# Analysis of total ozone trends in the Iberian Peninsula using satellite data

M. Antón[1,2], D. Bortoli[2], P.S. Kulkarni[2], M.J. Costa[2], A.F. Domingues[2], D. Loyola[3], A.M. Silva[2], L. Alados-Arboledas[1]

**Abstract** — This work is focused on the study of the total ozone column (TOC) trends over the Iberian Peninsula during the last 30 years (1979-2009). This analysis is carried out using satellite TOC data and it is divided into two sub-periods in order to detect changes in the ozone trend pattern: from 1979 to 1994 using the NASA Total Ozone Mapping Spectrometer (TOMS) and from 1995 to 2009 by means of the ESA Global Ozone Monitoring Experiment (GOME). The analysis of the long-term ozone trends is performed using annual mean time series derived from the average of the deseasonalized monthly TOC series. The results show that the ozone depletion was statically significant at the 95% confidence level during the first sub-period (1979-1994) in the entire region of study, with linear trends from -4.5 %/decade to -2.9 %/decade. These linear trends presented a clear dependence on latitude, being higher for the Northerner locations than for the Southerner. By contrast, the analysis of the second sub-period of study (1995-2009) presented positive ozone trends from +0.6 %/decade to +1.8 %/decade (only statically significant in four of nine locations of study), indicating that the ozone layer may be responding as expected to the controls on ozone-depleting substances imposed by the Montreal Protocol. Additionally, a seasonal trend analysis is performed using the average of the deseasonalized monthly values for each season of the year. The seasonal analysis showed that while the negative ozone trends during the first sub-period of study were statically significant in the springtime and summertime, the positive seasonal trends during the second sub-period did not show any statistically significance.

**Keywords** — GOME, Iberian Peninsula, TOMS, total ozone column, trend

## 1 INTRODUCTION

It is well known that the ozone plays a key role in the photo-chemical equilibrium of the atmosphere. In addition, the ozone layer has an outstanding role in protecting all living organisms from harmful ultraviolet (UV) radiation. Ozone is continually produced, destroyed and circulated in the Earth's atmosphere by a great variety of natural processes [1]. The ozone layer has additionally been clearly affected by human activities. In 1974, chlorofluorocarbons (CFCs, manufactured as refrigerants and aerosol propellants) were identified as the major source of stratospheric chlorine compounds [2]. Several studies showed that this chemical element could destroy ozone in the Earth's stratosphere [3]. This hypothesis was confirmed in 1985 by a measured ozone loss over Antarctica [4], what is now referred to as the ozone hole, and in 1988 by significant winter time decreases over northern mid-latitudes, later confirmed by satellite observations [5], [6]. Several studies using ground-based and satellite measurements have demonstrated that stratospheric ozone levels significantly declined until the mid-1990s in the middle and high-latitude regions of the two hemispheres, see [7]-[10].

As consequence of the Montreal Protocol from 1987 and its amendments [11] the production and release of CFCs and other ozone depleting substances (ODSs) containing chlorine and bromine decreased. This fact prevents a further thinning of the ozone layer and lead to a gradual recovery in the next decades [12]. Newman et al. [13] simulated a future world ("world avoided") where the ODSs were never regulated, and its production grew at an annual rate of 3%. They found by means of this "world avoided" simulation that 17% of the globally averaged column ozone would be destroyed by 2020 and 67% by 2065 in comparison to 1980. Close monitoring of the changes in the ozone layer has become a subject of major concern both by the scientific community and the general public. In this sense, sustainable global ozone observations with space-borne instruments play an essential role in explaining and understanding the global changes of stratospheric ozone and its impact on climate and human health [14].

The NASA Total Ozone Mapping Spectrometer (TOMS) installed on three successive satellites has provided daily images of the global ozone distribution with good spatial resolution from November 1978 until December 2005 [15], [16]. In this work, the TOMS TOC data have been inferred using the last available version (V8) of the TOMS retrieval algorithm [17], [18]. Extensive validation exercises on these TOMS TOC data using ground-based Brewer and Dobson spectrophotometers have

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1. *M. Antón and L. Alados-Arboledas are with Departamento de Fisica Aplicada, Universidad de Granada, Granada, Spain E-mail: manantон@unex.es*
2. *M. Antón, D. Bortoli, P.S. Kulkarni, M.J. Costa, A.F. Domingues and A.M Silva are with the Evora Geophysics Centre, University of Evora, Evora, Portugal.*
3. *D. Loyola is with Remote Sensing Technology Institute (IMF), the German Aerospace Center (DLR), Wessling, Germany*

shown an excellent agreement (at the 1% level) [19]-[22]. The ESA Global Ozone Monitoring Experiment (GOME) has been recording global TOC measurements since July 1995 [23], being currently operative. The current operational algorithm for the retrieval of TOC data using this satellite instrument is the GOME Data Processor Version (GDP) Version 4.x [24]. The accuracy of the total ozone columns retrieved by this GOME retrieval algorithm is, in average, very high (within a few percent) [21], [25]-[27].

