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Review Article

A Review Study on Ozone Phytotoxicity Metrics for Setting Critical Levels in Asia

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ABSTRACT

Ground-level ozone (O_3) can be a menace for vegetation, especially in Asia where O_3 levels have been dramatically increased over the past decades. To ensure food security and maintain forest ecosystem services, such as nutrient cycling, carbon sequestration and functional diversity of soil biota, in the over-populated Asia, environmental standards are needed. To set proper standards, dose-response relationships should be established from which critical levels are derived. The predictor of the response in the dose-response relationship is an O_3 metric that indicates the dose level to which the plant has been exposed. This study aimed to review the relevant scientific literature and summarize the O_3 metrics used worldwide to provide insights for Asia. A variety of O_3 metrics have been used, for which we discuss their strengths and weaknesses. The most widely used metrics are based only on O_3 levels. Such metrics have been adopted by several regulatory agencies in the global. However, they are biologically irrelevant because they ignore the plant physiological capacity. Adopting AOT40 (O_3 mixing ratios Accumulated Over the Threshold of 40 nmol mol^{-1}) as the default index for setting critical levels in Asia would be a poor policy with severe consequences at national and Pan-Asian level. Asian studies should focus on flux-based O_3 metrics to provide relevant bases for developing proper standards. However, given the technical requirements in calculating flux-based O_3 metrics, which can be an important limitation in developing countries, no-threshold cumulative exposure indices like AOT0 should always accompany flux-based indices.

Key words: AFst6, Air pollution, AOT40, Critical levels, Dose-response, DO$_3$SE, Flux, Metrics, MPOC, Ozone, PODY, SUM06, Threshold, Uptake, W126

1. INTRODUCTION

Ozone (O_3) levels at the lower troposphere throughout the Northern Hemisphere have been dramatically increased compared to the pre-industrial ones (Sicard *et al*., 2016; Saitanis *et al*., 2015; Kalabokas *et al*., 2013; Vingarzan, 2004; Akimoto, 2003). Notably, Asia, including China and India, is currently a “hot spot” of air pollution, with O_3 levels in some regions exceeding by far the critical levels (CL) of the standards set for protecting vegetation (Kitao *et al*., 2016; Feng *et al*., 2015; Kim *et al*., 2015; Komatsu *et al*., 2015; Verstraeten *et al*., 2015; Oksanen *et al*., 2013; Takigawa *et al*., 2009). Such increases in O_3 levels may detrimentally affect plants and thereby reduce yields, and alter their quality, of major crop plants used for the feeding needs of humanity and other animals (Osborne *et al*., 2016; Tian *et al*., 2016; Broberg *et al*., 2015; Feng *et al*., 2015, 2008; McGrath *et al*., 2015; Wilkinson *et al*., 2012; Morgan *et al*., 2003). It may thus be a challenge to adequately feed the increasing population of Asia under the scenario of O_3-induced losses in yields (Lu *et al*., 2015). O_3-induced losses in yields in Asia (Wang and Mauzerall, 2004) and globally (Avnery *et al*., 2011) associate with an economic cost of billions of US$. Furthermore, chronic exposure of natural vegetation to such potentially phytotoxic O_3 levels may pose a threat for the sustainability of plant communities and ecosystems (Agathokleous *et al*., 2016, 2015; Chappelka and Grulke, 2016; Koike *et al*., 2013; Lindroth, 2010; Cape, 2008; Ashmore, 2005; Matyssek and Innes, 1999).

The most critical information when studying O_3 effects on plants is the O_3 phytotoxicity metric utilized (Matyssek *et al*., 2007; Paoletti and Manning, 2007; Musselman *et al*., 2006). The O_3 metric is considered a versatile tool that can be utilized for: a) providing information about the exposure of plants to O_3; b) establishing dose-response relationships, and thus serving as a predictor of plant response along the full dose-response continuum; c) conducting risk assessment and setting CL for protecting vegetation against adverse effects caused by O_3; and d) communicating meaningful information to policy and decision makers for setting environmental standards. O_3 metrics should be thus carefully selected and used in a united manner so as to contribute in adopting appropriate and effective standards at national and Pan-Asian level.

In this study we aim to provide the bases for future research on the assessment of O_3 impacts on Asian vegetation. In wide regions of Asia, summertime precipitation may be high, a phenomenon which relates to Asian summer monsoons (ASM) (Shi *et al*., 2017; Ha *et al*., 2012; Chang, 2004). Despite ASM may contribute to decreasing O_3 mixing ratios, such a potential decrease is of a small order, and thus O_3 will remain at potentially phytotoxic levels (Surendran *et al*., 2016). Synchronous summertime precipitation and high O_3 mixing ratios may pose a risk for O_3 phytotoxicity in Asian vegetation due to a greater stomatal O_3 uptake. We thus review the relevant scientific peer-reviewed literature and summarize the current global knowledge on the O_3 metrics to provide insights for Asia.

2. O_3 PHYTOTOXICITY METRICS

In the present paper, mixing ratios with a unit of nmol mol^{-1} were used to express the level of O_3 in the air. Although the term of "concentration" is often used in many papers, it is inaccurately defined as in most cases. The definition of "concentration" is mass per volume. On the other hand, O_3 monitoring systems measure in mixing ratios defined as the abundance (number of O_3 moles) of O_3 relative to air (per mole of air).

