

Supplementary information to:

**Climatic, biological and land cover controls on  
the exchange of gas phase semivolatile chemical  
pollutants between forest canopies and the  
atmosphere**

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**Table SI1** List of symbols (with units) and acronyms

$A$	Amount of chemical in the throughfall (pg)	$k_{st}^*$	Chemical-specific conductance from diffusion through stomata ( $m\ s^{-1}$ )
$B$	Amount of chemical in the fraction of rainwater that impacts the canopy in a given layer (pg)	$k_w$	Conductance from forced advection ( $m\ s^{-1}$ )
$C_a$	Concentration of chemical in air ( $pg\ m^{-3}$ )	$K_{OA}$	Dimensionless octanol/air equilibrium partition coefficient
$C_c$	Overall concentration of chemical in the leaf (surface+reservoir) ( $pg\ g^{-1}\ dw$ )	$K_{OW}$	Dimensionless equilibrium octanol water partition coefficient
$C_r$	Concentration of chemical in the leaf reservoir ( $pg\ m^{-3}$ )	$K_{ra}$	Dimensionless leaf reservoir/air equilibrium partition coefficient
$C_s$	Concentration of chemical in the leaf surface ( $pg\ m^{-3}$ )	$K_{sa}$	Dimensionless leaf surface/air equilibrium partition coefficient
$C_w$	Concentration in rain ( $pg\ m^{-3}$ )	$L$	Leaf area index
$C_w^*$	Concentration in rain after impaction with the canopy ( $pg\ m^{-3}$ )	LM	Leuning Model
$d$	Thickness of leaf surface or reservoir compartment (m)	$MV$	Molecular volume ( $cm^3\ mol^{-1}$ )
$dt$	Incremental time (s)	$MW$	Molecular weight ( $g\ mol^{-1}$ )
$D_a$	Chemical molecular diffusivity in air ( $m^2\ s^{-1}$ )	$P(x')$	Polynomial function of the normalized values of wind speed, temperature or rainfall
$DWSR$	Down-welling short wave radiation ( $W\ m^{-2}$ )	PAH	polycyclic aromatic hydrocarbon
$E$	Latent heat of water evaporation ( $W\ m^{-2}$ )	PBDE	polybrominated diphenyl ether
$F_{dr}$	Degradation flux of chemical in leaf reservoir ( $pg\ m^{-2}\ s^{-1}$ )	PCBs	polychlorinated biphenyl
$F_{ds}$	Degradation flux of chemical in leaf surface ( $pg\ m^{-2}\ s^{-1}$ )	$r$	Rainfall (m)
$F_{ra}$	Air-leaf reservoir exchange flux by diffusion through stomata ( $pg\ m^{-2}\ s^{-1}$ )	$r_a$	Annual rainfall ( $m\ y^{-1}$ )
$F_{sa}$	Air-leaf surface exchange flux by gaseous partitioning ( $pg\ m^{-2}\ s^{-1}$ )	$\bar{r}_i$	Rainfall intensity ( $m\ s^{-1}$ )
$F_{sr}$	Leaf surface-leaf reservoir exchange flux by diffusion of chemical in leaf tissues ( $pg\ m^{-2}\ s^{-1}$ )	$R$	The ratio between $S_{c(M)}$ and $S_{c(R)}$
$F_{wet}$	Gaseous wet deposition flux ( $pg\ m^{-2}\ s^{-1}$ )	$S_c$	chemical storage in the whole canopy ( $ng\ m^{-2}$ )
$GP$	Gross photosynthesis ( $\mu mol\ CO_2\ m^{-2}\ s^{-1}$ )	$S_{c(M)}$	$S_c$ for the modified input scenario calculated at the end of the growth season
$H$	Henry's law constant ( $Pa\ m^3\ mol^{-1}$ )	$S_{c(R)}$	$S_c$ for the reference scenario calculated at the end of the growth season
$H'$	Dimensionless Henry's law constant	$S_i$	Layer specific chemical storage ( $ng\ m^{-2}$ )
$I$	Canopy interception (m)	$t_a$	Air temperature ( $^{\circ}C$ )
$I_{max}$	Maximum canopy interception (m)	$t_l$	Leaf temperature ( $^{\circ}C$ )
$k_{ac}$	Overall air canopy conductance ( $m\ s^{-1}$ )	$v_d$	Net deposition velocity ( $m\ s^{-1}$ )
$k_b$	Boundary layer conductance ( $m\ s^{-1}$ )	$U$	Wind speed ( $m\ s^{-1}$ )
$k_l$	Conductance within the leaf tissue ( $m\ s^{-1}$ )	$V_r$	Volume of leaf reservoir per unit of ground surface ( $m^3\ m^{-2}$ )
$k_{ra}$	Overall conductance between air and leaf reservoir ( $m\ s^{-1}$ )	$V_s$	Volume of leaf surface per unit of ground surface ( $m^3\ m^{-2}$ )
$k_{sa}$	Overall conductance between air and leaf surface ( $m\ s^{-1}$ )	$x'$	Normalized values of wind speed, temperature or rainfall. $x'=x/x_{max}$
$k_{st}$	Stomatal conductance ( $m\ s^{-1}$ )	$\delta$	Numerical coefficient used in Eqn. SI9
$k_h^*$	Chemical-specific conductance from free advection ( $m\ s^{-1}$ )	$\Delta H_{H_2O}$	enthalpy of evaporation for water at $15^{\circ}C$ ( $kJ\ kg^{-1}$ )
		$\Phi_R$	Transformation function for rainfall
		$\Phi_T$	Transformation function for temperature
		$\Phi_U$	Transformation function for wind speed

### Text SI1. Canopy interception of rainfall

The hydrological model calculates  $r$  at each canopy layer  $i$  as a function of layer-specific canopy interception per unit area ( $I_i$ , m) (namely, the volume of rain water per unit of ground area which can be intercepted by the foliage), as follows:

$$\begin{aligned} r_i &= r_0 - \sum_1^{i-1} I_i \cdot L_i & \text{if } L_i < 1 \\ r_i &= r_0 - \sum_1^{i-1} I_i & \text{if } L_i \geq 1 \end{aligned} \quad \text{SI1)}$$

where  $r_0$  (m) is the rainfall at the reference elevation.  $I_i$  is calculated as a function of time as the balance between rainfall intensity  $\bar{r}$  ( $\text{m s}^{-1}$ ) and the wet latent heat flux of each layer  $E_i$  (calculated by the canopy model [1]) as follows:

$$I_i = \int \left[ \frac{E_i}{\Delta H_{H_2O}} \cdot 10^{-6} - \bar{r}_i \right] dt \quad \text{SI2)}$$

with  $0 < I_i < I_{max}$

and  $\bar{r}_i = \frac{1}{dt} r_i$

where  $I_{max}$  is the maximum thickness of the water film that can be stored on the surface of the leaf (assumed to be  $10^{-4}$  m) and  $\Delta H_{H_2O}$  ( $\text{kJ kg}^{-1}$ ) is the enthalpy of phase change for water at  $15^\circ\text{C}$ . The numerical coefficient is needed to set consistent units.

## Text SI2. Gaseous wet deposition scheme

Wet deposition to canopy of gaseous chemicals dissolved in water drops is treated based on the rainfall interception scheme and the definition of  $r$  and  $I$  described in Text SI1. The initial condition assumes partitioning equilibrium between air and the raindrops above the forest canopy. After impaction with foliage, instantaneous re-equilibration of the chemical present in the volume  $r_0 L_0$  of rainwater that effectively impacts the uppermost canopy layer ( $i=0$ ) was assumed to occur.

We present in the following section a mathematical description of the wet deposition scheme applicable in the case  $L_i < 1$  in each layer of the canopy.

The exchange flux  $F_{ws}$  between rain and leaf surface at  $i=0$  can be described per each time step as follows:

$$F_{ws} = r_0 L_0 (C_{w(0)} - C_{w(0)}^*) \quad \text{SI3)}$$

where  $C_{w(0)}$  is the concentration of the chemical in rainwater before impaction with the canopy in the uppermost layer ( $i=0$ ) and  $C_{w(0)}^*$  is the concentration in the rainwater after impaction and re-equilibration with the leaf surface.

Equation SI3 can be rewritten expressing  $C_w$  and  $C_w^*$  as a function of  $C_a$  and  $C_s$ , respectively, as follows:

$$F_{ws} = r_0 L_0 \left( \frac{C_a}{H'} - \frac{C_{s(0)}}{K_{SA}} \right) \quad \text{SI4)}$$

where  $H'$  is the dimensionless Henry's law constant and  $K_{ow}$  is the equilibrium octanol/water partition coefficient.

