropics) 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

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4.2 Ozone measurements at LKO under Dütsch

In 1956, Dütsch was able to find resources to ensure~~put~~ the Umkehr ozone measurements in Arosa continued 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) at the time. A large majority ~~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 for the~~of~~ measurements at LKO (total ozone and Umkehr) continued as before (i.e., via KVV Arosa,~~ and~~ Arosa municipality and the Canton Grisons). In addition to the spectrophotometric measurements, Dütsch also initiated ozone sonde measurements, which made it possible to observe ~~allowed obtaining detailed information on the~~ ozone vertical profile in more detail. In 1966/67, these balloon measurements were operated by Dütsch ~~ing~~ from Kilchberg (close to Zürich), but~~and were taken over~~ in August 1968~~ by~~ MeteoSwiss took took over these observations and made them from Payerne, 140 km Southwest of Zürich~~,~~ on the Swiss plateau (Jeannet et al., 2007). In 2008 ~~Since then,~~ Payerne ~~has~~ beca~~o~~me a member of "The Global Climate Observing System (GCOS) Reference Upper-Air Network" (GRUAN) (fully certified in 2015), ~~which is~~ an international observing network ~~-~~under the auspices of WMO. GRUAN aims at~~ of sites~~ measuring essential climate variables providing long-term, high-quality climate data records from the surface, through the troposphere, and into the stratosphere~~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 (which was also used by Farman et al., 1985) 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 (see Fig. 6).

In 1973, the LKO measurements were moved from the "Florentinum" to "Haus Steinbruch" (see Fig. 2), just ~~at a distance of~~ a few hundred meters away. The working conditions at the LKO were much better at "Haus Steinbruch" than at the "Florentinum", however the running costs were higher more expensive (for more detail see Staehelin and Viatte, in prep.). In 1978, the first international intercomparison campaign of Dobson spectrophotometers took place in Arosa. This was organized by Dütsch under the auspices of the WMO. The results of this first intercomparison exercise at Arosa were not satisfying since, 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 into how necessary such comparisons were (and still are)are, fostering the excellent reputation of Swiss ozone research. As a result of these discrepancies Dütsch continued to apply the statistical Langley plot method to update the instrumental constants up to the begin of the 1990s.Dütsch continued to apply the statistical Langely plot method to update the instrumental constants.

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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 ozone chemistry 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) discovery that the chlorine and bromine species are very efficiently converted into ozone destroying forms on the surface of polar stratospheric cloud particles (Solomon et al., 1986), 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 fromby the Total Ozone Mapping Spectrometer (TOMS) instrument onboard the Nimbus 7 satellite available at the time 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. 76) 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 (cf. Fig. 5). To further support his main conclusion, Harris (1989) also used single other wavelengths 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).

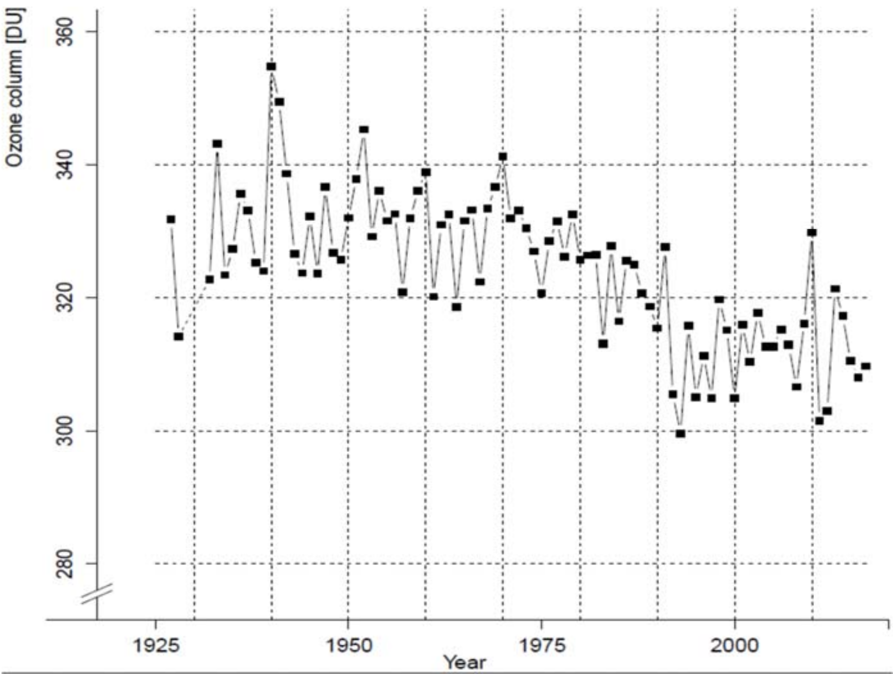

Figure 76. Annual mean total (column) ozone values measured at the world's longest continuous spectrophotometer site in Arosa, Switzerland, from 1926-present. The ozone column in Dobson units, where 100 DU correspond to a 1-mm thick slab of pure ozone gas at standard conditions (273.15 K, 1000 hPa).

