

Supplementary Materials: The following are available online at www.mdpi.com/10.3390/atmos12060789/s1

S1. Vegetation density for birch (*Betula*)

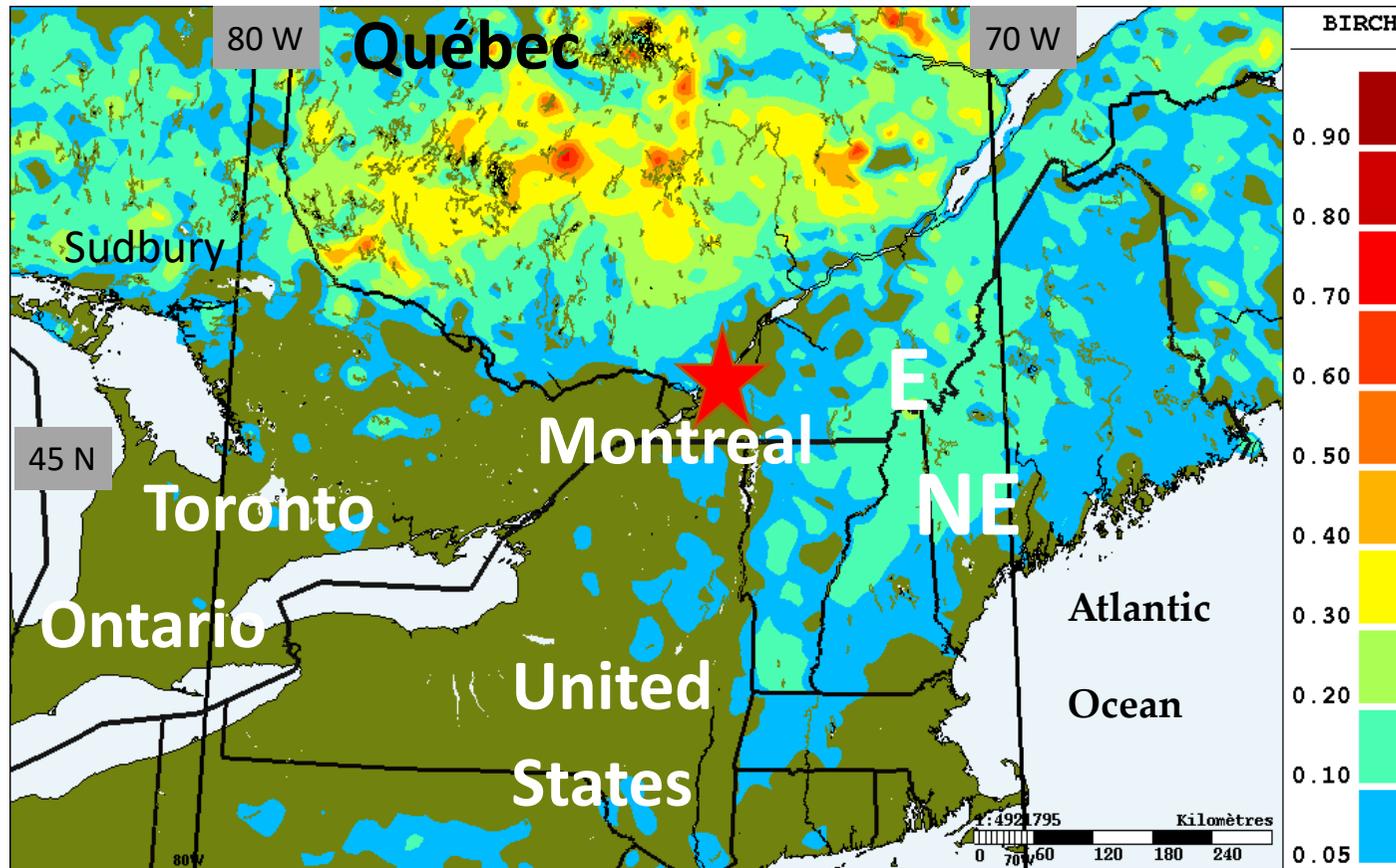


Figure S1: Vegetation density (Birch) over the eastern portion of North America.