In order to calculate  $F_{ws}$  for  $i=1 \dots 10$  it is necessary to know the value of  $C_w$  in each layer  $i$ . This value can be expressed in the following form:

$$C_{w(i)} = \frac{A + B}{r_i} \quad \text{SI5)}$$

where  $A$  is the amount (pg) of chemical in the throughfall (i.e., the volume of rainwater per unit of ground surface that did not impact the foliage in the overlying layer  $i-1$ ), and  $B$  is the amount of chemical in the fraction of water that impacted the foliage in  $i-1$  but that was not intercepted.  $A$  and  $B$  for each layer  $i$  are parameterized as follows:

$$A = C_{w(i-1)} \cdot r_{i-1} (1 - L_{i-1}) \quad \text{SI6)}$$

and

$$\begin{aligned} B &= C_{w(i-1)}^* (r_{i-1} L_{i-1} - I_{i-1}) & \text{with } (r_{i-1} L_{i-1} - I_{i-1}) > 0 \\ &\text{or} & \\ B &= 0 & \text{with } (r_{i-1} L_{i-1} - I_{i-1}) \leq 0 \end{aligned} \quad \text{SI7)}$$

where:

$$C_{w(i-1)}^* = \frac{C_{S(i-1)}}{K_{SA}} \quad \text{SI8)}$$

**Table SI2.** Selected LM vegetation input parameters.

	<b>Value</b>	<b>Units</b>
<b>Max leaf area index (<math>L_c</math>)</b>	5	-
<b><math>L_c</math> growth rate</b>	0.05	$\text{d}^{-1}$
<b><math>SLA</math></b>	0.022	$\text{m}^2 \text{g}^{-1}$
<b>Max canopy biomass (<math>LAI/SLA</math>)</b>	227	$\text{g m}^{-2}$
<b>Max <math>L_1</math> (Max <math>L</math> of Layer 1)</b>	0.6	-
<b>Max <math>L_2</math></b>	0.75	-
<b>Max <math>L_3</math></b>	0.75	-
<b>Max <math>L_4</math></b>	0.7	-
<b>Max <math>L_5</math></b>	0.6	-
<b>Max <math>L_6</math></b>	0.5	-
<b>Max <math>L_7</math></b>	0.4	-
<b>Max <math>L_8</math></b>	0.3	-
<b>Max <math>L_9</math></b>	0.2	-
<b>Max <math>L_{10}</math></b>	0.15	-

**Text SI3. Method for the accuracy test**

In order to test model accuracy we compared predicted  $C_c$  with the seasonal  $C_c$  trends of PCBs measured in an alpine broad-leaf deciduous forest [2] by reconfiguring the scenario for leaf appearance and gaseous concentration data. This forest has similar characteristics as the Harvard Forest in terms of vegetation parameters, climate and seasonality. Like Harvard Forest, the alpine forest is a mixed deciduous stand with net prevalence of broad-leaved species [2, 3] and  $L \approx 5$ . Seasonally-averaged temperature ( $14^\circ\text{C}$ ), minimum ( $-2.5^\circ\text{C}$ ) and maximum ( $23^\circ\text{C}$ )  $t_a$  were close to values for Harvard Forest in 2001 ( $14^\circ\text{C}$ ,  $-4^\circ\text{C}$  and  $24^\circ\text{C}$ , respectively). Cumulative rainfall between April and the end of October was 424 mm for the alpine site and 584 mm at Harvard Forest. Gaseous concentrations measured *in situ* in the alpine forest site were used as inputs to the model [2].  $K_{OA}$ ,  $H$  and enthalpies of phase change reported by Li et al. Ref. 14.



**Text SI4. Method for the assessment of biological and climatic forcing of air-canopy exchange of chemicals**

In order to assess the influence of climatic controls on air-canopy exchange, input data for  $t_a$ ,  $U$  and  $r$  from the reference Harvard Forest site were manipulated by means of transformation functions ( $\Phi_T$ ,  $\Phi_U$  and  $\Phi_r$ , respectively). These functions were designed to produce smooth translations of the mean seasonal values and were of the form

$$\Phi(x) = x + \delta \cdot P(x') \quad \text{SI9)}$$

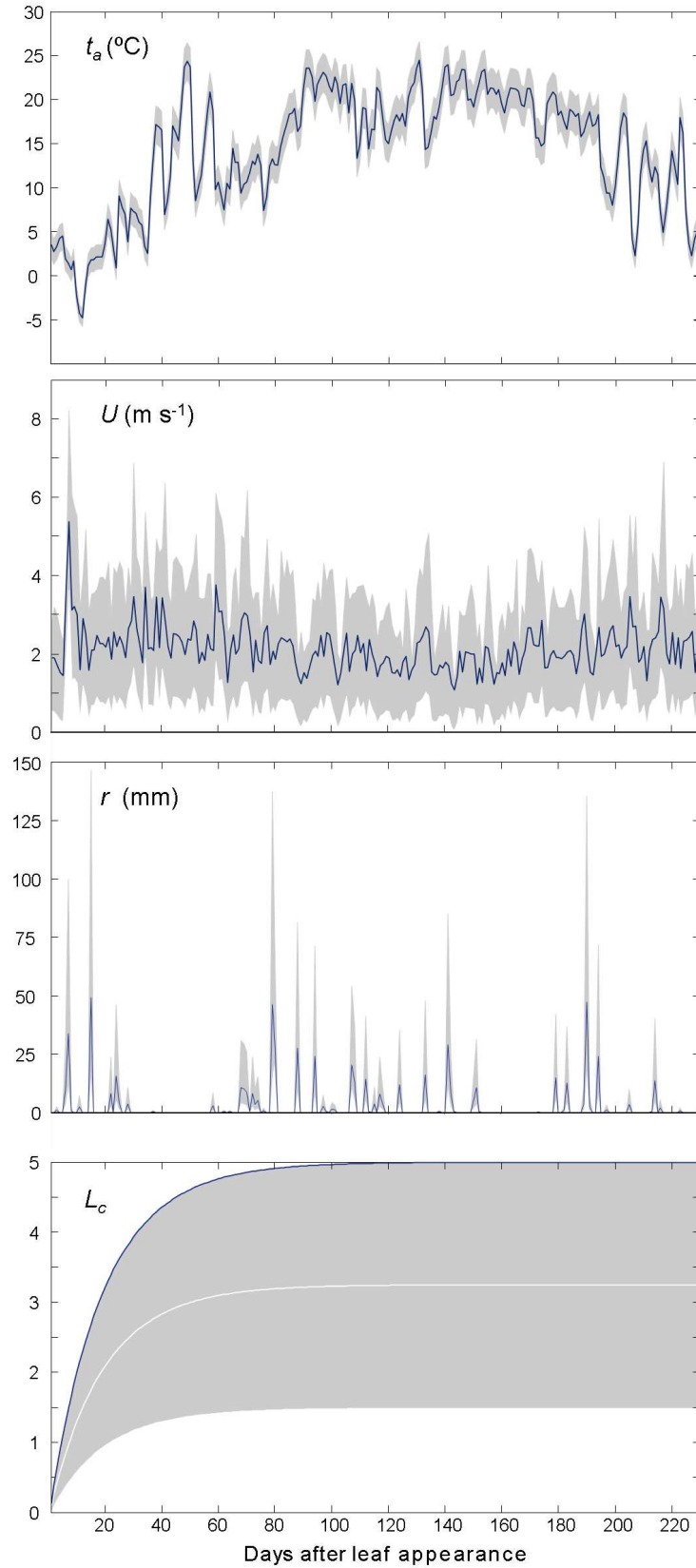
where  $x$  is the experimental value of the selected parameter,  $\delta$  is a numerical coefficient the sign and absolute value of which determine the direction and amplitude of the translation, respectively, and  $P(x')$  is a function of the normalized value of  $x$ , (being  $x' = x/x_{\max}$ ) defined as  $P(x') = ax'^3 + bx'^2 + cx' + d$ . Table SI3 reports the parameterizations of  $P(x')$ . This weighting method produced relatively small shifts in the measured reference values of the variables such that higher translation amplitude for values in the median range, intermediate translation amplitude for values in the highest range, and lower or no translation for values in the lower range of the variable were obtained. The approach prevented the generation of unlikely “extreme” meteorological events when shifting upward values of  $t_a$ ,  $U$  or  $r$ , and unrealistically frequent situations of  $U$  close to 0. In addition, it preserved covariance among the parameters. In this way a range of different climatic scenarios were produced by varying the parameters one by one, and realistic artificial scenarios used to conduct a sensitivity analysis were produced. To investigate biological controls on air-canopy exchange, the effect of varying canopy biomass,  $L_c$ , was investigated. The ranges defined by these transformations are represented in Figure SI2.