The main objective of this work is to quantify the decline and subsequent recovery of the TOC values in the Iberian Peninsula using satellite data recorded during the last 30 years (1979-2009). The long-term analysis is divided into two phases in order to detect changes in the ozone trend pattern: from 1979 to 1994 using TOMS data (period of strong increase in the ODSs concentration in the stratosphere) and from 1995 to 2009 by means of GOME data (slow decrease in the ODSs). Therefore, this paper is expected to contribute to improve the understanding of long-term TOC evolution in the Southwestern Europe. To our knowledge, only a limited number of papers have studied the long-term variability of TOC data in the Iberian Peninsula. For instance, Antón et al. [28] studied the long-term ozone changes over Portugal during the period 1979-1999 showing significant annual TOC trend of (−2.65±0.70)%/decade.

This paper is divided into four sections. The satellite TOC data are presented in Section 2. In Section 3, the methodology used to characterize the long-term TOC variability is explained. The results and its discussion are given in Section 4. The conclusions of the paper are summarized in Section 5.

## 2 TOTAL OZONE DATA

The NASA Goddard Space Flight Center (GSFC) TOMS instrument is a downward-viewing spectrometer which measures Earth-backscattered UV radiances [15], [16]. Six discrete 1-nm wavelength bands between 312 and 380 nm are measured at 35 scan positions and 3-degree intervals to continuously cover the regions between the orbital paths. The spatial resolution is 50 km by 50 km for the nadir view, and about 75 km by 200 km at the extreme cross-track scan positions. The TOMS retrieval algorithm (V.8) uses only two wavelengths (317.5 and 331.2 nm) to derive TOC data, while the other four wavelengths are used for error correction, and identification of aerosols and clouds [17], [18]. In addition, the TOMS algorithm uses the measured albedo at each cross-track position as input, and a modeled look-up-table. In this work we use the TOC data provided in a daily gridded format of 1° latitude by 1.25° longitude (level-3 data). The TOMS instrument was on-board three polar-orbiting sunsynchronous satellites: Nimbus-7 (1978–1993), Meteor-3 (1993–1994), and Earth Probe (EP) (1996–2005). In the present work, we use the TOMS data provided by the first two satellite instruments. Thus, we have a continuous period of TOC observations between January 1979 and November 1994. This defines our first sub-period of study for the analysis of the long-term TOC variability in the Iberian Peninsula.

ESA GOME instrument on board the Second European Remote Sensing Satellite (ERS-2) is a UV-visible-near infrared spectrometer which has 3584 spectral channels in the range from 240 to 793 nm with a spectral resolution of 0.2 to 0.4 nm [29]. Global coverage at the Equator is achieved with GOME within three days. The ground path (960 km) is divided into three ground pixels of 320 km (across orbit) × 40 km (along orbit). The GDP 4.x retrieval algorithm has two main steps to derive the total ozone column: the Differential Optical Absorption Spectroscopy (DOAS) least squares fitting for the ozone slant column, followed by the computation of a suitable Air Mass Factor (AMF) to make the conversion to the vertical column density [24]. GOME/ERS-2 is currently operative, providing TOC data since July 1995. The second sub-period of study used in this work to quantify the long-term ozone changes covers from July 1995 to December 2009.

Fig. 1. Spatial distribution of the nine locations selected to analyze the long-term TOC variability over the Iberian Peninsula.

## 3 STUDY AREA AND METHODOLOGY

The Iberian Peninsula covers around 583 000 square km of area mostly covered by Spain (85%) and Portugal (15%). We select nine locations in order to take into account the spatial TOC variability over the Iberian Peninsula. The spatial distribution of these selected locations is shown in Figure 1. It can be seen that the nine locations used to quantify the long-term ozone changes over the Iberian Peninsula are well distributed over this region. From North to South and from West to East, the nine locations are: Coruña (43.3° N, 8.4° W), Santander (43.5° N, 3.8°

W), Barcelona (41.4° N, 2.2° W), Figueira da Foz (40.1° N, 8.9° W), Madrid (40.4° N, 3.7° W), Valencia (39.5° N, 0.4° W), Faro (37.2° N, 7.9° W), Tarifa (36.0° N., 5.6° W), and Almería (36.8° N, 2.4° W).

