19^{th}–20^{th} century semi-quantitative surface ozone along subtropical Europe to tropical Africa Atlantic coasts

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Abstract. Tropospheric ozone (O_3) plays a key role in the climate system. Studying pre-industrial tropospheric O_3 implies two important challenges: *i*) the lack of observational records prior to the late 19^{th} century, which hampers understanding long-term climate trends, given O_3 crucial role, *ii*) and the uncertainties on their quantitative values in a non-polluted atmosphere across the planet. The ozonoscope was the first instrument used to measure ozone. It offers semi-quantitative estimates of surface O_3 when no other measurements were available. Despite their potential value, the digitisation, curation and publication of ozonoscope data remains largely unexplored. In this work, we initiate an effort to rescue surface O_3 ozonoscope records with a new data collection. We include data from 23 observatories covering Portugal and the African Atlantic regions, providing a latitudinal span from the extratropics in the northern hemisphere to the tropics in the southern hemisphere. This record represents the most extended ozonoscope data series to date, spanning 50 years of daily data and 58 years of monthly data, from 1855 to 1913.

1. Background & Summary

Tropospheric ozone (O_3) records for our planet before the end of the 19^{th} century are rare and sparse. It is not surprising given that O_3 was discovered by Schönbein in 1839 (Schönbein, 1840a, b) and it got little attention during the decades following its discovery. After 1860s, measuring it became common at meteorological stations. However, O_3 is a transcendental chemical for the understanding and study of the atmosphere. Tropospheric O_3 is a greenhouse gas and, at elevated concentrations, a pollutant harmful to human health, also affecting crops and ecosystems productivity (U.S. EPA, 2020). However, the study of tropospheric O_3 faces two important challenges: *i*) the lack of observational records prior to the late 19^{th} century, which hampers understanding long-term climate trends, *ii*) and the uncertainties on the quantitative values in a non-polluted atmosphere.

The first phase of the Tropospheric Ozone Assessment Report (TOAR) project (Schultz et al., 2017; Tarasick et al., 2019) developed a web-accessible database of surface O_3 observations, consisting in two main periods. The modern period, beginning around 1975 and spanning to the present, defined by widespread availability of sensitive UV photometers for surface O_3 measurements, and the historical period, covering 1877–1975, defined by the use of other techniques and the lack of UV photometers. The records available for the period previous to 1975 were evaluated using a set of four criteria to minimize uncertainties and biases between the measurement techniques available at that times, and the contemporary UV absorption standard. Those criteria are: the relationship of the measurement technique to the modern UV absorption standard, the absence of interfering pollutants, the representativeness of the well-mixed boundary layer, and expert judgement of their trustworthiness. The earliest surface O_3 measurements, corresponding to the 19^{th} century, extending until the early 20^{th} century using the test-paper method, also called "ozonoscope", were among the ones disregarded (Tarasick et al., 2019).

Considering the scientific questions motivating the TOAR, associated with the global distribution and trends of surface O_3 pollution (Gaudel et al., 2018), the decision not to include the 19^{th} century semi-quantitative O_3 measurements in the TOAR database is grounded. Yet, there are other scientific questions related to global distribution and surface O_3 pollution during the pre-industrial era, such as the atmospheric concentration in non-polluted areas and evaluation of the assumed O_3 concentrations, study of local sources of O_3, and better understanding of the role of such levels on the radiative balance. Answering such questions could benefit of using together the quantitative surface O_3 observations with semi-quantitative O_3 observations from ozonoscopes. Although the measurements with the ozonoscope were vulnerable to the influence of the humidity and oxidants in the air, those semi-quantitative O_3 observations will enable us to study semi-quantitatively climate variables under very low (or no exposure at all) anthropogenic activity when no other measurement was available (Bojkov, 1986), a gap in our knowledge of surface O_3 in the 19th century.

Efforts to recover some of those O_3 measurements have been performed in the past (Bojkov, 1986; Linvill et al., 1980; Anfossi et al., 1991; Sandroni et al., 1992; Sandroni and Anfossi, 1994; Marenco et al., 1994; Cartalis and Varotsos, 1994; Nolle et al., 2005); however, only a single sample of those surface O_3 datasets that we know of was digitized, published in a public data repository, and it was done during data recovery efforts focused on something other than ozone or atmospheric composition (Vaquero et al., 2022).

Here we introduce the rescued surface O_3 ozonoscope records covering Portugal and the African Atlantic oceanic sector from 23 observatories in four countries, as shown in Figure 1. The O_3 semi-quantitative observations provide a latitudinal coverage from extratropics in the northern hemisphere to tropics in the southern hemisphere. The observations were conducted following a standardized procedure with the same type of test-paper (Schönbein, 1850; Bérigny, 1858). The series of daily and monthly means of surface O_3 and humidity, and their corresponding metadata, have been digitized from the original documentary sources. They are representative of very different regions of the planet, such as tropics, oceans and coastal areas. One of them, from the Infante D. Luiz observatory, located in Lisbon, Portugal, provides almost fifty years of continuous daily data in the period 1863 to 1913 and nearly fifty-eight years of monthly means from 1855 to 1913, becoming the most extended and earlier surface O_3 ozonoscope data series known to date. Before this work, the longest and earliest reported series was the thirty-one years Montsouris observatory O_3 ozonoscope data series, which began in 1876 (Bojkov, 1986). Additionally, another ten of the daily records here recovered cover between twelve and seventeen years of data, while four monthly mean records extend between thirty-three and forty-two years. The difficulty to find records of meteorological variables covering oceanic regions for the preindustrial era, and in particular surface O_3 datasets, makes this contribution one of the most relevant features of the datasets recovered here, as it contains six datasets from islands in the East Atlantic (two at the Azores Island, one in the Madeira Island, two in Cape Verde and another in Saint Thomas & Prince). The data series have been tested for breakpoints and inhomogeneities, finding few of them; unfortunately, we have discovered scarce metadata that let us to provide context for the existing breakpoints; however, in several cases a change in the location of the observatory or instruments seems a plausible explanation.