The sensitivity analysis was performed by using the transformed arrays for  $t_a$ ,  $U$ ,  $r$  and  $L$  as independent variables to define a factorial experimental design aimed at investigating the influence of each factor on  $S_c$  calculated at the end of the growth season (before litterfall).

The factorial design was conducted by using three levels (lower limit, upper limit and experimental value in Table SI3) for each of the four selected parameters. Each combination of factors obtained from the experimental design was used to run simulations for a range of virtual chemicals having  $\text{Log}(K_{OA})$  between 6.5 and 11 and  $\text{Log}(H)$  between -0.2 and 2.3, which represents a wide range of physicochemical properties of POPs [4].

**Table SI3.** Range in parameters used in the factorial experimental design and parameters for the  $\Phi$ -functions.

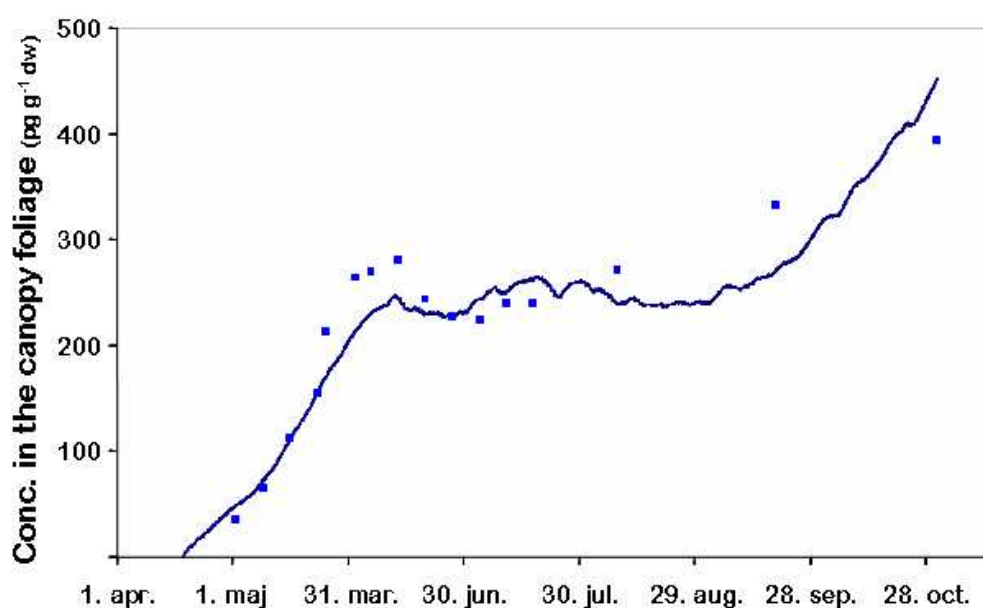
Parameter	Experimental value (Harvard Forest, 2001)	Lower limit	Upper limit	$\Phi$ parameter	
Average air temperature* ( $^{\circ}\text{C}$ )	14.3	12.3	16.3	$a=-1.039$ $c=0.978$ $\delta_{low}=-2.3$	$b=.274$ $d=0.531$ $\delta_{high}=2.3$
Average wind speed* ( $\text{m s}^{-1}$ )	2.1	1.3	4.2	$a=-2.237$ $c=1.817$ $\delta_{low}=-1.6$	$b=0.824$ $d=0$ $\delta_{high}=4$
Total precipitation* (m)	0.584	0.230	1.466	$a=-2.237$ $c=1.817$ $\delta_{low}=-4$	$b=0.824$ $d=0$ $\delta_{high}=10$
LAI ( $\text{m}^2 \text{m}^{-2}$ )	4.9	1.5	5		



**Figure SI1.** Environmental scenario data (daily averaged values). The blue line represents the values for the Harvard Forest reference scenario. Ranges represent low and high values assumed for the sensitivity analysis. The white line in the  $L_c$  plot represents the intermediate value used in the sensitivity analysis.

### Text SI5. Accuracy test results

The model successfully predicted  $C_c$  time trends and absolute values observed for PCBs with  $\text{Log}(K_{OA}) > 10$ . The relative deviation between predicted and measured values was always less than a factor of 3. As an example, Figure SI2 contains a plot of the trend in predicted and measured  $C_c$  for PCB 138. Compounds having lower values of  $\text{Log}(K_{OA})$  reached partitioning equilibrium relatively quickly (timescale of days to weeks), which caused their concentration in the foliage to be highly sensitive to variations in temperature and gaseous concentration, (with a timescale response of  $C_c$  on the order of hours to days), as suggested previously [3, 5]. Due to a lack of experimental input data of adequate resolution for compounds having  $\text{Log}(K_{OA}) \leq 10$  for the alpine scenario [2, 3], it was not possible to test accuracy for these compounds.



**Figure SI2.** Predicted (line) and measured (points) foliage concentration of PCB 138 for the alpine broad-leaved forest [2].

### **Text SI6. Distribution of chemicals in foliage and in the canopy**

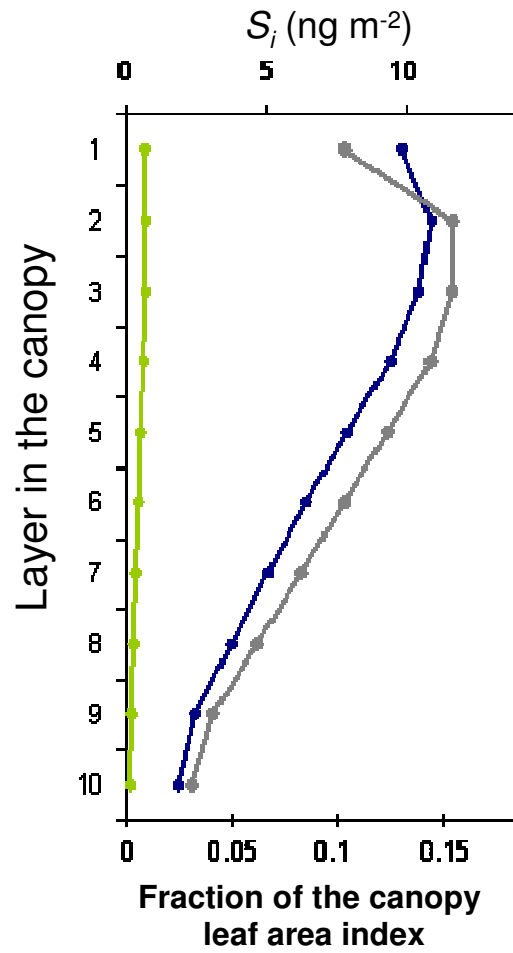
In fully mature foliage, the chemicals stored in the leaf surface represented only less than 10% of the total  $S_c$ . Despite our postulation of a value of the equilibrium partition coefficient of a factor eight higher for the surface compared to the reservoir, its storage capacity is hampered by its small compartment size, as previously observed by Moeckel et al. [6] for various plant species.

In Figure SI3,  $S_c$  vertical distribution of chemicals reflects the distribution of biomass with height, especially in mid-summer and toward the end of the season, when foliage capacity for chemicals is closest to being reached.