A series of metrics have been proposed and used for assessing the potential O_3 injury to vegetation, each of which has its own strengths and weaknesses (Table 1). These metrics are mainly used for setting CL for protecting vegetation based on the O_3 exposure (CLe_e) or the accumulated stomatal O_3 flux (CLe_f). The historical foundations of the basic metrics can be found in earlier articles (Musselman *et al*., 2006; Pleijel *et al*., 2004; Wang and Mauzerall, 2004; Danielsson *et al*., 2003; Grünhage *et al*., 1999; Fuhrer *et al*., 1997). Yet, explanations about specialized definitions which have been cited in this article can be found in Musselman *et al*. (2006).

In Asia, parameterization of models of stomatal conductance to water vapor (g_s), which can be used for CLe_f approaches, development of dose-response relationships, and derivation of CLe_e or CLe_f have been mainly conducted in China (Shang *et al*., 2017; Yuan *et al*., 2017; Zhang *et al*., 2017; Hu *et al*., 2015; Feng *et al*., 2012; Oue *et al*., 2011, 2009, 2008; Wang and Mauzerall, 2004) and Japan (Kinose *et al*., 2017, 2014; Kitao *et al*., 2016, 2014; Watanabe *et al*., 2016, 2012, 2011, 2010; Hoshika *et al*., 2015a, b, 2013b, 2012a; Azuchi *et al*., 2014; Yamaguchi *et al*., 2014; Watanabe and Yamaguchi, 2011) during the last decade. Amongst these studies, just few deal with the establishment of dose-response relationships and derivation of CL (Kinose *et al*., 2017; Shang *et al*., 2017; Yuan *et al*., 2017; Zhang *et al*., 2017; Hu *et al*., 2015; Yamaguchi *et al*., 2014; Feng *et al*., 2012; Watanabe *et al*., 2012, 2011, 2010; Wang and Mauzerall, 2004).

3. CLe_e

Exposure-based metrics were the first O_3 metrics used by the scientific community of air pollution. This category of metrics takes into account only the O_3 levels.

3.1 AOT40

The first and most widely accepted and used metric is the AOT*X* (O_3 mixing ratios Accumulated Over the Threshold of X nmol mol^{-1}), set for long-term O_3 exposures. AOT*X* is practically the sum of exceedances of daytime (solar radiation >50 W m^{-2}) hourly O_3 mixing ratios above a threshold of X nmol mol^{-1}, for a given exposure period (Grünhage *et al*., 1999; Fuhrer *et al*., 1997; Kärenlampi and Skärby, 1996). The X threshold is commonly set at 40 nmol mol^{-1} (AOT40), and the AOT40 is thus calculated according to the formula:

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

where: i = the running index and n = the number of hours with $[O_3] > 40$ nmol mol^{-1}. The units of AOT40 are nmol mol^{-1} h (classically ppb h). The historical foundations of AOT40 have been previously explained (Fuhrer *et al*., 1997). AOT40 CL have been adopted by the United States Environmental Protection Agency (USEPA), the United Nations Economic Commission for Europe (UN/ECE), and the World Meteorological Organization (WMO) (World Health Organization (WHO), 2000). The critical AOT40 level (= 5 or 10% yield reduction) for agricultural crops and semi-natural vegetation has been set at 3000 nmol mol^{-1} h. This level should not be exceeded during the running 3-month growing season of plants (e.g. May-July for central Europe), for protection against long-term ambient O_3 adverse effects (Kärenlampi and Skärby, 1996). For forests, the UN-ECE set the CL at 10000 nmol mol^{-1} h

Table 1. Strengths, weaknesses and caution points of the main metrics used for setting critical levels for protecting vegetation based on the O_3 exposure (CLe_e) or the accumulated stomatal O_3 flux (CLe_f).

	Strengths	Weaknesses	Cautions
		CLe_e	
AOT40	1) Easy to calculate, only O_3 levels data are needed (Fuhrer *et al*., 1997; Grünhage *et al*., 1999; Kärenlampi and Skärby, 1996). 2) It has been widely reported in the literature 3) Adopted by several regulatory agencies (without meaning that this is the appropriate policy)	1) O_3 levels are not weighed based on the plant ontogenic stage 2) Environmental constrains to O_3 uptake into leaf tissue are ignored; it is thus biologically unrealistic (Danielsson *et al*., 2003; Gerosa *et al*., 2005; Grünhage and Jäger, 2003; Harmens *et al*., 2007; Karlsson *et al*., 2007b; Matyssek *et al*., 2004; Paoletti and Manning, 2007; Sanz *et al*., 2016) 3) Ineffective in regions with low O_3 pollution 4) Highly overestimated O_3 effects in regions with dry climates (Gerosa *et al*., 2005)	1) O_3 levels should be those at the top of the plant canopy (Grünhage *et al*., 1999; Hicks *et al*., 1987) 2) Deposition models may be used to estimate the O_3 levels at the top of the plant canopy from the O_3 levels at a different height (Emberson *et al*., 2000a, 2000b; Simpson *et al*., 2012) 3) Improvements may be applied for toxicological effectiveness (Grünhage *et al*., 1999; Musselman *et al*., 2006) 4) Higher values may occur along coastlines (Anav *et al*., 2016) 5) Care should be exercised factors which may cause stomatal limitation, such as soil unavailability and acidification, to be controlled in experiments (Azuchi *et al*., 2014; Gerosa *et al*., 2005)
SUM06	1) Easy to calculate, only O_3 levels data are needed	Same as 1, 2, 3 and 4 in AOT40	Same as 1, 2 and 5 in AOT40
W126	1) Easy to calculate, only O_3 levels data are needed (Lefohn *et al*., 1988; Lefohn and Runeckles, 1987; Musselman *et al*., 2006; Wang and Mauzerall, 2004). 2) More realistic than AOT40 and SUM06 3) More effective than AOT40 and SUM06 in regions with low pollution 4) It has been adopted by some regulatory agencies	Same as 2 in AOT40	Same as 1, 2 and 5 in AOT40
		CLe_f	
DO_3SE	1) It takes into account environmental factors and vegetation properties (Anav *et al*., 2016; De Marco *et al*., 2016) 2) Species-specific responses to O_3 in multispecies cultures are meaningful and realistic, in contrast to CLe_e approaches, because the O_3 uptake into the leaf tissue depends on species-specific physiology (Calvete-Sogo *et al*., 2017)	1) More complex calculation than CLe_e approaches (Emberson *et al*., 2000a) 2) Several factors should be monitored and more data should be input in the model than CLe_e approaches (Anav *et al*., 2016; De Marco *et al*., 2016; Emberson *et al*., 2000a) 3) Efficiency depends on environmental conditions which affect plant physiology (Hu *et al*., 2015)	