In the next section, the Schönbein test paper method and its further improvement by Bérigny are briefly described, followed by the description of the data sources. Then, in Data Records, we describe the main features of the recovered datasets, both for the daily and monthly means of the Infante D. Luiz observatory and the other twenty-two observatories. The Technical Validation explains the homogeneity tests applied.

2. Methods

2.1. The test-paper method

The test-paper measurement method was based on the color change of an indicator test paper. The strip of blotting paper was coated with starched potassium iodide and then exposed to air between eight and twenty-four hours, protected from solar radiation and rain. After the exposure, the strip was moistened, developing a bluish color associated with the formation of a complex between starch and iodide, produced by the reaction between O_3 and iodide. The coloration depends on the O_3 concentration. Finally, the observed color was compared with a standard chromatic scale, graduated by Schönbein from 0 to 10, proportional to the O_3 content in the air (Schönbein, 1850; Ramirez-Gonzalez et al., 2020).

The method was criticized after it began to be used because the paper strip changes its color depending on the extent of the iodide reaction with ozone, but also with humidity and other atmospheric oxidants (Houzeau, 1857; Fox, 1873). When air reaches its water vapor saturation it causes the pre-dried paper to humidify increasing the rate of O_3 absorption (Kley et al., 1988; Volz and Kley, 1988). Those are the reasons for a non-linear correlation between the color changes and the ozone concentration.

Bérigny introduced the Schönbein's method in France in 1856 (Bérigny, 1856a, b, 1857). He also improved the method, defining the operating procedure, presenting a more precise chromatic scale graduated from 0 to 21 (Bérigny, 1858), and selecting the best quality of impregnated paper, the Berzelius paper manufactured by Jame, a chemist at Sedan (France) (Marenco et al., 1994). This scale was often referred to in logbooks containing measurements as “Jame de Sedan”.

More than one and half century after the test-paper method was introduced, numerous research has been conducted to understand the physical-chemical processes involved in the method and to deal with the associated interference problems (Marenco et al., 1994). Those studies for example estimated that the O_3 levels for 1880 to 1900 were approximately 10 ppb in the Great Lakes area of North America, with an annual cycle maximum in April-June, and the minimum in October-November (Bojkov, 1986). Another study using observations from Montevideo, Uruguay (1883-1885) and Cordoba, Argentina (1886-1892) also showed O_3 levels of the order of 5 – 10 ppb (Sandroni et al., 1992). It is beyond the scope of this article to discuss all the reported studies; we refer the readers to the review conducted by Marenco et al. (1994). Among the cited interferences present in the Schönbein method the one originated by the humidity has been the focus of multiple studies (Fox, 1873; Houzeau, 1857; Linvill et al., 1980; Bojkov, 1986; Marenco et al., 1994; Ramirez-Gonzalez et al., 2020). For this reason, the dependence of the ozonosonde values on humidity, here we also compile daily humidity values for the same days the O_3 observations were conducted at each site.

All the twenty-three sites, whose O_3 observations are reported here, followed a standardized procedure and used the same test-paper. The O_3 observations were conducted following the Schönbein method with the improvements introduced by Bérigny and using the Jame (de Sedan) paper. However, the observations in the Bérigny scale were converted to the decimal Schönbein scale, the one used for its processing and reporting (Fradesso da Silveira, 1865). For most of the observatories two strips of paper were exposed in the period of twenty four hours, reporting measurements each twelve hours. However, at some of the observatories only one strip of paper was exposed with the measurement lasting for twenty four hours, resulting in one daily observation. Further details on the exposure at each observatory are described below.

2.2. Data source: Annaes do Observatorio do Infante D. Luiz

The available information about the meteorological and magnetic observations conducted at the observatory Infante D. Luiz and its twenty-two associated observatories consist of the climatological tables reporting daily, monthly and seasonal means of the observed variables, usually consisting of reports of atmospheric pressure, rain, evaporation, temperature, vapor pressure, humidity, cloud coverage, wind direction and speed. Also, they included O_3, published for the first time in 1863, beginning the series of “Annaes do Observatorio do Infante D. Luiz” (hereinafter AOIDL) reports (see Figure 2 for an example) (Brito Capello, 1863). Only O_3 and humidity data were digitized in the context of the work presented here. O_3 observations were reported in that 1st volume only for the Infante D. Luiz observatory, consisting of the monthly and seasonal means of the diurnal, nightly and daily mean, from December 1855 to November 1863. The 2nd volume, the following year, began to include the daily diurnal and nightly O_3 observations at Infante D. Luiz from December 1863 to November 1864 (Fradesso da Silveira, 1864). The subsequent volumes of the AOIDL, continued including the daily diurnal and nightly O_3 observations at Infante D. Luiz until November 1913 (De Almeida Lima, 1913). The reports from 1914 for the Infante D. Luiz observatory still contained the diurnal and nightly daily observations for all the variables and the columns for O_3 were filled with 0,0 or “ – “ (De Almeida Lima, 1914). No information has been found for the end of the O_3 observations in 1913 at this observatory.