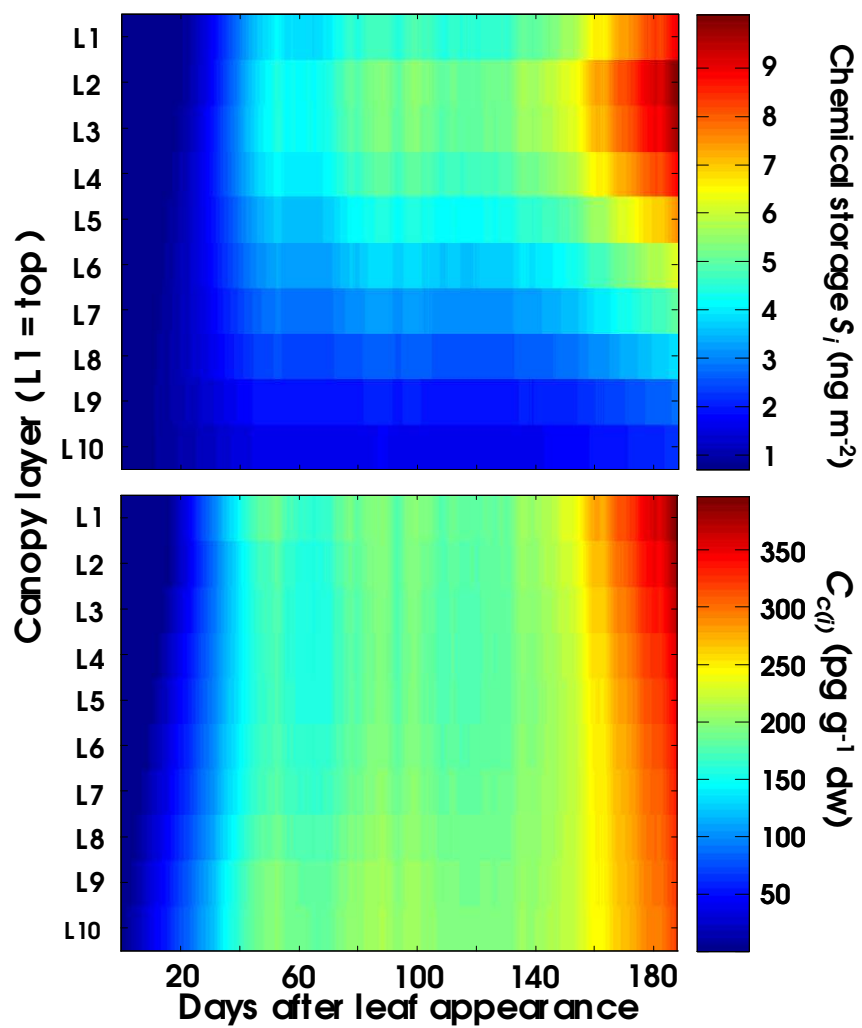
The gaseous air-leaf surface exchange followed by diffusion from cuticular waxes into the leaf reservoir represented the principal pathway for chemical accumulation. Direct uptake in the leaf reservoir through stomata had a limited effect on leaf exposure for chemicals with values of  $\text{Log}(K_{OA})$  and  $\text{Log}(H)$  in the selected range. The stomatal conductance typically represented about 20-30% of the total air-canopy conductance. In addition, we assumed stomata to be present on the lower side of the foliage only.

The model described here attempted to illustrate, for the first time, the vertical distribution of semivolatile chemicals in the foliage of a multi-layered canopy. For illustrative purposes, predicted  $C_{c(i)}$  and  $S_i$  for PCB 138 are shown in Figure SI4 vs. time. Differences in  $C_{c(i)}$  among layers of up to a factor of 3 in the early phase of leaf growth were observed due to the differing times of leaf appearance (and therefore exposure time) at different heights in the canopy. In the upper layers of the fully mature canopy, the relative variation in foliage concentration with height was less than a factor of 0.3.

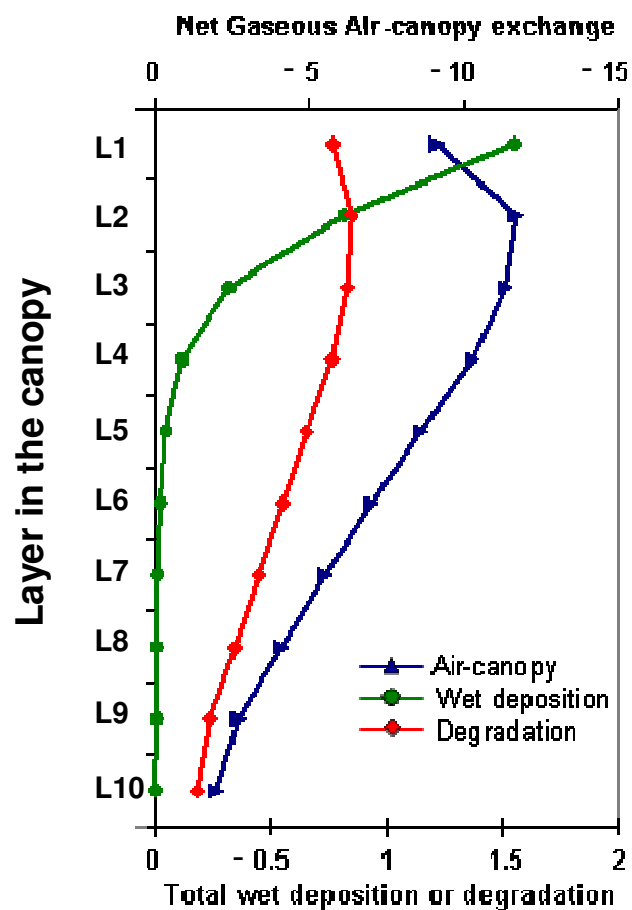
Figure SI5 shows the contribution of the yearly integrated fluxes, namely,  $F_{ca}$ , overall degradation fluxes ( $F_{ds} + F_{dr}$ ) and  $F_{wet}$ , in determining the value of  $S_c$  at the end of the season. The example refers to calculations performed for PCB 153. Yearly integrated wet deposition fluxes of dissolved chemicals typically accounted for less than 5% of total  $S_c$  only. The model demonstrated the contribution of canopy interception in limiting or delaying soil exposure to hydrophobic chemicals in rain. Typically, for compounds having  $6.5 < \text{Log}(K_{OA}) < 11$  and  $-0.3 < \text{Log}(H) < 2.3$ , between 50% and 100% of the seasonally integrated deposition of chemicals dissolved in rain was intercepted in the canopy within  $L > 3$ . Figure SI6 demonstrates the predicted canopy interception efficiency (as intercepted percentage of total wet deposition) as a function of leaf area index  $L$ .



**Figure SI3.** Distribution of  $L$  (grey line),  $S_i$  in the leaf reservoir (blue line),  $S_i$  in leaf surface (green line) vs. elevation at the end of the season.

