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Nineteenth- and twentieth-century semi-quantitative surface ozone along subtropical European 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 19th century, which hampers the understanding of long-term climate trends (given the crucial role of O_3), and (ii) and the uncertainties in quantitative tropospheric O_3 values in a non-polluted atmosphere across the planet. The ozonoscope was the first instrument used to measure ozone. It offered semi-quantitative estimates of surface O_3 when no other measurements were available. Despite their potential value, the digitization, curation, and publication of ozonoscope data remain largely unexplored. In this work, we initiate an effort to rescue surface O_3 ozonoscope records by compiling them 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 extensive ozonoscope data series to date, spanning 50 years of daily data and 58 years of monthly data, from 1855 to 1913. This record represents the most extensive ozonoscope data series to date, spanning 50 years of daily data and 58 years of monthly data, from 1855 to 1913. The data series is available from https://doi.org/10.1594/PANGAEA.969259 (Añel et al., 2024a) and https://doi.org/10.1594/PANGAEA.969241 (Añel et al., 2024b), respectively.

1 Background and summary

Tropospheric ozone (O_3) records for our planet before the end of the 19th century are rare and sparse. This is not surprising given that O_3 was discovered by Schönbein in 1839 (Schönbein, 1840a, b) and got little attention during the decades following its discovery. After the 1860s, measuring O_3 became common at meteorological stations. However, O_3 is a transcendental chemical with respect to the understanding and study of the atmosphere. Tropospheric O_3 is a greenhouse gas and, at elevated concentrations, a pollutant harmful to human health; moreover, it affects crops and ecosystem 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 19th century, which hampers the understanding of long-term climate trends, (ii) and the uncertainties in quantitative O_3 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 that consisted of two main periods: the modern period, beginning around 1975 and spanning to the present, defined by the widespread availability of sensitive ultraviolet (UV) photometers for surface O_3 measurements, and the historical period, covering the period from 1877 to 1975, defined by the use of other techniques and the lack of UV photometers. The records available for the period prior to 1975 were evaluated using a set of four criteria to minimize uncertainties and biases between the measurement techniques available at that time and the contemporary UV absorption standard. These criteria are as follows: 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 19th century and extending until the early 20th century using the test paper method, also called "ozonoscope" measurements, were among those disregarded (Tarasick et al., 2019).

Considering the scientific questions motivating the TOAR project, associated with the global distribution and trends of surface O_3 pollution (Gaudel et al., 2018), the decision not to include the 19th-century semi-quantitative O_3 measurements in the TOAR database is grounded. However, there are other scientific questions and uncertainties related to the global distribution of O_3 and to surface O_3 pollution during the pre-industrial era, such as the atmospheric concentration in non-polluted areas and the evaluation of the assumed O_3 concentrations, the study of local sources of O_3, and the understanding of the role of such O_3 levels in the radiative balance. Clarification of the abovementioned questions and uncertainties could benefit from the combined use of quantitative surface O_3 observations and semi-quantitative O_3 observations from ozonoscopes. Although ozonoscope measurements are vulnerable to the influence of humidity and oxidants in the air, these O_3 observations will enable us to semi-quantitatively study climate variables under conditions comprising very low (or no) exposure to anthropogenic activity during a period for which no other measurements are available (Bojkov, 1986), thereby filling a gap in our knowledge of surface O_3 in the 19th century.

Efforts to recover some of these 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, to our knowledge, only a single sample of these surface O_3 datasets has been digitized and published in a public data repository, and this was done during data recovery efforts focused on something other than O_3 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 Fig. 1. The semi-quantitative O_3 observations provide a latitudinal coverage from the extratropics in the Northern Hemisphere to the 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, as well as their corresponding metadata, have been digitized from the original documentary sources. They are representative of very different regions of the planet, such as the tropics, oceans, and coastal areas. One of them, from the Infante D. Luiz Observatory, located in Lisbon, Portugal, provides almost 50 years of continuous daily data for the period from 1863 to 1913 and nearly 58 years of monthly means for the period from 1855 to 1913, thereby constituting the most extensive and earliest surface O_3 ozonoscope data series known to date. Before this work, the longest and earliest reported series was the 31-year Montsouris Observatory O_3 ozonoscope data series, which began in 1876 (Bojkov, 1986). Additionally, another 10 of the daily records recovered here cover between 12 and 17 years of data, while 4 monthly mean records extend between 33 and 42 years. The difficulty involved with finding records of meteorological variables covering oceanic regions for the pre-industrial era, 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 eastern Atlantic (two from the Azores, one from Madeira, two from Cabo Verde, and one from Sao Tome and Principe). The data series have been tested for break points and inhomogeneities, although few were found. Unfortunately, we have discovered scarce metadata that allow us to provide context for the existing break points; however, in several cases, a change in the location of the observatory or instruments seems like a plausible explanation.