In addition to the Infante D. Luiz O_3 diurnal observations, the 2nd volume published in 1864 included decadal, monthly, and annual O_3 means from other observatories. The following volumes of the AOIDL continued reporting the monthly and seasonal means of the diurnal, nightly and daily mean for the Infante D. Luiz observatory (De Almeida Lima, 1913). Again, no information has been found for the interruption of the O_3 observations.

The reports of the decadal and monthly O_3 means at the associated twenty-two observatories continued after 1864, until 1905 (De Lina Vidal, 1905). Daily O_3 observations from some of those observatories conducted in December 1872 began to be reported intermittently, at least in the records we have already found, in the volume 11, corresponding to 1873 (Fradesso da Silveira, 1873). In the available AOIDL reports we found daily observations for some of the observatories ending in 1887, although the decadal and monthly means continued being reported. No reason has been found for the interruption of the reports; we speculate that the cost of publishing them could be a cause for it, a common reason in many cases. The rescued metadata comes mainly from the several sections (Introduction, Advertency, etc.) included randomly in the AOIDL (Brito Capello, 1863).

An advantage of this work is that all the ozonoscopes of the different observatories were calibrated in the Infante D. Luiz observatory. Figure 1 shows the geographical distribution of the observatories,and

Table 1 lists them, together with their geographical coordinates and their altitude, in decreasing latitude order to facilitate their identification on Figure 1. Figure 2 provides an example of the tables in the AOIDL containing the recovered O_3 data.

3. Data Records

3.1. Daily and monthly mean O_3 series from Infante D. Luiz observatory

The O_3 observations at the Infante D. Luiz observatory began in January 1855, together with a set of meteorological observations (Silvestre, 1881), and continued uninterrupted until October 13th, 1913 (De Almeida Lima, 1914). Between 1853 and September 1863 the station was in a building, with the coordinates 38° 43' 13" N, 9° 8' 20" W. The station was moved to a different building in October 1863, where it has remained until January 1st 1941, located at 38° 42' 59" N, 9° 8' 56" W (De Almeida Lima, 1918; Mendes Víctor, 2001) (at this date, the meteorological instruments were moved from the top of the main building to a new meteorological park next to this building, officially maintaining the same geographical coordinates). However, the measurement of its new location was conducted in 1879 and reported by first time in 1881 (Capello, 1881). No further changes were reported, at least until the end of the data series considered here. Therefore, we have assumed the last reported coordinates for the measurements conducted after October 1863.

Table 2 shows the yearly coverage of the rescued daily series, consisting of almost fifty years of data from 1863 to 1913. The daily observations from 1855 to 1862 were not included in the 1st AOIDL volume (Brito Capello, 1863). However, the monthly means of observations were included, as shown in Table 3. That is the reason for the difference between the number of years of data rescued for the daily and monthly means for the Infante D. Luiz observatory. Both series are by far the longest reported in the literature. They are also among the earliest.

3.2. Daily and monthly mean O_3 series from the other 22 observatories

For four of the stations reported in the AOIDL reports, Alcanhoes, Beja, Faro and Sao Fiel, from 1863 to 1914, we found only monthly mean values. This is why nineteen stations are listed in Table 2 (daily data) and twenty-three in Table 3 (monthly data). Also, Table 2 shows that although two daily O_3 observations were conducted at least at nine observatories, only the two daily observations at the Infante D. Luiz observatory were included in the cited AOIDL reports.

In the existing literature, daily and monthly mean O_3 observations at Campo Maior had already been recovered (Vaquero et al., 2022), and stored in the PANGAEA open access dataset repository (Vaquero et al., 2021). The monthly means series here reported match the one they reported. However, the daily O_3 observations (Brito Capello, 1877) did not contain the observations for the period 1863 to 1872, which we include in the recovered observations reported here. Also daily mean O_3 observations at the Porto observatory from 1861 to 1897 were reported (without making the dataset available) (Alvim-Ferraz et al., 2006). Table 2 shows we were only able to find and recover daily mean O_3 observations from Porto between 1872 and 1887; we were more successful regarding the monthly mean O_3 observations, shown in Table 2, recovering the period from 1862 to 1877 and the years 1897, 1900 and 1901. Daily mean O_3 observations from the Luanda observatory between 1890 and 1895 were used (again without making the dataset available) (Pavelin et al., 1999). On top of them, here we have been able to recover eight additional years of daily and monthly mean O_3 observations from the Luanda observatory, from 1880 to 1887. Neither the Porto's nor the Luanda's O_3 datasets described here had been reported or published in data repositories.

4. Technical Validation

4.1. Recovered datasets quality control

Each variable from the datasets was checked, assuring that they were in the range of its respective physically plausible magnitudes, the so-called limit test (Vaquero et al., 2022), and therefore the consistency of its recorded values.

The homogeneity of the recovered data was tested using the software Climatol 4.0.0 (Guijarro, 2023), which is based on the Standard Normal Homogeneity Test (SNHT) (Alexandersson, 1986). Climatol reconstructs each time series using the data from the neighboring stations and uses the reconstructed series as a reference to check homogeneity. Among the parameters that are set by the user, two are particularly relevant: the distance at which the weight of the reference stations is halved, and the threshold of the SNHT statistic above which an inhomogeneity is considered significant. The former parameter was set at 1000 km, the latter at 25 (the default value) for O_3 and 15 for RH. The measurements taken in Lisbon before 1863 and after 1905 were not checked because of the absence of reference stations. The full results of the homogeneity test are provided as Supplementary Information to this paper.