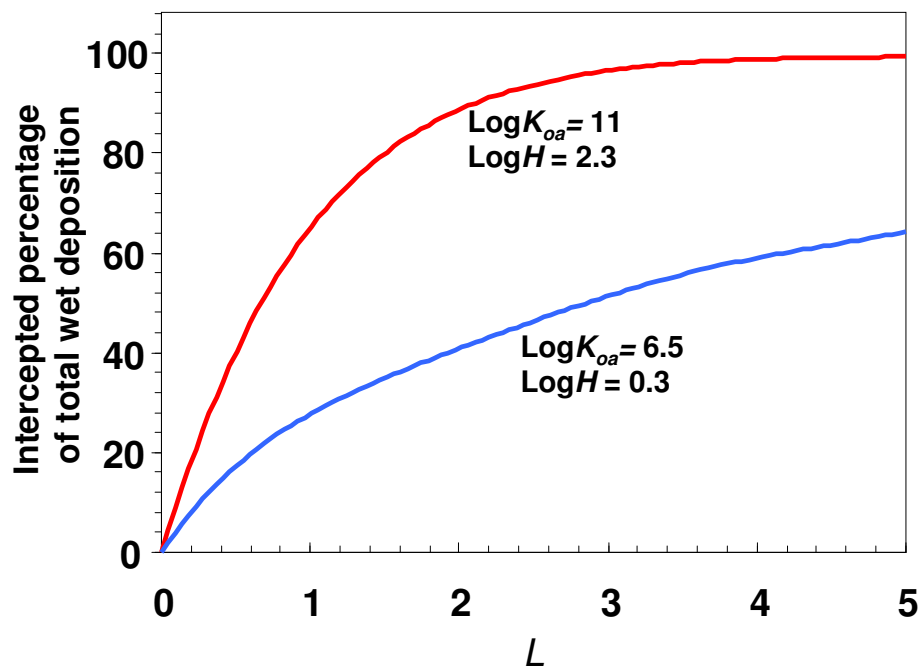


**Figure SI4.** Predicted vertical distribution of  $S_i$  and  $C_{c(i)}$  for PCB 138 vs. time.

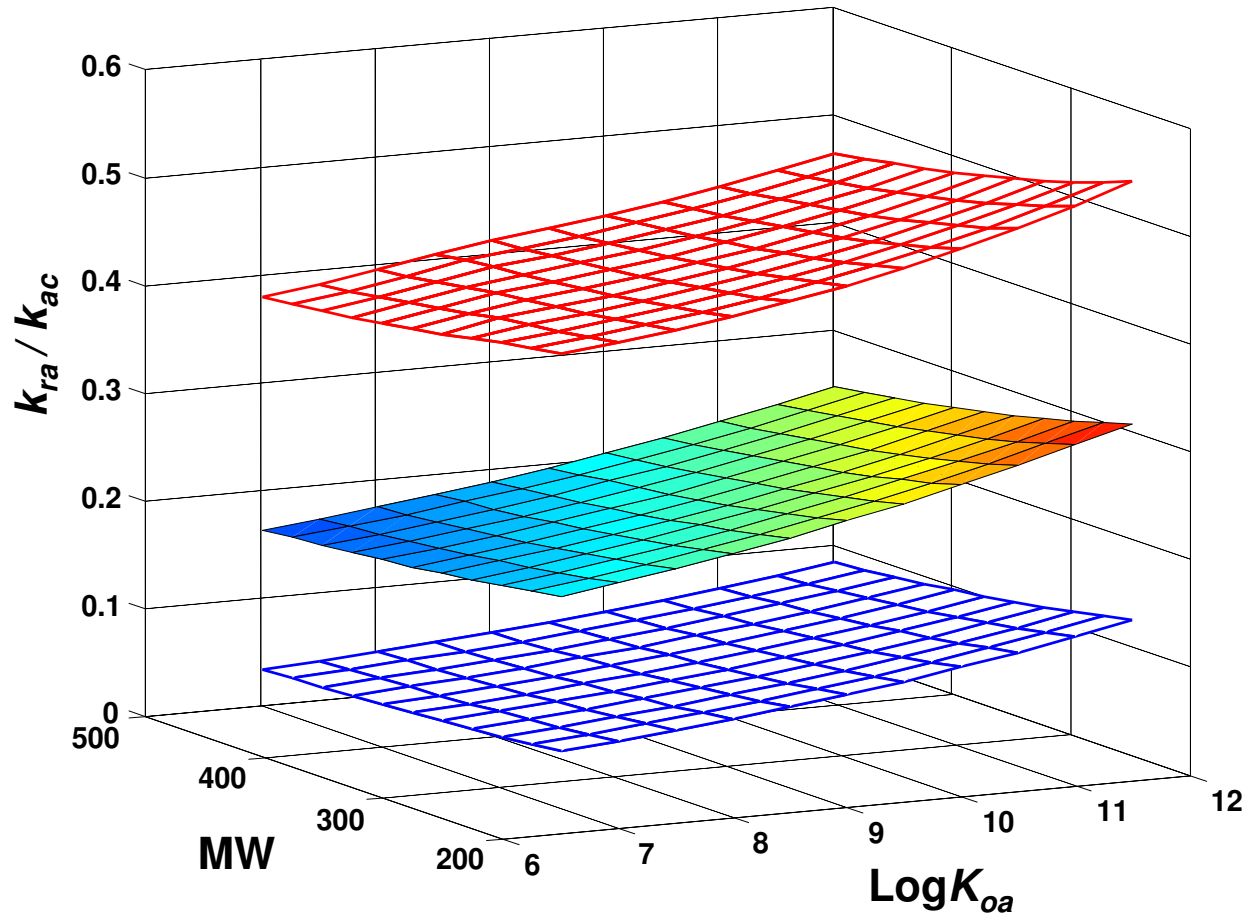


**Figure SI5.** Absolute values of the yearly integrated fluxes (ng m<sup>-2</sup> season<sup>-1</sup>) of air-canopy net exchange (upper scale), wet gaseous deposition, and degradation (lower scale), layer by layer in the canopy. Calculations refer to PCB 153 assuming  $C_a = 1$  pg m<sup>-3</sup>.

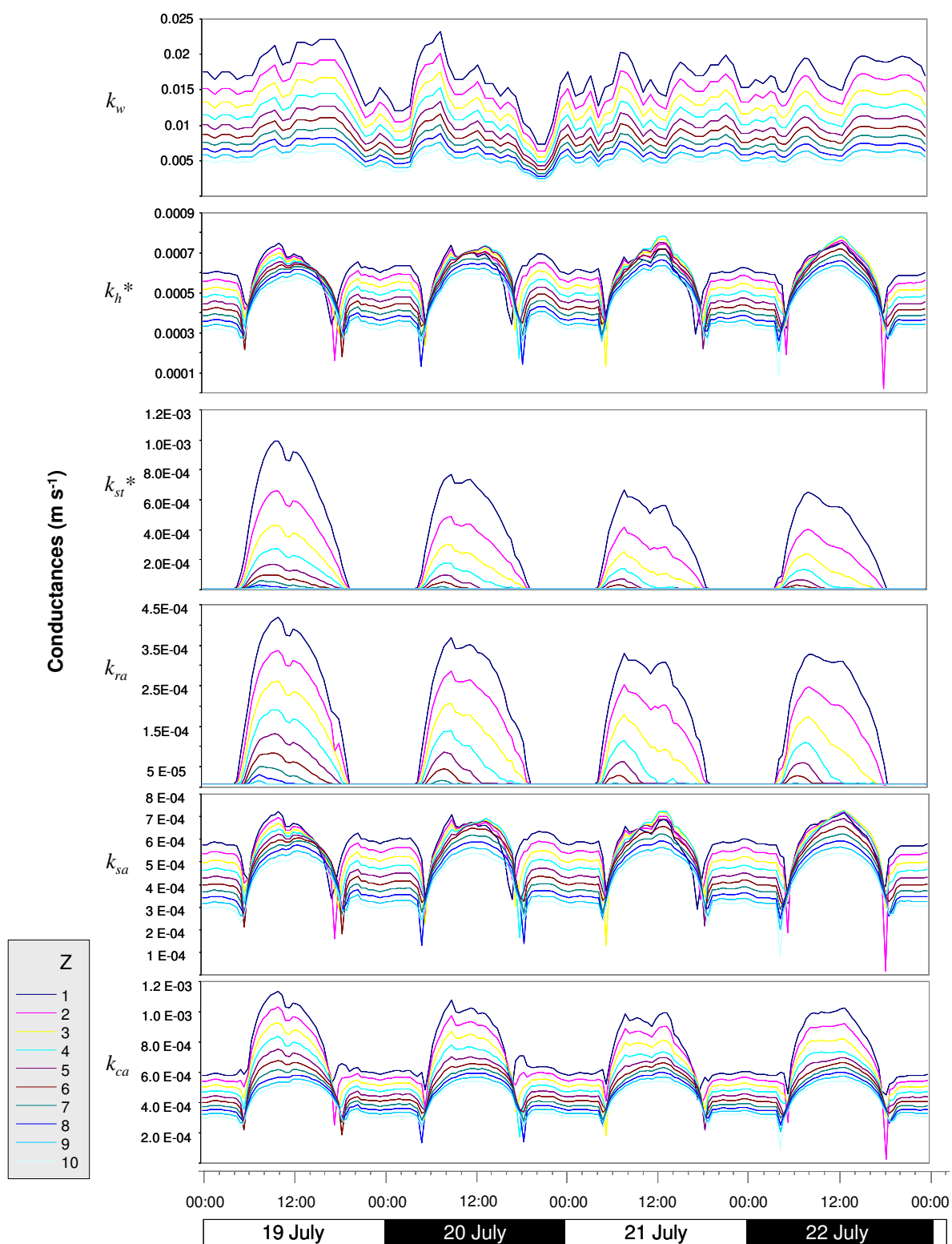




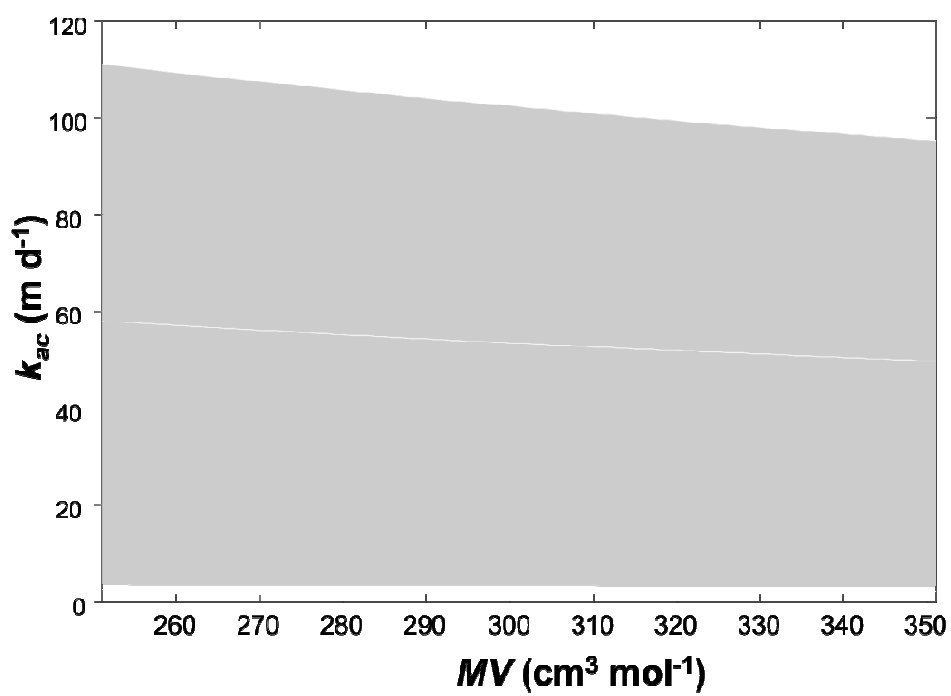
**Figure SI6.** Fraction of the total seasonal deposition intercepted by forest canopy as a function of leaf area index  $L$ . The lines refer to two extreme chemicals within the ranges of physical chemical properties considered in this study.



