



isoAOT40: An improved ozone exposure index based on the Annual Ozone Spectrum Profile (AO3SP)

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Abstract In recognition of the rising threats of ground-level ozone (O₃) pollution to forests, agricultural crops, and other types of vegetation, accurate and realistic risk assessment is urgently needed. The accumulated O₃ exposure over a concentration threshold of 40 nmol mol⁻¹ (AOT40) is the most commonly used metric to investigate O₃ exposure and its effects on vegetation and to conduct vegetation risk assessment. It is also used by international regulatory authorities for deriving critical levels and setting standards to protect vegetation against surface O₃. However, fixed periods of the growing season are used universally, yet growing seasons vary with latitudes and elevations, and the periods

of plant lifespan also differ among annual species. Here, we propose the concept of the Annual O₃ Spectrum Profile (AO₃SP) and apply it to calculate the profile of AOT40 throughout the year (AAOT40SP, Annual AOT40 Spectrum Profile) using the International Organization for Standardization (ISO) weeks as a shorter window ISO-based accumulated exposure. Using moving time periods of three (for crops) or six (for forests) months, the *iso*AOT40 behavior throughout the year can be examined as a diagnostic tool for O₃ risks in the short- or long-term during the lifecycle of local vegetation. From this analysis, AOT40 (*iso*AOT40) that is most representative for the local conditions and specific situations can be identified, depending on the exact growing season and lifecycle of the target vegetation. We applied this novel approach to data from five background monitoring stations located at different elevations in Cyprus. Our results show that the AAOT40SP approach can be used for improved and more realistic assessment of O₃ risks to vegetation. The AO₃SP approach can also be applied using metrics other than AOT40 (exposure- or flux-based), adding a new dimension to the way O₃ risk to vegetation is assessed.

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Introduction

Tropospheric ozone (O_3) concentrations have shown a widespread multi-fold increase in the northern hemisphere relative to their pre-industrial levels, and are continuously rising in several regions of the world (Diaz et al. 2020; Yin et al. 2020; Sicard 2021; Singh and Kavouras 2022). Tropospheric O_3 is a secondary air pollutant whose formation depends upon primary gases, e.g., volatile organic compounds (VOCs), and nitrogen oxides (NO_x), and meteorological conditions such as air temperature, relative humidity, and global radiation (Deroubaix et al. 2021; Cao et al. 2022; Cordero et al. 2022; Nguyen et al. 2022; Wang et al. 2022a, b; Ding et al. 2023). Hence, O_3 concentrations exhibit a considerable spatiotemporal variability, with peaks in O_3 exposures widely varying across space and time (Deroubaix et al. 2021; Cao et al. 2022; Cordero et al. 2022; Nguyen et al. 2022; Wang et al. 2022a, b; Ding et al. 2023). Since the 1990s, background O_3 concentrations have decreased in rural areas but increased in many urban areas in Europe and North America, mainly due to emission control policies that decreased local NO_x emissions; the increase in urban areas is due to local reduction of O_3 titration by NO (Diaz et al. 2020; Proietti et al. 2021; Sicard 2021). However, even if the O_3 mean concentrations have decreased in several rural areas, O_3 exposures remain as high as to be multi-fold (e.g., 2–8 times) the critical levels adopted by worldwide regulatory authorities for the protection of vegetation (Diaz et al. 2020; Proietti et al. 2021; Sicard 2021). Moreover, in contrast to the mean concentrations, low percentile and background concentrations increased, even in rural and remote stations, mainly due to climate change (Sicard 2021). Therefore, it is imperative to use accurate and improved O_3 metrics for continuous comprehensive O_3 risk assessments from local to regional scales.