4.2. Breakpoints

Forty-seven breakpoints were identified in the O_3 series and forty-six in the RH series (in 17 stations). Six breakpoints of the O_3 series coincide with those of the RH series. Additionally, in four of these stations the O_3 breakpoints happen later in the series than those of RH, within an interval of six months with respect to the RH observations (see Tables 4 and 5).

It is noteworthy that for Angra de Heroismo (on 1891-10-01) and Ponta Delgada (on 1867-12-01), the O_3 and RH breakpoints coincide in the same month.

The Montecorvo series has two simultaneous breakpoints for O_3 and RH, with a difference of 8 years. But in the first, the RH breakpoint (on 1879-10-01) precedes that of O_3. The other O_3 breakpoint (on 1887-09-01) happens after the RH breakpoint (on 1887-06-01).

In three of these cases, the coincidence of the interval between O_3 and RH is reduced to one or two months: Angra de Heroismo, Ponta Delgada and Villa Fernando, which could point out to a similar condition (e.g. moving the observatory or instruments) between both changes.

Despite the lack of metadata supporting it, simultaneous breakpoints in both O_3 and RH series could point out a change in the location of the station, as two independent instruments and data series simultaneously suffer alterations. We searched in the AOIDL for information to identify the possible causes of the breakpoints, but we only found information about the Loanda observatory, which is speculated was moved in 1881 (Raposo, 2017), when its RH data shows two breakpoints. For the remainder we did not found anything. There was a slight change in the geographical coordinates of the Infante D. Luiz observatory in 1879, as already described above, but it is unlikely it was a change in the site location. The only changes in the observations were found at São Tome: the initial twenty-four hour strip exposure between 3 PM of consecutive days were reported from March 1873 to January 1882 and in the February report the same year observations change to twelve hour strip exposures from 9 AM to 9 PM and 9 PM until 9 AM of the following day, depicted on Table 2. At this site two breakpoints in the O_3 series were detected in October 1874 and January 1886. No RH breakpoints were reported.

5. Data Availability

The surface O_3 semi-quantitative monthly (Añel et al., 2024b) and daily (Añel et al., 2024a) datasets recovered and reported here have been deposited at PANGAEA, and they are available at https://doi.org/10.1594/PANGAEA.969241 and https://doi.org/10.1594/PANGAEA.969259 respectively.

6. Code Availability

The Climatol 4.0.0 (Guijarro, 2023) software (DOI: 10.5281/zenodo.12786007) was used for the homogeneity test. For computational reproducibility (Añel, 2011, 2017) it is distributed as free software under the GPLv3 license and stored in a permanent Zenodo.org repository (https://zenodo.org/records/12786077).

7. Conclusions

We have recovered semi-quantitative surface O_3 ozonoscope records from the 19th century and the beginning of the 20th century in a new data collection. We include data from 23 observatories covering Portugal and the African Atlantic regions, providing a latitudinal span from the extratropics in the northern hemisphere to the tropics in the southern hemisphere. This record represents the most extended ozonoscope data series recovered to date, spanning 50 years of daily data and 58 years of monthly data, from 1855 to 1913. Moreover, with an small exception for part of a series of an observatory, the existence of the observations here recovered had not been noticed in the previous literature. This dataset presents only a small amount of inhomogeneities, and has the potential to eventually bring unvaluable information on pre-industrial O_3. For example, the fact that it includes daily nightly and diurnal observations may present an opportunity to apply to earlier times our understanding of ozone chemistry and surface ozone variability, which is still developing (Monks et al., 2021). It exist plenty of data from other observatories in logbooks that could be recovered (Bojkov, 1986; Möller, 2022), and such data and the work here published can contribute to a better understanding of pre-industrial O_3 and serve for future research on it.

Author contribution

J.A.A. devised the research, with the help of L.G. and A.S-L.; J.A.A. researched the books containing the datasets with the help of J.C.A.M., L.G., and M.A.V.; The datasets were digitised by J.A.A., J.C.A.M., and C.P.S.; Quality control on the data, including homogenization was performed by J.A.A., J.C.A.M, A.C.S., L.dlT. and Y.B. J.A.A., L.dlT. and L.G. secured the funding. J.A.A. and J.C.A.M. wrote the original draft. All authors have read and agreed to the published version of the manuscript.

Competing interests

The authors declare no competing interests regarding this paper.

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Figures

Figure 1: Map with the location of the observatories for which the data have been recovered.