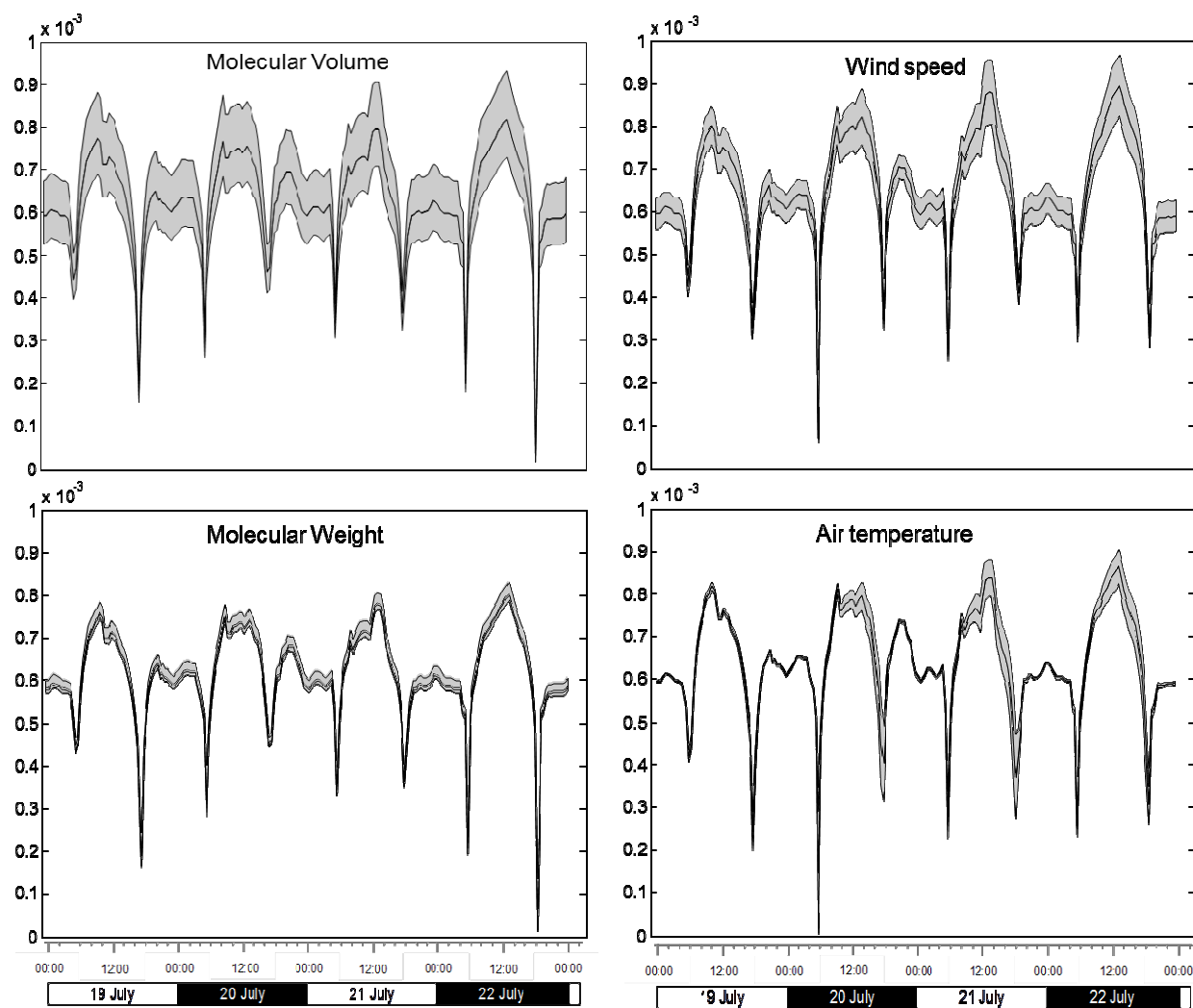
**Figure SI7.** Relative contribution of the conductance through stomata (namely the air-reservoir mass transfer coefficient,  $k_{ra}$ ) to the overall mass transfer coefficient between air and canopy ( $k_{ac}$ ) as a function of  $\text{Log}(K_{oa})$  and  $MW$ . The figure is for the mean seasonal value and ranges of variability.



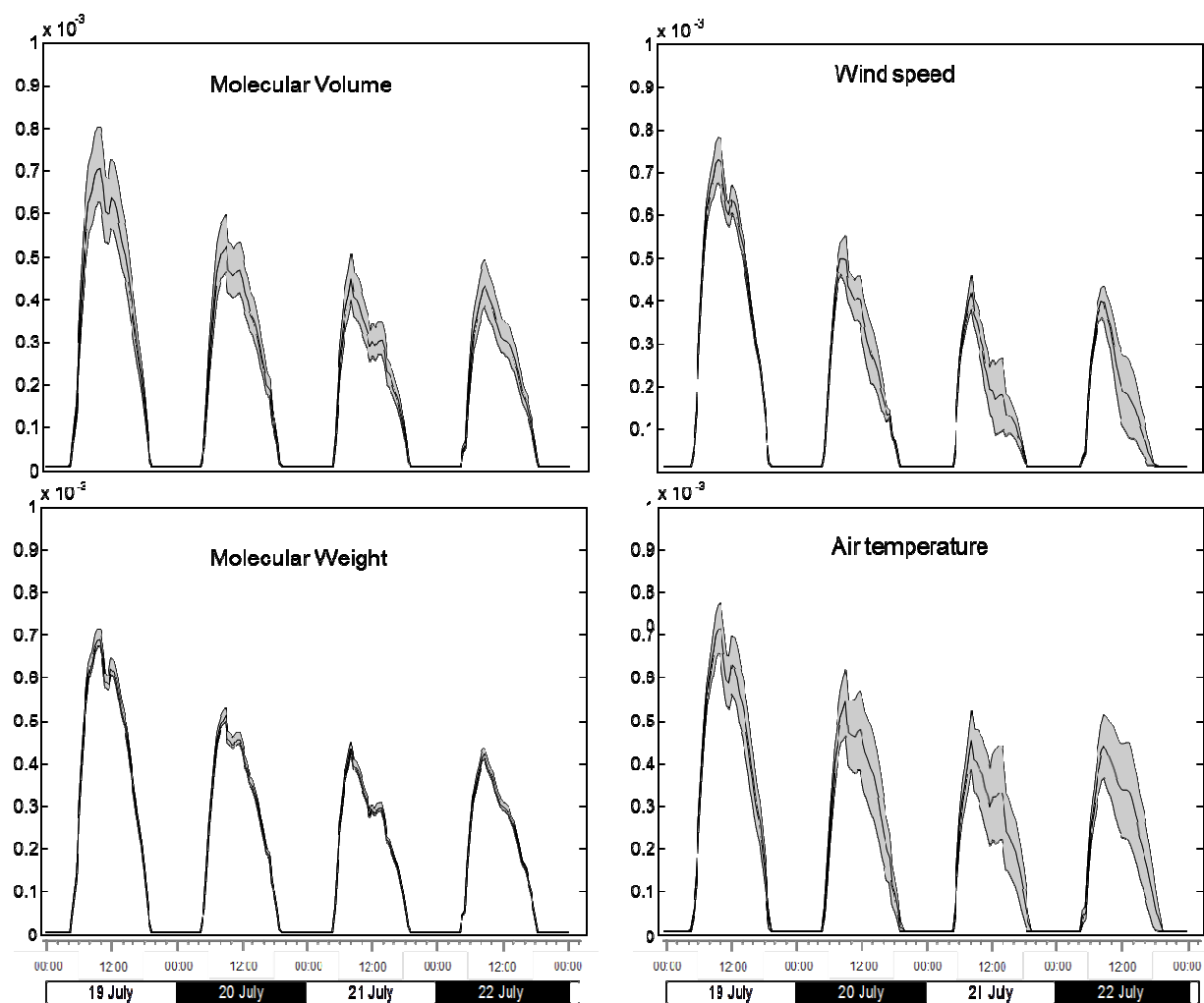
**Figure S18.** Conductance values for a compound with  $\text{Log}(K_{OA}) = 9.5$  and  $\text{MW} = 350 \text{ g mol}^{-1}$  during the period between 19 July and 22 July. Sub-plots show the conductance values for the different layers in the canopy.  $Z=1$  is the top layer.



