



Interception of wet deposited atmospheric pollutants by herbaceous vegetation: Data review and modelling



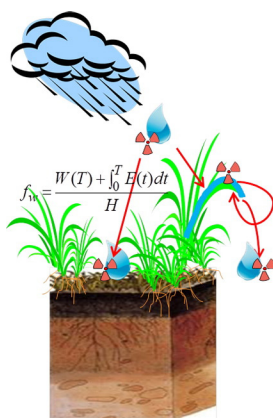
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HIGHLIGHTS

- Literature data on the interception of atmospheric pollutants by herbs were reviewed
- Predictive models were developed and evaluated in the Bayesian modelling framework
- Sensitivity of interception to environmental conditions was satisfactorily explained
- 81% of the observations were satisfactorily predicted by a semi-mechanistic model
- This model challenges empirical relationships currently used in risk assessment tools

GRAPHICAL ABSTRACT



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ABSTRACT

Better understanding and predicting interception of wet deposited pollutants by vegetation remains a key issue in risk assessment studies of atmospheric pollution. We develop different alternative models, following either empirical or semi-mechanistic descriptions, on the basis of an exhaustive dataset consisting of 440 observations obtained in controlled experiments, from 1970 to 2014, for a wide variety of herbaceous plants, radioactive substances and rainfall conditions. The predictive performances of the models and the uncertainty/variability of the parameters are evaluated under Hierarchical Bayesian modelling framework. It is demonstrated that the variability of the interception fraction is satisfactorily explained and quite accurately modelled by a process-based alternative in which absorption of ionic substances onto the foliage surfaces is determined by their electrical valence. Under this assumption, the 95% credible interval of the predicted interception fraction encompasses 81% of the observations, including situations where either plant biomass or rainfall intensity are unknown. This novel approach is a serious candidate to challenge existing empirical relationships in radiological or chemical risk assessment tools.

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Nomenclature

B	standing biomass density	$\text{kg} \cdot \text{d.w.} \cdot \text{m}^{-2}$
C_{Lai}	concentration of a substance fixed to the foliage surface	$\text{mol} \cdot \text{m}^{-2}$
C_{Rain}	concentration of a substance in rain water	$\text{mol} \cdot \text{l}^{-1}$
C_W	concentration of a substance in the aqueous film	$\text{mol} \cdot \text{l}^{-1}$
CR	concentration ratio at equilibrium	mm
D	drainage rate	$\text{mm} \cdot \text{h}^{-1}$
D_S	drainage rate at saturation	$\text{mm} \cdot \text{h}^{-1}$
E	evaporation rate	$\text{mm} \cdot \text{h}^{-1}$
E_0	potential evaporation rate	$\text{mm} \cdot \text{h}^{-1}$
EM, EM-Poly, EM-Cs, EM-PolyCs	equilibrium reversible models alternatives	
e	evaporation rate per unit surface of foliage	$\text{mm} \cdot \text{h}^{-1}$
f	interception fraction	unitless
f_0	unit interception fraction	unitless
f_B	mass interception fraction	$\text{m}^2 \cdot \text{kg}^{-1} \cdot \text{d.w.}$
f_{BO}	unit mass interception fraction	$\text{m}^2 \cdot \text{kg}^{-1} \cdot \text{d.w.}$
f_{LAI}	leaf area interception fraction	$\text{m}^2 \cdot \text{m}^{-2}$
f_W	water interception fraction	unitless
H	rainfall height	mm
HB, HI, H, HBI, HBI-Biomass, HBI-Cs, HBI-Poly, HBI-PolyCs	multi-linear regression models alternatives	
I	rainfall intensity	$\text{mm} \cdot \text{h}^{-1}$
J/K	absorption velocity	$\text{mm} \cdot \text{h}^{-1}$
KM, KM-Poly, KM-Cs, KM-PolyCs	kinetic irreversible models alternatives	
k	surface factor	unitless
k'	chemical affinity coefficient	unitless
LAI	single-sided leaf area index	$\text{m}^2 \cdot \text{m}^{-2}$
L	specific foliage storage	mm
MLR	multi-linear regression model	
PPLC	posterior predictive loss criterion	unitless
p	free throughfall coefficient	unitless
S	plant water storage capacity	mm
SLA	specific foliage area	$\text{m}^2 \cdot \text{kg}^{-1} \cdot \text{d.w.}$
SLA_{Leaf}	specific foliage area for leaves	$\text{m}^2 \cdot \text{kg}^{-1} \cdot \text{d.w.}$
SLA_{Stem}	specific foliage area for stems	$\text{m}^2 \cdot \text{kg}^{-1} \cdot \text{d.w.}$
T	exposure time	h
T^{abs}	absorption flux	$\text{mol} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$
T_S	saturation time	h
W	plant water storage	mm
α, β, γ	power coefficients of the multi-linear regression model	unitless
ε	residual variability	unitless
Σ	residual error	unitless

