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Operational chemical weather forecasting with the ECCC online Regional Air Quality Deterministic Prediction System version 023 (RAQDPS023) – Part 1: system description

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Supplement

S1 GEM-related meteorological components and attributes of RAQDPS023

S1.1 Dynamical core

The governing equations of atmospheric dynamics used by the GDPS 8.0.0, RDPS 8.0.0, and RAQDPS023 have been described by Côté et al. (1998) and Girard et al. (2014). Prognostic variables included the horizontal wind components, vertical velocity component in the hybrid ζ-coordinate (Sect. 2.1), virtual temperature, geopotential, specific humidity, and physics-related atmospheric tracers such as cloud condensate mixing ratio and turbulent kinetic energy (TKE). GEM was configured to be hydrostatic in all three forecast systems (e.g., CMC-RDPS-8.0.0, 2021b). Time integration of this dynamical core was performed using a two-time-level, iterative-implicit, three-dimensional semi-Lagrangian scheme (Côté et al., 1998; Girard et al., 2014; Husain and Girard, 2017). This semi-implicit, semi-Lagrangian integration is unconditionally stable for any time step, thus allowing the choice of time step to be based solely on accuracy requirements (Robert, 1981; Ritchie et al., 2022). A dynamics integration time step of 450 s was used by the GDPS 8.0.0 and one of 300 s was used by both the RDPS 8.0.0 and RAQDPS023. The improved trajectory calculations for semi-Lagrangian advection described by Husain and Girard (2017) were employed, which involve trapezoidal averaging of wind components along air-parcel trajectories and cubic interpolation at the departure positions. These calculations remove numerical inconsistencies in the discretization and interpolation used for the source terms between the dynamical and trajectory equations.

Semi-implicit, semi-Lagrangian time integration schemes are highly efficient by virtue of circumventing the stiffness of the dynamic equations, but semi-Lagrangian advection schemes are not inherently shape-preserving or mass-conserving (e.g., Staniforth and Côté, 1991; Hansen et al., 2011; de Grandpré et al., 2016). Shape preservation for two meteorological tracers (water vapour, cloud water) was implemented in all three forecast systems using a two-step, iterative locally-mass-conserving (ILMC) monotonicity correction for tracer advection (Sørensen et al., 2013). The ILMC scheme is an *a posteriori* filter that detects the lack of shape preservation of a forecasted solution, including negative concentrations, and corrects it by redistributing the overshoot or undershoot mass to the surrounding cells while preserving the mass of the forecasted solution. By imposing shape preservation, the creation of artificial local minima or maxima by the advection scheme was avoided. The Bermejo and Conde (2002) global mass fixer was then applied in the three forecast systems to impose mass conservation, where the LAM boundary-flux estimation was used for the two regional systems (Aranami et al., 2015). It is worth mentioning in this context that Staniforth and Côté (1991) observed that semi-Lagrangian advection schemes handle sharp discontinuities (e.g., clouds) better than Eulerian advection schemes and dispersion errors are localized around the discontinuity, which suggests that mass-fixing adjustments will tend to be localized as well. McTaggart-Cowan et al. (2019a) noted that the use of these two schemes in GEM improved the conservation of liquid water static energy and total water mixing ratio, with the result that total global precipitation in the GDPS was reduced by approximately 5%. A surface pressure adjustment was also made for moist-air mass conservation (McTaggart-Cowan et al., 2019a; CMC-RDPS-8.0.0, 2021b).

While semi-Lagrangian advection schemes have some inherent numerical diffusion (e.g., Staniforth and Côté, 1991), artificial horizontal diffusion was also implemented to damp spurious numerical noise at the shortest spatial scales (e.g., Mailhot et al., 2006). In all three systems a Del-4 operator was applied to momentum variables (10%) and a Del-6 operator (1%) was applied to potential temperature, but tracers were not affected. As well, a Del-2 operator with enhanced horizontal diffusion coefficients was applied to temperature and momentum variables in a sponge layer below the model lid (top eight levels) to prevent wave energy reflection from the rigid model lid (Mailhot et al., 2006; CMC-GDPS-8.0.0, 2021b; CMC-RDPS-8.0.0, 2021b).

S1.2 Land-surface and water-surface schemes

The ISBA (Interactions between Soil–Biosphere–Atmosphere) land-surface scheme was used in all three forecast systems to parameterize land surface processes and atmosphere-surface exchanges of heat and moisture (Noilhan and Planton, 1989; Bélair et al., 2003a,b). ISBA takes a mosaic approach that considers subgrid "tiles" of four major surface types: land; glacier; water; and sea ice. A GEM surface grid cell may be composed of up to four tiles to account for the presence of these different surface types. Ten prognostic variables are considered in the ISBA scheme for each land surface tile, including two surface temperature variables, three soil moisture variables, one vegetation canopy variable, and four variables describing snowpack characteristics. Each prognostic equation is solved based on an energy or water budget formulation with a force-restore term to moderate diurnal variations. Initial values of land surface temperature and moisture, deep-soil temperature and moisture, and snow depth, snow albedo and snow density were derived from analyses and then predicted using the ISBA scheme. Emissivities of bare soil and snow were derived from climatology and observations, respectively.

