

## Supplementary material

### Exploring phosphate effects on leaf flammability using a physical chemistry model

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### Model equations

For the equations that follow, definitions of symbols and their units are listed in Table S2, parameter values and associated literature sources appear in Table S3.

Decomposition reactions and evaporation of water were all governed by rate constants  $k$  of the form:  $k = A \exp[-E/(RT)]$ . Here  $A$  and  $E$  are two kinetic parameters, the pre-exponential constant and the activation energy, which are specific to each reaction.  $R$  is the universal gas constant and  $T$  is temperature. Changes in concentrations were governed by Eqns S1–S4:

$$\frac{\partial \rho_m}{\partial t} = -\rho_m k_m \quad (\text{S1})$$

$$\frac{\partial \rho_{\text{cel}}}{\partial t} = -(\rho_{\text{cel}} - \rho_{\text{ash}})k_v - (\rho_{\text{cel}} - \rho_{\text{ash}})k_c \quad (\text{S2})$$

$$\frac{\partial \rho_{\text{char}}}{\partial t} = X(\rho_{\text{cel}} - \rho_{\text{ash}})k_c - m_{\text{ox}} \quad (\text{S3})$$

$$\frac{\partial \rho_{\text{tar}}}{\partial t} = (\rho_{\text{cel}} - \rho_{\text{ash}})k_v - (\rho_{\text{tar}} - \rho_{\text{res}})k_{\text{ev}} \quad (\text{S4})$$

where  $\rho_m$ ,  $\rho_{\text{cel}}$ ,  $\rho_{\text{char}}$  and  $\rho_{\text{tar}}$  are densities of water, cellulose, char and tar, and  $\partial \rho / \partial t$  is the rate change with time. All the reaction rates scaled linearly with substrate density. The mineral content of the leaf was included in  $\rho_{\text{cel}}$  as an inert component  $\rho_{\text{ash}}$ . Likewise a small fraction of the tar products  $\rho_{\text{res}}$  was unavailable for vaporisation and remained as a residue. Factor  $X$  represented the fraction of charring products that were solid (c.f. light gases).  $m_{\text{ox}}$  governed the rate of char oxidation. The effect of adding phosphate was captured in altered activation energies and pre-exponential constants for the calculation of  $k_v$ ,  $k_c$  and  $m_{\text{ox}}$ .

Char oxidation can be limited either by chemical kinetics or by the diffusion of oxygen through the leaf boundary layer. This dual control can be represented as a circuit in which the oxidation rate passes

through two resistances (Boonmee and Quintiere 2005). The overall rate  $m_{\text{ox}}$  is then given by Eqns S5–S7:

$$m_{\text{ox}} = \frac{Y_{\text{OA}}}{R_{\text{diff}} + R_{\text{kin}}} \quad (\text{S5})$$

$$R_{\text{diff}} = \frac{(Y_{\text{OA}} - Y_{\text{OS}})c_g L}{2h \ln(1 + Y_{\text{OA}} / \nu)} \quad (\text{S6})$$

$$R_{\text{kin}} = \frac{1}{\rho_{\text{char}} k_{\text{ox}}} \quad (\text{S7})$$

where  $R_{\text{diff}}$  and  $R_{\text{kin}}$  are the resistances offered by diffusion and reaction kinetics.  $Y_{\text{O}}$  is the mass fraction of oxygen in air, in the ambient environment ( $Y_{\text{OA}}$ ) and at the leaf surface ( $Y_{\text{OS}}$ ).  $c_g$  is gas heat capacity,  $\nu$  is the stoichiometric mass of oxygen required to oxidise 1 g of carbon to  $\text{CO}_2$ ,  $L$  is leaf thickness and  $h$  is a heat transfer coefficient. The treatment in Eqns S5–S7 was drawn from a model of surface char oxidation in wood (Boonmee and Quintiere 2005), with three modifications. Since leaves are thin and structured to promote internal gas exchange, we allowed char oxidation to occur throughout the leaf thickness, rather than on a progressively receding surface. We neglected the effect of flammable gases exiting the leaf in hindering the inward diffusion of oxygen. Finally we assumed that the oxygen concentration at the leaf surface  $Y_{\text{OS}}$  was small compared to the ambient level, for the purpose of calculating the diffusion gradient associated with  $R_{\text{diff}}$ .