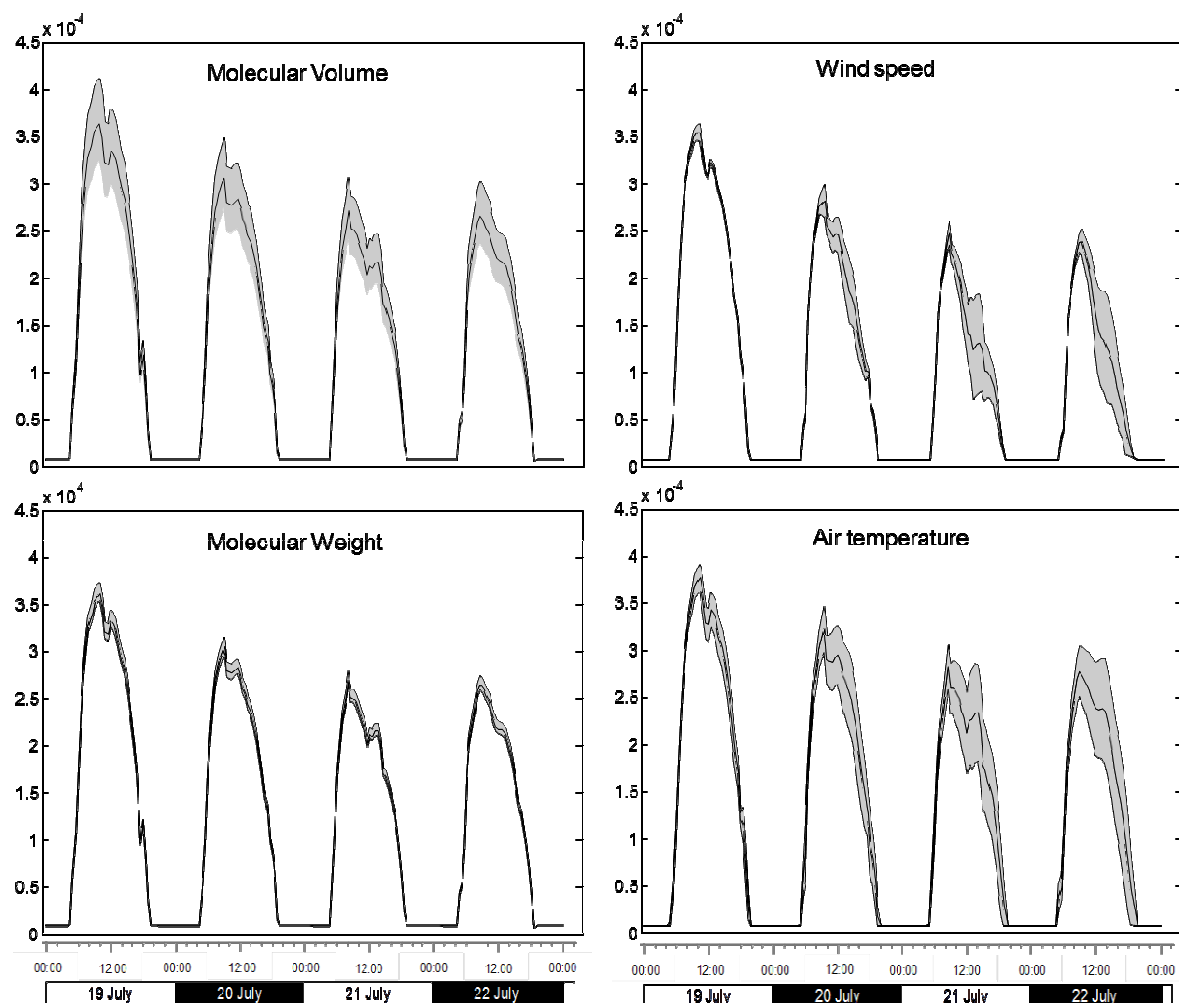
**Figure S19.** Mean seasonal value (white line) and ranges of variability of the overall air canopy mass transfer coefficient  $k_{ac}$  as a function of molecular volume.



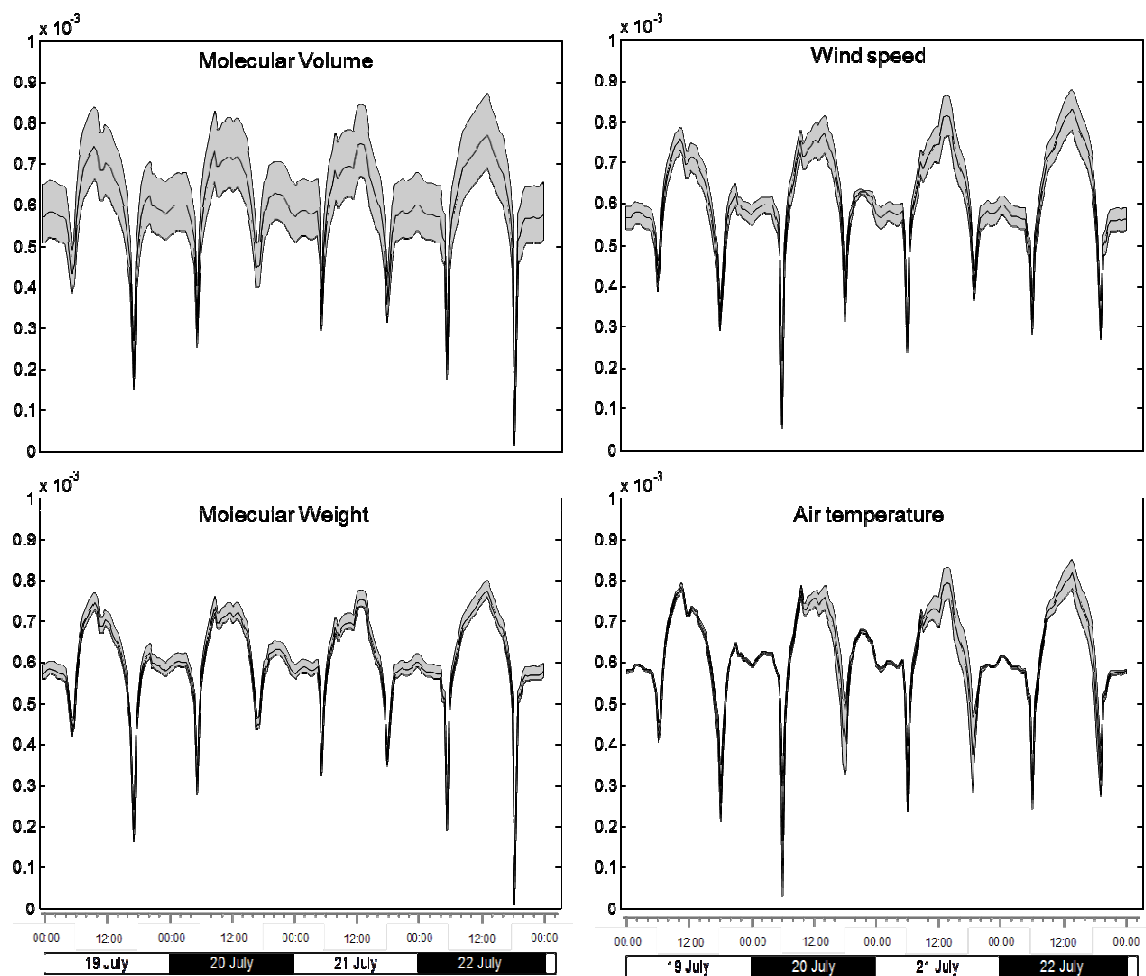
**Figure SI10a.** Sensitivity of canopy-averaged  $k_h$  to varying  $MV$  (range 240-350  $\text{cm}^3 \text{mol}^{-1}$ ),  $MW$  (range 200-450  $\text{g mol}^{-1}$ ), wind speed (range represented in Figure SI1) and air temperature (range represented in Figure SI1).



**Figure SI10b.** Sensitivity of  $k_{st}$  to varying  $MV$  (range 240-350  $\text{cm}^3 \text{mol}^{-1}$ ),  $MW$  (range 200-450  $\text{g mol}^{-1}$ ), wind speed (range represented in Figure SI1) and air temperature (range represented in Figure SI1).