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. 8~~7~~a). 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. 8~~7~~b). 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 8~~7~~b 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. 8~~7~~b 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.

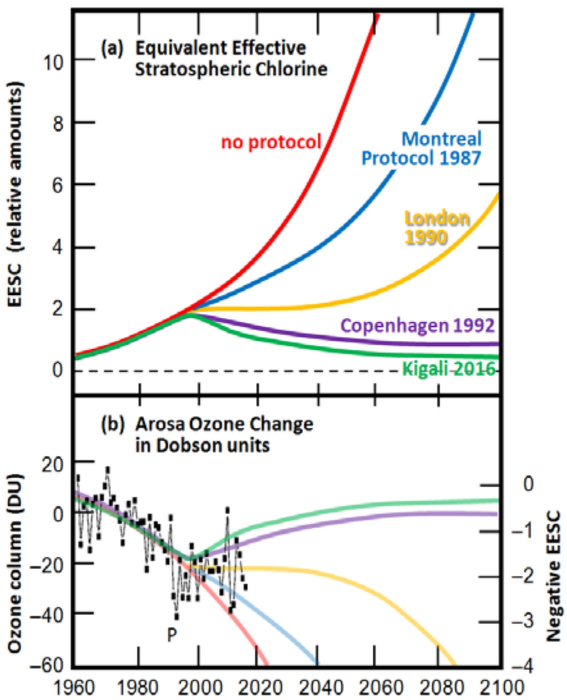

Figure 8~~7~~. (a) Relative abundance of Ozone Depleting Substances (ODSs, i.e. volatile halocarbons) expressed as equivalent effective stratospheric chlorine (EESC) for the mid-latitude stratosphere, shown for various scenarios (demonstrating the impact of the Montreal Protocol and its subsequent Adjustments 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. 7~~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 (ETH Zürich~~Z~~) 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. For more technical information including new electronics see Staehelin and Viatte, in prep. S~~S~~tübi et al. (2017a) demonstrated the excellent stability of the Arosa Brewer triad over the past 15 years.

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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 time series 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 time s~~s~~eries 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. The large data set of quasi-simultaneous measurements was particularly valuable for studying the effect of temperature dependence of ozone absorption cross-sections on total ozone measurements attributable to the different wavelengths used in Dobson and Brewer instruments (Scarnato et al., 2009, Redondas et al., 2014). These results were an important contribution to the GAW ACSO (Absorption Cross-Sections of Ozone) project in which available laboratory cross-sections of atmospheric ozone measurements were studied (ACSO, 2015; Orphal et al., 2016).

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. related to~~in connection with~~ tropopause altitude (Steinbrecht et al., 1998) and climate patterns (Steinbrecht et al., 2001). The unique length of the Arosa total ozone series was very valuable in demonstrating that the North Atlantic Oscillation (NAO) or Arctic Oscillation (AO) enhanced downward winter ozone trends in central Europe for the period up to the mid1990s ~~and the North Atlantic Oscillation (NAO) or Arctic Oscillation (AO)~~ (Appenzeller et al., 2000;~~ Steinbrecht et al., 2001;~~ Weiss et al., 2001). Brönnimann et al. (2004a, 2004b) also showed that the record high values of total ozone at Arosa that occurred in the early 1940s were due to an increase in strength of Brewer Dobson circulation caused by a very large ElNino/Southern Oscillation anomaly during that period.

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~~ attribution~~e~~ of extreme ozone values to events of various origins, ~~such as~~ dynamical features such~~actors~~ 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.

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6.2.5 Tropospheric ozone

The surface ozone measurements from Arosa are unique and very valuable for tropospheric chemistry studies. Surface ozone ~~was~~ measurements~~d~~ were begun already in the 1930s by Götz to quantify the contribution of tropospheric ozone to the total column, and were 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. This result was recently confirmed by Staehelin et al., 2017.