Table 1. Continued.

	Strengths	Weaknesses	Cautions
AFst*Y*	1) Same as 1 in AOT40 (Emberson *et al.*, 2007; Karlsson *et al.*, 2007b; Pleijel *et al.*, 2007) 2) More effective than the original DO_3SE because it implements the delimitation of the exposure period (Emberson *et al.*, 2007; Karlsson *et al.*, 2007b; Pleijel *et al.*, 2007) 3) Biologically more meaningful because plant ontogeny is considered with regards to max stomatal conductance (Pleijel *et al.*, 2007) 4) Same as 2 in DO_3SE	Same as 1 and 2 in DO_3SE	
POD_Y	1) Same as 1 in AOT40 (CLRTAP, 2015; Matyssek *et al.*, 2004) 2) Same as 2 in DO_3SE	Same as 1 and 2 in DO_3SE	1) Higher values may occur along coastlines (Anav *et al.*, 2016) 2) Soil water and nitrogen availability should be taken into consideration (Anav *et al.*, 2016; De Marco *et al.*, 2016) 3) Potential site-specificity (Azuchi *et al.*, 2014; De Marco *et al.*, 2016; González-Fernández *et al.*, 2017)

over the six months of the active growth of trees. In Europe, 9000 nmol mol^{-1} h (3-month, averaged over five years) have been proposed as target value, whereas 3000 nmol mol^{-1} h (3-month) have been proposed as long-term objective for protecting vegetation ("Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe," n.d.).

For acute O_3 toxicity, 5-day $AOT40_{VPD}$ CL, which take into consideration the vapor pressure deficit (VPD), have been proposed (Kärenlampi and Skärby, 1996). These levels are 500 nmol mol^{-1} h over five days with VPD greater than 1.5 kPa, or, 200 nmol mol^{-1} h over five days with VPD lower than 1.5 kPa.

A similar VPD correction can be used also for chronic exposures. Hourly O_3 mixing ratios $[O_3]$ can be multiplied by relevant hourly f_{VPD} factors (Spranger *et al.*, 2004) as follows,

$$f_{VPD} = 1 \ (VPD < 1.1\ kPa)$$
$$f_{VPD} = -1.1 * VPD + 2.2 \ (1.1\ kPa \leq VPD \leq 1.9\ kPa)$$
$$f_{VPD} = 0.02 \ (VPD > 1.9\ kPa)$$

AOT40 should be calculated using O_3 mixing ratios at the upper boundary of the quasi-laminar layer (top of the plant canopy) when the micro-meteorological approach of big-leaf is applied (Grünhage *et al.*, 1999; Hicks *et al.*, 1987); this requirement that has been often overlooked in the relevant literature. It has been demonstrated that failure to properly compute AOT40 with O_3 mixing ratios from a reference height may result in dramatic overestimates in potential yield which are unrealistic (Grünhage *et al.*, 1999). However, O_3 mixing ratio at measurement height can be effectively converted (approximation) to O_3 mixing ratio at the height of canopy top. This can be done using a relevant deposition model which is chosen based on the availability of meteorological data (Simpson *et al.*, 2012; Emberson *et al.*, 2000a, b).

In polluted regions, AOT40 critical values for chronic exposure can be exceeded even in individual months (Agathokleous *et al.*, 2017; Deb Roy *et al.*, 2009). Here we suggest that the AOT40 could be calculated on a monthly base and then a simple weighted correction could be applied for better reflecting the O_3 risk. This correction could be done by multiplying the total AOT40 value by the number of months AOT40 critical value was individually exceeded. For a final AOT40 value where the critical value was not exceeded in individual months, the AOT40 would be multiplied by 1; for a final AOT40 value where the critical value was exceeded in one individual month the AOT40 would be multiplied by 2; for a final AOT40 value where the critical value was exceeded in two individual months the AOT40 would be multiplied by 3; etc. This is because exceeding the AOT40 critical values in just a month is likely to exert more pressure on plants than it would happen if the AOT40 critical value is exceeded over three or six months.

