

1 **Stratospheric ozone measurements at Arosa (Switzerland):** 2 **History and scientific relevance**

3 Johannes Staehelin ¹⁾, Pierre Viatte ²⁾, Rene Stübi ²⁾, Fiona Tummon ¹⁾, Thomas Peter ¹⁾

4 ¹⁾ Institute for Atmospheric and Climate Science, ETHZ, Zürich

5 ²⁾ Federal Office of Meteorology and Climatology MeteoSwiss, Payerne

6 *Correspondence to:* Johannes Staehelin (johannes.staehelin@env.ethz.ch)

7 **Abstract.** In 1926 stratospheric ozone measurements were started at the Light Climatic Observatory (LKO) of
8 Arosa (Switzerland), marking the beginning of the world's longest series of total (or column) ozone measurements.
9 They were driven by the recognition atmospheric ozone is important for human health, as well as by scientific
10 curiosity about what was, at the time, an ill characterized atmospheric trace gas. From around the mid-1950s to
11 the beginning of the 1970s studies of high atmosphere circulation patterns that could improve weather forecasting
12 was justification for studying stratospheric ozone. In the mid-1970s, a paradigm shift occurred when it became
13 clear that the damaging effects of anthropogenic Ozone Depleting Substances (ODSs), such as long-lived
14 chlorofluorocarbons, needed to be documented. This justified continuing the ground-based measurements of
15 stratospheric ozone. Levels of ODSs peaked around the mid-1990s as a result of a global environmental policy to
16 protect the ozone layer, implemented through the 1987 Montreal Protocol and its subsequent amendments and
17 adjustments. Consequently, chemical destruction of stratospheric ozone started to slow around the mid-1990s. To
18 some extent, this raises the question as to whether continued ozone observation are indeed necessary. In the last
19 decade there has been a tendency to reduce the costs associated with making ozone measurements globally
20 including at Arosa. However, the large natural variability in ozone on diurnal, seasonal, and interannual scales
21 complicates the ability to demonstrate the success of the Montreal Protocol. And, chemistry-climate models predict
22 a "super-recovery" of the ozone layer at mid-latitudes in the second half of this century, i.e. an increase of ozone
23 concentrations beyond pre-1970 levels, as a consequence of ongoing climate change. These factors, and identifying
24 potentially unexpected stratospheric responses to climate change, support the continued need to document
25 stratospheric ozone changes. This is particularly valuable at the Arosa site, due to the unique length of the
26 observational record. This paper presents the evolution of the ozone layer, the history of international ozone
27 research, and discusses the justification for the measurements in the past, present and into future.

28

29 **1. Introduction**

30

31 The world's longest time series of total (or column) ozone observations is from Arosa in the Swiss Alps, made at
32 the "Light Climatic Observatory" (Lichtklimatisches Observatorium, LKO). This long total ozone dataset is
33 extremely valuable for long-term trend analyses of stratospheric ozone. In addition, other important ozone
34 measurements, such as Umkehr and surface ozone measurements were also made at Arosa. Since the 1970s, when
35 anthropogenic stratospheric ozone depletion became a subject of public concern, the measurements at LKO grew
36 in importance (Staehelin et al., 2016). A comprehensive report on the history of the LKO is presently in preparation
37 (Staehelin and Viatte, in prep.). Here we focus on the societal justification for these measurements over the long

38 history of the LKO, particularly highlighting the link to the development of international stratospheric ozone
39 research. This paper is based on the extensive correspondence by F. W. Paul Götz - ozone pioneer and founder of
40 the LKO - which is stored in the LKO archives located at MeteoSwiss in Payerne, Switzerland, on the annual
41 reports of the “Kur- und Verkehrsverein Arosa” (KVV Arosa, see below), and on other research. Following
42 Staehelin and Viatte (in prep.) we divide the history of LKO into five distinct periods (see Sections 2-6 below).
43 Section 7 looks at the potential pathways into the future of measurements at the LKO. Finally, a summary and
44 conclusions is presented in Section 8.

45 **2. Period 1921-1953: Friedrich Wilhelm Paul Götz**

46

47 **2.1. Therapy for tuberculosis prior to the availability of antibiotics**

48

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

60 The medical doctors of Davos and Arosa were convinced that the high altitude climate was an important factor for
61 optimal recovery from TB. To study this further, the potentially relevant environmental factors needed to be
62 investigated. Already in 1905, Turban proposed opening an institute aimed to study the scientific effectiveness of
63 the “rest cure therapy” of pulmonary TB (SFI, 1997). However, because of a lack of consensus among medical
64 doctors, this institute was founded only 17 years later in 1922. On 26 March 1922, the municipality of Davos
65 (“Landsgemeinde”) decided to create a foundation for an institute for high mountain physiology and tuberculosis
66 research (“Institut für Hochgebirgsphysiologie und Tuberkuloseforschung”, today the “Schweizerisches
67 Forschungsinstitut für Hochgebirgsklima und Medizin, SFI” in Davos). The resources for operating the institute
68 mainly originated from a small fee that was paid by all guests of staying in the town, who needed to register when
69 staying in Davos (a form of “tourist tax”).

70 At this point, Carl Dorno played an important role. He was a rich industrialist from Königsberg (Germany), who
71 came to Davos because his daughter suffered from pulmonary TB. She unfortunately passed away a few years
72 after arriving in Davos, but Dorno remained and founded an institute to study the environmental factors important
73 for treating TB using his own funds in 1907 (SFI, 1997). During the first World War and in the subsequent period
74 of inflation, Dorno lost most of his financial resources. On 18 February 1923, the municipality of Davos decided
75 to support the Observatory Dorno, the nucleus of the renowned Physical Meteorological Observatory Davos

76 (PMOD), which since 1971 also serves as the World Radiation Center (WRC) of the World Meteorological
 77 Organization (WMO), a center for international calibration of meteorological radiation standards within the global
 78 network. When Dorno retired as director in 1926, the institute was integrated as an independent department into
 79 the Swiss research institute for high mountain physiology and tuberculosis research in Davos and was financed by
 80 the Davos community, similar to the other institutes. Despite numerous studies, however, it was never shown that
 81 the Alpine climate was a superior environment for recovery from pulmonary TB (Schürer, 2017).

82

83 2.2. F.W.P. Götz and the foundation of the LKO (LKS)

84

85 Friedrich Wilhelm Paul Götz grew up in Southern Germany (Göppingen, close to Stuttgart) and went to Davos for
 86 the first time prior to the beginning of the First World War to recover from pulmonary TB, when he was working
 87 on his PhD thesis in astronomy (see Fig. 1). He stayed twice in the “Deutsche Heilstätte” sanatorium (1914-1915)
 88 after which he was released as “fit for work”. For the following years (1916-1919) he intermittently taught at the
 89 “Fridericianum” German school in Davos and later worked with Dorno (probably for some months) during the
 90 1919-1920 period. See Staehelin and Viatte (in prep.) for more details.

91



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
1931	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	Illness (including arteriosclerosis)
1954	Died on 29 Aug. in Chur (Switzerland)