S2. Details about the numerical platform GEM-MACH

The GEM-MACH model solves the standard advection-diffusion equation for pollutant and uses the semi-Lagrangian transport algorithm to disperse the pollutants in the horizontal according to the following equation:

$$\partial C/\partial t = -\nabla(\mathbf{v}C) + \nabla(\mathbf{K} \cdot \nabla C) + Q + S \quad (\text{S.1})$$

where C is the pollen concentration, \mathbf{v} represents the advecting velocity 3-D vector field or wind field, \mathbf{K} is a three-dimensional eddy diffusivity vector, and Q and S represent source (emission) and sink (removal) terms, respectively. Note that the mathematical operator ∇ expresses the spatial gradient. This equation means that the local change in pollen concentration (first term on the left hand side of the Eq. S.1) at a given point depends on the transport, diffusion (horizontal and vertical) and source and sinks respectively (right hand side of Eq. S.1). This equation is applied to every point of a discrete domain of 744 by 644 points covering most of North America. GEM-MACH (version 1.3.8) uses a simple 2-bin sectional representation of the particulate size representation (bin 1 is for 0-2.5 μm aerodynamic diameter and bin 2 is for 2.5-10 μm particles, that is fine and coarse particles respectively).

A number of air quality process representation exists in GEM-MACH, including those for gas-phase, aqueous-phase, and heterogeneous chemistry and for particular processes; nucleation, condensation, coagulation, inorganic, gas-particle, partitioning, sedimentation, in-cloud and below-scavenging and secondary organic aerosol (SOA) formation and, finally, and wet and dry deposition. Nine pollutants are simulated in the basic configuration of GEM-MACH: SO_4 , NO_3 , NH_4 , elemental carbon (EC), primary and secondary organic aerosols, crustal material, sea salt, and particle-bound water. SMOKE emissions processing system was used to produce anthropogenic input emission files from 2006 national inventories for Canada, 2005 (but projected to 2012) for U.S. and 1999 for Mexico. Biogenic emissions are estimated on-line using BEIS v3.09 algorithms. GEM-MACH-10 km is a limited-area configuration of GEM regional. The original domain covers most of North America. The model has 80 vertical levels from the surface up to 0.1 hPa (mesosphere). Both meteorological and chemical fields are simulated simultaneously. More details for GEM and MACH are presented in Robichaud and Comtois [54]. Figure S2 gives the basic components of the modelling system used here.