In the next section, the Schönbein test paper method and its further improvement by Bérigny are briefly described, followed by a description of the data sources. Then, in Sect. 3, 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 22 observatories. Section 4 explains the homogeneity tests applied.

2 Methods

2.1 The test paper method

The test paper measurement method was based on the colour change of an indicator test paper. A strip of blotting paper was coated with starched potassium iodide and then exposed to air for between 8 and 24 h (while protected from solar radiation and rain). After exposure, the strip was moistened and developed a bluish colour associated with the formation of

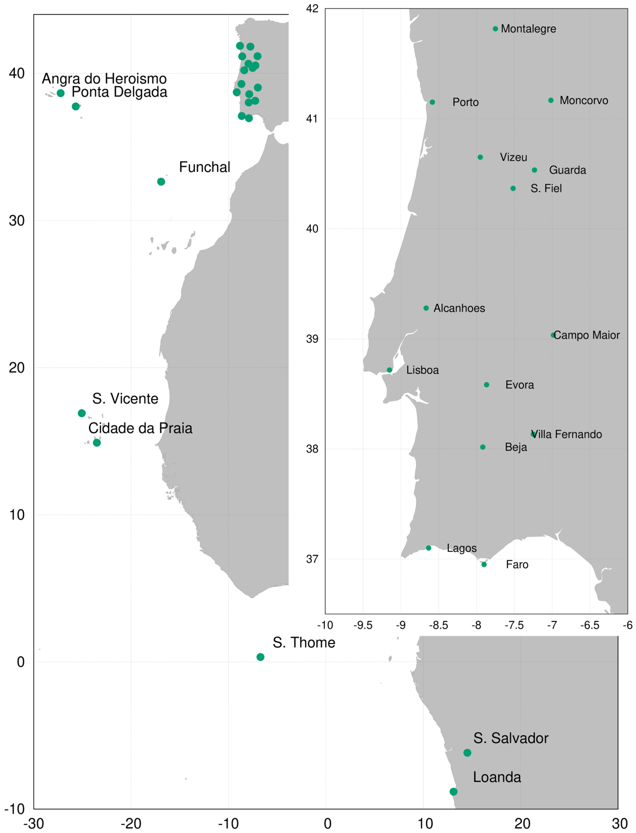

Figure 1. Map showing the location of the observatories for which data have been recovered.

a complex between starch and iodide, produced by the reaction between O_3 and iodide. The colouration depends on the O_3 concentration. Finally, the observed colour 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 colour depending on the extent of the iodide reaction with not only ozone but also humidity and other atmospheric oxidants (Houzeau, 1857; Fox, 1873). When air reaches water vapour saturation, it causes the pre-dried paper to humidify, thereby increasing the rate of O_3 absorption (Kley et al., 1988; Volz and Kley, 1988). These are the reasons for a non-linear correlation between the colour changes and the ozone concentration.

Bérigny introduced the Schönbein 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 impregnated paper (Berzelius paper, manufactured by Jame, a chemist in Sedan, France) (Marenco et al., 1994). This scale was often referred to in logbooks containing measurements as "Jame de Sedan".

In the more than 150 years since 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). These studies, for example, estimated that the O_3 levels for 1880–1900 were approximately 10 ppb in the Great Lakes area of North America, with an annual cycle maximum in April–June and a minimum in October–November (Bojkov, 1986). Another study using observations from Montevideo in Uruguay (1883–1885) and Cordoba in 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 of the reported studies; thus, we refer readers to the review conducted by Marenco et al. (1994). Among the cited interferences present in the Schönbein method, the interference caused by 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), we also compile daily humidity values for the same days that the O_3 observations were conducted at each site.

All 23 sites with O_3 observations 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 Jame (de Sedan) paper. However, observations in the Bérigny scale were converted to the decimal Schönbein scale for processing and reporting (Fradesso da Silveira, 1865). For most of the observatories, two strips of paper were exposed over a period of 24 h, reporting measurements each 12 h. However, at some of the observatories, only one strip of paper was exposed for 24 h, resulting in one daily observation. Further details on the exposure methods used at each observatory are described below.