AOT40 has been used in several studies because of its simplicity in calculation. However, AOT40 has been questioned because it considers only ambient O_3 levels but ignores environmental constraints to O_3 uptake through stomata like ambient temperature and water availability (Sanz *et al*., 2016; Harmens *et al*., 2007; Karlsson *et al*., 2007b; Paoletti and Manning, 2007; Gerosa *et al*., 2005; Matyssek *et al*., 2004; Danielsson *et al*., 2003; Grünhage and Jäger, 2003). For instance, a study in Japan suggested that an area with high O_3 exposure does not necessarily correspond to a high risk of O_3 impact because the impacts depend on the environmental conditions at the habitat, plant sensitivity to O_3 and plant physiological capacity (Watanabe *et al*., 2011). AOT40 may also be effective in highly polluted areas but ineffective in less polluted areas. One important limitation is that O_3 levels may exceed the threshold of 40 nmol mol^{-1} only after noon hours when g_s starts decreasing (Agathokleous *et al*., 2017; Cassimiro *et al*., 2016), thus less O_3 enters plant tissues despite higher O_3 levels. Still, as it takes into account only O_3 mixing ratios greater than 40 nmol mol^{-1}, it suggests there is no phytotoxicity at lower O_3 mixing ratios. However, recent progress in air pollution science suggests that O_3 can cause adverse effects to sensitive vegetation at levels lower that 40 nmol mol^{-1} e.g. (Sugai *et al*., 2018; Agathokleous *et al*., 2015; Grünhage *et al*., 2001). It is also known that vegetation may be "more sensitive to long-term exposure to modest O_3 levels (characterized by seasonal means) than frequent exposure to high O_3 levels (which are best captured by cumulative indices)" (Wang and Mauzerall, 2004). Hence, AOT40 may be unrealistic and lead to misleading assessment of O_3 impacts to sensitive vegetation which is negatively affected by O_3 levels below 40 nmol mol^{-1}. Adopting AOT40 as the default index for setting CL in Asia would be a poor policy with severe consequences at national and Pan-Asian level. It should be mentioned that thresholds higher than 40 nmol mol^{-1} have been considered as well. For instance, in the experiment of Feng *et al*. (2012), when AOT was calculated with thresholds ranging from 55 to 85 nmol mol^{-1}, it outperformed AOT40 in the exposure-response relationships. However, increasing the threshold does not provide a solution to the limitations explained above.

Instead of AOT40, AOT0 could be more effectively used (Azuchi *et al*., 2014). There is a preliminary evidence showing that AOT0 may predict the severity of the O_3-induced injury (number of injured leaves/total number of leaves on the injured plants × 100) better than AOT40 and Phytotoxic Ozone Dose (POD) indices (Cassimiro *et al*., 2016). In contrast to AOT40, AOT0 does not ignore low O_3 levels and could be thus more effective in any regions. Furthermore, since it takes into account low O_3 levels, AOT0 can yield more realistic exposure-response relationships when physiological endpoints are the matter of study; it is shown that physiological endpoints display dynamic responses to stressors which vary across the entire exposure-response continuum.

More details on useful improvements for toxicologically effective AOT40 can be found in earlier articles (Musselman *et al*., 2006; Grünhage *et al*., 1999).

AOT40 metric dominates the literature with Asian studies. Several experiments with crop plants conducted in Asia utilize AOT40 for deriving CLe_e. Experimenting with Chinese wheat cultivars (*Triticum aestivum* L. cvs.) in a Free Air Controlled Exposure (FACE) system, Oue *et al*. (2011) revealed cultivar-specific exposure responses of photosynthesis and stomatal aperture. In a further study with Chinese wheat cultivars (Feng *et al*., 2012), the slope between relative yield and AOT40 was significantly larger than European ones (Mills *et al*., 2007) even at shorter time period. This study also suggested that these Asian wheat cultivars are more sensitive to O_3 than North American ones. Two modern Indian wheat cultivars were also assessed as to their sensitivity to O_3 using open top chambers (OTCs) in Varanasi, India (Sarkar and Agrawal, 2010): AOT40 CLe_e for 5% reduction in yield was same to that for temperate wheat in Europe (Mills *et al*., 2007). In a different OTC experiment in China, four soybean cultivars (*Glycine max* L. Merr.), typical in Northeast China, were studied (Zhang *et al*., 2017): AOT40 CLe_e for 5% reduction in relative seed yield was similar to that for soybean in Europe (Mills *et al*., 2007). Regarding trees, Yamaguchi *et al*. (2011) summarized various study examples of O_3 impact assessment in tree seedlings using AOT40 in Japan and presented exposure-response relationships with AOT40 as predictor of relative whole-plant dry matter of Japanese larch (*Larix kaempferi* (Lamb.) Carr.) and Japanese beech (*Fagus crenata* Blume) seedlings under different soil nitrogen (NH_4NO_3) availability. A further OTC experiment was conducted with five poplar clones in China (Hu *et al*., 2015). The AOT40-based CLe_e derived from exposure-response relationship was 12,000 nmol mol^{-1} h for 5% reduction in total biomass across clones, whereas the AOT40 value in the experimental site in China was 40,500 nmol mol^{-1} h, which translates to 16.7% loss of total biomass (Hu *et al*., 2015). These values exceed by far the values set by worldwide regulatory agencies for protecting vegetation (discussed above). Two poplar clones were also studied in a different experiment employing five O_3 exposure levels in OTCs in China (Shang *et al.*, 2017). AOT40 CLe_e for 5% reduction in total biomass, photosynthetic parameters and leaf mass per area (LMA) for the two clones were 14,800, 4,000 and 5,800 nmol mol^{-1} h, respectively (Shang *et al.*, 2017).