In this work we are interested in long-term TOC trends and not day to day variations, therefore, we obtain the monthly TOC data, M(t), averaging the daily values measured by the two satellite instruments.

The fist step in the analysis of the long-term ozone trends is to deseasonalize the monthly TOC series for each individual location. Deseasonalization process of a given series is based on obtaining a good model of the seasonal component, and then subtracting it from the measured (monthly) series. In this work, the annual cycle of TOC data (seasonal pattern) was estimated from the best fit of monthly TOC values by least squares method using the following function [30]:

$$S(t) = a + b \cdot Sin(\omega \cdot t) + c \cdot Cos(\omega \cdot t) \quad (1)$$

where t is the time in months, $\omega = 2\pi/12$, a is the central TOC value, and the term $b \cdot \sin(\omega \cdot t) + c \cdot \cos(\omega \cdot t)$ represents the seasonal component of the TOC variability. The amplitude of this seasonal component was calculated as $\sqrt{b^2 + c^2}$.

Thus, the deseasonalized monthly time series is calculated as:

$$D(t) = M(t) - S(t) \quad (2)$$

The annual mean time series is derived from the average of these deseasonalized monthly values for each year. The long-term ozone trend over each individual location is studied from the linear regression analysis applied on this annual time series. Additionally, we work with four time series for the four seasons of the year. From the climate point of view, Iberian Peninsula has four seasons, winter (December, January, and February), spring (March, April, and May), summer (June, July, and August), and autumn (September, October, and November) [31]. These four seasonal time series are inferred from the average of the deseasonalized values for each season of the year. Thus, the long-term ozone trends are also calculated separately for each season by means of a linear regression analysis applied on each time series.

The entire period (1979-2009) was divided into two sub-periods (1979-1994 and 1995-2009) to analyse and compare the TOC trends over the Iberian Peninsula. TOMS and GOME TOC data are used to obtain the long-term ozone trends for the sub-periods 1979-1994 and 1995-2009, respectively.

## 4 RESULTS AND DISCUSSION

It is well known that the major component of long-term TOC variation at midlatitudes is its annual cycle, which is controlled by a balance between transport associated with the diabatic mean circulation of the stratosphere or Brewer–Dobson circulation and photochemical loss [32]. This component is clearly appreciated in the evolution of the monthly total ozone values over Madrid for the period 1979-2009 (Figure 2, top). A buildup of TOC values during winter and early spring can be seen, when the transport is dominant, while a decline is observed through late spring and summer, when transport decreases and photochemical loss dominates with the increase of solar radiation. To characterize the annual cycle, the monthly TOC series of each location were fitted to a periodic function (equation 1). The amplitude (±error) of this seasonal component for each individual location is shown in Table 1. For the first sub-period of study (1979-1994), the amplitude varies from (41.33±2.38) DU at Barcelona to (29.83±2.03) DU at Almería, indicating that the amplitude of the annual TOC cycle increases with latitude over the Iberian Peninsula. This behaviour is corroborated during the second sub-period (1995-2009) where the amplitude variation is between (32.0±1.7) DU at Barcelona and (24.6±1.7) DU at Tarifa. This clear dependence of seasonal amplitude with respect to latitude is associated with a greater wintertime transport in the higher latitudes due to larger planetary-wave amplitudes which are modulated by the tropical zonal winds [32], [33]. In addition, Table 1 also shows that the TOMS data used in the first sub-period present larger seasonal amplitude for all locations than the GOME data utilized in the second sub-period. This result could be related to the fact that GOME satellite observations do not completely cover the TOC variability over the Iberian Peninsula. Antón et al. [25] showed that while low ozone amounts (240–250 DU) are overestimated by the GOME instrument (1–4%), the higher ozone values (380–420 DU) are underestimated (2–4%), decreasing the amplitude of the seasonal component recorded by this satellite instrument. However, this seasonal behavior of GOME data does not affect to the long-term analysis of the data.