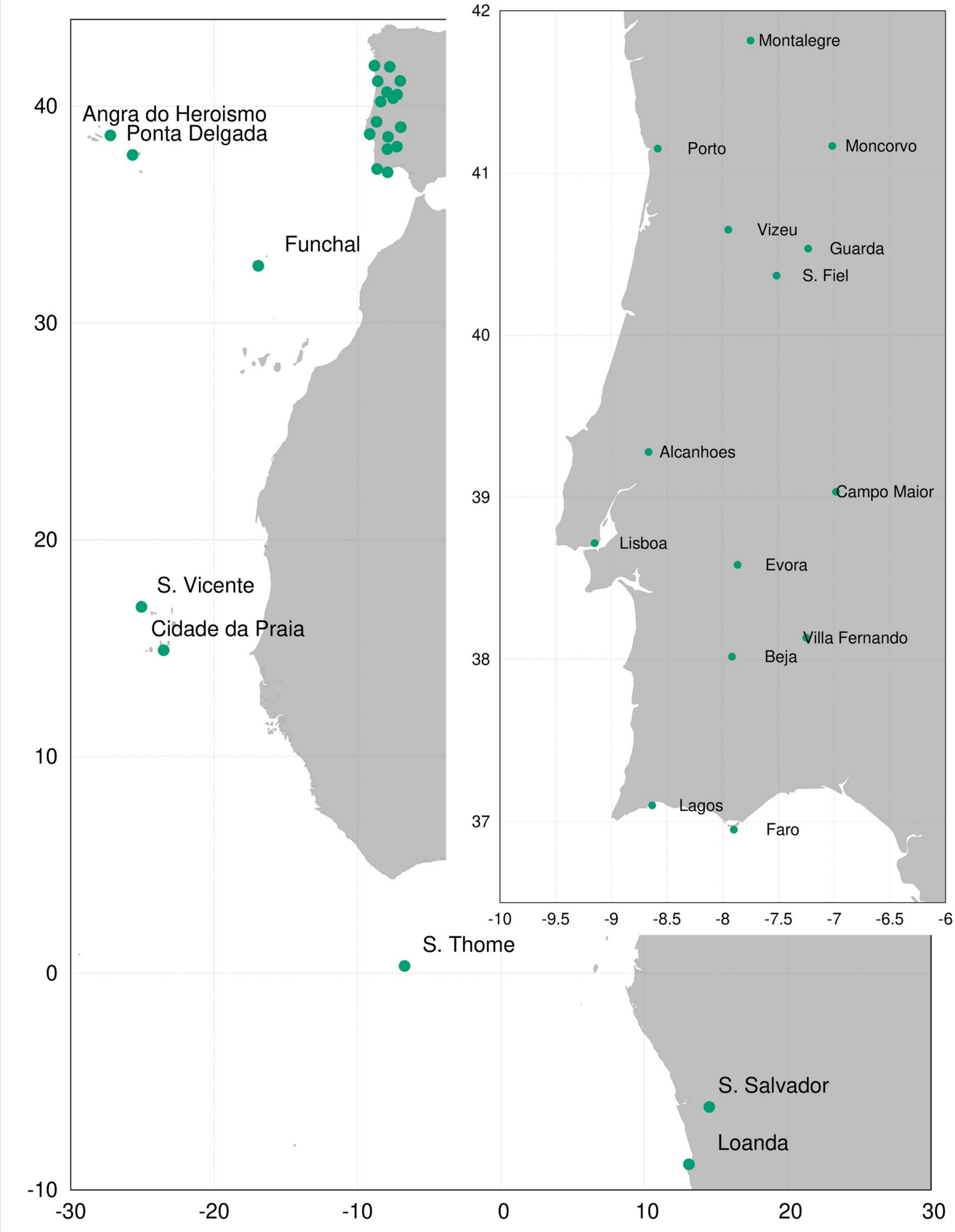

Figure 2: Example of the original tables from the AOIDL containing the recovered O_3 values on the right hand column. Source: Brito Capello (1877).