The treatment of glacier surface tiles closely followed that for land surfaces and combined a surface-energy-budget model with a two-layer force-restore term to predict glacier temperature profiles (McTaggart-Cowan et al., 2019a). Updated ice emissivities were based on observations.

The treatment of water surface tiles depends on the forecast system. The GDPS 8.0.0 was coupled to an ocean model whereas the RDPS 8.0.0 and RAQDPS023 obtained initial sea-surface temperatures from surface analyses and then employed the one-dimensional thermodynamic mixed-layer model of Zeng and Beljaars (2005) to predict the diurnal evolution of the water skin temperature and underlying mixed-layer temperature. This two-layer scheme provides a more realistic description of diurnal variations in water surface temperature and hence surface fluxes over water. Note that the GDPS 8.0.0 also used the mixed-layer scheme over lakes as did the RDPS 8.0.0 and RAQDPS023 (McTaggart-Cowan et al., 2019a; CMC-GDPS-8.0.0, 2021b; CMC-RDPS-8.0.0, 2021b). The surface roughness length for momentum over water surfaces was based on the Charnock (1955) scheme while roughness lengths over water for heat and moisture came from Deacu et al. (2012).

Sea-ice (and frozen lake) surfaces were parameterized using a one-dimensional heat diffusion model (Semtner, 1976) with a simple, three-level vertical discretization (McTaggart-Cowan et al., 2019a). Initial sea-ice thickness values were derived from climatology but then evolved in time, whereas sea-ice fractions were derived from surface analyses, were fixed in time, and were checked for consistency with the sea-ice thickness. A 3% lead fraction in sea ice was assumed over salt water (McTaggart-Cowan et al., 2019a).

Other geophysical variables needed by the ISBA land-surface scheme include surface roughness length (except over water), surface albedo, vegetation characteristics (vegetation type and fraction, leaf area index), soil texture and soil thermal and hydraulic coefficients, and glacier fraction. For grid cells with heterogeneous land surface cover (e.g., some bare soil/some vegetation or some land/some water), some grid-scale geophysical fields such as roughness length, albedo, and emissivity were determined as weighted averages of tile-specific geophysical fields.

S1.3 Surface-layer scheme

Monin-Obukhov surface-layer similarity theory was used in all three forecast systems to describe turbulent surface fluxes of momentum, heat, and moisture and state variable profiles in the model surface layer, which extends from the Earth’s surface to the first prognostic model level (located at approximately 10 m AGL for temperature and moisture and 20 m AGL for winds on the staggered Charney-Phillips vertical grid: Sect. 2.1). Separate profiles were calculated over each of the four surface types (tiles) and were then aggregated as a weighted average to provide a grid-cell value (McTaggart-Cowan et al., 2019a). Stability functions proposed by Delage and Girard (1992) and Beljaars and Holtslag (1991) were used for unstable and stable stratifications, respectively (CMC-GDPS-8.0.0, 2021b; CMC-RDPS-8.0.0, 2021b).

For stable conditions the following minimum Obukhov lengths were imposed to prevent surface-atmosphere decoupling (Foken, 2006; McTaggart-Cowan et al., 2019a): 20 m over soil surfaces; 5 m over glaciers; and 10 m over water surfaces and sea ice (CMC-GDPS-8.0.0, 2021a; CMC-RDPS-8.0.0, 2021a). The higher value over land surfaces is consistent with a dependence on surface roughness (van de Wiel et al., 2007; Zheng et al., 2017).

S1.4 Boundary-layer scheme and vertical diffusion

Vertical diffusion in the planetary boundary layer (PBL) and free troposphere was described in all three forecast systems by the vertical diffusion coefficient K_z, which is parameterized using a 1.5-order turbulence closure scheme based on a predictive equation for subgrid-scale (SGS) turbulent kinetic energy (Benoit et al., 1989; Bélair et al., 1999). A key parameter of the scheme is the gradient Richardson number Ri. Two turbulence length scales are also needed by this scheme. A turbulence-regime-specific mixing length λ, which reflects the significant eddy size, is needed to calculate both TKE and K_z, and a dissipation length scale λ_e is also needed to calculate TKE. The required lower boundary conditions are obtained from the surface-layer parameterization (see previous subsection). K_z from GEM was also needed by MACH (**Sect. 3.1**).

Note that a number of enhancements have been made to the original formulation of Benoit et al. (1989). First, λ was calculated in the GDPS 8.0.0, RDPS 8.0.0, and RAQDPS023 using a blended formulation (McTaggart-Cowan and Zadra, 2015) based on a local scheme for λ (Blackadar, 1962) and a non-local scheme for λ (Bougeault and Lacarrére, 1989). A treatment for Richardson number hysteresis across the temporal transitions from stable-to-unstable conditions and unstable-to-stable conditions that uses an Ri transition zone rather than a single critical value of Ri has been implemented (McTaggart-Cowan and Zadra, 2015). A 900 s relaxation time scale was also imposed on λ to dampen the effects of turbulent transitions, and an upper bound of 50 m was imposed on λ_e (Bélair et al., 1999; McTaggart-Cowan et al., 2019a).

