



# Multilayer modelling of ozone fluxes on winter wheat reveals large deposition on wet senescing leaves

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## ABSTRACT

Understanding how ozone is deposited on vegetation canopies is needed to perform tropospheric greenhouse gas budgets and evaluate the associated damage on vegetation. In this study, we propose a new multilayer scheme of ozone deposition on vegetation canopies that predicts stomatal, cuticular and soil deposition pathways separately. This mechanistic ozone deposition scheme is based on the multi-layer, multi-leaf mass and energy transfer model MuSICA. This model was chosen because it explicitly simulates the processes of rain interception, through fall and evaporation at different depths within the vegetation canopy, so that ozone deposition on wet leaf cuticles can be explicitly modelled with ozone dissolution, diffusion and chemical reaction inside the water films. The model was evaluated against a 3-year dataset of ozone, CO<sub>2</sub> and evapotranspiration flux measurements over a winter wheat field near Paris, France (ICOS Fr-GRI). Only periods with fully developed canopies (including senescence) were considered to minimise the contribution of soil deposition to the total ozone flux. Before senescence, the model could reproduce the measured ozone deposition rates as well as the CO<sub>2</sub> and water vapour fluxes. During senescence, large ozone deposition rates were observed under wet canopy conditions that could only be explained by first-order reaction rates in the water film of around 10<sup>5</sup> s<sup>-1</sup>. Such reaction rates are not compatible with the chemical composition of rainwater. We therefore hypothesise that, during senescence, the cell content leaks out of the leaves when they become wet, exposing anti-oxidants to ozone. These results provide for the first time a mechanistic explanation of the commonly observed increase in ozone deposition rates during rain or dew formation.

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## 1. Introduction

Tropospheric ozone (O<sub>3</sub>) is one of the major atmospheric pollutants. Worldwide, the concentration of tropospheric O<sub>3</sub> has doubled since preindustrial era (Anfossi and Sandroni, 1997; Vingarzan, 2004) and is expected to increase by 40–60% in the next century (Meehl et al., 2007). It is also known that the increase in O<sub>3</sub> disrupts plant physiology (Karnosky et al., 2007; Ren et al., 2007). In particular, it decreases plant uptake of carbon dioxide (Felzer et al., 2004) and therefore affects the total greenhouse gas budget of the atmosphere.

Ozone is deposited on vegetation canopies through four pathways: (1) stomatal absorption, (2) chemical destruction in the

canopy air, (3) soil deposition and (4) cuticle deposition on vegetation. The partitioning between these four deposition pathways has been studied for a range of ecosystems (Altimir et al., 2002; Kurpius and Goldstein, 2003; Lamaud et al., 2009; Massman et al., 1994; Stella et al., 2013, 2011b). In general, stomatal absorption is considered to represent the main sink for mature, productive canopies. However, on a yearly basis, ozone deposition rates on soil and leaf surfaces have been found to represent up to 70% to 80% of the cumulative flux in various ecosystems (Coyle et al., 2009; Stella et al., 2011a, 2013; Zhang et al., 2002a).

Continuous total O<sub>3</sub> fluxes can be easily estimated from micrometeorological flux measurements, but estimating the amount of ozone molecules taking each of the pathways is less direct. (1) Stomatal O<sub>3</sub> absorption is usually inferred from stomatal conductance for water vapour, and hence empirically derived from measurements of transpiration or total evapotranspiration. Such method can lead to large errors on the stomatal pathway during periods of

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low evaporative demand (e.g. at night) or an overestimation of this pathway when evaporation from soil or leaf surfaces becomes non negligible (e.g. after rainfall). In addition, stomatal O<sub>3</sub> absorption is usually taken proportional to stomatal conductance, assuming that O<sub>3</sub> is completely destroyed inside the leaf mesophyll and that the internal resistance of ozone transfer from the stomatal cavity to the sites of destruction is negligible compared to stomatal resistance. This can create an overestimated estimation of ozone absorption through stomata (Tuzet et al., 2011). At night the opposite is true as night-time stomatal ozone absorption is usually neglected (Altimir et al., 2004; Lamaud et al., 2009), despite the growing evidence of stomatal opening during the night, especially at low vapour pressure deficit (Barbour and Buckley, 2007; Caird et al., 2007). (2) Chemical destruction of ozone in the canopy air is usually neglected as well because the half-life time of O<sub>3</sub> in the atmosphere is typically longer than the travel time of gas molecules inside the canopy air (Galmarini et al., 1997; Stella et al., 2012). (3) Ozone deposition on bare agricultural soils can be measured using the eddy-covariance method during bare soil periods and the parameterisations obtained during those periods are usually assumed to remain valid in the presence of vegetation (Stella et al., 2011a). (4) Ozone deposition on plant cuticles is thus determined as the residual of total ozone deposition minus soil and stomatal absorption (Lamaud et al., 2009; Zhang et al., 2002b). Using this approach Coyle et al. (2009) found that O<sub>3</sub> deposition on plant cuticles was correlated with solar radiation or air temperature, while Zhang et al. (2002a) and Lamaud et al. (2009) found that it increased exponentially with air relative humidity. Although the effect of surface wetness on this residual conductance has been shown for a long time (Altimir et al., 2006; Fuentes and Gillespie, 1992; Fuentes et al., 1994; Grantz et al., 1995; Lamaud et al., 2002; Pleijel et al., 1995), currently no clear consensus exists on the drivers and processes governing O<sub>3</sub> deposition on plant cuticles. Existing parameterisations (Coyle et al., 2009; Lamaud et al., 2009; Zhang et al., 2002b) remain empirical and based on residual analysis. Moreover, they rely on the questionable assumption that the leaf-to-air vapour pressure deficit – required to estimate canopy stomatal conductance – can be estimated from an energy budget-computed with single-leaf models. These models ignore the surface temperature heterogeneity in a vegetation canopies with, e.g. sunlit and shaded leaves (Ding et al., 2014). Such an assumption could be at the origin of the relationship between the O<sub>3</sub> deposition residual and air temperature or relative humidity that was reported in the literature. Clearly a process-based approach that estimates O<sub>3</sub> deposition on plant cuticles and explicitly accounts for the spatial heterogeneity of plant surface properties within vegetation canopies is needed.