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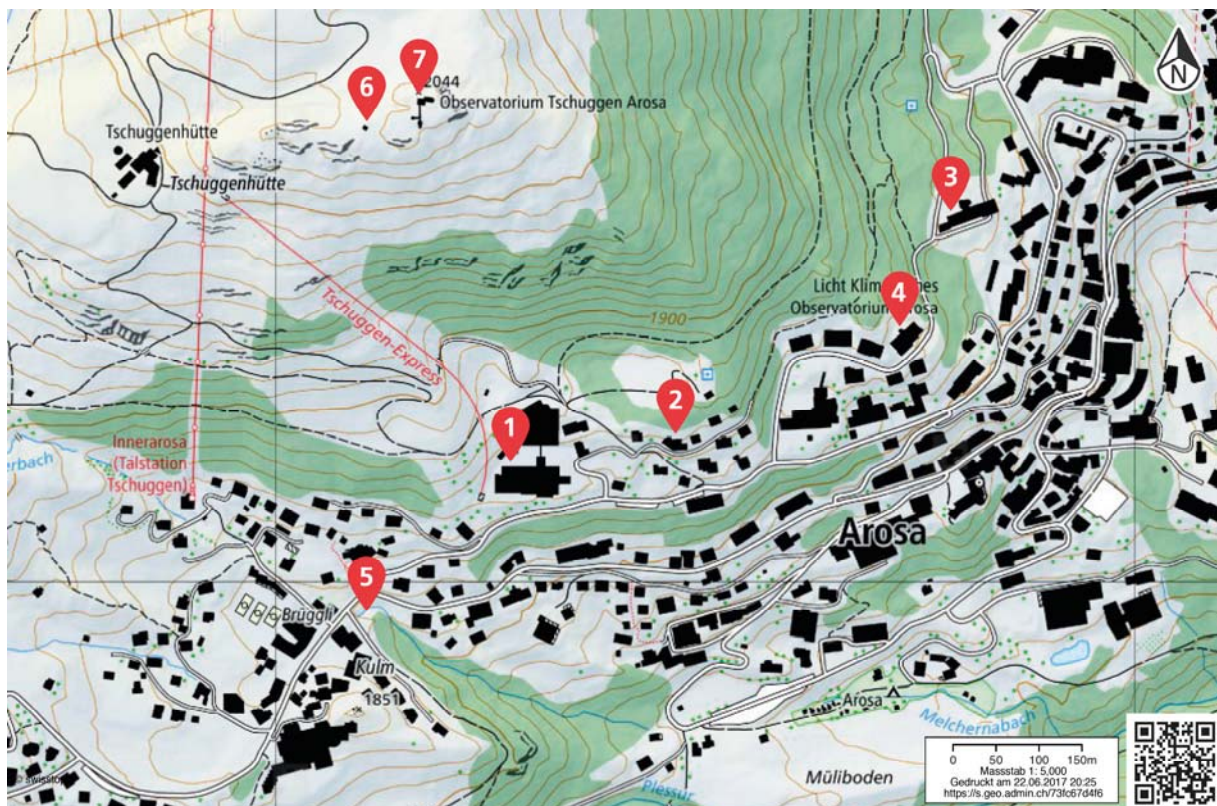
93 **Figure 1.** Biography of F.W. Paul Götz, founder of the Light Climatic Observatory in Arosa.

94

95 It appears that Götz was the main driver behind the initiative to make atmospheric measurements at Arosa. He
 96 likely first contacted the Arosa medical doctors and together they subsequently made a request to the managing
 97 committee of the KVV Arosa in March 1921 to initiate climate studies relevant for health. The KVV Arosa (Kur-
 98 und Verkehrsverein Arosa) was an organization that had a fairly large budget. It was supported mainly through
 99 the “tourist” tax, a fee paid by foreigners/guests staying in Arosa, which was also used to cover the costs of various
 100 other activities that nowadays are subject of communal responsibility. Götz’s request was supported by the General

101 Assembly of the KVV Arosa on 20 August 1921, and Götz was asked to found the “Light Climatic Station” (LKS),
 102 which later became known as the “Light Climatic Observatory (LKO)”. The objectives of the LKS were to
 103 complement the meteorological observations made at Arosa since 1884 by the Swiss national weather service (now
 104 “MeteoSwiss”) by measurements which were thought to be relevant for studying the recovery from pulmonary
 105 TB. Thus, in 1921 Arosa was the first municipality to finance an institute with the task of studying environmental
 106 factors favorable to curing (pulmonary) TB. The support Götz obtained from the KVV Arosa was rather modest
 107 and he later secured additional regular funding from, the Chur-Arosa railway company, the Arosa municipality
 108 and the canton of Grisons (for more detail see Stachelin and Viatte, in prep.). The LKS measurements were made
 109 on the roof of the Inner-Arosa Sanatorium, where nowadays the “Grand Hotel Tschuggen” is located (see Fig. 2).

110



111

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

116

117

118 For the first few years Götz was able to borrow an instrument from Dorno (who was based in Davos, see Section
 119 2.1) to measure “biologically active ultraviolet (UV) radiation”. This instrument had been adapted and used by
 120 Dorno and consisted of a photoelectric cell with a cadmium (Cd) cathode (Levy, 1932). Götz published several
 121 papers using measurements covering the period November 1921-May 1923 (Götz 1925, 1926a and b). He found
 122 the first indication of seasonal variability of stratospheric ozone in the northern mid-latitudes, with a minimum in
 123 autumn and maximum in spring. This turned out to be a very important result later contributing to develop a better
 124 understanding of stratospheric circulation patterns. This seasonal cycle represents one pillar on which the modern

125 understanding of the Brewer-Dobson circulation rests. In fact, Götz published this result earlier than the well-
126 known publication of Dobson and Harrison (1926). Dorno did not agree with Götz's Cd-cell results, and this led
127 to an open dispute published in the literature (Dorno, 1927). It seems likely that there were also some personal
128 difficulties between Dorno, who was 26 years older, and Götz, which surfaced with time. It also appears there
129 were issues between the physicians from Davos and Arosa, with the latter suggesting that the scientific studies
130 made in Arosa should be coordinated with those from Davos. They also asked that the institute for high mountain
131 physiology and tuberculosis research in Davos (Institut für Hochgebirgsphysiologie und Tuberkuloseforschung in
132 Davos) be renamed to include Arosa. These efforts failed probably since members of the Davos community wanted
133 a larger financial contribution from Arosa for the institute (based on the principle of equal duties, equal rights
134 ("gleiche Rechte, gleiche Pflichten")). The KVV Arosa was, however, not willing to pay the requested amount.

135



136

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

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140 **2.3. LKO under Götz**

141

142 1926 was an important year for Götz. After the sobering debate regarding cooperation between the Arosa and
 143 Davos medical doctors (for more details see Staehelin and Viatte, in prep.) Götz moved into the “Villa Firnelicht”
 144 (see Fig. 3), which is very close to the Inner-Arosa Sanatorium, where measurements had been previously
 145 performed (see Fig. 2). Evidence suggests that Götz used family resources to build the large house, probably the
 146 inheritance from his father, Paul Götz, who owned an ironmongery (“Eisenwarehandlung”) in Göppingen
 147 (Trenkel, 1954) and died in 1926. “Villa Firnelicht” offered space for atmospheric observations on the roof and a
 148 balcony. It hosted three apartments and was therefore too large for just Götz and his wife. When Götz moved into
 149 “Villa Firnelicht” the institute was renamed the “Light Climatic Observatory” (Lichtklimatisches Obervatorium
 150 (LKO)). Götz invited colleagues to come to the LKO for sabbatical-type collaborations and to make atmospheric
 151 observations.

152

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

Instrument	Characteristic	Ownership	Operation	1921-30	1931-40	1941-50	1951-60	1961-70	1971-80	1981-90	1991-2000	2001-10	2011-20
Spectrograph ⁽¹⁾	Photographic	LKO Arosa	Campaign ⁽²⁾		Occasionally used ⁽³⁾								

⁽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 operated 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	Standard Instr. TO									
				1921-30	1931-40	1941-50	1951-60	1961-70	1971-80	1981-90	1991-2000	2001-10	2011-20
D002 ⁽²⁾	Photographic	London Met Office	Daily ⁽¹⁾			Occasionally used							
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 ⁽³⁾	Photomultiplier	IOC/IMA ⁽⁴⁾	Daily ⁽¹⁾										

⁽¹⁾ In favorable weather conditions ⁽²⁾ Féry type spectrograph/name D2 given by Dütsch (not internationally 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	Standard Instr. UM									
				1921-30	1931-40	1941-50	1951-60	1961-70	1971-80	1981-90	1991-2000	2001-10	2011-20
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	1921-30	1931-40	1941-50	1951-60	1961-70	1971-80	1981-90	1991-2000	2001-10	2011-20
Br040	MarkII ⁽²⁾	MeteoSwiss	Daily										
Br072 ⁽³⁾	MarkII	MeteoSwiss	Daily										
Br156	MarkIII	MeteoSwiss	Daily										

⁽¹⁾ Up to 2005 Br40 mainly devoted to Total 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

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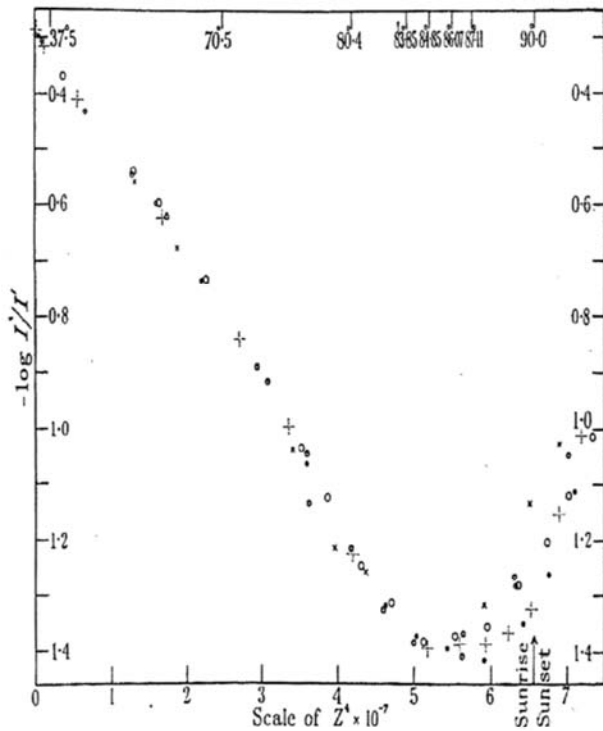
154 **Figure 4.** Sun photometers operated at Arosa from 1921-present (for more details see Staehelin and Viatte in
 155 prep.).