Char oxidation proceeds more slowly for chars derived from phosphate-rich cellulose (Shafizadeh *et al.* 1982). For the phosphate concentration considered here (0.67% w/w), the resulting rate constant  $k_{\text{ox}}$  measured using thermogravimetry was the same as for chars derived from untreated cellulose oxidising at a temperature 50°C cooler (Shafizadeh *et al.* 1982). Our model imposes an effective temperature =  $T - 50$  for calculation of  $k_{\text{ox}}$  for phosphate-rich material.

Fuel temperature was governed by the following energy balance:

$$\left[ c(\rho_{\text{cel}} + \rho_{\text{char}} + \rho_{\text{tar}}) + c_m \rho_m \right] \frac{\partial T}{\partial t} = \quad (\text{S8})$$

$$\begin{array}{cccccc} Q_m \rho_m k_m & + & Q_{\text{ev}} (\rho_{\text{tar}} - \rho_{\text{res}}) k_{\text{ev}} & + & Q_c (\rho_{\text{cel}} - \rho_{\text{ash}}) k_c & + & Q_{\text{ox}} m_{\text{ox}} & + & \partial / \partial x (\lambda \partial T / \partial x) \\ [1] & & [2] & & [3] & & [4] & & [5] \end{array}$$

On the left in Eqn S8,  $c$  and  $c_m$  are heat capacities of solid material and water, and  $\partial T / \partial t$  is the rate change in temperature with time. On the right side, the first four terms represent rates of heat release associated with endothermic evaporation of water [1], endothermic tar vaporisation [2], exothermic charring [3] and exothermic oxidation of chars [4]. Throughout,  $Q$  is the heat of reaction. Term [5]

represents conduction of heat through the leaf, where  $\lambda$  is thermal conductivity and  $x$  is the distance inward from the surface of the leaf.

### Representation of leaf chemistry and phosphate effects

Our simulations compare the fire behaviour of material containing 0 v. 0.67% phosphate % phosphate of dry WT. This spans 92% of leaf phosphate concentrations recorded in 752 species spread over six continents (Wright *et al.* 2004). In the experiments, and in the model, the chemistry of biomass was simplified to cellulose, water and diammonium phosphate. This compound provides a source of inorganic phosphate, without also supplying metal cations that exert separate retardant effects (Richards and Zheng 1991; Kandola *et al.* 1996; Di Blasi *et al.* 2008). The representation of leaf chemistry can be made more sophisticated by adding kinetics for the decomposition of lignin and hemicellulose; this is demonstrated for the third fire scenario (see *Simulated fire scenarios* in main document).

Some additional fire-retarding actions of phosphate additives were neglected in the model. First, they can act as flame inhibitors (rate constant *flame* in Fig 2); this effect is small at phosphate concentrations typical of untreated biomass (Camino and Costa 1988; Lu and Hamerton 2002; Scarff and Westoby 2008). Second, they can produce a foamed char with thermally insulating properties (Di Blasi *et al.* 2007). This was neglected since we are modelling leaves, which are thin enough that combustion outcomes are insensitive to changes in thermal conductivity. Finally, they can create a viscous coat that reduces the transfer of both heat and oxygen at the fuel surface (Di Blasi *et al.* 2007). This process was not modelled in view of the relatively low phosphate concentrations, thin construction and large internal surface area for gas exchange in leaves.

### Initial and boundary conditions

Model runs began with the leaf at ambient temperature, with a cellulose density of  $\rho_0 = 600 \text{ kg m}^{-3}$  (including a small mineral content  $\rho_{\text{ash}}$ ). A negligible quantity of char and tar were also present at the start, purely for practical reasons relating to speed of computation. The boundary conditions and the initial moisture content varied amongst the five different fire scenarios explored, which are described below. The problem domain was effectively one-dimensional, depicting thermal decomposition through the thickness of the leaf. The temperature boundary condition took this general form:

$$\lambda \frac{\partial T}{\partial x} = \underbrace{(1-z)h(T_a - T)}_{[1]} + \underbrace{\varphi \varepsilon \sigma (T_a^4 - T^4)}_{[2]} + \underbrace{z \zeta h_f (T_f - T)}_{[3]} \quad (S9)$$