The accumulated O_3 exposure over a concentration threshold of 40 nmol mol⁻¹ (AOT40) is the most widely used metric in the literature to evaluate O_3 exposure (Anav et al. 2016; Agathokleous et al. 2018; Lefohn et al. 2018; Mills et al. 2018; Blanco-Ward et al. 2021; Ascenso et al. 2021). For example, a search in the Web of Science Core Collection with the keyword “AOT40” produced 345 results (search on 12 July 2022). Owing to its easy and fast-forward calculation, as it requires only hourly O_3 concentration data, AOT40 is widely used, not only in scientific programs but also in worldwide regulatory standards for the protection of vegetation (Paoletti and Manning 2007). For the protection of agricultural crops, a critical level of 3,000 nmol mol⁻¹ h has been adopted by European Union (EU) legislative bodies (2008/50/CE Directive), whereas the critical level of 5,000 nmol mol⁻¹ h over the growing season is recommended for the protection of forests (UNECE 2017). Flux-based O_3 metrics are more biologically sound compared to

exposure-based metrics as they integrate vegetation characteristics and physiology (e.g., phenology, stomatal conductance; how much O_3 plants absorb), as well as other important environmental factors that modify the amount of O_3 entering plant tissues, such as soil water content (Matyssek et al. 2007; Paoletti and Manning 2007; B  ker et al. 2015; Anav et al. 2016; Agathokleous et al. 2018; Blanco-Ward et al. 2021; Paoletti et al. 2022). Although there is increasing interest in the use of flux-based metrics (Proietti et al. 2021 and 2022; Blanco-Ward et al. 2021; Shashikumar et al. 2022), their use is restrictive in underdeveloped and developing countries as well as in remote areas where physiological and environmental data needed are not readily available. In fact, a search in the Web of Science Core Collection with the keyword “PODy”, the metric proposed for O_3 fluxes, revealed only 44 results (search on 12 July 2022). Moreover, due to the complexity in its calculation and the many input data required, its adoption by worldwide regulatory agencies is also challenging. Therefore, the use of AOT40, and other exposure-based metrics, may prevail for some time.

Growing seasons shift across latitudes and elevations, and plant lifespans differ among annual species. However, fixed periods of the growing season are used for AOT40, for example from 1 April to 30 September for the protection of forest trees (AOT40f), and from 1 May to 31 July for agricultural crops (AOT40c) for latitudes around 45  N (2008/50/EC Directive; UNECE 2017). Hence, AOT40-based risk assessment, based on the proposed fixed integration periods currently adopted by worldwide regulatory guidelines, cannot fully depict the risk for local vegetation. For this reason, new developments are needed to improve the efficiency of such metrics and decrease the uncertainty in risk estimation, especially across larger geographical areas. Here, we propose $_{iso}$ AOT40, a modification of the classic AOT40, which can identify more realistic local O_3 risks to forests and other types of vegetation based on the ISO week system.

Methods

In this study, we used O_3 monitoring data from Cyprus, the third largest Mediterranean island with a land area of 9,250 km², as a case study. The Republic of Cyprus had a population of 918,100 in October 2021 (Ministry of Finance 2022). Mount Olympus (1,951 m a.s.l.), in the Troodos Mountains, is the highest elevation on the island. According to the K  ppen classification, the climate is Mediterranean, classified as both “hot semi-arid climate” (BSh) and “hot-summer Mediterranean climate” (Csa). Cyprus suffers O_3 episodes regularly (Kleanthous et al. 2014). Hourly O_3 data from five background stations were obtained from Cyprus’s Air Quality Section of the Department of Labor Inspection. The stations were scattered across the island

and subjected to different meteorological influences. Two were inland regional background stations at high (Troodos, 34.56 N–32.51 E, 1819 m a.s.l.) and middle (Agia Marina Xyliatou, 35.02 N–33.03 E, 532 m a.s.l.) elevations, which are representative stations in the European Monitoring and Evaluation Program (EMEP). Another was a rural station (Stavrovouni, 34.53 N–33.26 E, 650 m a.s.l.), while the remaining two were rural-marine stations, Ineia (34.57 N–32.22 E, 672 m a.s.l.) and Cavo Greco (34.57 N–34.04 E, 23 m a.s.l.). Data were available for the three-year period 2014–2016, except for Stavronouni, whose operation ceased in January 2016, and thus, for this station data for 2014–2015 were used. The wide horizontal and vertical (elevation) distribution of the stations permits the evaluation of how O₃ risk estimation may be modified by the O₃ exposure metric.