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

Available information on the meteorological and magnetic observations conducted at the Infante D. Luiz Observatory and its 22 associated observatories consists of climatological tables reporting daily, monthly, and seasonal means of the observed variables, usually consisting of reports of atmospheric pressure, rain, evaporation, temperature, vapour pressure, humidity, cloud coverage, and wind direction and speed. Moreover, O_3 observations were included, published for the first time in 1863, beginning the series of *Annaes do Observatorio do Infante D. Luiz* (hereinafter AOIDL) reports (see Fig. 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 first volume for the Infante D. Luiz Observatory alone, consisting of the monthly and seasonal means of the diurnal, nightly, and daily mean, from December 1855 to Novem-

PORTO

ALTITUDE DO BAROMETRO 85 METROS

Janeiro — 1878	Pressão atmospherica em millimetros 9 A. M.	Maxima (1)	Minima (1)	Media (2)	Chuva em millimetros	Evaporação em millimetros	Temperatura em graus centesimaes 9 A. M.	Maxima	Minima	Media (3)	Tensão do vapor atmospherico em millimetros 9 A. M.	Media (2)	Humidade relativa 9 A. M.	Media (2)	Quantidade de nuvens 0 a 10 9 A. M.	Media (1)	VENTO Direcção 9 A. M.	Predominante ou media (1)	Velocidade em kilometros 9 A. M. (1)	Media diurna	Maxima (1)	OZONE 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	–	2,1	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	5.3	6.4	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	4.1	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	81	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	[illegible]	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

Figure 2. Example of the original tables from the *Annaes do Observatorio do Infante D. Luiz* (AOIDL; source: Brito Capello, 1877). The recovered O_3 values are shown in the right-hand column.

ber 1863. The second volume, the following year, began to include daily diurnal and nightly O_3 observations at Infante D. Luiz Observatory from December 1863 to November 1864 (Fradesso da Silveira, 1864). The subsequent volumes of the AOIDL continued to include daily diurnal and nightly O_3 observations at Infante D. Luiz Observatory 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 of the variables, but the columns for O_3 were filled with 0.0 or "– " (De Almeida Lima, 1914). No information was found for the end of the O_3 observations in 1913 at this observatory.

In addition to the Infante D. Luiz Observatory O_3 diurnal observations, the second 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 with respect to the interruption in the O_3 observations.

The reports of the decadal and monthly O_3 means at the associated 22 observatories continued after 1864, until 1905 (De Lina Vidal, 1905). Daily O_3 observations from some of these observatories became intermittent in December 1872, at least in the records that we have found (in volume 11, corresponding to 1873) (Fradesso da Silveira, 1873). In the available AOIDL reports, we found that daily observations for some of the observatories ended in 1887, although the decadal and monthly means continued to be reported. No reason was found for the interruption in the reports, although we speculate that the cost of publishing them could be a cause, as this is a common reason in many cases. The rescued metadata mainly come from several sections (e.g. "Introduction" and "Advertency") included randomly in the AOIDL (Brito Capello, 1863).

An advantage of this work is that all of the ozonoscopes of the different observatories were calibrated at the Infante D. Luiz Observatory. Figure 1 shows the geographical distribution of the observatories, and Table 1 lists them, along with their geographical coordinates and their altitude, in order of decreasing latitude to facilitate their identification in Fig. 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, along with a set of meteorological observations (Silvestre, 1881), and continued uninterrupted until 13 October 1913 (De Almeida Lima, 1914). Between 1853 and September 1863, the station was situated in a building located at 38°43′13″ N, 9°8′20″ W. The station was moved to a different building in October 1863 (at 38°42′59″ N, 9°8′56″ W), where it remained until 1 January 1941 (De Almeida Lima, 1918; Mendes Víctor, 2001); on 1 January 1941, the meteorological instruments were moved from the top of the main building to a new meteo-