3. 2 SUM06 and W126

Other than AOT*X* CLe_e indices based on cumulative exposure have been also used. Two examples of indices are the sum of all hourly average O_3 mixing ratios ≥ 0.06 μmol mol^{-1} (SUM06), and the sigmoidally weighed W126 which includes lower O_3 levels but assigns greater weight to higher mean hourly O_3 levels (Lefohn *et al.*, 1988; Lefohn and Runeckles, 1987). The formulas are:

$SUM06 = \sum_{i=1}^{n} [O_3]_i$ for $[O_3] \geq 0.06$ μmol mol^{-1} and

$W126 = \sum_{i=1}^{n} w[O_3]_i$

For SUM06, $[O_3]$ is the hourly mean O_3 mixing ratio (μmol mol^{-1}), *i* the index and *n* the total number of hours in three consecutive months for which the SUM06 value is greatest. For W126, $[O_3]$ and *i* are same as for SUM06 but without a threshold value. W126 rather employs *w* which is the weighting factor for the ith hour:

$$w_i = 1/[1 + M \times exp(-A \times [O_3]_i)]$$

where *M* and *A* are the arbitrary constants 4403 and 0.123 and $[O_3]_i$ the O_3 mixing ratio *i*. The units are μmol mol^{-1} h (classically ppm h). More explanations on the calculations and the historical bases can be found elsewhere (Musselman *et al.*, 2006; Wang and Mauzerall, 2004; Lefohn *et al.*, 1988; Lefohn and Runeckles, 1987).

The SUM06 index has not prevailed in the literature. The W126 index has been adopted by regulatory agencies in North America, where the standard for protection has been set at or below a range of 13,000-17,000 nmol mol^{-1} h (3-year average). W126 is calculated based on a sigmoidally weighted sum of all hourly mixing ratios observed during a particular daily and seasonal time window, with each ratio having a weight increasing from 0 to 1 with increasing ratio ("National Ambient Air Quality Standards for Ozone; Final Rule," 2015).

Recently, Zhang *et al.* (2017) reported that a SUM06 CLe_e of 7,600 nmol mol^{-1} h and a W126 CLe_e of 6,800 nmol mol^{-1} h relates to 5% reduction in relative seed yield of soybean cultivated in Northeast China (explained above in AOT40).

3. 3 MPOC

An alternative approach is the Maximum Permissible Ozone Concentration (MPOC), which can be used mainly for trees and at national level (Grünhage *et al.*, 2001). This approach requires transforming O_3 mixing ratios from a reference height to the canopy top, and thus has been characterized as more realistic (Grünhage *et al.*, 2001). However, MPOC has not been prevailed and AOT40 has hitherto dominated the literature.

4. CLe_f

Plants have tiny pores, called stomata (from the Greek *στόματα*, "mouths"), through which exchange gases with the atmosphere (Cieslik *et al.*, 2009). Although plants have stomata on a series of organs, leaf epidermis accounts for the vast majority of the total number of stomata. The functioning of stomata defines the amount of O_3 that does enter into the plant tissues (Vaultier and Jolivet, 2015): The greater the number or size of stomata is, or the longer the stomata remain open, a greater amount of O_3 enters the plant tissues. This indicates that higher O_3 level does not always induce higher stomatal O_3 flux when stomata are closed (Kinose *et al.*, 2014; Kitao *et al.*, 2014; Gerosa *et al.*, 2005). Hence, O_3 metrics which take into account the O_3 dose or flux in plants have been developed.

CLe_f approaches are considered superior to CLe_e approaches (Karlsson *et al.*, 2007b). This assumption relies on underlining plant physiological mechanisms according to which plant response is driven by the O_3 dose absorbed into the leaf tissue. Therefore plant response has been defined as (Grünhage *et al.*, 1999):

$$\begin{aligned} plant\ response &= f_1(PAD(O_3)) \\ &= f_1\left(\int_{t_1}^{t_2} |f_{absorbed}(O_3)| \times dt\right) \end{aligned}$$

where PAD is the pollutant absorbed dose (Fowler and Cape, 1982).

However, CLe_f approaches are technically more difficult because of the information needed in the *information processing system*. One important aspect for the CLe_f approaches is the soil moisture which should be implemented when setting CL (De Marco *et al.*, 2016; Pleijel *et al.*, 2004).

