Parameterization of ozone and aerosol particle fluxes

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Abstract

This paper presents preliminary results on dry deposition of ozone and submicronic aerosol particles using the eddy-correlation technique. In the absence of vegetal activity, and over dry soil (the site is located in Northern Spain near Zaragoza) with low resistance for ozone removal, ozone fluxes are controlled by dynamic and convective turbulence. The ozone deposition velocity has been normalized by a new velocity scale which is a combination of the friction velocity and the convective velocity. This reduced deposition velocity is a function of the dimensionless stability index (z/L). A similarity equation is proposed allowing the determination of deposition velocities in good agreement with the experimental data. Friction velocity and atmospheric instability in the boundary layer seem to govern aerosol particle deposition.

1. Introduction.

The dry deposition of atmospheric constituents is controlled by dynamical processes and surface removal by both vegetation and soil (Whelpdale1). On vegetation, the process occurs through various pathways: outer leaves, inner canopy, soil; cuticle, stomata (Baldocchi et al.2). Partitioning ozone fluxes against its different components is rather difficult since only the total ozone fluxes are measured (Massman3).

Decomposition by contact is the only mechanism involved in ozone dry deposition over a dry soil so discrimination of the surface resistance using a canopy resistance model is no longer useful. Assuming that the daily variations of soil resistance to ozone removal during the campaign were not significant, it appears that the ozone deposition velocity depends on turbulent motion, characterized in the surface layer by friction velocity and sensible heat flux.

It is interesting to compare the soil-atmosphere exchange of ozone and
aerosol particles since both fluxes are downward, but are driven by different mechanisms. Deposition of aerosol particles occurs through impaction or gravitational sedimentation. This paper is not directly focused on parameterization of aerosol particle fluxes but it will be used to show that ozone and aerosol particle fluxes are not controlled by the same parameters.

The measurements have been carried out in July 1992. The site is located 80 Km Northeast from Zaragoza (Spain). The fluxes of momentum, sensible heat, ozone and aerosols were measured at a height of 7 m a.g.l. by the eddy-correlation method. The ozone analyzer is a chemiluminescence device (Güsten et al.*) and the aerosol analyzer was developed at the Laboratoire d’Aerologie by El Bakkali^5; its working principle is based on ionization of the particles by an electrical field. The time constants of both systems are lower than 0.2 s.

The fast-response ozone analyzer gives only relative values in the absence of an absolute concentration measurement; by dividing by their time average deposition velocities of ozone are directly obtained.

2. Site and meteorological conditions.

The site shows three different roughness length values, depending on wind direction. They correspond to a very smooth bare soil \((z_0=0.25\text{ cm})\) towards North-West), a rough bare soil \((z_0=1\text{ cm})\) towards South-East) and almond-tree rows (2 meters height) with very few foliar development (including a displacement height \(z_d=1.5\text{ m}\), \(z_0=10\text{ cm}\) towards South-West).

North-West is the direction of the prevailing regional wind called Cierzo, characterized by very high wind speeds (6-8 m/s). This occurred for days 11 and 12. SW winds are slower (often lower than 2 m/s) and occurred during days 16 to 20. These light winds led to very low Minion-Obukhov lengths. SE wind occurred after rotation at 18h for day 18, at 15h for day 19 and 12h for day 20. Wind intensity is increasing after this wind rotation.

Figure 1 shows the evolution of wind speed and direction, net radiation \((Rn)\) and soil heat flux \((G)\), temperature and turbulence rate for ozone.

Net radiation data clearly show that the sky was always cloudless during the campaign and the maximum intensity reaches about 400 Watt \(m^2\) at 13h. We observe the same regularity concerning the soil heat flux with maximum values of 150 Watt \(m^2\) at 13h. These values are similar to those found by Ide^6 for a semi-desert area in Niger.

3. Turbulent fluxes.

Figure 2 presents values of ozone deposition velocities \(V_d(O_3)\), friction velocities \((u^*)\), sensible heat fluxes \((H_0)\) and dimensionless stability index \((z/L)\).
Figure 1 - Meteorological conditions during the campaign.

Figure 2 - Turbulent fluxes for the campaign.
We notice a similarity in the daily evolution between ozone deposition velocity and friction velocity for days 16 and 18. For days 11 and 12, the daily trend of ozone deposition velocity is compares better with sensible heat flux; the deposition velocity values reach 0.008-0.007 m s\(^{-1}\). These values are in the same range as those found over a pine forest canopy in a previous study (Lamaud\(^7\)) and show clearly that ozone removal by dry soil is an efficient ozone removal process. For days 16 to 20, ozone deposition velocities decrease slightly between 0.002 and 0.006 m s\(^{-1}\). High deposition velocities were found for aerosol particles; they ranged from 0.002 to 0.008 m s\(^{-1}\).