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

	Observatory	Country	Region	Latitude	Longitude	Elevation (m)
1	Montalegre	Portugal	Iberian Peninsula	41°49′ N	7°45′ W	1027
2	Moncorvo	Portugal	Iberian Peninsula	41°10′ N	7°01′ W	415
3	Porto	Portugal	Iberian Peninsula	41°09′ N	8°34′ W	100
4	Vizeu	Portugal	Iberian Peninsula	40°39′ N	7°57′ W	494
5	Guarda	Portugal	Iberian Peninsula	40°32′ N	7°13′ W	1039
6	Serra da Estrela	Portugal	Iberian Peninsula	40°25′ N	7°34′ W	1450
7	São Fiel*	Portugal	Iberian Peninsula	40°22′ N	7°31′ W	516
8	Alcanhões*	Portugal	Iberian Peninsula	39°17′ N	8°40′ W	–
9	Campo Maior	Portugal	Iberian Peninsula	39°01′ N	6°58′ W	288
10	Infante D. Luiz	Portugal	Iberian Peninsula	38°43′ N	9°8′ W	95
11	Angra do Heroísmo	Portugal	Azores/Macaronesia	38°39′ N	27°13′ W	44
12	Evora	Portugal	Iberian Peninsula	38°34′ N	7°52′ W	313
13	Villa Fernando	Portugal	Iberian Peninsula	38°07′ N	7°15′ W	375
14	Beja*	Portugal	Iberian Peninsula	38°01′ N	7°55′ W	284
15	Ponta Delgada	Portugal	Azores/Macaronesia	37°45′ N	25°40′ W	20
16	Lagos	Portugal	Iberian Peninsula	37°06′ N	8°37′ W	13
17	Faro*	Portugal	Iberian Peninsula	36°57′ N	7°54′ W	14
18	Funchal	Portugal	Madeira/Macaronesia	32°37′ N	16°55′ W	25
19	São Vicente	Cabo Verde	Macaronesia	16°54′ N	25°04′ W	11
20	Cidade da Praia	Cabo Verde	Macaronesia	14°54′ N	23°31′ W	34
21	São Thome	Sao Tome and Principe	Gulf of Guinea	0°19′ N	6°43′ W	7
22	São Salvador do Congo	Angola	Continental Africa	6°10′ S	14°31′ E	559
23	Luanda	Angola	Continental Africa	8°49′ S	13°07′ E	59

rological park next to this building, officially maintaining the same geographical coordinates. However, measurements at this new location were first conducted in 1879 and were first reported in 1881 (Brito 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 50 years of data from 1863 to 1913. The daily observations from 1855 to 1862 were not included in the first AOIDL volume (Brito Capello, 1863); however, the monthly means of observations were included, as shown in Table 3. This 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, Alcanhões, Beja, Faro, and São Fiel, from 1863 to 1914, we found only monthly mean values. This is why 19 stations are listed in Table 2 (daily data), whereas 23 are listed in Table 3 (monthly data). Furthermore, Table 2 shows that, although two daily O_3 observations were conducted at at least 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 have been recovered (Vaquero et al., 2022) and stored in the PANGAEA open-access dataset repository (Vaquero et al., 2021). The monthly mean series reported here match the one that they reported. However, the daily O_3 observations (Brito Capello, 1877) did not contain the observations for the period from 1863 to 1872, which we include in the recovered observations reported here. Moreover, 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 that we were only able to find and recover daily mean O_3 observations from Porto between 1872 and 1887; however, we were more successful regarding the monthly mean O_3 observations, as 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). In addition to these observations, we were able to recover 8 additional years of daily and monthly mean O_3 observations from the Luanda Observatory, from 1880 to 1887. Neither the Porto nor the Luanda O_3 datasets described here had been reported or published in data repositories.

Table 2. Temporal coverage of the rescued daily O_3 observations for each of the 19 observatories (Infante D. Luiz Observatory and the other associated observatories). The stations are listed in decreasing order based on the available number of years with data. Blue cells correspond to the years with data rescued. The numbers inside the cells represent the number of observations conducted daily. One daily observation (1) consisted of a 24 h strip exposure, from 15:00 to 15:00 LT (local time) of the following day. Two daily observations (2) consisted of one 12 h strip exposure from 09:00 to 21:00 LT and another exposure from 21:00 to 09:00 LT of the following day. Blue cells without a number indicate that no information was found about the number of daily observations.