Lastly, the enhancement of turbulent diffusion near the top of the cloudy boundary layer due to updrafts, downdrafts, and related entrainment associated with boundary-layer clouds, including stratus, stratocumulus, and shallow cumulus clouds, was represented by an extension to the TKE scheme with statistical SGS cloudiness called the MoisTKE scheme (Bechtold and Siebesma, 1998; Mailhot and Bélair, 2000; Bélair et al., 2005; Mailhot et al., 2006). Three key parameters were considered: turbulent flux of total cloud water; SGS cloud fraction; and a nondimensional flux enhancement factor (Bélair et al., 2005). These cloud effects were activated only if the surface buoyancy flux was positive and the cloud layer resided below 1.5 times the PBL height (McTaggart-Cowan et al., 2019a).

PBL height, which is also an important quantity for air quality, was diagnosed every time step for use in the Kain-Fritsch deep convection scheme (Sect. S1.7) as well as the MoisTKE scheme. A bulk Richardson number (Ri_b) method was used in which PBL height was determined to be the lowest momentum vertical level at which Ri_b reaches a critical value of 0.25 by scanning the Ri_b profile upward from the surface (Seidel et al., 2012; McTaggart-Cowan et al., 2019a). PBL height from GEM was also needed by MACH (Sect. 3.13).

S1.5 Radiation scheme

Solar and infrared radiation were parameterized in all three forecast systems using a correlated-k distribution technique (Li and Barker, 2005). Nine prescribed longwave bands and four shortwave bands were considered. The updated version of this scheme that is now used by GEM included an improved treatment of the water vapour continuum and solar radiation absorption by methane (McTaggart-Cowan et al., 2019a). A monthly climatological O_3 distribution constructed from several data sets was used along with three-dimensional climatologies of other trace gases (CH_4, NO, CFC-11, CFC-12) appropriate for the year 2015 (McTaggart-Cowan et al., 2019a). A simple parameterization of methane oxidation by the hydroxyl radical (OH) was also considered as a source of water vapour in the middle atmosphere (McTaggart-Cowan et al., 2019a).

The GEM v5.1 code includes the addition of O_3 as a new prognostic tracer, which made it possible for the GDPS 8.0.0 to assimilate satellite O_3 measurements (CMC-GDPS-8.0.0, 2021a). In order to represent O_3 as a standalone chemical constituent (i.e., no explicit reactive chemistry), a linearized approach was used to parameterize the photochemical production and loss terms (McLinden et al., 2000). The use of this parameterization scheme, called LINOZ, in GEM has been shown to be both robust and very fast (de Grandpré et al., 2016).

S1.6 Grid-scale cloud and precipitation scheme

The Sundqvist grid-scale condensation scheme (Sundqvist et al., 1989; Pudykiewicz et al., 1992) was used in the GDPS 8.0.0, RDPS 8.0.0, and RAQDPS023 to represent grid-scale cloud and precipitation processes. Four water phases were considered: water vapour; cloud water; rain water; and snow water. One modification to the original scheme was to use an updated cloud-to-precipitable water autoconversion rate (McTaggart-Cowan et al., 2019a; CMC-GDPS-8.0.0, 2021b; CMC-RDPS-8.0.0, 2021b). Lastly, this grid-scale cloud scheme was coupled to the three SGS cloud schemes described next via the liquid and solid condensate mass detrained from convective updrafts in these cumulus parameterization schemes, which were added to the grid-scale condensate mass (McTaggart-Cowan et al., 2019a).

S1.7 Cumulus parameterization schemes

Three separate cumulus parameterization schemes were used in the GDPS 8.0.0, RDPS 8.0.0, and RAQDPS023 to represent three different modes of SGS unstable moist vertical motions, namely deep convection, shallow convection, and mid-level convection. The term “deep convection” denotes convective clouds connected to the PBL with depths of at least 3 km; the term “shallow convection” refers to the subset of convective clouds whose bases are located in the PBL but which have updraft depths between 0.5 and 3 km that penetrate the PBL capping inversion (thus distinguishing them from the boundary-layer clouds parameterized in the MoisTKE scheme described in Sect. S1.4); and the term “mid-level convection” (or elevated convection) refers to convective clouds whose bases are located above the PBL.

All three schemes are formulated to transport momentum as well as heat and moisture vertically. Each scheme is mass-flux-based, and their implementations share five important structural elements: trigger; updraft; downdraft; solver; and closure (McTaggart-Cowan et al., 2019a). Calculations were performed sequentially for these three schemes, starting with the deep convection scheme and finishing with the mid-level convection scheme.

For SGS deep convection a modified version of the Kain–Fritsch deep-convection scheme (Kain and Fritsch, 1990, 1992) was used, which has been designed for horizontal grid spacing approaching the "gray zone" over which deep convection is partially resolved (i.e., horizontal grid spacing on the order of 1–10 km). The modifications included a Lagrangian treatment of convective initiation, object-based clouds, and a vertical transfer of momentum. See McTaggart-Cowan et al. (2019a) for a summary and McTaggart-Cowan et al. (2019b) for a detailed description of these modifications. Note that the trigger-function parameters are sensitive to horizontal grid spacing, so different values were specified for the GDPS 8.0.0 versus the RDPS 8.0.0 and RAQDPS023 (see Table 2 of McTaggart-Cowan et al.. 2019a).