AOT40 was calculated by summing the hourly excesses of O₃ above 40 nmol mol⁻¹ during daylight hours using Eq. 1.

$$AOT40 = \sum_{i=1}^n ([O_3] - 40).dt, \text{ for } [O_3] > 40 \text{ nmol mol}^{-1} \quad (1)$$

where [O₃] is the hourly O₃ mixing ratio (nmol mol⁻¹), n is the number of hours in the calculation period, and dt is the time step (1 h).

The Annual O₃ Spectrum Profile (AO₃SP), the weekly O₃ exposures throughout a year, was then calculated as a thorough tool for risk assessment of local vegetation (Fig. 1). AAOT40SP, the _{iso}AOT40-based AO₃SP, is created by plotting the weekly AOT40 values (y axis) along the ISO week numbers (x axis). The ISO week date system is part of the ISO-8601 standard of the International Organization for Standardization. An ISO year has 52 or 53 ISO weeks, and the ISO week can be obtained from the common date ISOWEekNUM function syntax in MS Excel (versions 2013 and newer). The weeks begin on Monday, and the first week is the first week in a year that includes a Thursday. Therefore, the ISO week can be easily and rapidly obtained by MS Excel users.

For each station, and for all stations together, we created the annual profile of AAOT40SPc spectrum over 13-weeks (~3 months) for crops and AAOT40SPf spectrum over 24 weeks (~6 months) for forests, moving by weekly steps. Using the moving 3- or 6-month time periods, the behavior of _{iso}AOT40 throughout the year can be examined as a diagnostic tool for O₃ risks in the short- and long-term during the lifecycle of local vegetation. We identified the minimum

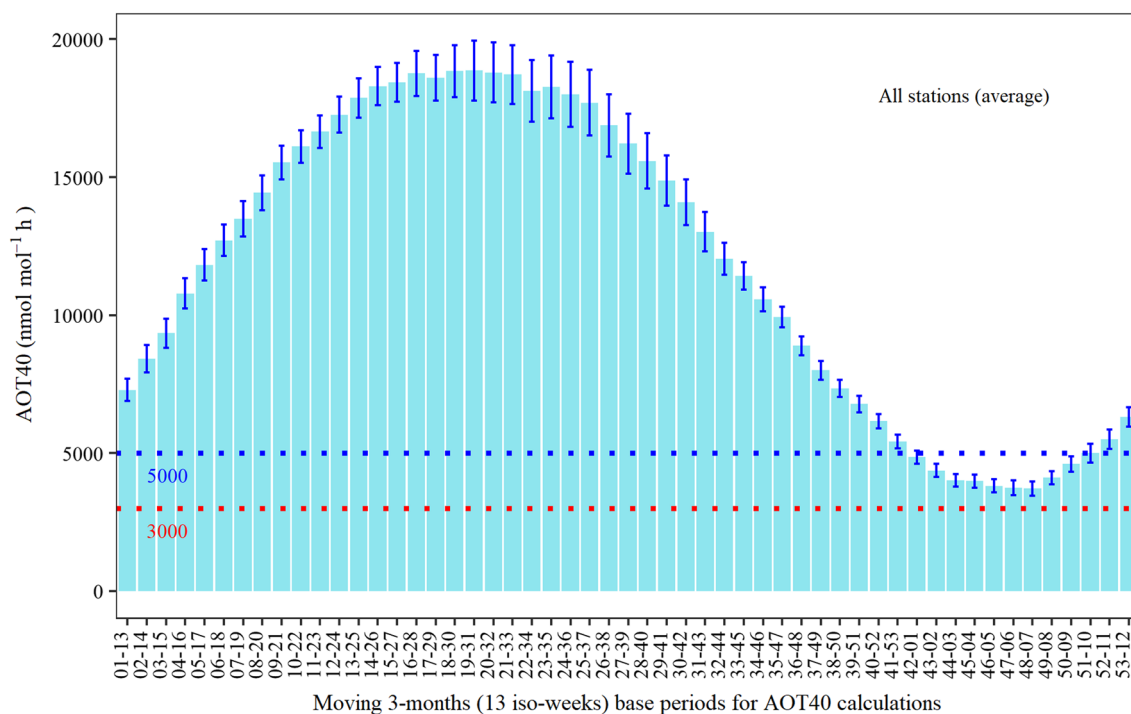


Fig. 1 Annual _{iso}AOT40 Spectrum Profile (AAOT40SP) over 3-months weekly (\pm standard deviation) moving periods. Calculations are based on average data from five monitoring stations across Cyprus over the period 2014–2016. The dashed red line indicates the recommended AOT40c thresholds for the protec-