Stations	1863	1864	1865	1866	1867	1868	1869	1870	1871	1872	1873	1874	1875	1876	1877	1878	1879	1880	1881	1882	1883	1884	1885	1886	1887	1888
Infante D. Luiz	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
Ponta Delgada										2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
Funchal										2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
Porto										2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	
Guarda										1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
Campo Maior										2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	
Angra do Heroismo										2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
Evora										1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
São Thome												1	1	1	1	1	1	1	1	1	1	2	2	2	2	2
Moncorvo																1	1	1	1	1	1	1	1	1	1	1
Lagos												1	1	1	1	1	1	1	1	1	1					
Montalegre																			1	1	1	1	1	1	1	1
Vizeu																			2	2	2	2	2	2	2	2
Luanda																					2	2	2	2	2	
Cidade da Praia																										
Serra da Estrella																					2	2	2	2	2	2
Villa Fernando																										
São Vicente																										
São Salvador																										

Stations	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
Infante D. Luiz	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
Ponta Delgada																									
Funchal																									
Porto																									
Guarda																									
Campo Maior																									
Angra do Heroismo																									
Evora																									
São Thome																									
Moncorvo																									
Lagos																									
Montalegre																									
Vizeu																									
Luanda																									
Cidade da Praia																									
Serra da Estrella																									
Villa Fernando																									
São Vicente																									
São Salvador																									

Table 3. Temporal coverage of the rescued monthly mean O_3 observations for each of the observatories, outlining the period rescued and the 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
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
Faro																				
Funchal										1	11	11	12	12	12	12	12	11	12	12
Guarda									12	12	12	12	12	12	12	12	12	12	12	12
Illa de S. Vicente																				
Infante D Luiz	1	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12
Lagos																		6	6	10
Loanda																				
Moncorvo																				
Montalegre																				
Ponta Delgada											10	12	12	12	11	10	12	12	11	12
Porto								1	12	12	12	12	12	11	12	12	12	12	12	12
S. Fiel																				
S. Thome																		5	8	12
Salvador do Congo																				
Serra da Estrela																				
Villa Fernando																				
Vizeu																				

Station	1875	1876	1877	1878	1879	1880	1881	1882	1883	1884	1885	1886	1887	1888	1889	1890	1891	1892	1893	1894
Alcanhoes																				
Angra do Heroismo														12	12	12	12	12	12	12
Beja																				
Campo Maior														10	11	12	12	12	12	
Cidade da Praia															11	12				
Evora	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12
Faro																				
Funchal	12	11	12	12	10	12	12	10	12	10	11	11	12	12	12	12	11	12		11
Guarda	12	11	12	12	12	12	11	12	12	12	12	12	12	12	12	12	12	12	12	12
Illa de S. Vicente										12	11	10	10	11	12	12		12	12	11
Infante D Luiz	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12
Lagos	12	12	11	12	11	12	12	5												
Loanda						12	12	12	12	12	12	12	12							
Moncorvo			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
Ponta Delgada	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12	12
Porto	12	11	12	12	12	12	12	12	12	12	11	12	12							
S. Fiel																				
S. Thome	12	8	12	12	12	12	12	10	10	6		12	12	12	12					
Salvador do Congo									6	4	11	11	12	8						
Serra da Estrela								11	12	12	12	11	11	12	12	12	11	10	11	12
Villa Fernando										7	12	12	12	12	12	12	12	12	12	12
Vizeu					12	12	12	9	12	12	12	12	12							

Station	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										
Beja			12	12	12	12	12	12	12	12	12								
Campo Maior																			
Cidade da Praia										12	12								
Evora	12	12	12	12	12	12			12	12									
Faro	12	12	12	12	12	11	11	11	12	12	12								
Funchal		12	12	12	11	11	12	12	12	12	12								
Guarda	12	12	12	11	12	12	12	12	12	12	12								
Illa de S. Vicente	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	10
Lagos										11	12								
Loanda																			
Moncorvo	12	12	12	12	12	12	12	12	12	12	12								
Montalegre	12	12	12	12	12	12	12	12	12	12	12								
Ponta Delgada	12	12	12	12	12	12	12	12	12										
Porto			12			12	12												
S. Fiel								11	12	12	12								
S. Thome																			
Salvador do Congo																			
Serra da Estrela	12	12	12	10	9	12	12	12	11	12	12								
Villa Fernando	12	12	12	12	12	12	11	11	12	12	12								
Vizeu																			

4 Technical validation

4.1 Quality control of the recovered datasets

Using the so-called limit test, each variable from the datasets was checked, assuring that it was in the range of its respective physically plausible magnitudes (Vaquero et al., 2022); thus, the consistency of its recorded values was also examined.