SGS shallow convection was represented with a mass-flux scheme based on Bechtold et al. (2001). Simple representations of cloud entrainment (Siebesma et al., 2003) and constant fractional detrainment (Siebesma and Cuijpers, 1995) were used in the GEM implementation along with a simple, non-iterative closure that assumes that the shallow convective clouds in a grid cell are in a state of quasi-equilibrium with boundary-layer forcing (Bechtold et al., 2014). Note that downdrafts and precipitation were not parameterized by this scheme.

Lastly, SGS mid-level convection was treated using components of the deep convection scheme if certain conditions were met (McTaggart-Cowan et al., 2019a). One condition was that a minimum upward grid-scale mass flux was present: the required threshold was higher for the GDPS 8.0.0 than the RDPS 8.0.0 / RAQDPS023 (1×10^7 kg s^{-1} vs. 7×10^6 kg s^{-1}). Another condition was that there must be a layer at least 500 m AGL that can build a cloud at least 2 km thick after the layer is raised to its lifting condensation level. Note that the deep and shallow convection schemes would not be activated in a low convective-available-potential-energy (CAPE) environment, but the mid-level convection scheme could be triggered even in low-CAPE environments.

S1.8 Gravity wave drag and low-level orographic blocking schemes

SGS orography is associated with two processes that must be parameterized: (i) vertical momentum transport due to the breaking of mountain-induced gravity waves ("mountain wave drag") and (ii) blocking of low-level upstream flows by mountain barriers. Parameterizations due to McFarlane (1987) for mountain wave drag and Lott and Miller (1997) for low-level blocking (with an enhanced drag coefficient: Wells et al., 2008; Vosper et al., 2009) were used by all three forecast systems (McTaggart-Cowan et al., 2019a; CMC-GDPS-8.0.0, 2021b; CMC-RDPS-8.0.0, 2021b). In addition, the Hines (1997a,b) Doppler spread parametrization for non-orographic gravity-wave drag due to imbalances and geostrophic adjustment associated with a variety of transient mesoscale meteorological features was also used to simulate the upper-level circulation accurately (Charron et al., 2002; McTaggart-Cowan et al., 2019a; CMC-GDPS-8.0.0, 2021b; CMC-RDPS-8.0.0, 2021b)

The geophysical variables required by these schemes include gridded orography plus SGS orographic parameters for gravity wave drag and low-level blocking. Note that it is a standard procedure to apply a smoothing operator to the model orography field in order to minimize stationary noise generation due to small-scale resolved orographic features. A numerical filter with a wavelength cutoff at 3Δx for the GDPS 8.0.0 and 5Δx for the RDPS 8.0.0 and RAQDPS023 was applied. SGS orographic parameters were computed using an analytically–extended reference orography spectrum and a 5 km cut-off for horizontal terrain scales (Beljaars et al., 2004; McTaggart-Cowan et al., 2019a; CMC-GDPS-8.0.0, 2021b; CMC-RDPS-8.0.0, 2021b).

S2 Overview of anthropogenic national emissions inventories pre-processing and processing

A number of adjustments had to be made to the projected national emissions inventories before they underwent the processing required to create the SET4.0.0 emissions input files. For the projected 2020 Canadian Air Pollutant Emissions Inventory (APEI), emissions from two emissions sectors, prescribed burning and agricultural-land wind erosion, were removed from the inventory because of insufficient information to allocate these emissions to specific days of the year or times of day. Projected 2020 aircraft in-flight cruise emissions were also removed because they would not materially affect short-term surface pollutant concentrations due to the altitude (6-10 km above sea level) at which they were emitted, whereas projected 2020 aircraft landing and takeoff emissions were kept. Projected 2020 residential wood combustion (RWC) emission values were replaced by 2010 APEI values due to concerns over some aspects of the revised estimation methodology used to calculate RWC emissions for the 2015 APEI (ECCC, 2013, 2016). In addition, projected 2020 emission values for one power plant and one large industrial facility that had been active in 2015 but that were known to have closed by 2020 were removed from the major-point-source emissions file (Moran et al., 2021).

Adjustments were also made to Canadian projected 2020 fugitive dust emissions from paved and unpaved roads and from construction activities. Fugitive dust emissions are the largest component of anthropogenic $PM_{2.5}$ and PM_{10} emissions, of which the main contributing emission sectors are vehicular traffic on paved and unpaved road surfaces and agricultural and construction activities. Emissions estimation methodologies are always subject to re-evaluation and improvement, and a new methodology to estimate fugitive dust emissions from paved and unpaved road surfaces was introduced by ECCC in 2020 for the 2018 APEI (ECCC, 2020). In order to account for these methodological changes in the projected 2020 APEI, whose base 2015 fugitive road dust emissions had been calculated using an older methodology (ECCC, 2016), the paved-road fugitive dust emissions from the projected 2020 inventory for each province and territory were scaled by a factor of 0.15 (i.e., decreased by 85%) and unpaved-road fugitive dust emissions were scaled by a factor of 1.11 (i.e., increased by 11%).

