Supplement (S1)

Key DO_3SE model formulations related to R_{sur} and g_{sto}

One of the unique features of the DO_3SE model is the method used to estimate R_{sur} (see eq 1, main text). Given the importance of both the formulations and parameterisation of this R_{sur} module to the estimates of deposition this Supplement (S1) provides a full description of this component of the CMAQ-DO_3SE model. The parameterisations for different land cover and species for the formulations provided below are given in Tables S1 and S2.

Phenology

Estimates of the start (*SGS*) and end (*EGS*) of the physiologically active growth period are made according to a latitude model (Eq. 3 and 4) for those forest trees that exhibit strong seasonality; other tree species are either assumed to have a year round growth period or physiological limits are assumed to be inferred by the function relating stomatal conductance to air temperature (f_T) (LRTAP Convention, 2008).

$$SGS = (lat - 50) * 1.5 + 105 \quad (3)$$

$$EGS = 297 - ((lat - 50) * 2) \quad (4)$$

where *lat* represents the latitude in degrees.

For croplands, represented by wheat, a six month period (from year day 92 to 270) was used as the growing season. This allowed capture of the full UK cropping season even though it is recognized that individual crops will tend to have shorter growing seasons (normally up to 3 months duration).

The Leaf Area Index (*LAI*), Stand Area Index (*SAI*) and f_{phen} functions are fitted within these growth periods. The calculation of R_{sur} described in Eq. (1) is dependent upon accurate estimates of *LAI* (described in Eq. 5 where *dd* represents the Julian day of the year) to scale from the leaf/needle to canopy level. The *LAI* scaling of the g_{sto} term employs a canopy light extinction model to estimate sunlit and shaded *LAI* canopy fractions and hence scales r_{sto} as a function of radiative penetration into the canopy (Norman, 1979).

$$LAI = LAI_{min} \text{ when } dd < SGS$$

$$LAI = (LAI_{max} - LAI_{min}) * ((dd - SGS) / LAI_s) + LAI_{min} \text{ when } SGS \leq dd < SGS + LAI_s$$

$$LAI = LAI_{max} \text{ when } SGS + L_s \geq dd < EGS - LAI_e$$

$LAI = (LAI_{max} - LAI_{min}) * ((EGS - dd) / LAI_e) + LAI_{min}$ when $EGS - LAI_e \geq dd < EGS,$

$LAI = LAI_{min}$ when $dd \geq EGS$ (5)

where LAI_{min} and LAI_{max} are the minimum and maximum values of seasonal LAI and LAI_s and LAI_e are the period in days required to cycle between these maximum and minimum values of seasonal LAI.

Canopy level f_{phen} for O_3 deposition and leaf level f_{phen} for O_3 damage estimates (i.e. estimates of g_{sto} used within the F_{st} calculations) were made using equation (6) with the exception of leaf level f_{phen} for croplands and wheat.

f_{phen}=$(1 - f_{phen\_a}) * (dd - SGS) / ((f_{phen\_c} + SGS) - SGS) + f_{phen\_a}$ when $SGS \leq dd > (SGS + f_{phen\_c}),$

f_{phen}=1 when $(SGS + f_{phen\_c}) > dd < (EGS – f_{phen\_d}),$

f_{phen}=$(1 - f_{phen\_b}) * ((EGS - dd) / (f_{phen\_d})) + f_{phen\_b}$ when $(EGS - f_{phen\_d}) < dd \geq EGS$ (6)

Where f_{phen_a} represents the minimum starting f_{phen} value (at the start of the growing season), f_{phen_c} represents the length of time to reach the maximum f_{phen} of the season and $f_{phen\ \_d}$ the length of time to decrease from this maximum to the minimum end of season value represented by f_{phen_b}. Outside the growing season f_{phen}=0

For wheat, the F_{st} accumulation period is defined as lasting a specific number of days (defined as between A_{start} and A_{end}) within the growth period calculated as in equations (7 and 8); since F_{st} is estimated for the flag leaf in wheat, the variation in g_{sto} with the age of the flag leaf (termed *leaf* f_{phen}) will be different to the canopy level f_{phen} and is estimated according to equation (9).

$A_{start} = (2.57 * lat + 40) -15$ (7)

$A_{end} = A_{start} + 55$ (8)

leaf $f_{phen} = (1 - f_{phen\_1}) * ((dd - A_{start}) / f_{phen\_3}) + f_{phen\_1}$

when $A_{start} \leq dd < (A_{start} + f_{phen\_3})$

leaf $f_{phen} = 1$

when $(A_{start} + f_{phen\_3}) \leq dd \leq (A_{end} – f_{phen\_4})$

leaf $f_{phen} = (1 - f_{phen\_2}) * ((A_{end} – dd) / f_{phen\_4}) + f_{phen\_2}$

when $(A_{end}-f_{phen\_4}) < dd \leq A_{end}$ (9)

where f_{phen_1} is 0.8, f_{phen_2} is 0.2 representing the starting and ending *leaf* f_{phen} values respectively; f_{phen_3} is 15 and f_{phen_4} is 40 representing the length of time in days for the increase and decrease in *leaf* f_{phen} respectively between A_{start} and A_{end}.