1. Introduction

Rainfall plays a crucial role in the deposition of airborne pollutants onto the ground surfaces and vegetation. Where rain occurs in the presence of airborne contamination, the wash-out (entrainment of particles or dissolution of gaseous species by falling droplets) and rain-out (formation of rain drops around particles) of material from the atmosphere can be responsible for deposit levels higher than those arising from dry deposition. A recent example was given by the Fukushima nuclear accident which induced considerable radioactive deposits in March 2011, even up to several tens of kilometers to the Northwest of the nuclear site. This heavy ground contamination was mainly attributed to the passage of heavy rainfall at time where high radioactive releases were dispersed in the atmospheric boundary layer (Katata et al., 2012a,b; Terada et al., 2012; Mathieu et al., 2012; Korsakissok et al., 2013; Draxler et al., 2014). Knowledge of the proportion of the deposit which is initially intercepted and retained by plants, also called wet interception fraction, is an important element of risk assessment studies, because it determines the level of contamination that can be expected in crops and other vegetation, for weeks, months or even years after deposition. In emergencies or post-accidental situations the consequences have to be evaluated quickly but as realistically as possible, it is of paramount

importance to take into consideration the type of deposited substances, the peculiarities of the precipitation event(s) that cause deposits and the characteristics of vegetation covers in the contaminated territories. Generic approaches which do not consider the specificities of the situation might lead to significant underestimations or overestimations, by more than one order of magnitude.

Rather comprehensive reviews of the mechanisms involved in the interception of wet-deposited radioactive contaminants by crops and herbaceous species were provided by Hoffman et al. (1992a,b), Kinnersley et al. (1997), Kinnersley and Scott (2001) and Pröhl (2009). The interception fraction, f (unitless), is defined as the ratio of the pollutant initially retained by the above-ground vegetation immediately after the deposition event and the total amount deposited onto the soil-plant system. This key parameter depends first on the capacity of a plant foliage (i.e. leaves, stems, etc.) to intercept and store precipitation. It is usually quantified by the water storage capacity, denoted S (mm of water), expressed in liters of water per unit surface area. As discussed for example by Keim et al. (2006a,b) and Muzyllo et al. (2009), storage capacity increases with above-ground plant biomass and, to some extent, also depends on the structure of the plant foliage as determined by the geometry and orientation with respect to the horizontal direction of the leaves (e.g. typical values for grass are slightly smaller than for other crop plants due to their preferential vertical orientation). Once this storage limit is reached, subsequent precipitation runs off the plant surface to the ground, therefore with increasing rainfall height, the intercepted water and contamination are expected to become a decreasing fraction of the total deposited amount. The effect of mechanical movement of the foliage, induced by wind or heavy rainfall intensities, may reduce water storage capacity, and should for this reason also be considered as a controlling factor. Secondly, the interception fraction depends on the ability of the plant foliage to fix or absorb the chemical substances contained in water droplets during the time of exposure from the plant. The physico-chemical affinity of the substances for the foliage surfaces is mainly governed by their electrical valence because foliage cuticle is negatively charged. As plant cuticle acts as a cation-exchanger at pH-values above 3 (Keppel, 1966; Franke, 1967; Schönherr, 1977; Schaefer and Reiners, 1990), it results in a strong absorption of cations and an inhibited absorption of anions. This has been confirmed by numerous experimental studies reported in the literature for a variety of plants and substances (e.g. as reviewed by Pröhl and Hoffman, 1996 and Pröhl, 2009). The experimental work by Kinnersley et al. (1997) who investigated the kinetics of Cesium attachment to crop leaf surfaces from a contaminated solution demonstrated that active chemical accumulation of Cesium occurred at plant surfaces and increased with the time of exposure, long after maximum water storage capacity had been reached. These results are compatible with Keppel's (1966) assertion that cuticle acts as a cation exchanger. These results actually suggest that, for any precipitation amount greater than the water storage capacity, the interception fraction should increase with rainfall duration or decrease with rainfall intensity. This is contradictory with the assertion by Hoffman et al. (1995) that rainfall intensity appeared to be of minor importance in determining interception.

A few operational models have been proposed in the literature that account for the dependence of the interception fraction upon the above-mentioned factors. The first approach was originally proposed by Hoffman et al. (1989, 1992a,b) based on an empirical regression analysis of detailed data from various herbaceous plant species and deposited substances (i.e. dissolved ions and micronic particles). They proposed to express the interception fraction, f , as a function of the standing biomass density, B (in kilograms of dry weight per square-meter, or $\text{kg} \cdot \text{d.w.} \cdot \text{m}^{-2}$), rain amount, H (in mm), and rain intensity, I (in $\text{mm} \cdot \text{h}^{-1}$), as follows:

$$f = f_0 \times \left(\frac{H}{1 \text{ mm}} \right)^\alpha \times \left(\frac{B}{1 \text{ kg d.w. m}^{-2}} \right)^\beta \times \left(\frac{I}{1 \text{ mm h}^{-1}} \right) \quad (1)$$

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