tion of crops (3,000 nmol mol⁻¹ h over three months), whereas the dashed blue line indicates the recommended threshold for forests (5,000 nmol mol⁻¹ h over six months) (2008/50/EC Directive; UNECE 2017)

and maximum values of $_{\text{iso}}\text{AOT40c}$ and $_{\text{iso}}\text{AOT40f}$ annual profiles and the corresponding ISO weeks. For the estimation of $_{\text{iso}}\text{AOT40}$, the average of three years was used, except for the Stavrovouni station that was based on two years. All data processing was done with MS Excel (Microsoft). The figures were produced with ggplot2 package of R language (Wickham 2016).

Results and discussion

This novel approach captured the temporal variability in AOT40 exposures well, and thus O_3 risks (Fig. 1).

The results of the station-specific analyses showed that $_{\text{iso}}\text{AOT40c}$ and $_{\text{iso}}\text{AOT40f}$ have exceeded the critical levels in all the stations all year round, with the exception of Stavrovouni station toward the end of the year (Fig. 2). Importantly, $_{\text{iso}}\text{AOT40}$ exposures were two to six times higher than the regulatory critical level for almost the entire year (Fig. 1). The AO_3SP analyses revealed that persistent high O_3 exposures are continuously threatening both annual and perennial species of the local flora throughout the year. In addition, the results show distinct AO_3SP s among the stations (Fig. 2). Temporal differences in $_{\text{iso}}\text{AOT40}$ peaks were observed across the different elevations (peaking later at

higher altitudes; Table 1). Air quality and particularly O_3 pollution often worsens at higher elevations due to higher stratospheric intrusion and weaker NO titration (Musselman et al. 1998; Musselman and Korfmacher 2014; Semple and Moore 2020). Furthermore, O_3 levels on Mt Troodos might be influenced more by forest fires occurring during the warmer seasons (Cristofanelli et al. 2007). As an example, the $_{\text{iso}}\text{AOT40}$ exposures peaked earlier in the year and showed less variability throughout the most part of the year at the low-elevation station compared to the high-elevation station (Fig. 2).

These results are fundamentally important because they show that using the traditional AOT40 with the specified time windows would not capture actual risks for many plants, especially in warm climates. For instance, in Cyprus, favorable weather conditions permit the cultivation of many crops especially vegetables throughout the year, whereas the peak of the warm season with heat waves may be avoided due to the harsh conditions for plants occurring concurrently with prolonged drought. For example, in July 2016 the total rainfall was 0–0.5 mm and the highest maximum temperature 29 °C–39 °C in areas where the O_3 monitoring stations operated (Meteorological Report for July 2015, Cyprus Department of Meteorology, <https://www.dom.org.cy/>). For this reason, vegetable