7. Future of ozone measurements at the LKO

7.1 International Demands

Policy makers ~~There is a~~ and the general public would like to see ~~demand of~~ proofs of the effectiveness of the Montreal Protocol and to better understand~~a~~ ~~heads-up of~~ how climate~~-related~~ change~~s~~ will affect the ozone layer, i.e. what are the impacts ~~are~~ of the ~~anticipated~~ stratospheric cooling and the anticipated enhanced Brewer-Dobson circulation on ozone, 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. Recent examples are~~, such~~ ~~as of~~ merged satellite datasets, and trend analysis techniques~~.~~ (s~~S~~ee 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 a recent~~the~~ ~~latest~~ analysis of upper stratospheric ozone trends confirming the expected increase in upper stratospheric ozone in extratropics. Finally, Ball et al (2018) showed that total ozone in the mid-latitudes has not increased as expected and their careful analysis of mostly satellite measurements indicated a downward trend in the lower stratosphere (15-22 km) which continued since 1987. The physical cause of this surprising trend is presently unknown and requires further study.

It is vital ~~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, which in turn modify~~ing~~ reaction rates and lead~~ing~~ to increasing extra-tropical stratospheric ozone concentrations. This is not the case over the poles, where ~~At the same time, however,~~ the ~~polar~~ stratosphere~~s~~ is~~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 production~~source~~ region to the extra-tropics (Butchart, 2014). Modification of the Brewer Dobson Circulation is expected to increase stratospheric ozone in the mid-latitudes to levels above~~-~~ those seen in the past; t~~levels of recovery in response to the decrease in ODSs alone. T~~this has been termed "super recovery". In~~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 in the stratosphere, and the latest period with the slow decrease in stratospheric ODS concentrations~~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 would result in reduced measurement costs in combination with ~~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 the PMOD in Davos. Within this activity~~program~~ the Dobson instruments are currently completely automated (comp. Fig. 4). However, before such a move is to take place, a multiannual ~~n 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. A~~However, the~~ relocation is particularly challenging as stratospheric recovery from ODS is expected to be slow (see Sec. 6.1) meaning~~leading to small~~ ozone changes will be small and thus~~requiring therefore~~ ~~measurements of~~ very high quality (i.e. very high stability) measurements are required. At present simultaneous total ozone measurements of Brewer instruments of Dav~~va~~os 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). Furthermore, they serve as conclusive test in evaluating numerical simulations such as Chemistry Climate models (CCMs) which are used to make projections of future ozone evolution (see e.g. Eyring et al., 2013, Arblaster et al., 2014). 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 the NAO/AO (Appenzeller et al., 2000) and El Nino Southern Oscillation (Brönnimann et al., 2004a and 2004b). Furthermore, the measurements were very valuable for ~~as well as~~ the evaluation of the (early part of the) Twentieth Century Reanalysis Project (Compo et al., 2011; Brönnimann and Compo, 2012).

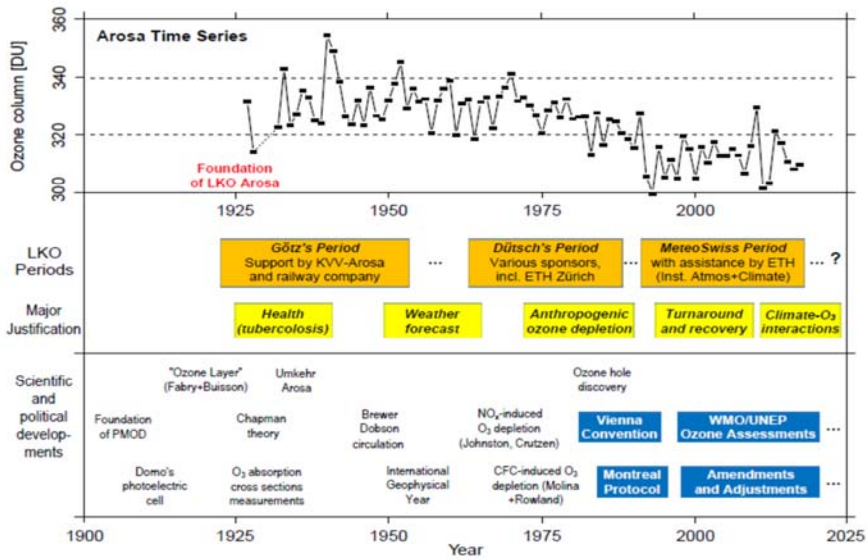

Figure 9. Historical overview of the successive periods of Light Climatic Observatory of Arosa (LKO). Total ozone measurements (top, annual means); different phases during the history of LKO including main sponsors (in orange), justification of measurements for society (in yellow); milestones in international ozone research, and international legislation (blue).