Stomatal conductance is the most critical parameter because it defines the O_3 dose entering into the plant tissues. Hence, extensive research has been done on modeling and parameterization of g_s at global level. The most common g_s model is the empirical multiplicative g_s model of Jarvis (Jarvis, 1976), which has been advanced for calculating stomatal O_3 flux (Hoshika *et* *al*., 2017b; Marzuoli *et al*., 2017; Kinose *et al*., 2014; Oue *et al*., 2011, 2009, 2008; Danielsson *et al*., 2003; Emberson *et al*., 2000a); Jarvis model is adopted by UNECE-CLRTAP. An alternative semi-empirical approach is that of Ball-Woodrow-Berry (BWB), which assumes that g_s is closely associated with photosynthesis and does not depend on maximum g_s (g_{max}) like Jarvis model (Ball *et al*., 1987). The BWB model is similarly effective with Jarvis model in O_3 risk assessment (Hoshika *et al*., 2017b; Kinose *et al*., 2017; Kitao *et al*., 2016). Improvements in BWB have been later proposed, such as including the leaf-to-air vapor pressure deficit (Leuning, 1995) and gross assimilation rate (Yu *et al*., 2001) rather than relative humidity and net photosynthetic rate. A potential O_3 avoidance mechanism of plants to avoid O_3 is by closing stomata, thereby decreasing g_s and consequently O_3 uptake. O_3 itself may induce stomatal closure (Shang *et al*., 2017; Hoshika *et al*., 2013b; Kitao *et al*., 2009; Wittig *et al*., 2007) or delay stomata response to environmental stimuli, so called stomatal sluggishness (Hoshika *et al*., 2013a, 2012b; Mills *et al*., 2009; McAinsh *et al*., 2002). However, other environmental factors than O_3, e.g. soil water unavailability, can affect stomata in a similar way with O_3 and at even a considerably greater extent than O_3 (Cotrozzi *et al*., 2017, 2016; Alexou, 2013; Hoshika *et al*., 2013a). Hence, when developing g_s models for estimating O_3 uptake, environmental factors inducing stomatal closure, such as N load-induced soil acidification (Azuchi *et al*., 2014) and soil water limitation (De Marco *et al*., 2016), should be taken into account. Soil water availability is a very critical driver of O_3 uptake by plants as can either increase or decrease g_s. Limitation to g_s by soil water unavailability may not occur in some Asian regions like Japan (Hiyama *et al*., 2005) but may commonly occur in other Asian regions like China (Tian *et al*., 2016; Qiu, 2010). As drought may greatly diminish the uptake of O_3 by plants, care should be exercised to take into account stomatal closure due to soil water limitation when conducting O_3 risk assessment in Asian regions where drought commonly occurs.

4.1 DO$_3$SE

The stomatal flux-based model called Deposition of O_3 and Stomatal Exchange (DO$_3$SE) has been proposed as an alternative to AOT40 (Emberson *et al*., 2000a). DO$_3$SE takes into account environmental factors and vegetation properties in addition to ambient O_3 levels (Anav *et al*., 2016; De Marco *et al*., 2016). Species-specific effects of soil water availability, air temperature, vapor pressure deficit, irradiation, plant phenology and stomatal functioning, are factors taken into account. DO$_3$SE, as it has been originally proposed, has not prevailed in the literature.

4.2 AFst*Y*

The cumulative leaf uptake of O_3 per total leaf area over time (CUO) has been proposed as an effective adjustment of the Emberson calibration (Emberson *et al*., 2000a) of the multiplicative g_s for crops and trees, especially for a cumulated period lasted from anthesis to harvest for crops (Karlsson *et al*., 2004a, 2004b; Danielsson *et al*., 2003). Based on the CUO concept, the index of accumulated stomatal flux above a flux rate threshold *Y* (AFst*Y*) has been developed which, in contrast to AOT40, implements the delimitation of the exposure period (Emberson *et al*., 2007; Karlsson *et al*., 2007b; Pleijel *et al*., 2007). AFst*Y* is calculated (Emberson *et al*., 2007) as:

$$\mathrm{AFst}Y = \sum_{i=1}^{n} [Fst_i - Y] \text{ for } Fst_Y \geq Y \,\mathrm{nmol\,m^{-2}}\, PLA\, \mathrm{s^{-1}}$$

where *Y* is a defined flux rate threshold, *n* is the number of hours within the accumulation period, Fst_i the hourly mean O_3 flux (nmol O_3 m^{-2} PLA s^{-1}), and PLA the projected plant leaf area. AFst*Y* is more plant-relevant than AOT and takes into account the influence of plant ontogeny on g_{max} (Pleijel *et al*., 2007). Accumulated stomatal O_3 flux above a flux rate threshold of 6 nmol O_3 m^{-2} projected sunlit leaf area s^{-1}, based on O_3 flux hourly values ($AF_{st}6$) or above a flux rate threshold of 1.6 nmol O_3 m^{-2} projected sunlit leaf area s^{-1} ($AF_{st}1.6$) have been applied to crops (Fuhrer, 2009; Oue *et al*., 2008; Pleijel *et al*., 2007) or trees (Emberson *et al*., 2007; Karlsson *et al*., 2007b).