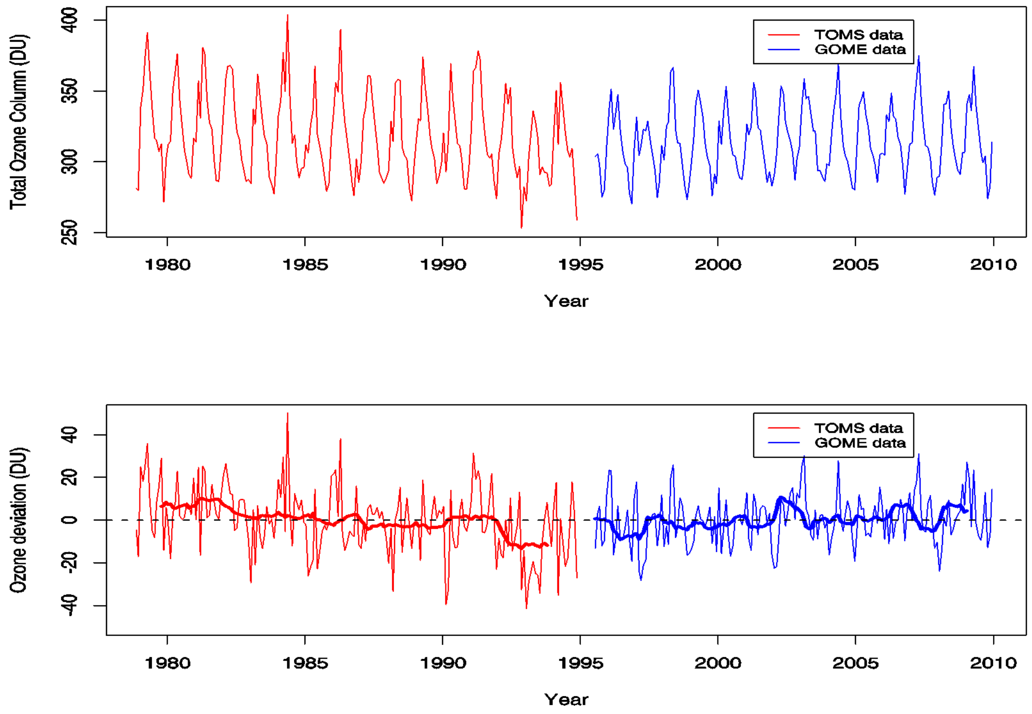

Fig. 2. Top: Time series of the monthly TOC values over Madrid during the period 1979-2009. Bottom: Evolution of the deseasonalized monthly time series over Madrid during the period 1979-2009.

In order to analyze the long-term ozone trend, the major component of natural ozone variations (seasonal component) must be subtracted from the monthly total ozone dataset (equation 2). Figure 2 (bottom) shows the evolution of this deseasonalized monthly time series over Madrid for the period 1979-2009. In addition, following the approach of Hadjinicolaou et al. [34], this time series was smoothed with a 24-month running averages for removing the Quasi-Biennial Oscillation (QBO) effect and in this way makes long-term ozone changes more prominent. A distinct ozone decline can be clearly appreciated for the sub-period 1979-1994 using TOMS data. By contrast, it can be seen a more stable behaviour during the second sub-period 1995-2009 using GOME data, showing even a slight increase in TOC values. Similar evolutions of the deseasonalized monthly series are obtained for the rest of stations at the Iberian Peninsula.

Table 1. Amplitude of the seasonal component (± error) for the nine locations at the Iberian Peninsula.

| | ***Amplitude 1979-1994 (DU)*** | ***Amplitude 1995-2009 (DU)*** |
|---|---|---|
| Coruña | 40.9±2.3 | 32.0±2.0 |
| Santander | 41.3±2.4 | 31.7±1.8 |
| Barcelona | 37.8±2.4 | 32.0±1.7 |
| Figueira | 32.4±2.3 | 29.6±1.7 |
| Madrid | 36.2±2.2 | 28.6±1.7 |
| Valencia | 34.7±2.2 | 29.4±1.8 |
| Faro | 32.4±2.0 | 26.3±1.7 |
| Tarifa | 30.1±2.3 | 24.6±1.7 |
| Almería | 29.8±2.0 | 25.7±1.2 |

The next paragraphs deal with the quantification and explanation of these ozone long-term changes observed in each sub-period of study.