156

157 Hartley (1881) was the first to postulate that atmospheric ozone is responsible for absorbing solar light in the UV-
158 B spectrum. Because the amount of biologically active UV-radiation is determined by stratospheric ozone levels,
159 Götz devoted a large part of his time to stratospheric ozone research (see Staehelin and Viatte, in prep.). He realized
160 that studying stratospheric ozone required suitable instrumentation and using resources from the KVV Arosa he
161 mandated the Schmidt-Haensch company based in Berlin (Germany) to construct a Buisson-Fabry type of a sun
162 spectrophotometer, with a design supervised by him. The instrument was delivered and used by Götz in his
163 expedition to Spitzbergen (see below), but it is unknown to us why it was subsequently only rarely used. In 1926
164 Götz started a very fruitful collaboration with Gordon Dobson, a British physicist and meteorologist at the
165 University of Oxford, who had just developed his first spectrophotometer (Walshaw, 1989). Götz began
166 continuous total ozone measurements at Arosa using an instrument called a Fery spectrograph, which was
167 developed by Dobson (Staehelin et al., 1998a). Later, Götz used improved sun spectrophotometers also constructed
168 by Dobson (abbreviated as Dx, where x is the fabrication number; see Fig. 4). Dobson was very interested in the
169 favorable climate and good weather and working conditions at the LKO. Thus, he arranged that the instruments
170 were formally made available to the LKO through the International Association of Meteorology and Atmospheric
171 Sciences (IAMAS, an association of the International Union of Geodesy and Geophysics (IUGG)). This allowed
172 Götz to make total ozone observations at Arosa for many years, since it would have been very difficult for him to
173 buy such spectrophotometers. After 1948 these instruments were formally borrowed through the International
174 Ozone Commission (IO3C) of the IAMAS. The sun photometers constructed by Dobson measure the intensity of
175 solar radiation at wavelength pairs in the range of 300-340 nm at the Earth's surface. Three different types of
176 instruments were constructed by Dobson (Dobson, 1968) which are shortly characterized in Fig. 5. In order to
177 minimize the falsifying effects of atmospheric aerosols on total ozone measurements the two wavelengths pairs
178 method was introduced during the International Geophysical Year (1958).

179 Götz became one of the leading ozone researchers. In the second half of the 1920s and the first half of the 1930s
180 a key research question was how ozone is distributed in the vertical. Surface measurements e.g, from Arosa
181 indicated low tropospheric ozone concentrations and rather unprecise measurements suggested ozone maxima in
182 the mid-latitudes (in partial pressure) at altitudes of around 40-50 km (see Dobson, 1968). The Umkehr method
183 developed by Götz et al., 1934 (see Fig. 5), however, showed maximum concentrations rather at 20-22 km. This
184 was considered a scientific breakthrough providing the first reliable information about the vertical ozone profile.
185 This method is based on the "Umkehr effect", which Götz discovered during his expedition to Spitzbergen in 1929
186 (Götz, 1931). The first series of Umkehr measurements (besides a limited number of observations made in Oxford
187 in 1931) was performed together with Dobson and his coworker Meetham on the roof of the "Villa Firnelicht" in
188 1932/33 (Götz et al., 1934).

189 Götz was active in the international research community, as a member of the International Radiation Commission
190 from 1932-1936 (Int. Rad. Com., 2008) and as a member of the International Ozone commission (IO3C) created
191 in 1948, when it was formally established at the Seventh IUGG Assembly, until 1954 (see Bojkov, 2012). Götz's
192 research interests were broad, concerning many aspects of weather and climate, and led him to publish two books
193 on focusing to the statistical analysis of radiation measurement and meteorological observations made at Arosa
194 (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

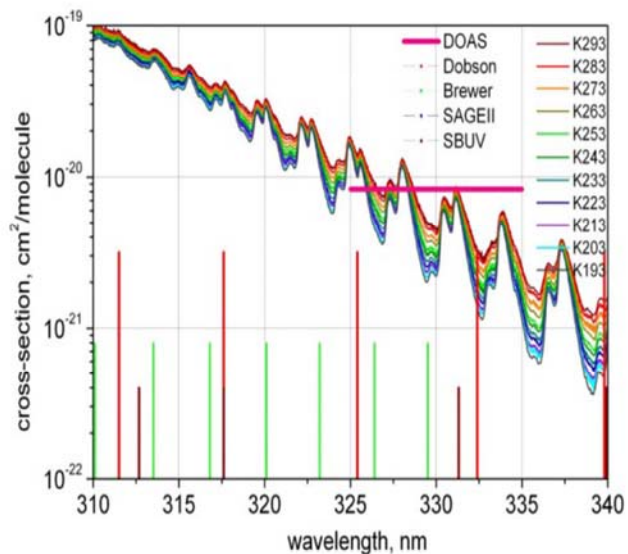
photomultipliers:

wavelengths pairs: A: 305.5/325.4;

B: 308.8/329.1; C: 311.45/332.4;

D: 317.6/339.8.

Since 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 5. Ozone observations by instruments designed by Dobson.

195

196

197 During World War II, the KVV Arosa's financial support for the LKO was substantially decreased and Götz
 198 considered leaving Switzerland. Karl Wilhelm Franz Linke, professor and director of the Institute for Meteorology
 199 and Geophysics of the Goethe University of Frankfurt am Main (Germany) made him two offers to move to

200 Frankfurt. At the same time Heinrich von Ficker, professor at the University of Vienna and director of the Central
 201 Institute for Meteorology and Geodynamics, asked Götz to become professor in Vienna (Austria). However, Götz
 202 decided to stay in Arosa (in the Swiss Alps). If he had moved to Frankfurt or Vienna during World War II, the
 203 column ozone measurements made at LKO would likely have come to an end after just about one decade of
 204 measurements.

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

219 In the last years of his life Götz suffered from health problems (including arteriosclerosis) (Trenkel, 1954) and he
 220 died at the age of 63 in 1954. Dr. Gertrud Perl was his main assistant from 1948 onwards and she continued making
 221 measurements even after Götz’s death, but because of difficulties with Götz’s wife, who owned “Villa Firnelicht”
 222 the LKO had to move to the Florentinum Sanatorium (see Fig. 2) at the end of 1953. Unfortunately, the Dobson
 223 instrument was damaged during transport to the Florentinum, so that there are a few months of data missing from
 224 the Arosa total ozone time series during this period.

225 **3. Period 1954-1962: First intermediate period**

226

227 After Götz’s death, it was uncertain for several years whether the measurements at LKO would continue. Jean
 228 Lugeon, the director of MeteoSwiss (Meteorologische Zentralanstalt at the time), supported the ozone
 229 measurements at Arosa during this critical period. He knew Götz personally, since they had taught together at the
 230 University of Zürich, and was aware of the scientific value of the measurements. He was also the coordinator of
 231 the Swiss contribution to the International Geophysical Year (IGY) in 1958, in which the total ozone measurements
 232 at Arosa were recognized as a geophysically significant data set. For a few years, the Swiss National Science
 233 Foundation (SNSF) contributed to Perl’s salary in addition to the support received from the KVV Arosa, the Arosa
 234 municipality and the canton Grisons. From 1957 onwards, the Arosa total ozone measurements were additionally
 235 supported by MeteoSwiss. Hans-Ulrich Dütsch, a former graduate student of Götz (see Sect. 4.1), also played an
 236 important role for the continuation of ozone measurements at Arosa. He wrote a letter to the head (minister)
 237 councilor of the Swiss Federal Department of Home Affairs in Bern. In his response we read that MeteoSwiss

238 could be mandated to assume the responsibility for the Arosa ozone measurements based on several resolutions of
239 the World Meteorological Organization (WMO), which advised that national meteorological services undertake
240 ozone measurements. It was suggested that the Federal Meteorological Commission (“Eidgenössische
241 Meteorologische Kommission”), the committee responsible for overseeing MeteoSwiss, should consider this in a
242 comprehensive way, also looking at additional options, such as moving the LKO measurements to nearby Davos.
243 Dütsch disagreed with the move to Davos, since he feared that this might lead to a serious discontinuity in the
244 ongoing Umkehr measurements that were started in 1956 by Dütsch (see Section 4.2), because of larger aerosol
245 contamination in Davos. In the end, the LKO stayed independent and was not integrated into MeteoSwiss, but
246 MeteoSwiss and KVV Arosa provided financial support and measurements were continued at Arosa.