One useful check on any Canadian CAC annual emissions inventory is to compare per capita Canadian and U.S. emissions for the same emissions source sectors for the same year or closest year (since U.S. National Emissions Inventories (NEIs) are only available every three years). Given that differences in population have been controlled for by considering per capita values, one expects the per capita emissions values to be broadly similar given socioeconomic and emission control similarities between the two countries. When this comparison was made for the scaled 2015 Canadian fugitive dust emissions from paved and unpaved road surfaces and from agricultural activities and residential construction, the agreement with the closest U.S. CAC inventory, the 2014 U.S. NEI (https://www.epa.gov/air-emissions-inventories/2014-national-emissions-inventory-nei-data), was reasonable. For 2015 fugitive dust emissions from heavy construction, however, per capita Canadian emissions were 40 times higher than 2014 per capita U.S. emissions. Moreover, the estimation methodology used to estimate 2015 Canadian heavy construction emissions had not changed since 2012 and heavy construction emissions in the APEI had been held constant since 2013, which suggested a high level of uncertainty for these emissions. It was thus decided to scale Canadian projected 2020 fugitive dust emissions for heavy construction by a factor of 0.05 so that 2015 per capita Canadian emissions from this sector would be within a factor of two of the 2014 per capita U.S. emissions.

The Canadian projected 2020 APEI then underwent two final preparatory steps by ECCC before the emissions processing step. First, the point source file was separated into two files, a major-point-source file and a minor-point-source file, where a 20-m stack height threshold was applied to assign individual point sources to the major- or minor-point-source files based on stack height. Similarly, the area source file was separated by emissions source type into multiple files, including on-road mobile sources, off-road mobile sources, marine sources, agricultural sources, other fugitive dust area sources, and other non-fugitive-dust area sources. This separation by source type made it possible to apply special treatments such as meteorological modulation to an emissions subset (Sect. 3.12), and also to perform quality-control checks more easily after the emissions processing step. The second final preparatory step was to convert the Canadian projected 2020 APEI into a different format that would be readable by the emissions processing system. This step included augmentation of the APEI to include some non-standard data such as point source stack characteristics (Sect. 3.13) and facility operating schedules (e.g., Sassi et al., 2021).

Several minor adjustments were also required to the projected 2023 U.S. NEI for emissions from commercial marine vessels. These emissions had been pre-processed by the U.S. EPA for three non-overlapping modelling grids used by the EPA: (i) emissions within U.S. federal waters for the continental U.S. on a 12-km horizontal grid; (ii) emissions for Alaska on a 9-km grid; and (iii) emissions for other Pacific and Atlantic Ocean areas on a 4-km grid. However, there were spatial gaps among these three emissions regions, which were filled by inserting emissions from another marine emissions data set that was used for Phase 4 of the Air Quality Model International Initiative (AQMEII-4) project (Galmarini et al., 2021). The projected 2023 U.S. inventory also contained marine emissions over some Canadian waters: these emissions were removed to avoid double counting with the Canadian inventory. Note that the

EPA had created an emissions-processing version of the projected 2023 U.S. NEI and had also separated the surface emissions into multiple files by source type for easier processing and tracking (U.S. EPA, 2019).

S2.1 Emissions processing

Once the above inventory adjustments had been made, the three projected national inventories were processed using version 4.7 of the SMOKE (Sparse Matrix Operation Kernel Estimation) emissions processing system software (see https://www.cmascenter.org/smoke/) to generate the SET4.0.0 set of major-point-source emissions files and gridded surface emissions files. Two types of emissions files were needed because the separate major-point-source files contained emissions for large individual sources such as power plants and smelters for which plume rise was calculated by GEM-MACH (Sect. 3.13), whereas the surface emissions were the sum of area sources, on-road mobile sources, off-road mobile sources, and minor point sources for which plume rise was not relevant. Note that there were also two types of surface emissions identified in the SET4.0.0 set, one type identifying fugitive-dust area emissions and the other type identifying all other surface emissions. This made it possible to apply meteorological modulation only to fugitive-dust area emissions (cf. Sect. 3.12).

The SMOKE emissions processing system must perform four major processing tasks: (i) temporal allocation, where annual/monthly inventory emissions were allocated to specific hours of specific days and months of the year based on a set of source-sector-specific monthly, day-of-the-week (DOW), and diurnal temporal profiles; (ii) spatial allocation, where emissions reported by jurisdiction such as a province or county were allocated to the group of model grid cells that overlay that jurisdiction in as realistic a pattern as possible using source-sector-specific gridded spatial surrogate fields; (iii) chemical speciation, where four of the CACs contained in the inventories (NO_x, VOC, $PM_{2.5}$, PM_{10}) were separated into more detailed model species using source-sector-specific speciation profiles; and (iv) PM_x size disaggregation ($x = 2.5$ or 10), where PM_x emissions from the inventory were allocated to different model PM size sections or modes using source-sector-specific PM size distribution profiles (e.g., Dickson and Oliver, 1991; Houyoux et al., 2000; Moran et al., 2013; Matthias et al., 2018; Zhang et al., 2018; Boutzis et al., 2020).