4.3 POD$_Y$

The air pollution scientific community and the UN/ECE have put effort in the past decade to redefine O_3 CL for protecting vegetation using POD, based on the cumulated stomatal O_3 (CLRTAP, 2015; Matyssek *et al*., 2004). The effort of UN/ECE is being a part of the Convention on Long-Range Transboundary Air Pollution (CLRTAP), which is implemented by the European Monitoring and Evaluation Programme (EMEP) and directed by the UN/ECE. The mission of CLRTAP is to protect the human environment against air pollution and to gradually reduce and prevent air pollution, including long-range trans-boundary air pollution. POD$_Y$ (POD above a threshold flux of *Y* nmol m^{-2} PLA s^{-1}) has been widely used for deriving CL for crop plants, pastures and trees (Calvete-Sogo *et al*., 2017; Marzuoli *et al*., 2017; Cassimiro *et al*., 2016; De Marco *et al*., 2016; Kitao *et al*., 2016; Sanz *et al*., 2016; Bagard *et al*., 2015; Büker *et al*., 2015; Danielsson *et al*., 2013; Grünhage *et al*., 2012; Mills *et al*., 2011). It has been shown that POD$_0$ may predict more effectively the incidence of the injury (number of injured plants/total number of plants × 100), severity of the injury and leaf abscission than POD_1, POD_2, POD_3, POD_4, POD_5 and POD_6 (Cassimiro *et al.*, 2016). POD approach has been advanced to the point that it has been used to estimate O_3 impacts at forest level (Kitao *et al.*, 2016) or setting O_3 CL for multi-species canopies of Mediterranean annual pastures (Calvete-Sogo *et al.*, 2017). These efforts are very important because (i) plant response to O_3 may vary between monospecific and multispecies cultures and (ii) the target of ecotoxicology is communities and populations and not individuals. The availability of nitrogen (Calvete-Sogo *et al.*, 2017) and water (De Marco *et al.*, 2016) should be taken into account when working with POD_Y because soil nitrogen and water availabilities influence the flux-response relationships and modify the CLe_f.

In O_3 risk assessments using high spatial resolution over Europe for the years 2000-2005, it was found that the AOT40-based risk assessment showed a good consistency compared to *in situ* data and other model-based datasets, but stomatal O_3-uptake-based assessment showed different spatial patterns compared to other model-based datasets (Anav *et al.*, 2016).

POD CLe_f for agricultural and horticultural crops, forest trees and (semi-)natural vegetation, based on a series of experiments conducted in Europe, have been previously summarized (Mills *et al.*, 2011). It has been shown that POD_6 values of 1, 2, 2, 5 and 2 mmol m^{-2} were needed to reduce by 5% the grain yield of wheat, 1,000 grain weight of wheat, protein yield of wheat, tuber yield of potato, and fruit yield of tomato, respectively (Mills *et al.*, 2011). Also, POD_1 values of 4, 8, and 2 mmol m^{-2} were needed to reduce by 4% the annual whole tree biomass of beech and birch, 2% the annual whole tree biomass of Norway spruce, and 10% the above-ground biomass of *Trifolium* spp (Mills *et al.*, 2011). An analysis of published data indicated POD_1 CLe_f (95% CI) of 12.2 (8.9, 15.5), 7.2 (1.1, 13.3) and 4.6 (2.7, 6.5) mmol O_3 m^{-2} for a 10% loss of above-ground biomass, reproductive capacity and consumable food value, respectively, of Mediterranean annual Dehesa-type pastures (Sanz *et al.*, 2016). Recently, a POD_6 CLe_f of 1 mmol O_3 m^{-2} has been also reported for a 15% loss of marketable yield in lettuce (Marzuoli *et al.*, 2017).

There is however a lack of experimental evidence on observation-based CLe_f in Asia, and, hence, more studies are needed to reach the point of generalizations. Regarding crop plants, POD was estimated in Japanese rice (*Oryza sativa* L. cv. Koshihikari) using a variety of flux thresholds and phenological integration periods (Yamaguchi *et al.*, 2014). This study suggested that the Koshihikari yields can be assessed using a threshold of 10 nmol O_3 m^{-2} PLA s^{-1} (POD_{10}) and an integration period of −300 to 100°C days from anthesis. In the experiment with winter wheat in subtropical China discussed in section III, dose-response relationships were tested for POD values with threshold *Y* ranging from 0 to 24 nmol O_3 m^{-2} PLA s^{-1} (Feng *et al.*, 2012). Cultivar Yangfumai 2 displayed a greater sensitivity of relative yield to POD_Y than the other three cultivars tested, but only for *Y* values ≥ 13 nmol O_3 m^{-2} PLA s^{-1}. Overall, $POD_{11\text{-}20}$ was a better predictor of relative yield than AOT40 (Feng *et al.*, 2012). The authors suggested a threshold of 12 nmol O_3 m^{-2} PLA s^{-1} for wheat flux-response relationships in subtropical China. Zhang *et al.* (2017) improved g_s model to include the effect of leaf age. They found that the POD_6 and $POD_{9.6}$ CLe_f for 5% reduction of relative seed yield of soybean in China were 1.8 and 0.9 mmol O_3 m^{-2}, and $POD_{9.6}$ yielded better dose-response relationships than the other tested metrics (explained above in section III). In a different study, simulated O_3 was used to evaluate the relative yield loss of rice in a domain of Southern Vietnam using AOT40, mean 7 h daytime O_3 level (M7), POD_0 and POD_{10} metrics (Danh *et al.*, 2016): This study revealed a prediction of greater rice production loss by POD_{10}, compared to AOT40, M7 and POD_0, in agreement with previous findings in Japan (Yamaguchi *et al.*, 2014). This may be the first attempt for assessing O_3-induced crop yield loss with different O_3 exposure metrics in Asian developing countries and is encouraging for further studies. Asian studies implementing POD are fewer for trees than crop plants. In the experiment with poplars in China mentioned above (section III), POD_1 dose-response relationship indicated a 2% biomass loss at 2.1 mmol m^{-2} and a 4% loss at 4.8 mmol m^{-2}, whereas POD_7 yielded a 5% reduction in total biomass at 3.8 mmol m^{-2} and was more effective predictor than AOT40 and POD_1 (Hu *et al.*, 2015); POD_1 is recommended by CLRTAP (2015) for protecting against O_3-induced biomass loss of trees. Based on the findings of Hu *et al.* (2015), Shang *et al.* (2017) used POD_7 for deriving O_3 CLe_f for an experiment with two poplar clones (explained in section III). CLe_f for 5% reduction in total biomass, photosynthetic parameters and leaf mass per area (LMA) for the two clones were 9.8, 3 and 4 mmol O_3 m^{-2} PLA, respectively; coefficients of determination (R^2) were similar between AOT40- and POD_7-based dose-response relationships for all the response variables (Shang *et al.*, 2017). Based on these clones, POD_7 has been also recommended over AOT40 for large-scale risk assessment of isoprene emission to O_3 in poplar (Yuan *et al.*, 2017).