Term [1] represents the outward heat flux normal to the leaf surface. Term [2] describes convective heat transfer between the leaf and the ambient environment;  $h$  is a heat transfer coefficient and  $T_a$  is ambient temperature. The variable  $z$  assumes a value of one when ignition has occurred, decreasing to

zero otherwise. So the expression  $(1 - z)$  switches off convective exchange with the ambient air when there is a flame. Term [3] represents radiant heat exchange with the environment, which was typically calculated for a number of surfaces (e.g. furnace walls, approaching wildfire front, ambient laboratory or forest environment), each assigned a configuration factor  $\phi$  representing the view of each surface from the leaf. The emissivity of each surface is given by  $\varepsilon$ , the model assumes unit absorptivity at the leaf surface. The Stefan–Boltzmann constant is signified by  $\sigma$ . Term [4] models heat transfer from the flame to the leaf, where  $h_f$  is a heat transfer coefficient and  $T_f$  is flame temperature. This term is also controlled by the ignition variable  $z$  which increases from zero to one when the supply of tar evaporating from each leaf surface exceeds  $2.5 \text{ g m}^{-2} \text{ s}^{-1}$  (Drysdale 1998). This area-specific supply rate  $\dot{m}''_{\text{tar}}$  of tars to the flame is determined as:

$$\dot{m}''_{\text{tar}} = \frac{1}{2} \int (\rho_{\text{tar}} - \rho_{\text{res}}) k_{\text{ev}} dx \quad (\text{S10})$$

In Eqn 9 Term [4],  $\zeta$  simulates reduced heat transfer from the flame at high rates of mass loss, due to blowing of the flame away from the leaf face, given by:

$$\zeta = \frac{\dot{m}''_{\text{g}} c_{\text{g}} / h_f}{\exp(\dot{m}''_{\text{g}} c_{\text{g}} / h_f - 1)} \quad (\text{S11})$$

$$\dot{m}''_{\text{g}} = \frac{1}{2} \int (\rho_{\text{cel}} - \rho_{\text{ash}}) (k_{\text{v}} + (1 - X) k_{\text{c}}) dx \quad (\text{S12})$$

where  $c_{\text{g}}$  is gas heat capacity and  $\dot{m}''_{\text{g}}$  is the mass flux of tars and light gases from one face of the leaf (Ohlemiller and Shields 1993).

The gas phase reactions were not modelled explicitly. Once ignition occurred, the flame was modelled as a flat sheet with a temperature fixed at 1200 K (Albini *et al.* 1995). Flame height was given by:

$$F = 0.018 \dot{m}''_{\text{tar}} Q_{\text{cel}} w \quad (\text{S13})$$

where  $F$  is flame height,  $Q_{\text{cel}}$  is the heat of combustion of cellulose and  $w$  is the width of the sample. This correlation was developed for flames burning on vertical surfaces (Tsai and Drysdale 2002).

### Parameterisation

To make the model as general as possible, all 28 parameter values were sourced from generic literature values (see Tables S2, S3). The model thus parameterised performed well in predicting ignition times. However it was necessary to tune two parameters to improve flame height predictions; the tuned values were then used throughout all simulations, including prediction of ignition time. These parameters were  $h_f$ , the heat transfer coefficient from the flame to the fuel element; and  $Q_{\text{c}}$ , the exothermicity of char formation. The need to tune  $h_f$  most likely reflects conditions unique to the flame height experiment

(*paper combustion*), where the wire mesh sample holder interfered with heat transfer from the flame to the sample. Tuning of  $Q_c$  implies a shift in the enthalpy of the charring reaction, which can reflect whether, as the char is formed, hydrogen is liberated as  $H_2O$  or  $H_2$  and oxygen as  $H_2O$  or  $CO_2$  (Milosavljevic *et al.* 1996). The value of  $Q_c$  was only important for the behaviour of phosphate treated material, since in untreated samples charring was minimal. The need to tune  $Q_c$  suggests that phosphate addition changes the composition of the light gases evolved during charring.