Each SET4.0.0 emissions file generated by SMOKE was specific to a particular day of the week and month of the year, and each file contained 24 hours of gridded emission fields of model species (where NO_x emissions were split into emissions of NO, NO_2, and HONO, $PM_{2.5}$ and PM_{10} emissions were each split into emissions of six model PM chemical components, and VOC emissions were split into emissions of 11 model lumped VOC species for the ADOM-2 gas-phase chemistry mechanism) (Stroud et al., 2008; Zhang et al., 2018; see Table 3).

For the first emissions processing task, temporal allocation, SMOKE used 82 monthly temporal profiles, 22 DOW profiles, and 29 diurnal profiles to process the Canadian emissions, 9,971 monthly profiles, 12 DOW profiles, and 95 diurnal profiles to process the U.S. emissions, and 161 monthly profiles, 15 DOW profiles, and 49 diurnal profiles to process the Mexican emissions (CMC-RAQDPS-023, 2021). For the second task, spatial allocation, SMOKE employed 55 source-sector-specific gridded spatial surrogate fields to allocate emissions for each Canadian province or territory to one or more grid cells on the RAQDPS023 10-km continental Yin grid (Fig. 2), 42 gridded spatial surrogate fields for each U.S. county, and 15 gridded spatial surrogate fields for each Mexican county (CMC-RAQDPS-023, 2021). The gridded surface emissions from these different jurisdictions and source types were then summed for each model grid cell. Major-point-source emissions, on the other hand, were assigned to a single model grid cell based on their latitude-longitude coordinates while remaining in separate records for subsequent plume-rise calculations (Sect. 3.13). For the third task, chemical speciation, SMOKE used 331 VOC and 85 PM speciation profiles from the U.S. EPA's SPECIATE 4.5 database (https://www.epa.gov/air-emissions-modeling/speciate-1; Reff et al., 2009; Simon et al., 2010). Only two speciation profiles were used to speciate NO_x emissions: one profile for on-road and off-road sources, which included a small HONO component as well as NO and NO_2 components, and a generic profile with only NO and NO_2 components for point and area sources. Lastly, the fourth task, PM size disaggregation, was handled simply by assigning $PM_{2.5}$ emissions to the first RAQDPS023 PM size bin and assigning PM_{cf} emissions, calculated as the difference between PM_{10} and $PM_{2.5}$ emissions, to the second RAQDPS023 PM size bin. Following the chemical speciation and PM size disaggregation steps the SET4.0.0 output files produced by SMOKE contained hourly gridded emission fields of 18 gases and 12 particle size bin/chemical components for a total of 30 emitted model species: SO2, (H2)SO4, NO, NO2, HONO, CO, C3H8, ALKA, ETHE, ALKE, TOLU, AROM, HCHO, ALD2, MEK, CRES, ISOP, NH3, SU1, SU2, NI1, NI2, AM1, AM2, EC1, EC2, PC1, PC2, CM1, and CM2.

It should be noted that two more adjustments were made to all fugitive dust emissions *after* SMOKE processing. One adjustment was done immediately as a (static) post-processing step, where a transportable-fraction (TF) scaling factor field was applied to account for "home-grid-cell" SGS particle settling and particle removal by interception and impaction by vegetation (e.g., Pace, 2005). The TF scaling factor ranged between 0 and 1 and was dependent on the fractional coverage of different land-use classes in each grid cell, where forest classes had a TF value of zero and agricultural classes had a value of 0.75. As described in Sect. 3.12 the other (dynamic) adjustment was to apply meteorological modulation at each chemistry time step during each RAQDPS023 simulation to account for the day-specific dampening effects of precipitation and snow cover on fugitive dust emissions. In order to accommodate the model calculations required for meteorological modulation and plume rise (Sects. 3.12 and 3.13), three sets of emissions fields must be read by the RAQDPS023 each hour from two separate day-specific files: (i) major-point source emissions; and (ii) surface emissions files consisting of fugitive dust emissions (type F) and other surface emissions (type E). SET4.0.0 thus consisted of two input files per day of a typical week of each month, or 168 files in total. More information related to the SMOKE processing required to create GEM-MACH emissions files may be found in Zhang et al. (2018) and Sassi et al. (2021).

S3 Description of Canadian Forest Fire Emissions Prediction System (CFFEPS) version 4.1

CFFEPS v4.1 consists of a fire-growth module, a fire emissions module, and a thermodynamic-based module to estimate the vertical penetration height of a smoke plume (Chen et al., 2019a,b). Outputs from the Canadian Wildland

Fire Information System (CWFIS), which is an operational product developed and managed by the Canadian Forest Service (CFS), underpin the CFFEPS fire-growth module. The CWFIS is itself composed of three major subsystems: (i) the Canadian Forest Fire Weather Index (FWI) subsystem, which accounts for the influence of four surface weather elements (dry-bulb temperature, relative humidity, wind speed, and 24 hour accumulated precipitation) on the relative risk for wildfire occurrence; (ii) the Fire Monitoring, Mapping, and Modeling (Fire M3) subsystem, which identifies and locates active fires ("hotspots") using NRT satellite imagery; and (iii) the Canadian Forest Fire Behaviour Prediction (FBP) subsystem, which estimates fire fuel consumption and fire intensity (https://cwfis.cfs.nrcan.gc.ca/home).