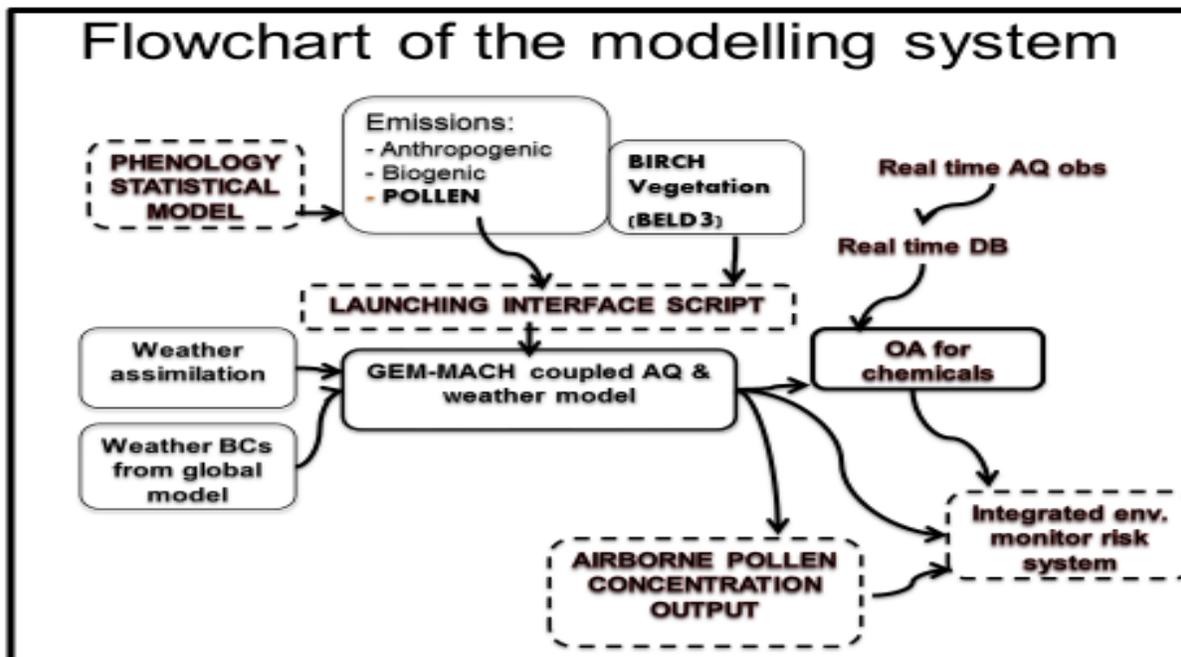


Figure S.2. Components of the atmospheric modelling system including pollen simulation. Note. BC stands for boundary conditions, DB for database, OA for Objective analysis (model-data fusion) and AQ for air quality.

S.2.1. Emission modelling

The pollen emission module is one of the most important but most uncertain component of the modelling system shown in Fig. S.2. Determining the emission of pollen grains involves the knowledge of source areas, timing of flowering and intensity of pollen release through anthers. According to Laaidi et al. [66], Helbig et al. [104] and de Weger et al. [67], meteorological factors have the most impact in controlling anther dehiscence. Relatively high temperature, low humidity and moderate wind speed favoring a passive dehydration, which leads to bursting of the anthers and dehiscence [67,104]. In a broad sense, it is useful to identify three classes of factors, which promote emission [66]. Among the primary factors, we distinguish the heat sum of temperature (degree-days of growth), the solar radiation and the soil humidity, which all need to be adequate to predict time of flowering of the plant and later pollen release. These factors control the start of the season and the first pollen release of the season. Other meteorological factors including teleconnections indices, such as NAO (North Atlantic Oscillation) and ENSO (El Niño Southern Oscillation) are statistically related to birch starting of the pollen season as discussed by Robichaud and Comtois [25]. The so-called secondary factors control anther release or dehiscence on a day-to-day basis. Dry air will favor dehiscence whereas rain or very high relative humidity would stop pollen release. Finally, tertiary factors control the transport and dispersion of pollen. They are the wind, turbulence, topography and the rain washout.

A coherent modelling of the flux pollen release is given by Helbig et al. [104] and adopted here (i.e, Figures 6-8) but with some minor modifications described below. This pollen release model has been used by several authors [54,61,64]. The emission is given by:

$$\text{Emission} = F_e(t,i,j) \times SA(i,j) \times D(i,j) \tag{S.2}$$

where $SA(i,j)$ is the surface area of a model grid cell (about 100 km² for GEM-MACH) and D is the birch coverage density (a fraction between 0 and 1; according to Fig. S1). $F_e(t,i,j)$ emission flux closely follows that of Helbig et al. [104]. In that formulation, F_e is evaluated at the top of vegetation and is proportional to the product of a characteristic concentration and a characteristic velocity (u^*):

$$F_e(t,i,j) = C_e(t) \times c^* \times K_e(u^*, T, RH) \times u^* \quad (S.3)$$

where u^* , the friction velocity, is given by the GEM model averaged over a tile of 10 km by 10 km. $C_e(t)$ is a specific factor describing the probability of emission of etamins. In Helbig et al. [104], the value of C_e is given as a function of time:

$$\begin{aligned} C_e(t) &= 4 \times 10^{-4} \times (d/S - d^2/S^2) \quad \text{during the pollen season} \\ C_e &= C_o \quad \text{before and after the season} \end{aligned} \quad (S.4)$$

where d is the number of days since the beginning of the pollen season and S , the length of the season and C_o , a very small value. Note that the pollen season is defined here by 2.5-97.5% of pollen sum over the whole period. The parameter c^* is independent of time and space and is expressed by:

$$c^* = Q_p / (LAI \times h_c) \quad (S.5)$$

where LAI is the mean leaf area index, Q_p the total pollen quantity within a season per unit area (pollen grains /m²) and h_c the canopy height. As pointed out by Siljamo [64], Q_p , the total amount of pollen in catkins is a very uncertain parameter based on meteorological variations and pollen release of the previous year. The parameter K_e is a switch which has the value zero, if the velocity friction wind (u^*) is below a certain threshold defined in Eq. S.6. Finally, c^* and u^* are characteristic values of pollen concentration (in particulates per cubic meter) and wind velocity at the surface (in m/s). There are no units for C_e and K_e so that the units of F_e are in grains/m²/s. As in Helbig et al. [104], we formulate K_e as being dependent on u^* , temperature, relative humidity and surface wind velocity:

$$\begin{aligned} K_e &= (1 - u_{th}^*/u^*) \quad \text{if } u^* > u_{th}^* \\ K_e &= 0 \quad \text{otherwise} \end{aligned} \quad (S.6)$$

with the threshold value $u_{th}^* = u_{th}^t \alpha$. Following Helbig et al. [104], the value of u_{th}^t is taken as 0.333 and the expression for α is given below:

$$\alpha = 3 / (\alpha_v + \alpha_T + \alpha_D) \quad (S.7)$$

with the threshold wind, temperature and dew point temperature, which respectively are:

$$\alpha_v = V/V_{th}, \quad \alpha_T = T/T_{th}, \quad \alpha_D = D/D_{th}. \quad (S.8)$$

The expression for α is also taken the same as in Helbig et al. [104] except for a small modification. Here we have used the dew-point depression $T - T_d$ instead of the relative humidity for convenience. Table S.2 summarizes parameters adopted for the basic simulation and the threshold values for meteorological parameters as defined in Eqs. S.7 and S.8. The pollen density is taken as 800 kg/m³, which determines the sedimentation velocity. The reader is referred to Helbig et al. [104] for more details concerning other parameters of the emission model. Sensitivity tests have confirmed that most of the parameters taken from Helbig et al. [104] for alder are appropriate for *Betula* birch except for the threshold temperature which is taken higher for *Betula*. For temperature threshold, the value of 11 °C was taken instead of 8 °C, the former reducing the error when compared to observations (results not shown). The quantity of emitted pollen for each grid squared area is computed by multiplying F_e by the grid area and by the birch

density (fraction from 0 to 1, see Fig. S1) as given by Eq. S.2. The pollen pool (Q_p) was taken as 2.0 billion per square meter as evaluated in Robichaud and Comtois [54] which is considered a typical value for the family of Betulacea [104]. The emission intensity of the pollen puff is controlled by equations S.2 through S.8.

Table S.1 Parameters used in the pollen modelling simulation (*H04*: [104], *S06*: [61], *S13*: [64]).

Parameter	Symbol	Value	Units	Reference
Total emission per unit surface	Q_p	2.0E09	Pollen/m ²	<i>H04</i> Taken as same as alder
Length duration	S	42	days	[25,104]
Average height	Hc	22.5	meters	[104]
Mean leaf area index	LAI	2	-	
Pollen density	ρ_{pollen}	800	kg/m ³	
Pollen aerodynamic Diameter	D_a	22	microns	Taken as same for alder (<i>H04</i>)
Sedimentation velocity	V_d	1.3	cm/s	(1.2 cm/s is used in <i>S06</i>)
Temperature Threshold ¹	T_{th}	11	°C	For alder, (<i>H04</i>) proposed 8 °C
Dew point depression threshold ¹	D_{th}	5	°C	Adapted from <i>H04</i> (Ute=80% R.H. corresponds to T-Td of 5 deg. C.
Wind threshold ¹	V_{th}	2.9	m/s	Taken as for alder (<i>H04</i>)
Threshold friction velocity ¹	u^*_t	0.323	m/s	Taken as <i>H04</i>
Precipitation threshold ²	P_{th}	0.5	mm/h	<i>S13</i>
Vertical correlation length	L_z	10	Hybrid levels	Set to average height of boundary layer

¹No emission below this threshold, ²No emission above this threshold.