Intercomparison and sensitivity analysis of gas-phase dry deposition schemes

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Abstract

The dry deposition of gaseous species, such as ozone, sulphur dioxide and nitrogen oxides, is a major pathway in its removal from the atmosphere. Chemical Transport Models (CTM) commonly use a resistance approach to determine the deposition velocity of these gases. The objective of this study is twofold, first to compare different dry deposition schemes used in state-of-the-art CTMs, and second to evaluate the sensitivity of such schemes to key input parameters (meteorological conditions, soil type, leaf area index, reference values for non-stomatal resistances).

The canopy resistance accounts for most of the resistance in deposition velocity, therefore its formulation has been used for the intercomparison in the present study. The selected schemes are based on the formulations of Wesely [1], Emberson et al. [2] or Zhang et al. [3] with subsequent modifications accounting for the effect of phenology, photoactive radiation, vapor pressure deficit and soil-water potential. The stomatal resistance is the component of the canopy resistance related to the uptake of gases by the leaves. As it is commonly calculated for ozone and then extrapolated to other gases, the parameterization for ozone has been studied. The sensitivity analyses have been carried out by modifying the input parameters shared by most of the schemes. They are related mainly to the physiology of the vegetation and the process of gas exchange at the stomata. Meteorological inputs derived from ERA5 are used in the intercomparison. The simulated ozone deposition velocities are compared with available observational data. The dry deposition schemes show strong differences in the seasonal cycle, both in the vegetated and non-vegetated land use classes. The identification of critical parameters helps constraining dry deposition schemes incorporated in atmospheric models.

Keywords

Dry deposition velocity; Parametrization; Ozone

Introduction

Air quality is a major concern nowadays, as air pollution is responsible for negative impacts on human health and ecosystems. Aerosol and gas phase pollutants are routinely monitored in Europe through the observations of air quality networks.

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Besides, the diagnosis and prediction of air pollutant levels are commonly supported by numerical models, which allow defining and assessing air quality management strategies.

Chemical Transport Models (CTMs) represent the life cycle of air pollutants in the atmosphere, including emission, transport, chemical transformation, and wet and dry deposition processes. The dry deposition of gaseous species, such as ozone, sulphur dioxide or nitrogen oxides, is a major pathway in its removal from the atmosphere, but the mechanism that guides this process is not fully understood yet.

CTMs commonly use a resistance approach to determine the deposition velocity of trace gases, defining various in-series or in-parallel deposition pathways. These pathways consider an aerodynamic resistance, a common or surface-dependent quasi-laminar resistance and a canopy resistance.

The objective of this study is twofold, first to compare different dry deposition schemes used in state-of-the-art CTMs, and second to evaluate the sensitivity of such schemes to key input parameters.

Methodology

The intercomparison in the present study focuses on the formulation of the canopy resistance among models, as it is the most influencing resistance. We selected seven schemes (A to G) used in eulerian air quality models developed at Europe and North America, which ultimately rely on three main formulations (Table I). Schemes A to C follow the Wesely [1] dry deposition formulation, which defines the stomatal, cuticular, lower canopy and ground pathways. The scheme A represents the annual cycle defining two seasons, and the scheme B considers four seasons. The scheme C corrects a reference value for the stomatal resistance through an effective Leaf Area Index (LAI), which depends on LAI, zenith angle and cloud fraction. The other schemes (D to G) exclude the lower canopy pathway, but comprise modifications accounting for the effect of phenology, photoactive radiation, vapor pressure deficit and soil-water potential. The schemes D to F are based on Emberson et al. [2], which considers as input parameters the LAI and the surface area index (SAI), while the scheme G, based on Zhang et al. [3], only includes the former. As many of the models calculate the canopy resistance for ozone and then extrapolate the results to other gases, the parameterization for ozone is studied here.

The seven schemes for ozone dry deposition velocity were implemented as box models. The sensitivity analyses cover the year 2018 and meteorological inputs, derived from ERA5 [4], were obtained for a European location not directly affected by coastal phenomena (47°N, 4°E). All the schemes use tabulated resistances that depend on the land use classification, therefore several executions were made to consider a diversity of land uses. The input parameters considered to test the sensitivity of the deposition velocity were those shared by most of the schemes, namely: temperature at 2 meters above ground, relative humidity at 2 meters above ground, solar radiation, LAI, and the cuticular and ground surface resistances. These are related mainly to the physiology of the vegetation and the gas exchange process at the stomata.

Additionally, simulations of ozone deposition velocities were conducted and compared with available observational data over Europe and North America, on a monthly basis, considering the local meteorology and the land use reported at these sites [5-8]. The observation periods were 11 months in 2011 at Belgium (51°06’44’’N, 3°51’02’’E), 8 months between 2012 and 2013 at France (48°51’N, 1°58’E), 33 months between 2000 and 2003 at Norway (60°22’N, 11°4’E), and 60 months between 2008 and 2013 at Canada (44°19'N, 79°56'W).

Table I. Dry deposition schemes’ characteristics. T: temperature, SR: solar radiation, eLAI: effective leaf area index, PAR: photoactive radiation, VPD: vapor pressure deficit, PHE: phenology, SWP: soil-water potential.

Dry deposition scheme	Dry deposition formulation	Deposition pathways	Stomatal resistance’s correction factors	Tabulated resistance values
A	Wesely	4	T, SR	By land use and season (2)
B	Wesely	4	T, SR	By land use and season (4)
C	Wesely	4	T, eLAI	By land use
D	Emberson	3	T, PAR, VPD, PHE	By land use
E	Emberson	3	T, PAR, VPD, PHE, SWP	By land use
F	Emberson	3	T, PAR, VPD	By land use
G	Zhang	3	T, PAR, VPD, SWP	By land use

Results and Discussion

The baseline simulations of the dry deposition velocity show a high variability for the annual cycle among the schemes used, with peaks occurring at different times (Fig. 1). However, the schemes also share some common traits, for instance on vegetated surfaces, the deposition velocity is higher in spring-summer, while on non-vegetated surfaces it follows a constant annual profile (*data not shown*).