247 **4. Period 1962-1985: Hans-Ulrich Dütsch**

248

249 **4.1. Dütsch and international ozone science**

250

251 After Dütsch completed his PhD thesis in 1946 (title: “Photochemische Theorie des atmosphärischen Ozons unter
252 Berücksichtigung von Nichtgleichgewichtszuständen und Luftbewegungen”, Photochemical theory of
253 atmospheric ozone under consideration of non-equilibrium states and airflow), he first worked as a physics teacher
254 (mainly) at a high school (Gymnasium) in Zürich. However, he remained interested in ozone research and
255 eventually decided to pursue a career in science (see Fig. 6). From 1962-1965 he lived with his family in Boulder
256 (Colorado, USA) working as a researcher at the newly founded National Center for Atmospheric Research
257 (NCAR). Together with Carl Mateer, Dütsch was the first to use modern computers to retrieve vertical ozone
258 profiles with the Umkehr method.

259 In 1965 Dütsch was appointed as full professor at the ETH Zürich (ETHZ), where he served as director of the
260 Laboratory of Atmospheric Physics (LAP, merged in 2001 with the Institute of Climate Sciences to become
261 today’s Institute for Atmospheric and Climate Science (IAC)). Dütsch’s research continued to focus on ozone,
262 and he continued, pursued and extended the Swiss ozone measurements (see Section 4.2).

263 During Dütsch’s first years at ETHZ the main motivation for atmospheric ozone measurements at Arosa and
264 Payerne was improving understanding of the “high atmosphere” circulation patterns with the aim of providing
265 improved weather forecasts.. Publications using measurements from the nearby Hohenpeissenberg Observatory
266 (located in Bavaria, Southern Germany) revealed links between ozone levels and synoptic weather types
267 (Hartmannsgruber, 1973; Attmannspacher and Hartmannsgruber, 1973, 1975) and the relationship between the
268 vertical distribution of ozone and synoptic meteorological conditions become an important research topic in the
269 1960s and the early 1970s (see Breiland, 1964).

270 Stratospheric ozone depletion resulting from anthropogenic emissions was first publicized in the 1970s. Molina
271 and Rowland (1974) as well as Stolarski and Cicerone (1974) independently discovered that chlorine radicals
272 destroy stratospheric ozone in a chain reaction. Furthermore, Molina and Rowland postulated that
273 chlorofluorocarbons were a possible source gas for stratospheric chlorine. The chemical industry, particularly
274 market leader DuPont, strongly objected to the view of Molina and Rowland. DuPont went so far as to launch an
275 advertisement in the New York Times in 1975 stating that “Should reputable evidence show that some

276 fluorocarbons cause a health hazard through depletion of the ozone layer, we are prepared to stop production of
 277 the offending compounds". This provided a new justification for making high quality total ozone measurements,
 278 namely as a basis for reliable long-term trend analysis. This was a new challenge for ground-based total ozone
 279 measurements since stratospheric ozone in the extra tropics can vary by as much as $\pm 20\%$ from day to day,
 280 whereas anthropogenic stratospheric ozone changes were (and still are) on the order of only a few percent per
 281 decade.

282

283



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 Götz
1947-1962	High school (Gymnasium) teacher in physics in Zurich, continuing ozone research
1950	Visiting scientist 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. at the Swiss Federal Institute of Technology Zürich (ETH Zürich)
2004	Died on 27 Dec. in Zürich (Switzerland)

284

285 **Figure 6.** Biography of Hans-Ulrich Dütsch.

286

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

295

296 **4.2 Ozone measurements at LKO under Dütsch**

297

298 In 1956, Dütsch was able to find resources to ensure the Umkehr ozone measurements in Arosa continued on a
 299 regular, operational basis. When Gertrud Perl had to leave Arosa in 1962 because of health problems, Dütsch took

300 the responsibility and scientific leadership of the LKO, although he was still living in Boulder (CO, USA) at the
301 time. A large majority of the observations, particularly the Umkehr measurements, were performed by students,
302 under the tutelage of Perl and others, until Kurt Aeschbacher became responsible for the LKO measurements in
303 1964, remaining so until November 2001. When Dütsch became professor at ETHZ in 1965, financial support for
304 the measurements at LKO (total ozone and Umkehr) continued as before (i.e., via KVV Arosa, Arosa municipality
305 and the Canton Grisons). In addition to the spectrophotometric measurements, Dütsch also initiated ozone sonde
306 measurements, which made it possible to observe ozone vertical profile in more detail. In 1966/67, these balloon
307 measurements were operated by Dütsch from Kilchberg (close to Zürich), but in August 1968 MeteoSwiss took
308 took over these observations and made them from Payerne, 140 km Southwest of Zürich on the Swiss plateau
309 (Jeannet et al., 2007). In 2008 Payerne became a member of “The Global Climate Observing System (GCOS)
310 Reference Upper-Air Network” (GRUAN) (fully certified in 2015), an international observing network under the
311 auspices of WMO. GRUAN aims at measuring essential climate variables providing long-term, high-quality
312 climate data records from the surface, through the troposphere, and into the stratosphere.

313 When Dütsch was responsible for the LKO, total ozone and Umkehr measurements were routinely performed
314 using two Dobson spectrophotometers (see Fig. 4). To obtain the total ozone, only direct sun observations were
315 performed. Dütsch applied the statistical Langley plot method to update the instrumental constants of the Dobson
316 instruments every year (Dütsch, 1984). To apply the statistical Langley plot method (which was also used by
317 Farman et al., 1985) a large number of ozone observations with different solar angles is required and therefore the
318 observers need to choose suitable meteorological conditions, e.g. cloud free conditions lasting for at least several
319 minutes. Each year Dütsch went to Arosa for several days to check all the total ozone measurements for reliability
320 and to apply the statistical Langley plot method. This led to small corrections being made to the total ozone
321 measurements for the previous year and some small changes to the instrumental constants for the following year.
322 Students, who usually stayed in Arosa for several months at a time, made the Umkehr measurements, which need
323 to be started prior to sunrise every morning (see Fig. 5).

324 In 1973, the LKO measurements were moved from the “Florentinum” to “Haus Steinbruch” (see Fig. 2), just a
325 few hundred meters away. The working conditions at the LKO were much better at “Haus Steinbruch” than at the
326 “Florentinum”, however the running costs were higher (for more detail see Staehelin and Viatte, in prep.). In 1978,
327 the first international intercomparison campaign of Dobson spectrophotometers took place in Arosa. This was
328 organized by Dütsch under the auspices of the WMO. The results of this first intercomparison exercise at Arosa
329 were not satisfying since “differences between (standard) instruments led to a debate as to which should be used
330 as the standard for the intercomparison” (see Staehelin et al., 1998a). However, this debate deepened the insight
331 into how necessary such comparisons were (and still are), fostering the excellent reputation of Swiss ozone
332 research. As a result of these discrepancies Dütsch continued to apply the statistical Langley plot method to update
333 the instrumental constants up to the begin of the 1990s.