PORTO

ALTITUDE DO BAROMETRO 85 METROS

Janeiro — 1876	Pressão atmospherica em millimetros				Chuva em millimetros	Evaporação em millimetros	Temperatura em graus centesimaes				Tensão do vapor atmospherico em millimetros		Humidade relativa		Quantidade de nuvens 0 a 10		VENTO: Direcção		VENTO: Velocidade em kilometros			OZONE
	9 A. M.	Maxima (1)	Minima (1)	Media (2)			9 A. M.	Maxima	Minima	Media (3)	9 A. M.	Media (2)	9 A. M.	Media (2)	9 A. M.	Media (1)	9 A. M.	Predominante ou media (1)	9 A. M. (1)	Media diurna	Maxima (1)	Media
1	761,7	761,7	761.1	761.4	0,0	–	3,2	13,0	2.3	7,6	4,9	6.0	86	81	2	1,7	E.	ESE.	fra.	–	–	2,7
2	63,2	63.2	62.5	62,9	0,0	–	2,4	12.4	2.2	7,3	5,1	5,8	93	81	2	3.7	ESE.	ESE.	–	–	–	4,0
3	61.3	61.3	59,1	60.2	0,0	–	3,0	11,2	2,3	6,8	4.7	5.7	83	82	5	5,0	E.	ESE.	fr.	–	–	3,8
4	56,9	56.9	55,0	56,0	0.0	–	5,2	11.3	0,1	5.7	4.5	5,2	69	66	10	10,0	ESE.	ESE.	fr.	–	–	4.0
5	51.9	54.9	54.3	54,6	0,0	–	7.1	11,0	2.4	6.7	5,4	6,1	71	72	10	10,0	E.	E.	fra.	–	–	4,2
6	51.7	51,7	52,9	53,8	0.0	–	8,2	14.2	3.2	8.7	6,0	6,8	75	70	2	2,0	SE.	SE	fra.	–	–	3,5
7	50,1	50.1	47,6	48,9	0,0	–	5.1	13.2	1.1	7 1	5.5	6.1	78	77	2	1,3	SE.	SSE.	fra.	–	–	3,3
8	52.6	53,6	52.6	53.1	0.0	–	1.2	12,2	0,1	6,2	5,0	5,4	80	76	1	1,7	NE.	NNW.	fra.	–	–	4,0
9	50,3	50,3	48,8	49.7	14.6	–	7,0	11.1	1.4	6.2	6,7	6,9	91	88	10	7.0	S.	SSW.	–	–	–	3,5
10	52.1	52,4	51.6	52,0	0,8	–	4.2	10,1	0,2	5.2	4.4	5.1	70	71	2	1.0	NNE.	NNE.	fr.	–	–	5,2
11	56,3	756.3	756,0	756,2	0,0	–	4.4	11,0	0,3	5,6	5,0	5,5	80	72	0	0,0	ENE.	NNE.	fra.	–	–	1.8
12	61,3	61.3	60.2	60.8	0.0	–	4.2	10.4	0,0	5.2	5,1	5,8	83	78	2	2.0	E.	NNE.	fra.	–	–	1.0
13	56,7	56,7	53.9	55.3	0.0	–	5,4	10,2	2,2	6.2	6 4	6,5	97	84	10	7.0	NNW.	NNW.	fra.	–	–	1.2
14	53.5	53,5	52.6	53,1	0,0	–	7,1	12.1	1.4	6.8	5,5	6.1	74	70	10	4,7	NNE.	NNE.	fr.	–	–	4,5
15	57,6	57,6	57.3	57,5	0,0	–	6.3	11,4	2.0	6,7	5.9	6.5	82	77	2	1,0	ESE.	ESE.	fra.	–	–	2,8
16	62.2	62,6	62.1	62.4	0,0	–	7.2	12.4	3,0	7.7	5.5	6.3	74	71	0	0.0	ENE.	ESE.	fr.	–	–	3.0
17	66,8	66,8	65.2	66,0	0.0	–	7,0	13,0	2.4	7.7	5,6	6,6	74	72	2	0,7	NNE.	NNE.	fra.	–	–	3,7
18	65,9	65.9	63.9	64.9	0,0	–	6,4	13,2	2,2	7.7	5,7	6.7	79	74	0	0,0	E.	S.	–	–	–	2,8
19	62.5	62.5	59.9	61.2	0,0	–	5.3	12.3	1.1	6.7	5.3	6,2	81	74	1	1,7	E.	ESE.	fra.	–	–	1,0
20	56,4	56.4	51.3	55.4	0.0	–	5.2	11.4	2.3	6.8	4 8	6.0	71	78	5	7.0	ESE.	SSE.	–	–	–	3.7
21	54,7	751.7	751.0	751.4	5.5	–	6.1	11,1	2.4	6 8	5.9	7.1	85	87	10	10,0	E.	SSE.	fra.	–	–	1,5
22	60.0	60.0	59.1	59.5	3,8	–	8.4	13.1	3.3	6.7	7.1	7.5	86	80	2	1.7	NE.	NNE.	fra.	–	–	1,3
23	62,5	62,5	61.6	62.1	0.0	–	8,3	13,6	3.4	8.5	5.7	6.7	70	69	0	0.7	ESE.	E.	for.	–	–	4,2
24	63,8	63,8	61.6	62.7	0.0	–	9.3	14.3	5.1	9.7	6,1	7.2	73	70	0	0,0	ESE.	ESE.	fr.	–	–	3,0
25	59,8	59,8	58.7	59,3	0.0	–	9.3	15,1	6.0	10.5	6,5	7,5	76	72	10	7,0	ESE.	ESE.	fra.	–	–	3,8
26	62,3	62,4	61.8	62.1	0.0	–	11.2	17,0	7.0	12.0	7,6	9.2	77	78	6	7.0	E.	ESE.	fra.	–	–	1,0
27	61,7	61,7	58.2	60,0	0,0	–	10.1	15,3	5.1	10,2	7.0	8,2	76	77	0	4.7	ESE.	SSW.	fr.	–	–	1,5
28	56,4	57,6	56,4	57,0	15.4	–	11.1	16,1	6.4	11,3	8,6	9,4	87	84	2	4.3	ESE.	SSW.	fra.	–	–	5,2
29	61,9	61.9	61.2	61,6	0.0	–	9,1	15.0	5.1	10.0	7,4	8,0	86	82	4	7,7	E.	SSW.	fra.	–	–	1.3
30	62.8	62,8	61.3	62,0	0.0	–	8.4	16.2	4 3	10,3	7.0	8,5	86	82	3	3,3	E.	E.	fra.	–	–	4.2
31	62,4	62.4	61.3	61,9	0,0	-	10.0	17,0	6,0	11,5	7.2	8.3	79	74	10	7.3	E.	ESE.	fra.	–	–	3.3
Medias	758,89	Maxima 766.8	Minima 747,6	758.33	Total 10,1	–	6.8	12,9	2,8	7,8	5.9	6.7	79.7	76,5	4.0	4,0	E.	ESE.	–	–	–	3,0

Tables

Table 1: List of observatories including country, region, latitude, longitude, and elevation. They are listed in decreasing latitude order. Only monthly mean O_3 series are available from the four stations flagged with (*).

	Observatory	**Country**	**Region**	**Lat.**	**Long.**	**E (m)**
1	Montalegre	Portugal	Iberian Pen.	41.82	-7.75	1027
2	Moncorvo	Portugal	Iberian Pen.	41.17	-7.02	415
3	Porto	Portugal	Iberian Pen.	41.15	-8.58	100
4	Vizeu	Portugal	Iberian Pen.	40.65	-7.95	494
5	Guarda	Portugal	Iberian Pen.	40.53	-7.23	1039
6	Serra da Estrela	Portugal	Iberian Pen.	40.42	-7.58	1450
7	S. Fiel (*)	Portugal	Iberian Pen.	40.37	-7.52	516
8	Alcanhoes (*)	Portugal	Iberian Pen.	39.28	-8.67	—-
9	Campo Maior	Portugal	Iberian Pen.	39.03	-6.98	288
10	Infante D. Luiz	Portugal	Iberian Pen.	38.72	-9.23	95
11	Angra do Heroismo	Portugal	Azores/ Macaronesia	38.65	-27.23	44
12	Evora	Portugal	Iberian Pen.	38.58	-7.87	313
13	Villa Fernando	Portugal	Iberian Pen.	38.13	-7.25	375
14	Beja (*)	Portugal	Iberian Pen.	38.02	-7.92	284
15	Ponta Delgada	Portugal	Azores/ Macaronesia	37.75	-25.68	20
16	Lagos	Portugal	Iberian Pen.	37.10	-8.63	13
17	Faro (*)	Portugal	Iberian Pen.	36.95	-7.90	14
18	Funchal	Portugal	Madeira/ Macaronesia	32.63	-16.92	25
19	S. Vicente	Cape Verde	Macaronesia	16.90	-25.07	11
20	Cidade da Praia	Cape Verde	Macaronesia	14.90	-23.52	34
21	S. Thome	Saint Thomas & Prince	Gulf of Guinea	0.33	-6.72	7
22	S. Salvador do Congo	Angola	Continental Africa	-6.17	14.53	559
23	Loanda	Angola	Continental Africa	-8.82	13.12	59