The evaluation of dry deposition velocities against observations is a challenging task, due to the scarcity of measurements. For this work, we use as a reference the retrieved ozone dry deposition velocities in four sites in Europe and North America. The evaluation with data from France, Norway and Canada suggests that, even if all schemes appropriately follow the deposition velocity seasonality, they show both under- and over- estimations of the retrieved values, and this happens both in winter and summer. The closer values between observational and simulated data are obtained with the schemes based on either Wesley [1] or Emberson et al. [2] formulations. The simulated values did not match the observations at the site in Belgium, which falls in a land use class characterized as temperate crop (Fig. 1). In fact, almost none of the schemes captured the observed seasonal cycle, showing negative correlation coefficients.

Then, the sensitivity analyses allowed us to determine the key parameters that modulate the deposition velocity, selected as those that increased at least two times this variable (Table II). On any land use, temperature, and on vegetated surfaces the LAI, if used in the scheme, are critical parameters in the modulation of the dry deposition velocity of ozone. The reference values for cuticular and ground surface resistances are key parameters as well, on vegetated and non-vegetated surfaces, respectively. Therefore, an accurate definition of these key parameters would allow increasing the robustness of the models. Some of them are inherent to the models' formulation, i.e., the cuticular and ground surface resistances are usually tabulated values. Others come either from tabulated values or external data sources, e.g., the LAI. Given its relevance in defining the resistance for deposition, observation-constrained data (e.g., from remote sensing) would be recommended.

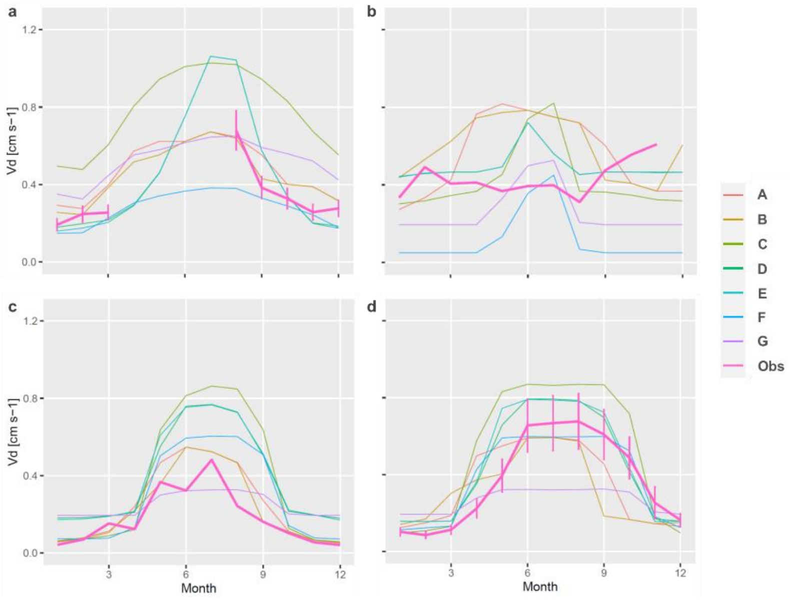

Fig. 1. Monthly mean ozone dry deposition velocity (Vd, cm s^{-1}) simulated in the box models (A-G) compared to observational data (Obs, thick pink line), over **(a)** grassland in France, **(b)** crop in Belgium, **(c)** coniferous forest in Norway and **(d)** coniferous forest in Canada. If available, mean±sd are shown in the observational data with error bars.

Table II. Sensitivity analysis of meteorological and vegetation related parameters. The change in dry deposition velocity equal to or higher than 2 times is indicated for the corresponding land use categories (V: vegetated surfaces; NV: non-vegetated surfaces).

Dry deposition scheme	Temperature (15/-15 °C)	Relative humidity (90/10 %)	Solar radiation (700/100 W m^{-2})	Leaf area index (5/0 m^2 m^{-2})	Cuticular resistance (200/9000 s m^{-1})	Ground surface resistance (100/3500 s m^{-1})
A	V, NV	-	V	-	V	V, NV
B	V, NV	-	V	-	V	V, NV
C	V, NV	-	-	V	V	V, NV
D	V, NV	-	-	V	V	V, NV
E	V, NV	V	-	V	V	V, NV
F	V	-	-	V	V	V, NV
G	V, NV	-	-	V	V	V, NV

Conclusions

State of the art Chemical Transport Models commonly represent the dry deposition of gas-phase species through resistance approaches and define deposition pathways to the vegetation and to the ground surface. These dry deposition schemes rely on three different main formulations, with subsequent modifications. Regardless of their common approach, the deposition velocities derived from these schemes show high variability in magnitude and seasonality over vegetated surfaces. For ozone deposition velocity, and by extension for other gases, temperature and LAI constitute the key input parameters.

The evaluation of the dry deposition schemes performance is challenging due to the scarcity of observational data. The comparison conducted in this work, with the available information, shows that the schemes are better at reproducing the seasonality of the ozone deposition velocity than its magnitude, with under- or over-estimations depending on the scheme and period.

In addition to the refinement of the key input parameters that have proven relevant to define the dry deposition velocity, there is a need for more observational data to better constrain the process representation in air quality models.

To complement the analysis presented here, additional sensitivity tests will be performed with a 3D eulerian air quality model to assess the impact of the deposition parameters on the modelled air quality levels.

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