**Table S1. Effect of phosphate on the maximum gap that can be bridged by flames: model predictions from horizontal fuel gap scenario**

Scenario	Maximum gap (m)	
	0% phosphate	0.67% phosphate
1-m flames, 0.3 mm-thick horizontal leaf	0.41	0.40
1-m flames, 1.0 mm-thick horizontal leaf	0.51	0.49
5-m flames, 0.3 mm-thick horizontal leaf	2.1	2.0
5-m flames, 1.0 mm-thick horizontal leaf	2.6	2.4
1-m flames, 0.3 mm-thick vertical leaf	1.7	1.6
1-m flames, 1.0 mm-thick vertical leaf	1.9 <sup>A</sup>	1.7 <sup>B</sup>
5-m flames, 0.3 mm-thick horizontal leaf	8.4	7.9
5-m flames, 1.0 mm-thick vertical leaf	12.1	11.1

<sup>A</sup>With intermittent flashing at 2.0–2.4 m, but no sustained ignition.

<sup>B</sup>With intermittent flashing at 1.8–2.3 m, but no sustained ignition.

**Table S2. Nomenclature (SI units used throughout)**

$A$	pre-exponential constant ( $s^{-1}$ )
$c$	specific heat ( $J\ kg^{-1}\ K^{-1}$ )
$E$	activation energy ( $J\ mol^{-1}$ )
$F$	flame height (m)
$h$	heat transfer coefficient ( $W\ m^{-2}\ K^{-1}$ )
$H$	height above fire (m)
$H'$	virtual height above fire (m)
$k$	rate constant ( $s^{-1}$ )
$L$	leaf thickness (m)
$m$	overall rate constant ( $s^{-1}$ ) (controlled by diffusion + kinetic rates)
$\dot{m}_g''$	mass flux rate per unit area of leaf surface ( $kg\ m^{-2}\ s^{-1}$ )
$Q$	heat of reaction ( $J\ kg^{-1}$ )
$R$	universal gas constant ( $J\ mol^{-1}\ K^{-1}$ )
$R_{diff}$	resistance due to gas diffusion rate ( $m^3\ kg^{-1}\ s^{-1}$ )
$R_{kin}$	resistance due to kinetic rate ( $m^3\ kg^{-1}\ s^{-1}$ )
$T$	temperature (K)
$t$	time (s)
$w$	sample width (m)
$X$	solid fraction of char products ( $kg\ kg^{-1}$ )
$x$	distance from leaf surface (m)
$Y$	mass fraction in air ( $kg\ kg^{-1}$ )
$z$	pseudo-binary variable signifying ignition (ND)
Greek symbols	
$\epsilon$	emissivity (ND)
$\zeta$	blowing coefficient (ND)
$\lambda$	thermal conductivity ( $W\ m^{-1}\ K^{-1}$ )

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$\nu$	stoichiometric mass of oxygen ( $\text{kg kg}^{-1}$ )
$\rho$	density ( $\text{kg m}^{-3}$ )
$\sigma$	Stefan–Boltzmann constant ( $\text{W m}^{-2} \text{K}^{-4}$ )
$\phi$	configuration factor (ND)
Subscripts	
a	ambient
ash	ash content
c	char production
cel	cellulose
char	char
$\Delta\alpha$	increase from ambient near fire
diff	determined by diffusion rate
ev	tar evaporation
f	flame
g	gas
kin	determined by kinetic rate
OA	oxygen in ambient air
OS	oxygen at leaf surface
ox	char oxidation
res	residue from tar evaporation
tar	tar
v	tar production

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**Table S3. Parameter values and literature sources**