Table 2: Temporal coverage of the rescued daily O_3 for each of the 19 observatories (Infante D. Luiz and associated observatories). The stations are listed in decreasing order of the available number of years with data. Blue cells correspond to the years with data rescued. The numbers inside the cells represents the number of observations conducted daily. One daily observation (1) consisted in a 24-hour strip exposure, between 3 PM of consecutive days. Two daily observations (2) consisted in a 12-hour strip exposure from 9 AM to 9 PM and 9 PM until 9 AM of the following day. Blue cells without number indicate that no information was found about the number of daily observations.

Stations	Infante D. Luiz	Ponta Delgada	Funchal	Porto	Guarda	Campo Maior	Angra do Heroismo	Evora	São Thome	Moncorvo	Lagos	Montalegre	Vizeu	Loanda	Cidade da Praia	Serra da Estrela	Villa Fernando	São Vicente	São Salvador
1863	2																		
1864	2																		
1865	2																		
1866	2																		
1867	2																		
1868	2																		
1869	2																		
1870	2																		
1871	2																		
1872	2	2	2	2	1	2	2	1											
1873	2	2	2	2	1	2	2	1											
1874	2	2	2	2	1	2	2	1	1		1								
1875	2	2	2	2	1	2	2	1	1		1								
1876	2	2	2	2	1	2	2	1	1		1								
1877	2	2	2	2	1	2	2	1	1		1								
1878	2	2	2	2	1	2	2	1	1	1	1								
1879	2	2	2	2	1	2	2	1	1	1	1								
1880	2	2	2	2	1	2	2	1	1	1	1								
1881	2	2	2	2	1	2	2	1	1	1	1	1	2						
1882	2	2	2	2	1	2	2	1	1	1	1	1	2						
1883	2	2	2	2	1	2	2	1	1	1	1	1	2	2		2			
1884	2	2	2	2	1	2	2	1	2	1		1	2	2		2			
1885	2	2	2	2	1	2	2	1	2	1		1	2	2		2			
1886	2	2	2	2	1	2	2	1	2	1		1	2	2		2			
1887	2	2	2	2	1	2	2	1	2	1		1	2	2		2			
1888	2	2	2		1		2	1	2	1		1	2			2			
1889	2																		
1890	2																		
1891	2																		
1892	2																		
1893	2																		
1894	2																		
1895	2																		
1896	2																		
1897	2																		
1898	2																		
1899	2																		
1900	2																		
1901	2																		
1902	2																		
1903	2																		
1904	2																		
1905	2																		
1906	2																		
1907	2																		
1908	2																		
1909	2																		
1910	2																		
1911	2																		
1912	2																		
1913	2																		

Table 3: Temporal coverage of the rescued monthly mean O_3 for each of the observatories: period rescued and available data. Brown cells correspond to the years with data rescued. The numbers inside the cells represent the number of monthly means available per year.

Station	1855	1856	1857	1858	1859	1860	1861	1862	1863	1864	1865	1866	1867	1868	1869	1870	1871	1872	1873	1874	1875	1876	1877	1878	1879	1880	1881	1882	1883	1884
Alcanhoes															9	12	3													
Angra do Heroismo												3	11	12	12	11	12	11												
Beja																														
Campo Maior									1	12	12	12	12	12	12	12	12	11												
Cidade da Praia										1	12	5																		
Evora															1	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12
Faro																														
Funchal										1	11	11	12	12	12	12	12	11	12	12	12	11	12	12	10	12	12	10	12	10
Guarda									12	12	12	12	12	12	12	12	12	12	12	12	12	11	12	12	12	12	11	12	12	12
Illa de S. Vicente																														12
Infante D Luiz	1	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12
Lagos																		6	6	10	12	12	11	12	11	12	12	5		
Loanda																										12	12	12	12	12
Moncorvo																							12	12	12	12	12	12	12	12
Montalegre																									12	12	12	12	12	12
Ponta Delgada											10	12	12	12	11	10	12	12	11	12	12	12	12	12	12	12	12	12	12	12
Porto								1	12	12	12	12	12	11	12	12	12	12	12	12	12	11	12	12	12	12	12	12	12	12
S. Fiel																														
S. Thome																		5	8	12	12	8	12	12	12	12	12	10	10	6
Salvador do Congo																													6	4
Serra da Estrela																												11	12	12
Villa Fernando																														7
Vizeu																									12	12	12	9	12	12