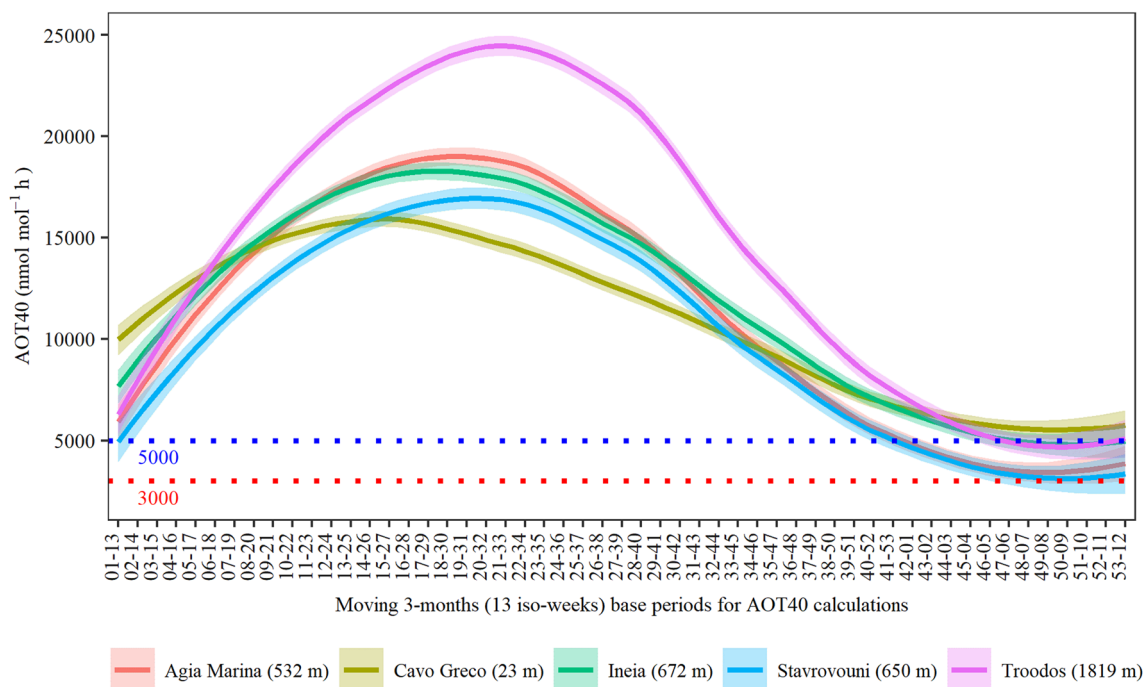


Fig. 2 Annual $_{\text{iso}}\text{AOT40}$ Spectrum Profile (AAOT40SP) over 3-months weekly moving periods (\pm confidence interval) in Cyprus over the period 2014–2016. A loess smoothing method has been applied for clarity. Calculations are based on data per monitoring station. The dashed red line indicates the recommended AOT40c

thresholds for the protection of crops (3,000 $\text{nmol mol}^{-1} \text{h}$ over three months), whereas the dashed blue line indicates the recommended threshold for forests (5,000 $\text{nmol mol}^{-1} \text{h}$ over six months) (2008/50/EC Directive; UNECE 2017)

Table 1 Minimum and maximum values of $_{\text{iso}}\text{AOT40c}$ and $_{\text{iso}}\text{AOT40f}$ annual profiles and the corresponding iso-weeks of their appearance (wk) in parentheses, in Cyprus (2014–2016)

Station	Elevation (m)	max $_{\text{iso}}\text{AOT40c}$ (nmol mol ⁻¹ h)	min AOT40c (nmol mol ⁻¹ h)	max AOT40f (nmol mol ⁻¹ h)	min AOT40f (nmol mol ⁻¹ h)
Cavo Greco	23	16,815 (wk 14–26)	4,937 (wk 44–04)	27,995 (wk 12–35)	13,089 (wk 36–07)
Agia Marina	532	19,286 (wk 19–31)	3,193 (wk 46–06)	32,905 (wk 12–35)	9,392 (wk 40–11)
Stavrovouni	650	17,081 (wk 18–30)	2,897 (wk 47–07)	29,200 (wk 14–37)	8,658 (wk 39–10)
Ineia	672	18,833 (wk 16–28)	4,492 (wk 44–04)	31,987 (wk 13–36)	12,358 (wk 40–11)
Troodos	1,819	25,040 (wk 24–36)	4,800 (wk 48–08)	41,998 (wk 14–37)	12,088 (wk 40–11)
Average \pm SD		19,411 \pm 3,324	4,064 \pm 950	32,817 \pm 5,507	11,117 \pm 1,962