Such a process-based approach was attempted by Tuzet et al. (2011) using a two-layer vegetation canopy model and an O<sub>3</sub> deposition model on plant surfaces based on reaction-diffusion equations inside cuticles of dry leaves. In this model some “reaction sites” were assumed to be responsible for ozone destruction inside the leaf cuticles and their number was dynamically computed and systematically reset to its maximum value during rain events in order to reproduce the very dynamic response of observed O<sub>3</sub> deposition rates during such events and its subsequent decrease during canopy drying. The role of reaction-diffusion processes inside the water films present on wet leaf surfaces during and after rain events could not be evaluated because rain interception and evaporation were not embedded in the model.

In this study, we will test the hypothesis that ozone deposition on leaf cuticles occurs only when the cuticles are wet. For this we developed a mechanistic ozone deposition scheme based on the multi-layer, multi-leaf mass and energy transfer model MuSICA (Ogée et al., 2003). This model was chosen notably because it explicitly simulates the processes of rain interception, through fall and

evaporation at different depths within the vegetation canopy. For this new ozone deposition scheme stomatal ozone uptake is parameterised separately for sunlit and shaded leaves in different canopy layers, while ozone deposition on wet leaf cuticles is explicitly modelled with ozone dissolution, diffusion and chemical reaction inside the water films. The model is evaluated using water, CO<sub>2</sub> and energy fluxes and ozone deposition velocities measured above a winter wheat crop over three growing seasons. A sensitivity analysis to the model parameters governing the water film thickness dynamics and the chemical reaction rates in water is also presented, and the seasonal dynamics of simulated stomatal and non-stomatal deposition rates are then discussed with respect to other canopy-scale parameterisations.

## 2. Material and methods

### 2.1. Model description

#### 2.1.1. The MuSICA soil-vegetation-atmosphere model

The MuSICA model (Ogée et al., 2003) is a multi-layer, multi-leaf, soil-vegetation-atmosphere transfer model computing the exchanges of energy, CO<sub>2</sub>, water and their stable isotopes in the soil-vegetation-atmosphere continuum. The model considers several vegetation classes in each layer according to their light regime (sunlit or shaded), age (days or years depending on the species) and water status (wet or dry). The terrain is assumed to be flat (no horizontal advection) and the vegetation horizontally homogeneous (no clumping beyond the shoot/whirl level).

The radiative transfer model is based on the radiosity method and supports multiple species in each vegetation layer (Sinoquet and Bonhomme, 1992; Sinoquet et al., 2001), with either needle leaves (Smolander and Stenberg, 2003) or broad leaves (Myneni et al., 1989). Turbulent transfer inside the canopy is described by the Lagrangian one-dimensional turbulent dispersion model of Raupach (1989) with the parameterisations of Massman and Weil (1999). This dispersion model is coupled to an energy, water and CO<sub>2</sub> leaf-air exchange model for each leaf type in each layer, that combines a photosynthesis model (Farquhar et al., 1980), a stomatal conductance model (Ball et al., 1987; Leuning, 1995), a boundary-layer conductance model (Grant, 1984; Nikolov et al., 1995) and a leaf energy budget model. In the soil, a multilayer coupled heat and water transport scheme is used (Braud et al., 1995) that takes into account root water uptake and redistribution (Domec et al., 2012). Leaf water potential is also estimated by mass balance between root water uptake, leaf transpiration and internal water storage (Williams et al., 2001). When leaf water potential falls below a species-specific threshold, stomatal conductance, leaf photosynthetic capacity and root hydraulic conductivity are down regulated to empirically simulate the response of plant function to drought (McDowell et al., 2013).

Canopy rain interception and water storage on leaf surfaces are computed in each vegetation layer using a water balance equation and the concept of maximum storage capacity (Rutter et al., 1971). The leaf fraction covered with liquid water in vegetation layer *j* ( $p_{wet,j}$ ) is then computed from the corresponding water storage ( $W_{f,j}$ , kg m<sup>-2</sup> of ground area) as:

$$p_{wet,j} = \left( \frac{W_{f,j}}{W_{f,max,j}} \right)^\mu \quad (1)$$

where  $W_{f,max,j}$  (kg m<sup>-2</sup> of ground area) denotes the maximum water storage capacity of vegetation layer *j* (assumed proportional to leaf area, i.e.  $W_{f,max,j} = W_{f,max}L_j$ ) and  $\mu$  is a parameter related to the wettability of the leaf cuticle, ranging from 0 (full wettability) to 1 (constant thickness). When condensation occurs Eq. (1) is replaced by  $p_{wet,j} = 1$ , assuming that dew forms on the entire

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