Station	1885	1886	1887	1888	1889	1890	1891	1892	1893	1894	1895	1896	1897	1898	1899	1900	1901	1902	1903	1904	1905	1906	1907	1908	1909	1910	1911	1912	1913
Alcanhoes																													
Angra do Heroismo				12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12										
Beja													12	12	12	12	12	12	12	12	12								
Campo Maior				10	11	12	12	12	12																				
Cidade da Praia					11	12														12	12								
Evora	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12			12	12									
Faro											12	12	12	12	12	11	11	11	12	12	12								
Funchal	11	11	12	12	12	12	11	12		11		12	12	12	11	11	12	12	12	12	12								
Guarda	12	12	12	12	12	12	12	12	12	12	12	12	12	11	12	12	12	12	12	12	12								
Illa de S. Vicente	11	10	10	11	12	12		12	12	11	12	12	12	12	12	12													
Infante D Luiz	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	10
Lagos																				11	12								
Loanda	12	12	12																										
Moncorvo	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12								
Montalegre	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12								
Ponta Delgada	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12										
Porto	11	12	12										12			12	12												
S. Fiel																		11	12	12	12								
S. Thome		12	12	12	12																								
Salvador do Congo	11	11	12	8																									
Serra da Estrela	12	11	11	12	12	12	11	10	11	12	12	12	12	10	9	12	12	12	11	12	12								
Villa Fernando	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	11	11	12	12	12	12								
Vizeu	12	12	12																										

Table 4: Observatory, date and SNHT value for the O_3 breakpoints.

Observatory	Date	SNHT
Angra do Heroismo	1891-10-01	59.8
Beja	1902-11-01	31.2
Campo Maior	1866-05-01	25.8
Campo Maior	1888-01-01	27.6
Campo Maior	1891-12-01	39.2
Evora	1874-01-01	28.4
Evora	1890-07-01	66.7
Evora	1899-11-01	45.4
Funchal	1870-09-01	28.2
Funchal	1879-11-01	25.7
Funchal	1885-04-01	48.2
Funchal	1901-01-01	33.4
Guarda	1864-01-01	73.9
Guarda	1867-11-01	25.6
Guarda	1871-08-01	35.8
Guarda	1885-05-01	55.5
Guarda	1887-10-01	26.7
Guarda	1896-07-01	55.7
Infante D. Luiz	1866-06-01	30.9
Infante D. Luiz	1875-10-01	30.6
Infante D. Luiz	1879-04-01	70.6
Infante D. Luiz	1883-05-01	54.4
Infante D. Luiz	1889-12-01	27.0
Lagos	1904-02-01	65.2
Moncorvo	1878-11-01	36.8
Moncorvo	1879-08-01	43.9
Moncorvo	1887-09-01	87.0
Moncorvo	1904-10-01	26.9
Montalegre	1880-06-01	27.8
Montalegre	1892-05-01	69.9
Ponta Delgada	1867-12-01	88.6
Ponta Delgada	1898-09-01	40.1
Ponta Delgada	1902-06-01	25.1
Porto	1863-12-01	91.6
Porto	1886-10-01	37.5
Porto	1900-06-01	30.3
Porto	1900-12-01	26.4
S. Thome	1874-10-01	28.7
S. Thome	1886-01-01	76.6
S. Vicente	1886-07-01	36.9
S. Vicente	1892-02-01	32.2
S. Vicente	1895-04-01	36.8
Serra da Estrela	1889-08-01	26.6
Villa Fernando	1890-02-01	62.8
Villa Fernando	1896-07-01	75.2
Villa Fernando	1903-02-01	30.8
Vizeu	1882-10-01	31.6

Table 5: Observatory, date and SNHT value for the RH breakpoints.

Observatory	Date	SNHT
Angra do Heroismo	1888-05-01	55.1
Angra do Heroismo	1891-10-01	53.0
Angra do Heroismo	1902-05-01	15.7
Campo Maior	1864-07-01	16.5
Campo Maior	1872-11-01	31.5
Campo Maior	1890-06-01	17.4
Cidade da Praia	1904-06-01	21.7
Evora	1873-10-01	28.2
Evora	1878-04-01	37.0
Evora	1883-10-01	15.5
Evora	1888-12-01	39.5
Evora	1904-03-01	15.1
Faro	1904-04-01	15.6
Funchal	1871-10-01	24.8
Funchal	1884-06-01	16.6
Funchal	1888-11-01	32.1
Funchal	1894-03-01	39.3
Funchal	1896-11-01	18.6
Guarda	1879-08-01	69.1
Guarda	1887-09-01	20.3
Guarda	1895-10-01	16.2
Guarda	1904-04-01	17.8
Infante D Luiz	1863-10-01	28.4
Infante D Luiz	1866-09-01	15.3
Infante D Luiz	1873-12-01	18.1
Infante D Luiz	1891-09-01	21.9
Loanda	1881-05-01	19.4
Loanda	1884-10-01	17.4
Moncorvo	1879-10-01	27.9
Moncorvo	1887-06-01	59.1
Montalegre	1894-11-01	15.2
Ponta Delgada	1867-12-01	65.1
Ponta Delgada	1887-04-01	19.3
Ponta Delgada	1894-03-01	28.3
Ponta Delgada	1896-08-01	18.9
Porto	1882-03-01	27.4
Porto	1883-09-01	17.1
Porto	1885-01-01	63.9
Porto	1885-08-01	15.9
Porto	1887-06-01	78.3
S. Fiel	1902-07-01	16.0
S. Fiel	1904-10-01	18.1
S. Salvador do Congo	1885-11-01	21.1
S. Vicente	1889-03-01	24.2
S. Vicente	1890-12-01	30.2
Vizeu	1882-04-01	16.8