The homogeneity of the recovered data was tested using the Climatol software (version 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 neighbouring stations, and it employs the reconstructed series as a reference to check homogeneity. Among the parameters that are set by the user, two are particularly relevant: (1) the distance at which the weight of the reference stations is halved and (2) the threshold of the SNHT statistic above which an inhomogeneity is considered significant. The former parameter was set at 1000 km, and the latter was set 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 in the Supplement.

Table 4. Observatory, date, and SNHT value for the O_3 break points.

Observatory	Date (yyyy-mm-dd)	SNHT
Angra do Heroísmo	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
São Thome	1874-10-01	28.7
São Thome	1886-01-01	76.6
São Vicente	1886-07-01	36.9
São Vicente	1892-02-01	32.2
São 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

4.2 Break points

A total of 47 break points were identified in the O_3 series, while 46 break points were identified in the RH series (at 17 stations). Six break points in the O_3 series coincide with those in the RH series. Additionally, at four of these stations, the O_3 break points happen later in the series than those of RH, within an interval of 6 months with respect to the RH observations (see Tables 4 and 5).

It is noteworthy that the O_3 and RH break points coincide in the same month for Angra de Heroísmo (on 1 October 1891) and Ponta Delgada (on 1 December 1867).

The Montecorvo series has two simultaneous break points for O_3 and RH, with a difference of 8 years. However, in the first, the RH break point (on 1 October 1879) precedes that of O_3. The other O_3 break point (on 1 September 1887) happens after the RH break point (on 1 June 1887).

In three of these cases, the coincidence of the interval between O_3 and RH is reduced to 1 or 2 months: Angra de Heroísmo, Ponta Delgada, and Villa Fernando, which could point to a similar condition (e.g. moving the observatory or instruments) between both changes.

Despite the lack of metadata supporting it, simultaneous break points in both the O_3 and RH series could point to a change in the location of the station, as two independent instruments and data series simultaneously suffer alterations. We searched the AOIDL for information to identify the possible causes of the break points, but we only found information about the Luanda Observatory, which is speculated to have been moved in 1881 (Raposo, 2017), when its RH data show two break points. For the remainder, we did not find 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 that it was a change in the site location. The only changes in the observations were found at São Tome: initial 24 h strip exposure (from 15:00 LT to 15:00 LT on consecutive days, where LT denotes local time) observations were reported from March 1873 to January 1882; however, in the February report the same year, observations changed to 12 h strip exposures (from 09:00 to 21:00 LT and from 21:00 to 09:00 LT of the following day), as depicted on Table 2. At this site, two break points in the O_3 series were detected – in October 1874 and January 1886. No RH break points were reported.

5 Data availability

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

6 Code availability

The Climatol (version 4.0.0; Guijarro, 2023) software (https://doi.org/10.5281/zenodo.12786077, Guijarro and Añel, 2024) was used for the homogeneity test. For

Table 5. Observatory, date, and SNHT value for the RH break points.

Observatory	Date (yyyy-mm-dd)	SNHT
Angra do Heroísmo	1888-05-01	55.1
Angra do Heroísmo	1891-10-01	53.0
Angra do Heroísmo	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
Luanda	1881-05-01	19.4
Luanda	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
São Fiel	1902-07-01	16.0
São Fiel	1904-10-01	18.1
São Salvador do Congo	1885-11-01	21.1
São Vicente	1889-03-01	24.2
São Vicente	1890-12-01	30.2
Vizeu	1882-04-01	16.8

computational reproducibility (Añel, 2011, 2017), it is distributed as free software under the GPLv3 licence and stored in a permanent Zenodo repository (https://doi.org/10.5281/zenodo.12786077, Guijarro and Añel, 2024).

7 Conclusions

We have recovered semi-quantitative surface O_3 ozonoscope records from the 19th century and the beginning of the 20th century and compiled them 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 extensive ozonoscope data series recovered to date, spanning 50 years of daily data and 58 years of monthly data, from 1855 to 1913. Moreover, with the small exception of part of an observatory series, the existence of the observations recovered here has not been previously noted in the literature. This dataset presents only a small number of inhomogeneities and has the potential to eventually provide invaluable information on pre-industrial O_3. For example, the fact that it includes daily nightly and diurnal observations may present an opportunity to apply our understanding of ozone chemistry and surface ozone variability, which is still developing, to earlier time periods (Monks et al., 2021). Plenty of data that could be recovered from other observatories exist in logbooks (Bojkov, 1986; Möller, 2022), and such data and the work published here can contribute to a better understanding of pre-industrial O_3 and be employed in future research.

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