334 **5. Period 1985-1988: Second intermediate period**

335

336 **5.1. International development and the importance of the Arosa total ozone time series**

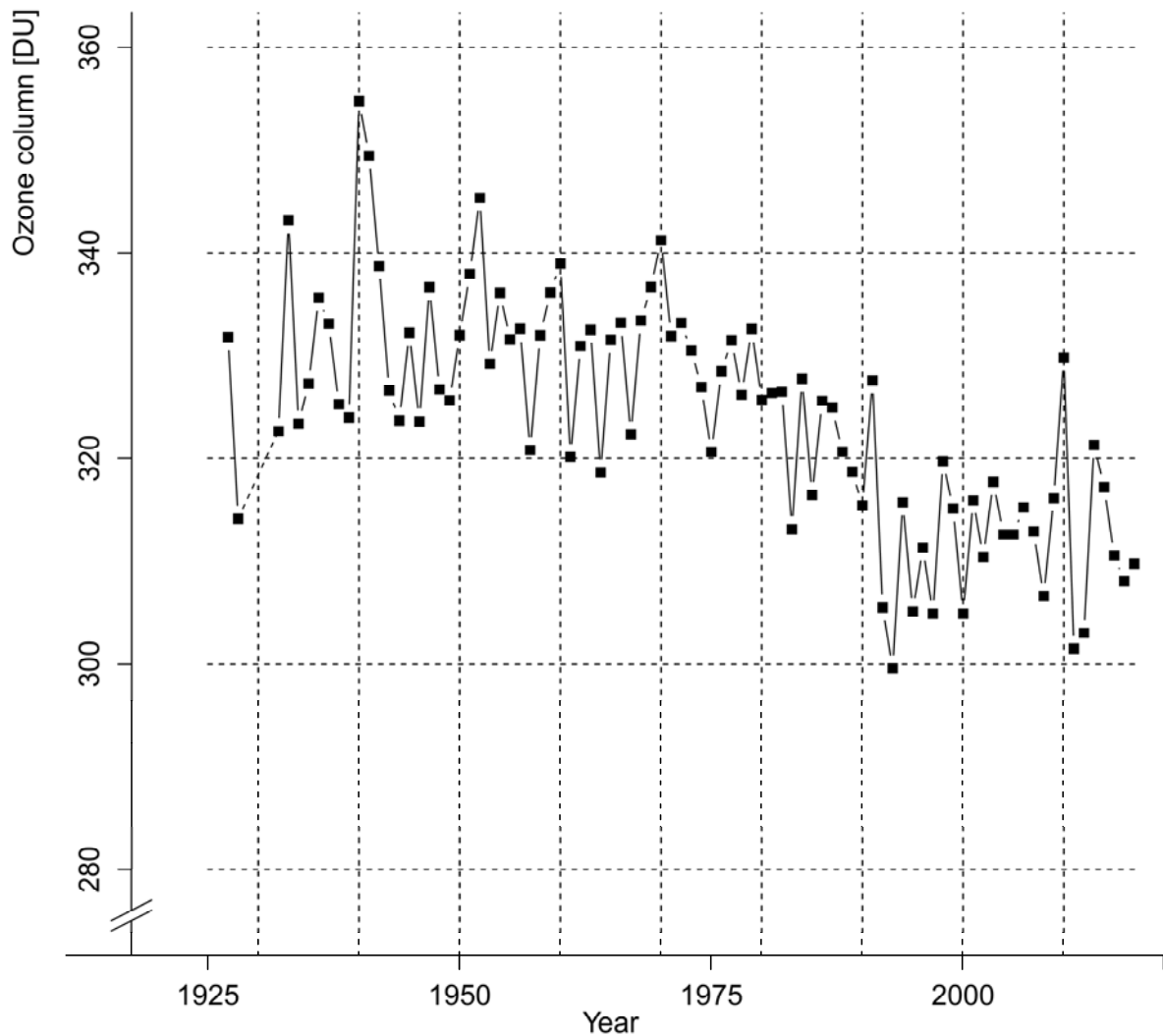
337

338 In the early 1980s, as new information about ozone chemistry reaction rate constants became available, it seemed
339 that chemical ozone depletion by ODSs was considerably less than had been predicted in the late 1970s (Benedick,
340 1991). However, in 1985 the Antarctic ozone hole was discovered (Farman et al., 1985), and the international
341 ozone research community was able to demonstrate that the ozone hole was caused by the chlorine and bromine
342 in halocarbons, which were largely of anthropogenic origin. New insight came through the discovery that the
343 chlorine and bromine species are very efficiently converted into ozone destroying forms on the surface of polar
344 stratospheric cloud particles (Solomon et al., 1986), acting as efficient catalysts in the cold polar stratospheric
345 vortex (for reviews see Rowland, 1991; Peter, 1997; Solomon, 1999).

346 In the mid-latitudes, the first analysis based on the relatively short record of measurements from the Total Ozone
347 Mapping Spectrometer (TOMS) instrument onboard the Nimbus 7 satellite available at the time also showed rapid
348 ozone decline (Heath, 1988). However, ground-based total ozone measurements such as those made using Dobson
349 instruments did not confirm the large downward trends suggested by the satellite data. This discrepancy led to the
350 1988 publication of the International Ozone Trend Panel report (IOTP, 1988). The report demonstrated that TOMS
351 data available at the time were not reliable enough for trend analysis because of inappropriate treatment of the
352 degradation of the diffuser plate. Later these data were reanalyzed more extensively using additional wavelengths
353 in the retrieval algorithms and results were significantly improved (Stolarski et al., 1991). It turned out that also
354 some of the data from the ground-based instruments were not of high enough quality to carry out reliable long-
355 term trend analyses. This was attributed to calibration issues with the Dobson instruments, which showed frequent
356 sudden changes when compared to TOMS overpass data (IOTP, 1988). Rumen Bojkov, Secretary of the IO3C
357 (1984-2000), used TOMS data to provide “provisionally revised” ground based measurements, which, however,
358 had weaknesses such as not correcting for sulfur dioxide (SO₂) interferences leading to potential errors in ozone
359 trends based on Dobson series (e.g., De Muer and De Backer, 1992).

360 The most important application of the long-term measurements from Arosa (see Fig. 7) was probably their use in
361 the 1988 IOTP report. The Arosa time series was the only Dobson dataset that required no correction and was
362 much longer than any of the other ground-based measurement records. Results from Neil Harris’s PhD thesis were
363 published in the IOTP and showed, for the first time, significant decreases in stratospheric ozone in the northern
364 mid-latitude winter season (Harris, 1989). He used two different approaches, namely (1) dividing the individual
365 records into two periods of similar length using measurements going back to 1957 and (2) developing a novel
366 multiple linear regression model taking into account trends for different months. In this model the downward trend
367 started in 1970, and the analyses also showed that the negative trend was not sensitive to the start year. At present,
368 standard Dobson measurements are based on observations of two (AD) wavelength pairs, which allow to minimize
369 the interference by aerosols, a technique introduced during the International Geophysical Year (IGY) in 1957-58
370 (cf. Fig. 5). To further support his main conclusion, Harris (1989) also used single other wavelengths pair (C) data
371 from Arosa, which are available as representative (homogenized) measurements since 1931. Again, he found
372 similar negative total ozone trends as at most other sites in the northern mid-latitudes (IOTP, 1988).

373



374

375 **Figure 7.** Annual mean total (column) ozone values measured at the world's longest continuous spectrophotometer
 376 site in Arosa, Switzerland, from 1926-present. The ozone column in Dobson units, where 100 DU correspond to a
 377 1-mm thick slab of pure ozone gas at standard conditions (273.15 K, 1000 hPa).

378

379 5.2. Continuation of measurements at the LKO

380

381 After Dütsch's retirement in 1985, the continuation of Swiss long-term ozone measurements again became
 382 uncertain. The professor succeeding Dütsch focused on another research topic, and consequently the ETH Zürich
 383 argued that the continuation of operational ozone measurements did not fall under the responsibility of a university.
 384 Conversely, MeteoSwiss, which already was responsible for the ozonesonde measurements since 1968, argued
 385 that such long-term measurements needed scientific analysis by a well-qualified scientist, which MeteoSwiss was
 386 not able to support (a hiring freeze for permanent positions existed at the federal level at the time). Dütsch again
 387 wrote a letter to the responsible minister of the Federal government to point out the importance of the Arosa ozone
 388 measurements. Representatives from the Swiss Federal Office for the Environment (the "Swiss EPA") argued that
 389 ozone research in Switzerland needed to be continued since expert ozone researchers served a vital role to provide
 390 advice to policy makers regarding both stratospheric (in terms of the Vienna Convention and Montreal Protocol)

391 and tropospheric ozone. Subsequently, a commission of the Swiss academy of Natural Sciences was tasked to
392 analyze the situation. Government representatives as well as Swiss ozone researchers were invited to their meeting.
393 Again, it was considered whether it made sense to move the LKO measurements to Davos (PMOD), but no
394 decision was made in this regard. Nevertheless, MeteoSwiss and ETH Zürich (i.e. IAC, Institute for Atmospheric
395 and Climate Science since 2001, at the time Laboratory of Atmospheric Physics (LAPETH) agreed to continue the
396 measurements, with the former officially accepting to take responsibility for the continuation of the ozone
397 measurements at Arosa (total ozone and Umkehr) as well as the ozonesondes launched from Payerne, and the IAC
398 at ETH Zürich consenting to continue ozone research. The agreement - implying that the person responsible for
399 the LKO operations was moved to a MeteoSwiss position, whereas the IAC filled a scientific position with a major
400 focus on ozone research became effective at the beginning of 1988.