~~Fig.~~The reasons for continuing the Arosa measurements have changed many times over the past decades and initially it was certainly never imagined that such a long record could be established. Fig. 9 provides a historical overview of international ozone research in connection with the different phases of the LKO indicating also various funding periods. Justification for the LKO measurements for society can be summarized as~~underwent a number of different phases~~

(1) study of environmental factors possibly important for the recovery from pulmonary TB~~tuberculosis~~ (relevant from the beginning until around World War II),
(2) investigating~~study~~ of air quality as~~being an~~ important natural resource in resort areas (as discussed in the second part of World War II)
(3) improving our understanding of atmospheric physics to improve weather forecasts (important in the 1960s and early 1970s)
(4) quantification of anthropogenic ozone destruction by ODSs (mid-1970s to mid-1990s)
(5) documentating the effectiveness of the Montreal Protocol in saving ozone (since around the middle of the 1990s)
(6) understanding the mutual relationship between climate change and global ozone depletion and the effectiveness of the Montreal protocol (this century).

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A key element for the success of LKO measurements and continuation was the motivation of the scientists involved, Götz's early initiative and Dütsch's persistence.

From our experience the following points were most relevant for successful operation of LKO for the last decades:

- Redundancy allows increasing credibility of measurements, which is particularly important for reliable long-term trend analysis. At Arosa, 3 Dobson and 3 Brewer spectrophotometers were simultaneously operated since 1998, which helped to obtain important scientific results regarding Dobson and Brewer spectrophotometers relevant in the broader context of atmospheric ozone measurements.
- Regular comparison of station instruments with standard spectrophotometers operated under the umbrella of WMO are important for high-quality measurements and comparability of ozone measurements within a particular network.
- Scientific analysis and use of stratospheric ozone measurements in scientific publications and model intercomparisons not only enhances visibility of the measurements within the community, but also is a quality assessment, which might motivate scientists and technicians operating the measurements.
- Finally, reliable techniques are important for high quality stratospheric ozone measurements including automation to lower manpower costs and to make measurements less dependent on the skills of an individual operator.

It is difficult to obtain funding for 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 is something that has been experienced by several other networks as well, for example NDACC. The success of the measures induced by the Montreal Protocol probably contributed to the decrease in number of ozone measurements submitted to the World Ozone and Ultraviolet Data Center (WOUDC, presently operated by Environment and Climate Change Canada) over the past few years (Geir Braathen, personal communication). This might be exacerbated in future as monitoring costs come under further 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. On the other hand, 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. It is important to stress the relevance of scientific activities using long-term observations. Excellent collaboration has existed between MeteoSwiss and the IAC (ETHZ) for the past three decades. However, this particular type of cooperation will be less feasible in future, as the required permanent scientific positions will typically no longer be available at universities. In other countries the research aspects are often 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). This problem still waits for proper solution for the Swiss long-term ozone measurements.

From the very beginning, the ozone measurements from Arosa (initiated by the fruitful collaboration between Götz and Dobson) have been an important contribution to both, the global network of ozone measurements and to ozone research. During the early part of the record, the International Ozone Commission (IO3C) of IAMAS coordinated the ozone measurements. Since the 1970s WMO has taken the lead, first in the framework of the Global Ozone Observing System (GOO3S) and later the Global Atmosphere Watch (GAW) programme (SAG-ozone) became responsible for overseeing and coordinating stratospheric ozone measurements to obtain and maintain high quality data suitable for long-term trend analysis. GAW might continue these activities in collaboration with other networks, such as NDACC, the present Brewer COST network, and the IO3C in order to (i) maintain and extend high quality records of ground-based ozone stations and (ii) to continue comparisons of Dobson and Brewer measurements with other/new instruments such as SAOZ and PANDORA. GAW might represent the ground-based community as partners with the satellite community, for example, in the Copernicus project and contribute to research programs and initiatives as illustrated by the long history of ozone research connected with LKO started by the pioneers Götz and Dütsch and continued more recently by MeteoSwiss and ETHZ under the auspices of WMO, IGACO-O3/UV, ACSO, and SPARC.

Beyond any doubts the Montreal Protocol (including enforcements) was very successful to protect the ozone layer over densely populated areas from large damage by manmade chemicals as shown by extended numerical simulations (Newmann et al., 2009). In future, when the ~~if~~ stratosphere~~ric ozone~~ is expected to gradually recover~~s~~ ~~as expected~~ from~~in response to~~ the decreasing burden of ODSs, continued observations will not only be required to document the expected increase in stratospheric ozone~~be necessary~~, but rather to document the effects of climate change on stratospheric ozone, as predicted to by CCMs, i.e., through enhancement of the Brewer Dobson Circulation and possible other effects connected with climate change (Ball et al., 2018).

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