5. CLe_e OR CLe_f?

Both CLe_e and CLe_f approaches may overestimate

O_3 effects on vegetation because plant detoxification mechanisms are not taken into account (Musselman *et al*., 2006). In both cases it is assumed that sensitivity and adaptive capacity are equal across species, which is a challenge in assessing species vulnerability to O_3 (Butt *et al*., 2016; Loibl *et al*., 2004). While CLe_f approach is biologically more meaningful and realistic than CLe_e approach, flux-related assessments taking into account physiological defense capacity could better predict O_3 effects because physiological response to O_3 can be modified by global change (Tausz *et al*., 2007; Loibl *et al*., 2004; Matyssek *et al*., 2004). In this sense, quantitative understanding of effective flux (Kinose *et al*., 2017) should be developed before adapting flux-based models by regulatory agencies (Musselman *et al*., 2006). Nevertheless, while CLe_f approaches are biologically more meaningful and are considered superior to CLe_e approaches, their calculation requires plant physiological data which are not always available. This is an important limitation especially in developing Asian countries where there may be no access to instruments for such physiological measurements. Hence, both CLe_e and CLe_f approaches are useful for assessing O_3 impacts on vegetation with a Pan-Asian perspective. Although biologically more relevant, CLe_f approaches should be accompanied by a standard CLe_e approach (e.g. AOT0) in order to provide a scientific base for mutual understanding and benefit among Asian countries.

6. CONCLUSIONS AND PERSPECTIVES

A variety of O_3 metrics has been used by the air pollution research community. Considering the strengths and weaknesses of the common indices, it could be suggested that AOT40 may be used for highly polluted regions whereas AOT0, W126 or a CLe_f approach can be used in regions with low O_3 levels. However, although AOT40 has been widely used and adopted by worldwide regulatory agencies, the current scientific literature suggests it is inefficient and should be replaced by other indices, like AOT0 (or SUM00: sum of all hourly average O_3 levels), which do not set a threshold.

To facilitate the future developments in science and foster correct policy and regulations at national and Pan-Asian level, it is recommended to set as a standard practice to report more than one indices and never AOT40 alone. It is recommended AOT0, which is easy to calculate, to always accompany AOT40 or flux-based metrics.

O_3 effects on plants do not depend only on O_3 exposure characteristics but also on plant physiology and biological plasticity, something that should be considered when selecting O_3 phytotoxicity metrics. In this context, CLe_e metrics may totally fail to realistically predict plant response (or O_3 effects on plants). For a more realistic assessment of species vulnerability and risk in the future, implementation of plant detoxification capacity should be set as a research priority. This will be a challenging task for the coming decades because of (a) the complexity of detoxification mechanisms; (b) the current incomplete understanding of O_3 mode of action in plants, or the plant response to O_3; and (c) the limited knowledge about O_3 effects on native or local cultivated plants in wide regions of Asia. Nevertheless, earlier trials where photosynthesis (Hoshika *et al*., 2017a; Kinose *et al*., 2017; Li *et al*., 2016; Oue *et al*., 2011; Kolb and Matyssek, 2001) and LMA (or its inverse specific leaf area; SLA) (Li *et al*., 2016; Wieser *et al*., 2002) were used as proxies can serve as a basis for further developments at Pan-Asian level.

More Asian studies are needed to improve the current limited understanding and contribute in setting national and Pan-Asian standards for the protection of crop plants and native vegetation. This is particularly important for Asian tropical regions as relevant experimental studies on plant response to O_3 remain very limited.

In contrast to closed exposure systems, FACE systems allow interaction of plants with the natural environment and thus provide experimental conditions close to natural ones. FACE systems (Kobayashi, 2015) can offer an interface for realistic derivation of CLe and a substantial base for adopting regulatory measures in Asia. As FACE systems cannot reduce ambient O_3 concentration, they should be established in O_3-clean areas or alternative methodologies to exclude potential background O_3 stress should be considered (Paoletti *et al*., 2017).

Environmental conditions, growing medium characteristics and plant competition (inter or intra) are likely to drive dose response relationships (González-Fernández *et al*., 2017; De Marco *et al*., 2016; Azuchi *et al*., 2014). Policy and decision makers and stakeholders of Asian regulatory agencies should consider site-specific differences in dose response relationships before adopting O_3 CLe_f. In this framework, multi-site assessments are required for adoption of representative O_3 CLe_f.

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CONFLICT OF INTEREST

The authors declare that their research has no conflict of interest.

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