The average of the five stations is considered the average at the national level. AOT40f: 6-month time window for the protection of forest trees. AOT40c: 3-month time window for the protection of agricultural crops. Min: minimum value along the moving 3-month base periods. Max: maximum value along the moving 3-month base periods. As a point of reference, the 1st of May is ISO week 18, 18, and 17 in 2014, 2015, and 2016, respectively, and the 31st of July is ISO week 31, 31, and 30 in 2014, 2015, and 2016, respectively (AOT40c time window). The 1st of April is ISO week 14, 14, and 13 in 2014, 2015, and 2016, respectively, whereas the 30th of September is ISO week 40, 40, and 39 in 2014, 2015, and 2016, respectively (AOT40f time window). S.D.: standard deviation

planting is often regulated to avoid harsh conditions in critical months of July–August. Many vegetables and other cultivated plants, e.g., beans, cantaloupe, carrot, lettuce, pea, potato, radish, and spinach, are widely sown in early January to February, and their cultivation ends before July. Therefore, including July in the calculation of AOT40 would not provide accurate O₃ risk estimates for several crops. Moreover, because of the weather conditions in Cyprus, many of the plants are cultivated again in the fall, sown mainly in July–August or September. Hence, the traditional AOT40 would be irrelevant yet the AAOT40SPc spectrum analysis reveals O₃ exposures exceeding the critical levels, which can be captured by the $_{\text{iso}}\text{AOT40c}$. Table grapes are also cultivated on lower mountain slopes and along the coastline (Markou and Stavri 2006), where the $_{\text{iso}}\text{AOT40}$ exposures peak earlier in the year compared to higher elevations. Given that the maturity of grapes varies by area, Cypriot grape vines supply table grapes to Europe from June to September (Markou and Stavri 2006). Therefore, these cultivations, which are economically and socially important at a national level (Markou 1998), can be exposed to $_{\text{iso}}\text{AOT40}$ peaks that occur earlier than the traditional accumulation period according to the E.U. Directive. Moreover, aromatic herbs and vegetables are often cultivated off-season (Markou and Stavri 2006), such as by seeding on fields much earlier in winter under plastic films. The O₃ risks of such cultivations outside the typical growing season would be misrepresented by the traditional AOT40.

Finally, the growing season of trees is prolonged in Cyprus, with many species being physiologically active almost the entire year; thus, the traditional AOT40 would miss a considerable O₃ risk. Hence, consideration of plant phenology during the specific growing season is important for more accurate AOT40-based risk assessment, and the

current framework may be combined with other traditional methods for more integrated risk assessment.

The ISO weeks of the maximum $_{\text{iso}}\text{AOT40c}$ mismatched the ISO weeks of the traditional AOT40c (17–30 or 18–31, depending on year). Specifically, the mismatch was up to two weeks for the three middle elevation stations i.e., Agia Marina, Stavrovouni, and Ineia (532–672 m a.s.l.), shifting however, 3–4 weeks earlier for the lowest elevation station of Cavo Greco (23 m a.s.l.) and 6–7 weeks later for the highest elevation station of Troodos (1,819 m a.s.l.) (Fig. 2; Table 1). These results indicate that the traditional AOT40c may not reflect the actual O₃ risks, which can be underestimated at the lowest and highest elevation stations due to inaccurate delimitation of the accumulation period (May–July), while some crops are grown year round on the island and are exposed to higher AOT40 than those of May–July. However, the ISO weeks of the maximum $_{\text{iso}}\text{AOT40f}$ had lower deviation from the ISO weeks of the traditional AOT40c (Table 1). Therefore, the 6-month time window for forests may be less sensitive than the 3-month time window for crops because it captures much of the period that is most conducive to increased O₃ concentrations. Nevertheless, the AO₃SPs indicated that AOT40f is still insufficient to identify considerable risks for vegetation grown outside its specific time window and/or for a longer time. The benefit of this analysis is higher for the Mediterranean area, with warmer conditions and a prolonged growing season, relative to other areas of Europe.

Conclusion

The proposed AAOT40SP can be used for more realistic assessment of vegetation risks to O₃, and the same AO₃SP approach can be applied for O₃ metrics other than AOT40.

Thus, the AAOT40SP is proposed as an easy-to-use and flexible approach, adaptive to different geographical areas (in latitudes and altitudes) and to any plant species (in life span and cultivation periods). Different geographical areas and different plant species can vary considerably in their AAOT40SP, and new comprehensive studies are needed to reveal how the AAOT40SP varies spatially and across plant taxonomic or functional groups.

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