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

402

403 **6.1. International Development: The Montreal Protocol**

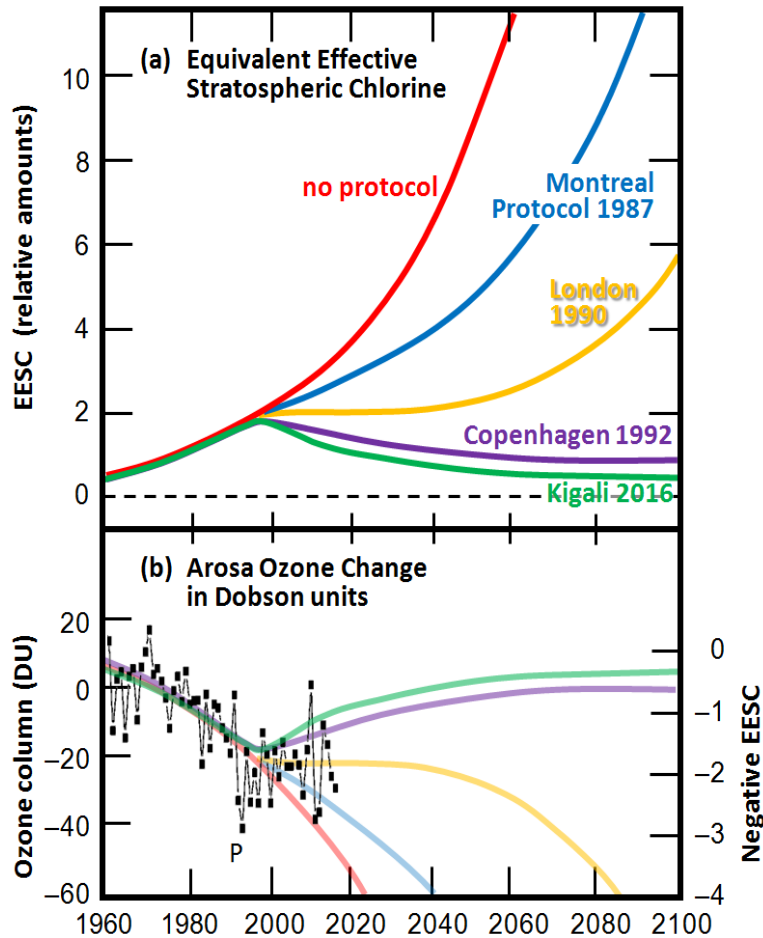
404

405 Since 1988, the most important justification for ozone measurements at LKO Arosa (total ozone und Umkehr) and
406 ozone sonde launches in Payerne has been the documentation of the effect of ODSs on the stratospheric ozone
407 layer and the effectiveness of the Montreal Protocol. Chemical ozone depletion by ODSs is expected to evolve
408 very similar to the evolution of Equivalent Effective Stratospheric Chlorine (EESC). EESC provides an estimate
409 of the total amount of halogens in the stratosphere, calculated from emission of chlorofluorocarbon and related
410 halogenated compounds into the troposphere (lower atmosphere) and their efficiency in contributing to
411 stratospheric ozone depletion (hence “effective”), and by taking the higher ozone destructiveness of bromine
412 appropriately into account (hence “equivalent”). EESC peaked in the second half of the 1990s and subsequently
413 showed a slow decrease, which is attributable to the Montreal Protocol, but in its slowness dictated by the long
414 lifetimes of the emitted substances (see Fig. 8a). Total ozone measurements at Arosa are broadly consistent with
415 long-term evolution of EESC (Staehelin et al., 2016) showing record low values in the early 1990s (Fig. 8b, cf.
416 Fig 7). The recovery of the ozone layer is a slow process and the signal of any sort of turnaround in the Arosa total
417 ozone time series is still indistinct. Figure 8b shows the large interannual variability of the annual means, which is
418 normal for a single measurement station and renders an attribution of the change in the downward trend difficult.
419 While model results suggest that the Montreal Protocol and its amendments and adjustments have helped to avoid
420 millions of additional skin cancer cases, Fig. 8b indicates that the global network of ozone station measurements
421 needs to remain strong in order to achieve a clear detection of the trend reversal and a proper attribution of the
422 reasons.

423

424

425



426

427 **Figure 8.** (a) Relative abundance of Ozone Depleting Substances (ODSs, i.e. volatile halocarbons) expressed as
 428 equivalent effective stratospheric chlorine (EESC) for the mid-latitude stratosphere, shown for various scenarios
 429 (demonstrating the impact of the Montreal Protocol and its subsequent Adjustments and Amendments). EESC can
 430 be viewed as a measure of chemical ozone depletion by ODSs and takes into account the temporal emission of the
 431 individual ODS species as well as their ozone depleting potential. (b) Arosa annual mean ozone columns (black
 432 symbols, as in Fig. 7) in comparison with the scenarios in (a). "P" marks the eruption of Mt. Pinatubo in 1991,
 433 which has aggravated the ozone loss.

434

435 6.2 LKO and related activities

436

437 6.2.1 Cooperation between MeteoSwiss and IAC (ETHZ)

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

445

446 **6.2.2 Renewal of the LKO infrastructure**

447 When Meteoswiss become responsible for the LKO ozone measurements in 1988, the instrument infrastructure
448 required renewal and extension. This was completed under the leadership of Bruno Hoegger and included
449 constructing a spectrodome to house the two Dobson spectrophotometers as well as semi-automation of the
450 Dobson total ozone measurements and full automation of the Dobson Umkehr measurements (Hoegger et al.,
451 1992). Three Brewer instruments were also purchased between 1988 and 1998, thus allowing increased reliability
452 of the Arosa total ozone series by complementing the Dobson Umkehr measurements and by providing
453 instrumental redundancy (see Fig. 4). Furthermore, UVB measurements were added. For more technical
454 information including new electronics see Staehelin and Viatte, in prep. Stübi et al. (2017a) demonstrated the
455 excellent stability of the Arosa Brewer triad over the past 15 years.

456

457 **6.2.3 Homogenization of the Arosa total ozone and Umkehr timeseries**

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

465 **6.2.4 Foci of scientific studies since the 1990s**

466 The comparison of the unique Arosa total ozone time series from Dobson and Brewer instruments has allowed
467 studies of the differences between the two instrument types (Staehelin et al., 1998a; Scarnato et al., 2009, 2010)
468 as well as their long-term behavior since they are calibrated in different networks. The large data set of quasi-
469 simultaneous measurements was particularly valuable for studying the effect of temperature dependence of ozone
470 absorption cross-sections on total ozone measurements attributable to the different wavelengths used in Dobson
471 and Brewer instruments (Scarnato et al., 2009, Redondas et al., 2014). These results were an important contribution
472 to the GAW ACSO (Absorption Cross-Sections of Ozone) project in which available laboratory cross-sections of
473 atmospheric ozone measurements were studied (ACSO, 2015; Orphal et al., 2016).

474

475

476 In the 1990s, quantification of the downward ozone trends was the main reason for making long-term stratospheric
477 measurements (comp. Section 5.1, and Staehelin et al., 1998b, 2001). These trends were seen as a consequence of
478 increasing ODS concentrations. Subsequent studies were also devoted to understanding the potential contribution
479 of other processes enhancing the observed downward trends, including long-term climate variability, e.g. related
480 to tropopause altitude (Steinbrecht et al., 1998) and climate patterns (Steinbrecht et al., 2001). The unique length
481 of the Arosa total ozone series was very valuable in demonstrating that the North Atlantic Oscillation (NAO) or
482 Arctic Oscillation (AO) enhanced downward winter ozone trends in central Europe for the period up to the
483 mid-1990s (Appenzeller et al., 2000; Weiss et al., 2001). Brönnimann et al. (2004a, 2004b) also showed that the
484 record high values of total ozone at Arosa that occurred in the early 1940s were due to an increase in strength of
485 Brewer Dobson circulation caused by a very large El Niño/Southern Oscillation anomaly during that period.