Input data needed by the FWI subsystem include observing station measurements of 2 m dry-bulb temperature and relative humidity, 2 m dew point, 10 m wind speed, surface pressure, and past 24 hour rainfall from about 2500 Canadian and U.S. weather stations plus 72 hour ECCC weather forecasts for the same station locations. The six primary outputs of the FWI subsystem are numerical moisture codes for the forest-floor fine-fuel layer (i.e., leaf litter, needles, cured grass, small twigs; 1.2 cm deep), duff layer (layer of partially/fully decomposed organic materials beneath the fine-fuel layer; 7 cm deep), and deep duff layer (beneath the duff layer; ~18 cm deep), an initial spread index (a function of surface wind speed and fine-fuel moisture content), total subsurface fuel available for combustion, and a combined FWI numerical value (Van Wagner, 1987; https://cwfis.cfs.nrcan.gc.ca/background/summary/fwi).

As of 2021 the fire hotspot data needed by the Fire M3 subsystem are obtained from the 4-µm and 11-µm infrared channel imagery of two radiometer instruments carried on four U.S. polar-orbiting satellites: (i) the Moderate Resolution Imaging Spectroradiometer (MODIS) on two NASA satellites; and (ii) the Visible Infrared Imaging Radiometer Suite (VIIRS) on two more NASA satellites (https://cwfis.cfs.nrcan.gc.ca/background/dsm/fm3). One to two retrievals covering all of Canada, including remote areas, are available from each satellite each day. However, hotspot detection may not be possible due to cloud or thick smoke obscuration of the surface or if fire size or intensity is small, so not every fire, even large fires, will be detected every day. In addition, internal data filtering and measurements at other wavelengths are applied to remove false positives (e.g., industrial sources, roofs of large buildings, cloud edges). To support ECCC the CFS has extended CWFIS capabilities to provide fire hotspots over the U.S. as well as Canada throughout the year instead of just the May to September period, which historically was the typical wildfire season in Canada. Note that the satellite measurements have a time lag or latency between one and seven hours due to the time required for image acquisition and distribution, hence the NRT descriptor.

The third major CWFIS component is the FBP subsystem, which uses FWI system outputs together with fuel type and topography to predict fire rate of spread (ROS; m min^{-1}), fire intensity (kW m^{-1}), crown fraction burned and fire type (crown fire or surface fire), and total fuel consumption (kg dry biomass m^{-2}) at hotspot locations (https://cwfis.cfs.nrcan.gc.ca/background/summary/fbp). Sixteen fuel types such as Boreal Spruce and Ponderosa Pine–Douglas Fir are considered (e.g., https://cwfis.cfs.nrcan.gc.ca/background/fueltypes/c2).

S3.1 Forest fire emission forecasts

CFFEPS v4.1 includes the source code for the FWI and FBP subsystems, which allows meteorological forecast fields from GEM to be considered for every hour and not just for noontime as in the CFS operational version of the CWFIS. Hourly biomass burning (BB) emissions are then estimated in CFFEPS v4.1 using a bottom-up, process-based approach, where emissions per fire (or hotspot) per day are the product of estimated burn area per hotspot, estimated fuel consumption per unit area, and emission factors for different chemical species and combustion stages (Chen et al., 2019a,b). Hotspot and associated fuel type information for the previous 24 hours is provided by the CWFIS Fire M3 subsystem. Initial daily burn area per hotspot is a prescribed value by province and fuel type that is updated periodically by CFS based on their analysis of CWFIS historical averages of reported burn areas by eco-region and fuel type; it is assumed to remain constant for each day of the forecast (i.e., persistence). Fuel consumption values are estimated by fuel type via the FBP subsystem driven with forecast meteorology. The incorporation of the FBP subsystem allows CFFEPS v4.1 to account for spatially variable biomass/fuel inputs using forest composition data from Canada's National Forest Inventory (Beaudoin et al., 2014, 2018) (Chen and Menelaou, 2021).

To calculate fire emissions, CFFEPS v4.1 uses FBP fuel combustion estimates to account for emissions from three combustion stages: flaming, smoldering, and residual combustion (Chen et al., 2019b). A look-up table is used to allocate fuel consumption between these different stages (Table 2 of Chen et al., 2019b), which have different burn durations: 15 minutes for flaming followed by several hours for smoldering and several hours for residual combustion, depending on the thickness of the forest-floor organic duff layer (Chen et al., 2019b). Different emission factors, which are constant parameters representing grams of emissions per kilogram of fuel consumed, are applied to estimate emissions from a wildfire for each combustion stage for eight chemical species: CO, CH_4, NO_X, NH_3, SO_2, non-methane hydrocarbons (NMHC), $PM_{2.5}$, and PM_{10} (Table 3 of Chen et al., 2019b). Chemical speciation is further applied for NMHC emissions (cf. Sect. S2) to calculate ADOM-2 VOC species emissions using the U.S. EPA SPECIATEv4.5 speciation profiles for flaming (#95425) and smoldering and residual combustion (#95428) (https://www.epa.gov/air-emissions-modeling/speciate-1; Chen et al., 2019a). Similarly, $PM_{2.5}$ and PM_{10} emissions from CFFEPS are subdivided into six PM chemical components with the following EPA SPECIATEv4.5 wildfire PM speciation profile: POM (78.5%), EC (9.5%), CM (9.7%), SU (1.3%), NI (0.1%), and AM (0.9%) (Chen et al., 2019a).