The influence of O_3 on senescence (f_{O3}) is given by equation (10), this is used only for croplands and wheat, for all other cover types is assumed that f_{O3} is equal to 1.

$$f_{O3} = ((1 + (POD_0 / 11.5)^{10})^{-1}) \quad (10)$$

The influence of the four environmental variables (irradiance (f_{light}), temperature (f_T), vapour pressure deficit (f_D) and soil water (f_{SW}) on g_{sto} is given in equations (11 to 14) where *PPFD* represents photosynthetic photon flux density in μmol m^{-2} s^{-1}; T represents surface air temperature in ^oC and T_{min} and T_{max} represent the T at which relative g is at a minimum and maximum respectively; D represents atmospheric water vapour deficit in kPa and D_{min} and D_{max} represent the D at which relative g is at a minimum and maximum respectively; and ψ_{pdleaf} represents the pre-dawn leaf water potential estimated according to methods described in (Büker et al., 2012).

$$f_{light}=1-\exp(-\alpha * PPFD) \quad (11)$$

$$f_T = \max\{f_{min}, (T - T_{min}) / (T_{opt} - T_{min}) * [(T_{max} - T) / (T_{max} - T_{opt})]^{bt}\}$$

$$bt = (T_{max} - T_{opt}) / (T_{opt} - T_{min}) \quad (12)$$

$$f_D = \max\{f_{min}, \min\{1, (1 - f_{min}) * (D_{min} - D) / (D_{min} - D_{max}) + f_{min}\}\},$$

$$\text{if } \Sigma D \geq D\_crit, \text{ then } g_{sto}\_hour\_n{+}1 \leq g_{sto}\_hour\_n \quad (13)$$

where $g_{sto}\_hour\_n$ and $g_{sto}\_hour\_n+1$ are the g_{sto} values for hour n and hour $n+1$ respectively calculated according to the g_{sto} equation.

$$f_{SW} = \min\{1,\max\{f_{min},0.355(-\psi_{pdleaf})^{-0.706}\}\} \quad (14)$$

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List of Tables and Figures

Table S1. Land cover and species-specific parameterisation of the seasonal components of the R_{sur} and g_{sto} modules of DO_3SE model (LRTAP Convention, 2008; Simpson et al., 2003).

	SGS	EGS	LAI_{max}	LAI_{min}	LAI_s	LAI_e	f_{phen_a}	f_{phen_b}	f_{phen_c}	f_{phen_d}
Land cover type										
Coniferous forest (based on Scots pine)	0	365	4.5	4.5	-	-	0.8	0.8	40	40
Deciduous forest (based on Beech)	*Lat**	*Lat**	4	0	15	30	0.3	0.3	15	20
Mixed forest (based on Scots pine & beech)	0	365	4	4	-	-	0.8	0.8	40	40
Cropland (based on wheat)	*Lat***	*Lat***	3.5	0	70	22	0.1	0.1	0	45
Productive grassland (based on perennial ryegrass)	0	365	2	3.5	140	135	1	1	-	-
Semi-natural vegetation	0	365	2	3	192	96	1	1	-	-

Species										
Beech	*Lat**	*Lat**	4	0	15	30	0.3	0.3	15	20
Wheat	*Lat***	*Lat***	3.5	0	70	22	0.1	0.1	0	45
Perennial ryegrass	0	365	2	3.5	140	135	1	1	-	-

Table S2. Land cover and species-specific parameterisation of the diurnal components of the R_{sur} and g_{sto} model (LRTAP Convention, 2008; Simpson et al., 2003).

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	g_{max} mmol O_3 m^{-2} PLA s^{-1}	f_{min}	f_{O3}	α	T_{min}	T_{opt}	T_{max}	D_{max}	D_{min}	D_{crit}
Land cover type										
Coniferous forest (based on Scots pine)	180	0.1	1	0.006	0	20	36	0.6	2.8	-
Deciduous forest (based on Beech)	150	0.1	1	0.006	0	21	35	1.0	3.25	-
Mixed forest (based on Scots pine & beech)	165	0.1	1	0.006	0	20	35	0.8	3.0	-
Cropland (based on wheat)	450	0.01	***	0.0105	12	26	40	1.2	3.2	8
Productive grassland (based on perennial ryegrass)	295	0.01	1	0.009	12	26	40	1.3	3.0	-

Semi-natural vegetation	60	0.01	1	0.009	12	26	40	88.8	-99.9	-
Species										
Beech	150	0.1	1	0.006	0	21	0.3	1.0	3.25	-
Wheat	450	0.01	***	0.0105	12	26	0.1	1.2	3.2	-
Perennial ryegrass	295	0.01	1	0.009	12	26	1	1.3	3.0	-