486 The unique length and high quality of the Arosa total ozone and Umkehr measurements also meant they were
487 important for the EU project CANDIDOZ (Chemical and Dynamical Influences on Decadal Ozone Change; Zanis
488 et al., 2006; Brunner et al., 2006; Harris et al., 2008). Later, as the ODS concentrations have decreased,
489 documentation of the “turn around” in stratospheric ozone trends became more and more important (e.g. Mäder et
490 al., 2010). The Arosa time series was also used to introduce the concept of extreme value theory in ozone science
491 (Rieder et al., 2010a, b). This allowed attribution of extreme ozone values to events of various origins, dynamical
492 features such as ENSO or NAO, or chemical factors, such as cold Arctic vortex ozone losses, or major volcanic
493 eruptions of the 20th century, e.g. Mt. Pinatubo.

494

495 **6.2.5 Tropospheric ozone**

496 The surface ozone measurements from Arosa are unique and very valuable for tropospheric chemistry studies.
497 Surface ozone measurements were begun already in the 1930s by Götzt to quantify the contribution of tropospheric
498 ozone to the total column, and were later continued by the careful and representative surface ozone measurements
499 made in the 1950s (Götzt and Volz, 1951; Perl, 1961). Thanks to these measurements it was possible to show that
500 surface ozone concentrations increased by more than a factor of two from the 1950s to 1990 (Staehelin et al.,
501 1994). This has commonly been attributed to the large increase in ozone precursor emissions (nitrogen oxides,
502 volatile hydrocarbons, and carbon monoxide) resulting from the strong economic growth in industrialized
503 countries following World War II. The surface ozone measurements made at Arosa and Jungfraujoch were pillars
504 in the studies of Parrish et al., (2012, 2013), which contributed to an important report by the Task Force of the
505 Hemispheric Transport of Air Pollution (HTAP). HTAP was organized in 2005 under the auspices of the United
506 Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution
507 (LRTAP Convention) to study intercontinental transport of ozone in northern mid-latitudes. Based on these data,
508 Parrish et al. (2014) compared three state-of-the-art chemistry climate models (CCMs) to show that simulated
509 surface (baseline) ozone trends over Europe were about a factor two smaller than those seen in the available
510 observations. This result was recently confirmed by Staehelin et al., 2017.

511 **7. Future of ozone measurements at the LKO**

512 **7.1 International Demands**

513

514 Policy makers and the general public would like to see proofs of the effectiveness of the Montreal Protocol and to
515 better understand how climate change will affect the ozone layer, i.e. what are the impacts of the stratospheric
516 cooling and the anticipated enhanced Brewer-Dobson circulation on ozone, and what this means for polar, mid-
517 latitude and tropical ozone.

518 Recovery of the stratospheric ozone layer in response to the reduction of ODS concentrations controlled by the
519 Montreal Protocol is slow (see Sect. 6.1) and requires continued long-term stratospheric ozone observations. ODSs
520 most directly impact ozone in the upper stratosphere, where photolysis leads to the release of halogen radicals
521 from these species. Extensive data analyses carried out under the auspices of the SI2N activity commonly
522 sponsored by SPARC (Stratosphere-troposphere Processes and their Role on Climate), IO3C, IGACO-O3/UV
523 (Integrated Global Atmospheric Composition Changes), and NDACC (Network for Detection of Atmospheric
524 Composition Changes) highlighted issues related to the availability and uncertainty of measurements. Recent
525 examples are merged satellite datasets, and trend analysis techniques (see the special journal issue jointly organized
526 between Atmospheric Chemistry and Physics, Atmospheric Measurement Techniques, and Earth System Science
527 Data: Changes in the vertical distribution of ozone – the SI2N report). Steinbrecht et al. (2017) presented a recent
528 analysis of upper stratospheric ozone trends confirming the expected increase in upper stratospheric ozone in
529 extratropics. Finally, Ball et al (2018) showed that total ozone in the mid-latitudes has not increased as expected
530 and their careful analysis of mostly satellite measurements indicated a downward trend in the lower stratosphere
531 (15-22 km) which continued since 1987. The physical cause of this surprising trend is presently unknown and
532 requires further study.

533 It is vital to continue high quality stratospheric ozone measurements to be able to follow the slow recovery of the
534 ozone layer in response to the changing burden of stratospheric ODSs, including nitrous oxide (N₂O), which is
535 likely to become the dominant species for stratospheric ozone depletion in future (Ravishankara et al., 2009;
536 Portmann et al., 2012).

537 Climate change will modify the distribution of stratospheric ozone in different ways (see e.g. Arblaster et al.,
538 2014). Increasing greenhouse gases cause decreasing stratospheric temperatures, which in turn modify reaction
539 rates and lead to increasing extra-tropical stratospheric ozone concentrations. This is not the case over the poles,
540 where the stratosphere is not expected to cool on average. Furthermore climate change is expected to enhance the
541 Brewer Dobson Circulation which transports ozone from the main tropical production region to the extra-tropics
542 (Butchart, 2014). Modification of the Brewer Dobson Circulation is expected to increase stratospheric ozone in
543 the mid-latitudes to levels above those seen in the past; this has been termed “super recovery”. In contrast, the
544 enhanced transport out of the tropics is expected to result in a decrease in stratospheric ozone in these regions. The
545 enhancement of the Brewer Dobson Circulation is, however, still under debate, with state-of-the-art CCMs
546 projecting an increase but only controversial observational evidence being available. Importantly, the expected
547 enhancement depends strongly on the climate change scenario investigated, thus it is essential that high quality
548 measurements are continued.

549 The unique length of the Arosa timeseries is particularly useful for documenting the effects of climate change on
550 ozone since the dataset covers a period of almost 40 years when the stratosphere was relatively undisturbed by
551 anthropogenic influence, about 25 years in which anthropogenic ODSs increased in concentration in the
552 stratosphere, and the latest period with the slow decrease in stratospheric ODS concentrations. The Arosa

553 timeseries will therefore play a crucial role in the coming decades to further document ozone changes in the
554 Northern mid-latitudes, including the predicted “super recovery” expected to become important around 2030 (e.g.
555 Hegglin et al., 2015).

556

557 **7.2 Continuation of measurements at the LKO**

558

559 The MeteoSwiss board of directors decided in 2015 to explore the possibility of moving the Arosa measurements
560 to the PMOD in Davos. Such a move would result in reduced measurement costs in combination with the advantage
561 of the excellent technical infrastructure and expertise that is available at the PMOD in Davos. Within this activity
562 the Dobson instruments are currently completely automated (comp. Fig. 4). However, before such a move is to
563 take place, a multiannual period of overlapping measurements at both sites (Arosa and Davos) is essential. A break
564 in the world’s longest total ozone time series would be very unfortunate. A relocation is particularly challenging
565 as stratospheric recovery from ODS is expected to be slow (see Sec. 6.1) meaning ozone changes will be small
566 and thus very high quality (i.e. very high stability) measurements are required. At present simultaneous total ozone
567 measurements of Brewer instruments of Davos and Arosa have been analyzed and presented (Stübi et al., 2017b).