The annual mean time series for each location is derived from the deseasonalized monthly values as was explained in Section 3. Table 2 shows the trend results of these mean annual time series together with their uncertainty (the standard error), expressed in Dobson Units (DU) and in percentage with respect to the averaged TOC value for each location and each sub-period. Those trends statistically significant at the 95% confidence level are marked with asterisk. For the first sub-period (1979-1994), the ozone trends vary from (-4.5±1.1) %/decade (Santander) to (-2.9±1.0) %/decade (Faro), being all them statically significant. These clear negative trends are mainly due to the increase of the atmosphere contamination by ODSs during this sub-period. WMO reports on the state of ozone layer [12], [35], [36] have underlined that this ODSs increase was mostly responsible for the ozone depletion in the extratropics in the 1980s and early 1990s. Additionally, several studies have confirmed that the long-term TOC variability over South Europe is partially controlled by climate variability described mainly by the North Atlantic Oscillation (NAO) [37], [38]. The NAO induces a clear signature on the tropospheric storm tracks and hence, on synoptic-scale TOC fluctuations, which significantly impinge in monthly and seasonal TOC levels. Therefore, the NAO's tendency to remain in its high phase in late eighties and nineties may also account for part of the decreasing trends over the

Iberian Peninsula [39]. Table 2 also shows that the negative ozone trend during this first sub-period present clear latitudinal dependence, being the trends larger for Northerner latitudes over the region of study. This result is in agreement with previously published works on regions located at higher latitudes than the Iberian Peninsula, e.g., [34], [40]-[43], which showed greater ozone trends from 1979 to middle 1990s.

Table 2. Linear trends in DU units per decade and percent per decade (± error) for the nine locations at the Iberian Peninsula. Periods 1979-1994 (TOMS data), and 1995-2009 (GOME data). Trends statistically significant at the 95% confidence level are marked with asterisk

| | ***Trend 1979-1994*** | ***Trend 1995-2009*** |
|---|---|---|
| Coruña | -13.6±3.2 DU/dec*<br>-4.2±1.0 %/dec* | +2.0±3.1 DU/dec<br>+0.6±1.0 %/dec |
| Sant. | -14.7±3.8 DU/dec*<br>-4.5±1.1 %/dec* | +5.8±2.1DU/dec*<br>+1.8±0.7 %/dec* |
| Barce. | -13.1±3.7 DU/dec*<br>-4.1±1.1 %/dec* | +5.5±1.9 DU/dec*<br>+1.7±0.6 %/dec* |
| Figue. | -11.7±3.2 DU/dec*<br>-3.6±1.0 %/dec* | +2.3±2.0 DU/dec<br>+0.7±0.6 %/dec |
| Madrid | -11.8±3.5 DU/dec*<br>-3.0±1.1 %/dec* | +4.6±1.5 DU/dec*<br>+1.5±0.5 %/dec* |
| Valen. | -11.7±3.9 DU/dec*<br>-3.6±1.2 %/dec* | +4.2±2.2 DU/dec<br>+1.3±0.) %/dec |
| Faro | -9.3±3.3)DU/dec*<br>-2.9±1.0 %/dec* | +2.3±1.3 DU/dec<br>+0.7±0.4 %/dec |
| Tarifa | -9.4±3.5 DU/dec*<br>-3.0±1.1 %/dec* | +3.5±1.4 DU/dec*<br>+1.1±0.5 %/dec* |
| Almer. | -10.2±3.8 DU/dec*<br>-3.2±1.2 %/dec* | +3.0±1.8 DU/dec<br>+1.0±0.6 %/dec |