	Symbol	Parameter	Value	Source
Kinetic parameters				
pre-exponential constants	$A_c$	charring	for $P = 0$ , $3.57 \times 10^{11} \text{ s}^{-1}$ for $P = 0.22$ , $5.32 \times 10^{11} \text{ s}^{-1}$	Scarff and Westoby (2008)
	$A_v$	tar production	for $P = 0$ , $1.74 \times 10^{17} \text{ s}^{-1}$ for $P = 0.22$ , $6.41 \times 10^{16} \text{ s}^{-1}$	Scarff and Westoby (2008)
activation energies	$A_{ev}$	tar evaporation	$3.33 \times 10^{11} \text{ s}^{-1}$	Suuberg <i>et al.</i> (1996)
	$A_{ox}$	char oxidation	$1.14 \times 10^4 \text{ s}^{-1} \text{ atm}^{-1}$	Di Blasi <i>et al.</i> (1999)
	$A_m$	water vaporisation	$3.41 \times 10^4 \text{ s}^{-1}$	Gray <i>et al.</i> (2002)
	$E_c$	charring	$162 \times 10^3 \text{ J mol}^{-1}$	Scarff and Westoby (2008)
	$E_v$	tar production	$230 \times 10^3 \text{ J mol}^{-1}$	Scarff and Westoby (2008)
	$E_{ev}$	tar evaporation	$141 \times 10^3 \text{ J mol}^{-1}$	Suuberg <i>et al.</i> (1996)
heats of reaction <sup>A</sup>	$E_{ox}$	char oxidation	$71.8 \times 10^3 \text{ J mol}^{-1}$	Di Blasi <i>et al.</i> (1999)
	$E_m$	water vaporisation	$42 \times 10^3 \text{ J mol}^{-1}$	Gray <i>et al.</i> (2002)
	$Q_{cel}$	combustion of cellulose	$1.71 \times 10^6 \text{ J kg}^{-1}$	Milosavljevic <i>et al.</i> (1996)
	$Q_c$	charring	$7 \times 10^5 \text{ J kg}^{-1}$	Milosavljevic <i>et al.</i> (1996)
	$Q_v$	tar production	$1.75 \times 10^5 \text{ J kg}^{-1}$ (tuned value)	Milosavljevic <i>et al.</i> (1996)
	$Q_{ev}$	tar evaporation	$0 \text{ J kg}^{-1}$	Milosavljevic <i>et al.</i> (1996)
stoichiometric ratios (non-dimensional)	$Q_{ox}$	char oxidation	$-5.38 \times 10^5 \text{ J kg}^{-1}$	Milosavljevic <i>et al.</i> (1996)
	$Q_m$	water vaporisation	$3.2 \times 10^7 \text{ J kg}^{-1}$	Raveendran and Ganesh (1996)
	$X$	solid fraction of char products	$-2.33 \times 10^6 \text{ J kg}^{-1}$ (from $E_m$ above)	Bradbury <i>et al.</i> (1979)
	$\nu$	oxygen : carbon required for char oxidation	0.35	Bradbury <i>et al.</i> (1979)
Thermal properties				
	$\nu$	oxygen : carbon required for char oxidation	2.67	(based on $\text{C} + \text{O}_2 \rightarrow \text{CO}_2$ )
	$c$	specific heat of leaf	$1.38 \times 10^3 \text{ J kg}^{-1} \text{ K}^{-1}$	Touloukian and Buyco (1970)
	$c_m$	specific heat of water	$4.19 \times 10^3 \text{ J kg}^{-1} \text{ K}^{-1}$	Gray <i>et al.</i> (2002)
	$h$	heat transfer coefficient	$20 \text{ W m}^{-2} \text{ K}^{-1}$	Drysdale (1998) <sup>B</sup>
	$h_f$	heat transfer coefficient from flame	$50 \text{ W m}^{-2} \text{ K}^{-1}$ $10 \text{ W m}^{-2} \text{ K}^{-1}$ (tuned value)	Ohlemiller and Shields (1991)
	$T_f$	flame temperature	1200 K	Albini (1985)
	$\lambda$	thermal conductivity	$5 \times 10^{-2} \text{ W m}^{-2} \text{ K}^{-1}$	Touloukian <i>et al.</i> (1970)

	Symbol	Parameter	Value	Source
Gas properties during char oxidation				
	$Y_{OA}$	mass fraction of oxygen in ambient air	0.233	Boonmee and Quintiere (2005) <sup>c</sup>
	$Y_{OS}$	mass fraction of oxygen in air at leaf surface	0	
Physical constants				
	$R$	universal gas constant	$8.314 \text{ J mol}^{-1} \text{ K}^{-1}$	Drysdale (1998)
	$\sigma$	Stefan–Boltzmann constant	$5.67 \times 10^{-8} \text{ W m}^{-2} \text{ K}^{-4}$	Drysdale (1998)

<sup>A</sup>Positive values indicate exothermic reaction.

<sup>B</sup>Calculated from table 2.4 in Drysdale (1998), based on natural, turbulent convection past a vertical surface.

<sup>C</sup>When the kinetically controlled rate of char oxidation is rapid, and the reaction rate is controlled by the rate of oxygen diffusion,  $Y_{OS}$  is assumed to be small.



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