568 **8. Summary and Conclusions**

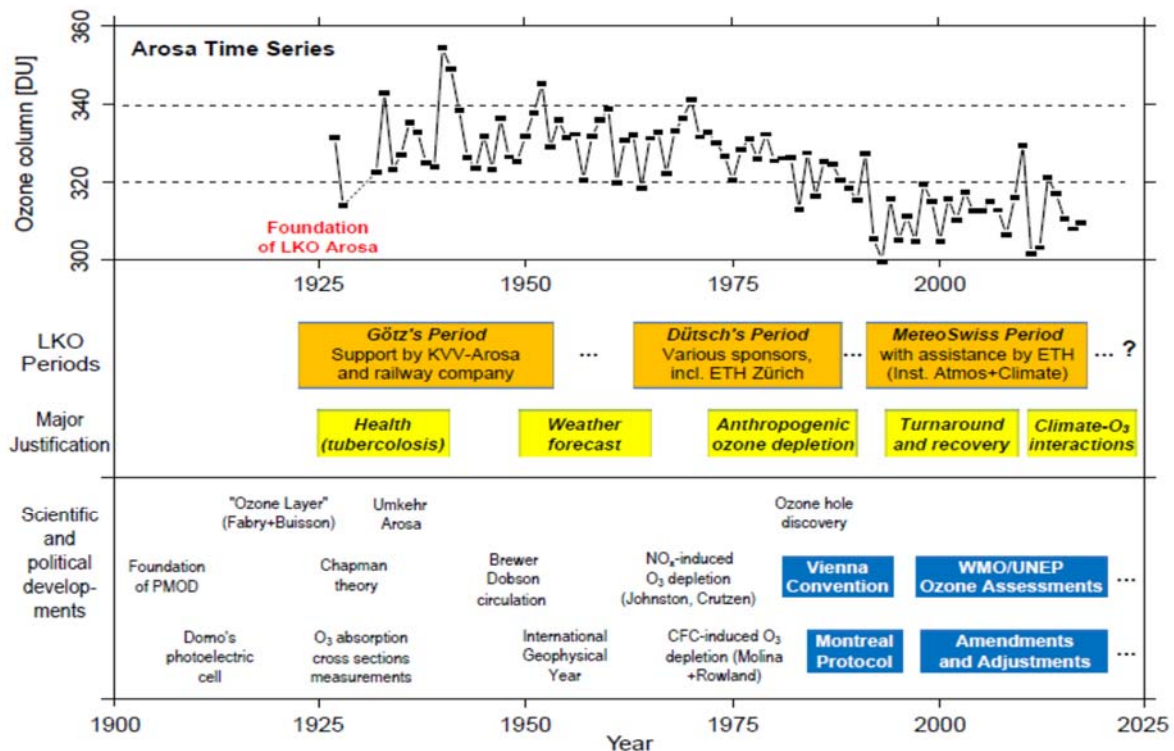
569

570 Homogenous long-term records such as the total ozone record from Arosa are very valuable for trend analyses in
571 climate science. Reliable long-term, ground-based total ozone measurements are also crucial for validation of
572 ozone observations from space, particularly in terms of validating the long-term stability of merged satellite
573 datasets (e.g. Labow et al., 2013). Furthermore, they serve as a baseline for evaluating numerical simulations such
574 as Chemistry Climate models (CCMs), which are used to make projections of future ozone evolution (see e.g.
575 Eyring et al., 2013, Arblaster et al., 2014). The extraordinary length of the Arosa record was important for a wide
576 range of studies, including the analysis of stratospheric ozone related to long-term climate variability such as the
577 NAO/AO (Appenzeller et al., 2000) and El Nino Southern Oscillation (Brönnimann et al., 2004a and 2004b).
578 Furthermore, the measurements have been very valuable for the evaluation of the (early part of the) Twentieth
579 Century Reanalysis Project (Compo et al., 2011; Brönnimann and Compo, 2012).

580 The reasons for continuing the Arosa measurements have changed many times over past decades, and it was never
581 imagined that such a long record could be established. Fig. 9 provides a historical overview of international ozone
582 research in connection with the different phases of the LKO, which also indicates various funding periods. The
583 justification for the LKO measurements for society can be summarized as

- 584 (1) to study environmental factors potentially important for the medical recovery from pulmonary TB
585 (relevant from the beginning until around World War II),
- 586 (2) to investigate air quality as an important natural resource in resort areas (as discussed in the second half
587 of World War II)
- 588 (3) to improve our understanding of atmospheric physics for improved weather forecasts (important in the
589 1960s and early 1970s)

- 590 (4) to quantify anthropogenic ozone destruction by ODSs (mid-1970s to mid-1990s)
- 591 (5) to document the effectiveness of the Montreal Protocol in saving ozone (since around the middle of the
- 592 1990s)
- 593 (6) to understand the mutual relationship between climate change and global ozone depletion, and the
- 594 effectiveness of the Montreal protocol (this century)
- 595



596

597 **Figure 9.** Historical overview of the successive periods of Light Climatic Observatory of Arosa (LKO). Total

598 ozone measurements (top, annual means); different phases during the history of LKO including main sponsors (in

599 orange), justification of measurements for society (in yellow); milestones in international ozone research, and

600 international legislation (blue).

601

602 A key element for the success of LKO measurements and its continuation was the motivation of the scientists

603 involved, i.e. Götz's early initiative and Dütsch's persistence.

604 From our experience the following issues were most relevant for the successful operation of LKO over the last

605 decades:

- 606 - Redundancy allows for increased credibility of measurements, which is particularly important for reliable
- 607 long-term trend analysis. At Arosa, 3 Dobson and 3 Brewer spectrophotometers were simultaneously
- 608 operated since 1998, which helps to obtain important scientific results regarding Dobson and Brewer
- 609 spectrophotometers relevant within the broader context of atmospheric ozone measurements.

- 610 - Regular comparison of station instruments with standard spectrophotometers operated under the WMO
611 umbrella are important for high-quality measurements and consistency of ozone measurements within a
612 particular network.
- 613 - Scientific analysis and use of stratospheric ozone measurements in scientific publications and model
614 intercomparisons not only enhances visibility of the measurements within the community, but also is a
615 quality assessment, which might motivate scientists and technicians operating the measurements.
- 616 - Reliable techniques are important for high quality stratospheric ozone measurements including
617 automation to reduce manpower costs and to make measurements less dependent on the skills of an
618 individual operator.

619 It is difficult to obtain funding for continuous observations through normal science funding agencies such as the
620 Swiss National Science Foundation (SNSF), since an additional few years of measurements usually do not result
621 in novel scientific conclusions. This is the experience within other networks as well, for example NDACC. The
622 success of the Montreal Protocol measures probably contributed to the decrease in the number of ozone
623 measurements submitted to the World Ozone and Ultraviolet Data Center (WOUDC, presently operated by
624 Environment and Climate Change Canada) over the past few years (Geir Braathen, personal communication). This
625 might be exacerbated in the future as monitoring costs come under further pressure in many countries. However,
626 we believe that such routine measurements are the responsibility of developed countries. Institutions like national
627 meteorological services, although they also may experience financial shortfalls, are ideally suited to carry out these
628 types of measurements since they are (in contrast to universities) capable of making long-term commitments and
629 have the possibility to hire permanent staff. On the other hand, universities have the advantage of being able to
630 focus on particular issues (e.g. through PhD theses) for a limited time, resulting in articles in peer-reviewed
631 journals. It is important to stress the relevance of scientific activities using long-term observations. Excellent
632 collaboration has existed between MeteoSwiss and the IAC (ETHZ) for the past three decades. However, this
633 particular type of cooperation will be less feasible in future, as the required permanent scientific positions will
634 typically no longer be available at universities. In other countries the research aspects are often integrated in the
635 same institution (e.g. the German Weather Service (DWD) in Germany or the “Centre National de la Recherche
636 Scientifique (CNRS)” in France). This problem still awaits a proper solution for the Swiss long-term ozone
637 measurements.

638 From the very beginning, the ozone measurements from Arosa (initiated by the fruitful collaboration between Götz
639 and Dobson) have been an important contribution both to the global network of ozone measurements and to ozone
640 research. During the early part of the record, the International Ozone Commission (IO3C) of IAMAS coordinated
641 the ozone measurements. Since the 1970s WMO has taken the lead, first in the framework of the Global Ozone
642 Observing System (GOO3S), later the Global Atmosphere Watch (GAW) programme (SAG-ozone) became
643 responsible for overseeing and coordinating stratospheric ozone measurements to obtain and maintain high quality
644 data suitable for long-term trend analysis. GAW might continue these activities in collaboration with other
645 networks, such as NDACC, the present Brewer COST network, and the IO3C in order to (i) maintain and extend
646 high quality records of ground-based ozone stations and (ii) to continue comparisons of Dobson and Brewer
647 measurements with other/new instruments such as SAOZ and PANDORA. GAW might represent the ground-
648 based community as partners to the satellite community, for example within the Copernicus project and GAW also
649 can contribute to research programs and initiatives, illustrated by the long history of ozone research connected

650 with the LKO started by the pioneers Götz and Dütsch and continued more recently by MeteoSwiss and ETHZ
651 under the auspices of WMO, IGACO-O3/UV, ACSO, and SPARC.

652 Beyond any doubt the Montreal Protocol (including enforcements) has been very successful for the protection of
653 the ozone layer over densely populated areas, avoiding large damage by manmade chemicals as shown by extended
654 numerical simulations (Newmann et al., 2009). In the future, when the stratosphere is expected to gradually recover
655 from the decreasing burden of ODSs, continued observations will not only be required to document the expected
656 increase in stratospheric ozone, but also to document the effects of climate change on stratospheric ozone, as
657 predicted to happen by CCMs, i.e. through enhancement of the Brewer Dobson Circulation and possible other
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