The ozone trends for the second sub-period (1995-2009) are all positive with values between (+0.62±0.97) %/decade (Coruña) and (+1.82±0.67) %/decade (Santander). These trends are only statically significant in four locations (Santander, Barcelona, Madrid and Tarifa). Moreover, it is not appreciated any dependence on latitude. All these results are in accordance with previous studies. Thus, Vyushin et al. [44] showed positive ozone trends but not statistically significant (in monthly mean satellite data for 5° latitudinal zones) for the period 1996-2005. Yang et al. [45] found statistically significant positive ozone trend of about 1%/decade in the period 1996–2007 for the averaged 50°S–50°N satellite data. Krzyscin [46] showed statistically significant positive trends (about 0.5–1%/decade) in the period 1996–2008 for the ozone data averaged over the globe (boreal spring), and in 50°S–50 °N zone (boreal summer and whole year). The ozone recovery is partially explained by the control of the production (and hence their emissions into the atmosphere) of the ODSs as result of the implementation of a series of international agreements—the Montreal Protocol and its amendments— which began in 1989 [35]. Because of the long lifetimes of ODSs, their atmospheric abundances continued to increase in the early 1990s even as their emissions were decreasing. This fact explains that the change of ozone trend is not observed until middle 1990s. Nevertheless, several studies have shown that the recent positive ozone trends in Northern middle latitudes are larger than those expected from the decline in ODSs, e.g, [47],[48]. Thus, in addition to the depletion in ODSs, another reason that could partially explain the slightly positive trend found over the Iberian Peninsula during the second sub-period may be the significant increase in tropospheric ozone in this region since 1996 up to now. This fact has been reported by Kulkarni et al. [49], who quantify an increase of tropospheric ozone almost 5% to 24% over the Iberian Peninsula between the period 1979–1993 and 1996–2005. Therefore, this increase of the tropospheric ozone could add to the stratospheric ozone recovery over the study region, producing the positive TOC trend found in this work during 1995-2009.

The use of more sophisticated trend models that include effects of the solar cycle, the QBO, volcanic eruptions, etc., should result in smaller uncertainties in the ozone trend estimations. Nevertheless, it is well documented that the trends derived from a linear regression analysis give approximately the same results for the major long-term ozone changes [8], [50], [51].

Figure 3 shows the evolution of the annual mean time series from the deseasonalized monthly values for Santander, Madrid and Tarifa. The linear regression trends have also been added to the plot which illustrates the apparent change in trend that has occurred over the Iberian Peninsula. The TOC trends are clearly negative during the sub-period 1979-1994, and slightly positive during 1995-2009 as has been explained above. The strong TOC reduction obtained in 1993 is related to the effects of the Mount Pinatubo eruption in June 199, e.g. [52], [53]. Figure 3 also shows large interannual variability of deseasonalized TOC values, which may make the trend analysis notably sensitive to the length of the data set [54]. Interannual fluctuations of TOC over northern midlatitudes are caused by several factors, such as the regional dynamical structure of the atmosphere (e.g., NAO), variability of the stratospheric circulation patterns related to anomalously fluxes of planetary-wave activity from the troposphere, volcanic eruptions, solar flux and the equatorial QBO [10], [37], [38], [55], [56].

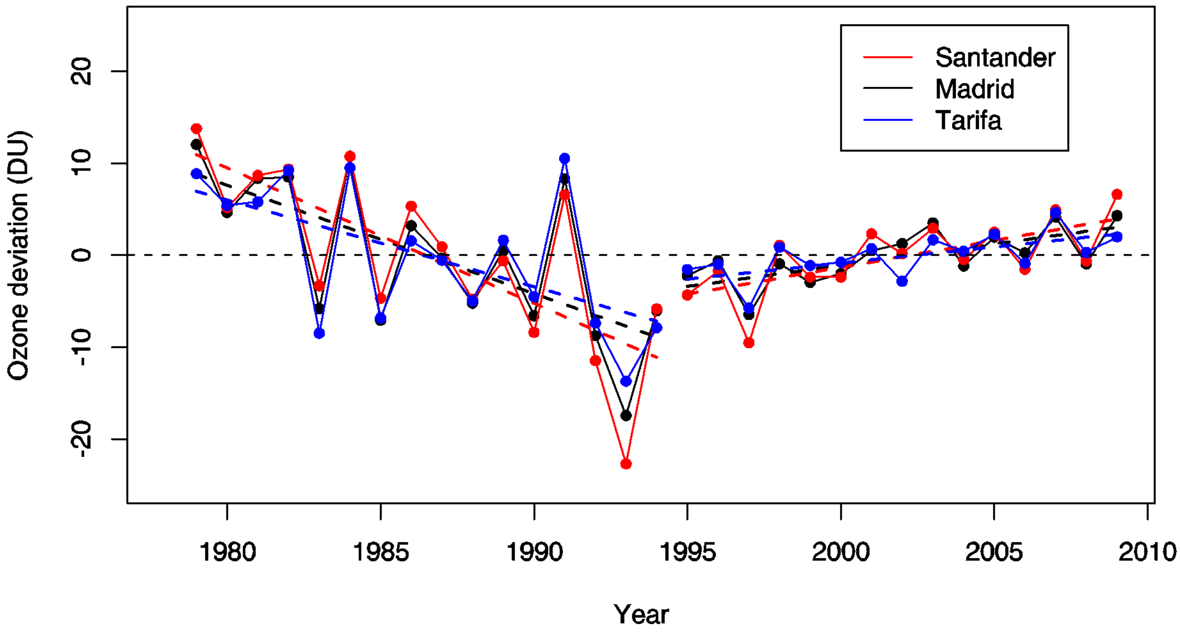

Fig. 3. Evolution of the annual mean time series from the deseasonalized monthly values for Santander, Madrid and Tarifa during the period 1979-2009. The linear regression trends (dotted-lines) for the sub-periods 1979-1994 and 1995-2009 have also been added to the plot.