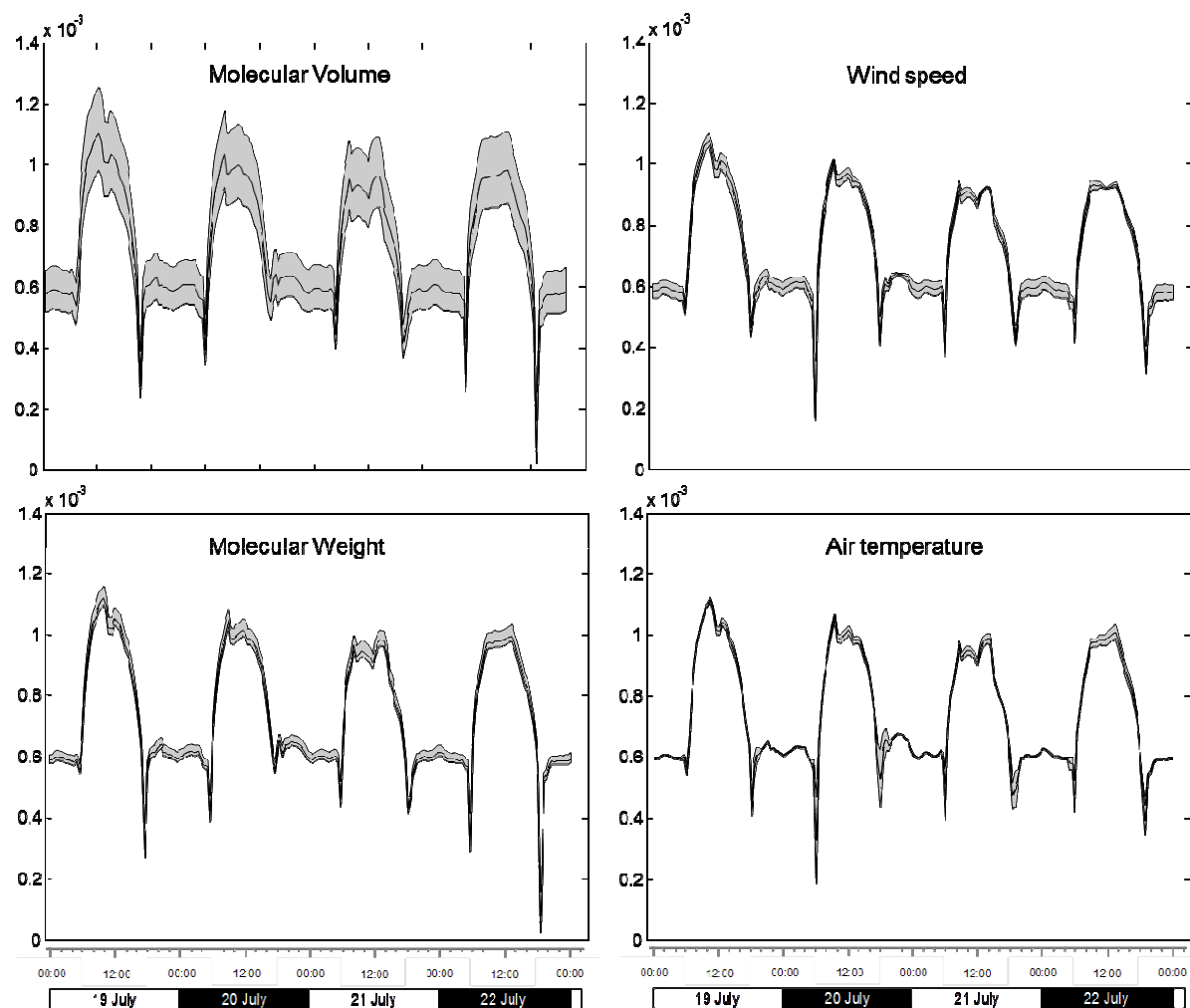


**Figure SI10c.** Sensitivity of  $k_{ra}$  to varying  $MV$  (range  $240\text{-}350\text{ cm}^3\text{ mol}^{-1}$ ),  $MW$  (range  $200\text{-}450\text{ g mol}^{-1}$ ), wind speed (range represented in Figure SI1) and air temperature (range represented in Figure SI1).

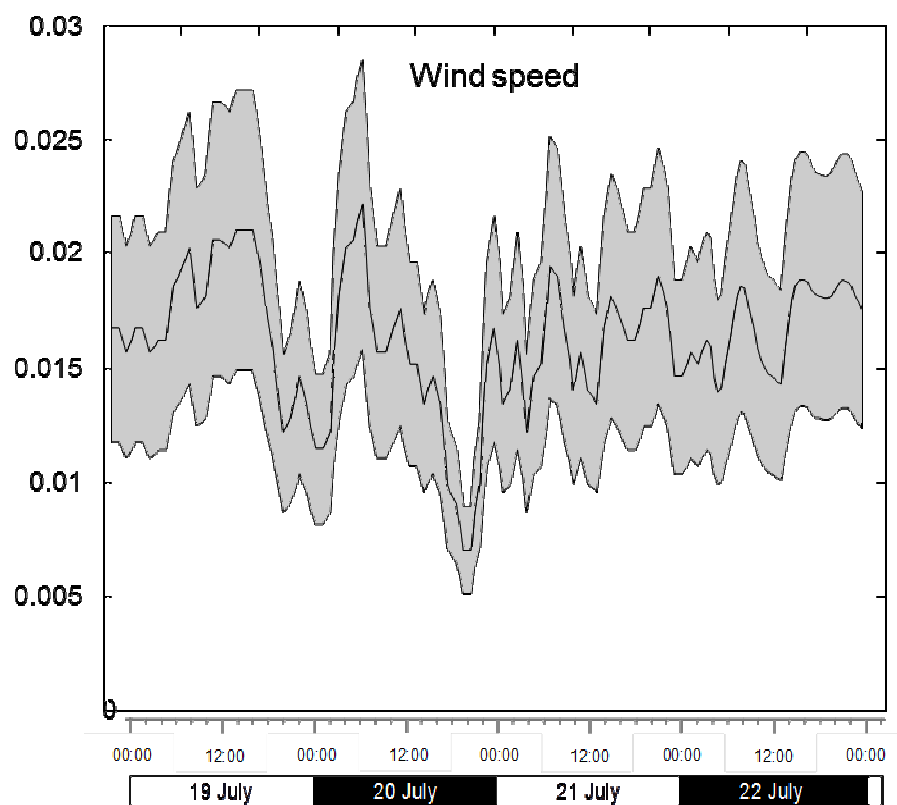


**Figure SI10d.** Sensitivity of  $k_{sa}$  to varying  $MV$  (range 240-350  $\text{cm}^3 \text{mol}^{-1}$ ),  $MW$  (range 200-450  $\text{g mol}^{-1}$ ), wind speed (range represented in Figure SI1) and air temperature (range represented in Figure SI1).





**Figure SI10e.** Sensitivity of  $k_{acHHHH}$  to varying  $MV$  (range 240-350 cm<sup>3</sup> mol<sup>-1</sup>),  $MW$  (range 200-450 g mol<sup>-1</sup>), wind speed (range represented in Figure SI1) and air temperature (range represented in Figure SI1).



**Figure SI10f.** Sensitivity of  $k_w$  to varying wind speed (range represented in Figure SI1).

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**Table SI4.** Regression analysis parameterization results for the function:  
 $\text{Log}S_c = a \cdot t_a + b \cdot L + c \cdot r + d \cdot U + e$  for compounds having the indicated combinations of  $\text{Log}(K_{OA})$  and  $\text{Log}(H)$ . The coefficients for the parameters with no statistically significant influence on  $\log(S_c)$  are not reported.

	<b>LogK<sub>OA</sub>=6.5 LogH=-0.3</b>	<b>LogK<sub>OA</sub>=6.5 LogH=2.3</b>	<b>LogK<sub>OA</sub>=9 LogH=-0.3</b>	<b>LogK<sub>OA</sub>=9 LogH=2.3</b>	<b>LogK<sub>OA</sub>=11 LogH=-0.3</b>	<b>LogK<sub>OA</sub>=11 LogH=2.3</b>
<b>a</b>	-0.032	-0.034	-0.03	-0.03	-0.013	-0.018
<b>b</b>	0.136	0.147	0.142	0.147	0.135	0.147
<b>c</b>	0.033	-	0.018	-	0.056	-
<b>d</b>	-0.009	-	-	-	-	-
<b>e</b>	1.784	1.757	3.583	3.56	4.25	4.27
<b>adj R<sup>2</sup></b>	0.98	0.99	0.99	0.99	0.98	0.99
<b>P</b>	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
<b>St Err</b>	0.034	0.025	0.024	0.024	0.029	0.025

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