4. Parameterization.

Wesely et al.,\(^8\), using the similarity theory, found that the deposition velocity for sulfate particles, when normalized by \(u^*\), was dependant on the Monin-Obukhov length scale \(L\). They found that \(V_d/u^*\) is almost constant at 0.002 for neutral and stable atmospheric conditions. The ratio \(V_d/u^*\) increases with increasing instability and this trend can be parametrized by:

\[
\frac{V_d}{u^*} = a + b \left( \frac{z_i}{L} \right)^{2/3}
\]

where \(z_i\) is the boundary layer height.

Figure 3 shows the results obtained during the Zaragoza campaign for aerosols. Very similar values (0.002) were found for neutral and stable conditions. As the observations of the boundary layer height were not available during this campaign, we used four values of \(z_i\), ranging from 200 m to 1500 m using \(a=0.002\) and \(b=0.0009\) as found by Wesely et al,\(^8\) Simulations are in agreement with experimental data (Lamaud\(^7\)).

![Figure 3 - Evolution of the deposition velocity for aerosol particle normalized by \(u^*\) versus \(z/L\) for the campaign.](image-url)
Figure 4 - Evolution of ozone deposition velocity normalized by u* versus z/L for the campaign.

Using a similar approach for ozone (see Figure 4), two separated tendencies appear. The upper branch of the curve corresponds to days 11, 12 and to the end of the day 19's afternoon. For other days (lower branch), the ratio $V_d/u^*$ seems to be constant against increasing z/L and its value is about 0.0013. At neutral and stable conditions, data are scattered without any clear tendency.

This means that the ozone deposition velocity is not controlled by turbulent kinetic energy as it seems to be the case for the deposition velocity of aerosol particles.

Since the correlation between sensible heat flux and ozone deposition velocity is high and most of the atmospheric conditions for days 16 to 20 are strongly unstable, we normalized the ozone deposition velocity by a combination of the convective velocity scale which is expressed by:

$$w_f = (\beta \overline{w'/u'^*}} z)^{1/3}$$

and with the friction velocity $u^*$, leading to a new velocity scale, $U_f$, which takes into account the two processes generating atmospheric turbulence.
Figure 5 - Evolution of $V_d/U_f$ versus $z/L$ for all the runs computed during the campaign in logarithmic representation.

\[ U_f = \frac{w_f^2}{u^*} \]  

(3)

Figure 5 shows the ratio $V_d/U_f$ versus $z/L$ in logarithmic representation in order to deduce the similarity power law. A linear regression procedure led to the following equation:

\[ V_d = U_f \left( 0.00158 \ |z/L|^{-0.6} \right) \]  

(4)

In order to evaluate the sensibility of the parameterization, we used the above equation to calculate the ozone deposition velocity. Figure 6 presents the scatter between measured and calculated values for ozone deposition velocity. The dashed lines correspond to +/- 25% differences with the 1:1 line. Most of the points are included in this range and the most scattered results concern day 16.

Figures 7 and 8 show a comparison between calculated and observed values for 2 days. Day 11 is characterized by high measured values of $V_d (O_3)$ and a strong diurnal variation. The computed values of ozone deposition velocity are in good agreement with the measured ones over the whole day and even small variations are reproduced. Day 20 shows increasing values of measured ozone deposition velocity during the day. The comparison with the calculated values is rather good for unstable conditions but diverges for neutral and stable atmospheric conditions. For these points, the dependency with $u^*$
Figure 6 - Comparison between measured and calculated ozone deposition velocities for the unstable atmospheric conditions.

Figure 7 - Daily evolution of measured and calculated ozone deposition velocity for day 11.

amplify the calculated values since the friction velocity increases strongly with the rotation of the wind without any correspondence in the measured values.
We report in Table I the mean difference per day between the measured and calculated ozone deposition velocity.

![Graph showing daily evolution of measured and calculated ozone deposition velocity for day 20.](image)

**Figure 8 -** Daily evolution of measured and calculated ozone deposition velocity for day 20.

**Table I -** Percentage of the mean difference between measured and calculated ozone deposition velocities for 7 days of the campaign.

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<th>day</th>
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