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Finally, we obtain the long-term ozone trend separately for each season and location from the deseasonalized monthly values. The results obtained for Santander, Madrid and Tarifa are shown in Table 3. It can be seen that the most seasonal ozone trends obtained during the first sub-period are negative in agreement with the strong decline derived from the annual mean time series. However, only spring and summer show statistical significance at the 95% confidence level. The TOC declines during spring show large values between (-4.6±2.2)%/decade for Tarifa and (-8.0±1.6)%/decade for Santander which is consistent with other findings for Northern Midlatitudes [28], [41], [47]. In addition, the significant negative summer trend (~ -3 %/decade) could justify the observed increase of UV radiation level in the midlatitudes of the Northern Hemisphere in summer months during the past decades, see [57], [58] It should be noted that the significant ozone trends derived from spring and summer confirm the latitude dependence (Northerner trends larger than Southerner trends) observed in the annual ozone trends shown above. By contrast, most seasonal ozone trends obtained during the second sub-period are positive but not significant at 95% in accordance with the results inferred from the annual trend analysis. Nevertheless, it can be seen that the spring ozone trends present the largest positive values, opposing to the great negative trends found for this season during the first sub-period.

Table 3. Linear trends in percent per decade (± error) for each season at Santander, Madrid and tarifa. Periods 1979-1994 (TOMS data), and 1995-2009 (GOME data). Trends statistically significant at the 95% confidence level are marked with asterisk.

| | **Trend 1979-1994 (%/decade)** | | |
|---|---|---|---|
| | **Santander** | **Madrid** | **Tarifa** |
| **Winter** | -4.8±2.4 | -3.9±2.2 | -3.3±2.0 |
| **Spring** | -8.0±1.6* | -6.3±1.9* | -4.6±2.2* |
| **Summer** | -3.9±1.1* | -3.0±1.1* | -2.5±1.1* |
| **Autumn** | -1.5±0.8 | -1.4±0.8 | +1.4±0.9 |
| | **Trend 1995-2009 (%/decade)** | | |
| | **Santander** | **Madrid** | **Tarifa** |
| **Winter** | +1.4±2.8 | -0.1±2.9 | +0.9±2.5 |
| **Spring** | +3.1±2.0 | +3.2±1.8 | +1.9±1.9 |
| **Summer** | +0.3±1.0 | +0.1±0.9 | -0.3±0.8 |
| **Autumn** | +0.9±1.2 | +1.9±0.9* | +1.7±0.8* |

## 5 CONCLUSIONS

Some important conclusions may be drawn from the analysis of the ozone trends using satellite data over the Iberian Peninsula for two sub-periods (1979-1994 and 1995-2009). The ozone depletion was statistically significant during the first sub-period in this entire region, with linear trends from -4.5 %/decade to -2.9 %/decade. These linear trends presented a clear dependence on latitude, being higher for the Northerner locations than for the Southerner. In addition, the ozone depletion had a distinct seasonal signature, with statistically significant rates observed only during springtime and summertime. By contrast, the analysis of the second sub-period of study (1995-2009) shows positive ozone trends from +0.6 %/decade to +1.8 %/decade, indicating a slow ozone recovery over the Iberian Peninsula. However, it can be noted that these positive trends were statistically significant only in four of nine selected locations. The seasonal analysis of this second sub-period showed that the ozone recovery has been more rapid in the springtime but without statistically significance.

Overall we would like to emphasize the remarkable success of the Montreal Protocol for controlling production of ozone depleting substances. Nevertheless, even though the abundance of these substances has significantly diminished in the atmosphere, other anthropogenic effects could complicate the ozone recovery process and may result in considerably altered ozone levels in the future. Therefore, the sustainable measurements of accurate total ozone data with ground-based and satellite instruments are essential to monitor the recovery of ozone layer in the next decades.

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