Fire emission forecasts were calculated by CFFEPS v4.1 prior to each RAQDPS-FW023 forecast simulation using the latest hotspot information for the past 24 hours from the operational CFS version of CWFIS. To obtain 72 hour forecasts of hourly BB emission values, current hotspots were assumed to persist for the next 72 hours and hourly changes in biomass fuel consumption were parameterized for a 24-hour period based on burn duration for the three combustion stages, fire ROS, and the average diurnal variation of the moisture content of the forest-floor litter layer, which increases as moisture content decreases (Chen et al., 2019b).

In the previous version of CFFEPS, v2.06, the diurnal distribution of daily burn area, which is used to allocate daily to hourly emissions, was estimated by a fixed weighted-normalization approach using a predefined diurnal profile, with peak fire growth at 16:00 local standard time (LST) and minimal near constant growth between 0:00 to 8:00 LST (Chen et al., 2019b). In CFFEPS v4.1, however, the diurnal distribution of daily burn area was determined based on a weighted moving average of the ratio of hourly fire ROS as estimated by the FBP subsystem to the sum of hourly ROS values for the previous 24 hours (Chen and Menelaou, 2021). This new scheme should be more realistic as it is based on hourly forecast meteorology rather than a static temporal profile.

S3.2 Plume rise

Plume-rise schemes for smokestacks like the Briggs (1984) scheme (cf. Sect. 3.13) have also been used to estimate wildfire plume rise, but wildland fires are much broader than smokestacks and they lack any initial vertical velocity; instead, they generate their own buoyant plume. CFFEPS v4.1 uses an alternate thermodynamic approach described by Anderson et al. (2011) to parameterize wildfire plume rise. Hourly plume injection height is estimated based on a fire energy balance such that the buoyancy of a smoke plume is thermodynamically balanced with the forecast vertical temperature profile of the atmospheric column above the fire while accounting for fuel moisture evaporation and condensation.

In earlier versions of CFFEPS, the plume-rise scheme was based on lifting the smoke plume dry adiabatically at a constant rate at hotspot locations and then equilibrating the buoyant plume with the local environmental lapse rate derived from a simplified vertical temperature profile determined by temperatures on five pressure levels: surface, 850, 700, 500, and 250 hPa (Chen et al., 2019b). CFFEPS v4.1 instead uses the higher-resolution forecast vertical temperature profiles that are available on the RAQDPS-FW023 84-layer vertical grid (Sect. 2.1). The thermodynamic calculation of plume heat flux in CFFEPS v4.1 was also updated to account for the heat lost due to the latent heat of evaporation of fuel moisture during combustion. At the same time, as the plume passes the atmospheric lifting condensation level, the latent heat of condensation of plume water vapour is added to the energy of the plume aloft to obtain the final injection height within the atmospheric column. These detailed physical processes are considered hourly for each hotspot as the fire thermal energy and the atmospheric vertical temperature profile change, and the final hourly plume injection height is used to prescribe the vertical distribution of emissions within the column below the plume injection height (Chen et al., 2019b; Chen and Menelaou, 2021).

S3.3 Outputs and file structure

CFFEPS v4.1 outputs are saved in one file similar in format to the anthropogenic major-point-source emissions input file. The outputs are estimated hourly plume injection height (ZPLM meters), plume vertical distribution ratio (RSMOKE), and hourly speciated emissions (g s^{-1}) over the 72-hour forecast period at each hotspot location as specified by latitude/longitude. There are 21 ADOM-2 gas-phase and 12 particle-phase emitted species whose emissions are output (identified by a leading "E"): ECO, ECO2, ENO, ENO2, ENH3, ESO2, ESO4, EA2, EA3, EALD,

EARO, EC38, ECRE, EETH, EHCH, EISO, EMEK, ETOL, EOTH, EC1, EC2, EAM1, EAM2, ECM1, ECM2, EEC1, EEC2, ENI1, ENI2, EPC1, EPC2, ESU1, and ESU2 (cf. Table 3). Note that ECO2, EC1 (methane), and EC2 (ethane) are not needed by the ADOM-2 gas-phase chemistry mechanism but may be used in other chemistry parameterizations. This BB emissions file was input directly to the RAQDPS-FW023, which then allocated the emissions to model surface and elevated grid cells.

S3.4 Limitations

CFFEPS v4.1 has a number of limitations that should be kept in mind (Chen et al., 2019a). First, as already noted above some fires may not be detected due to (i) their small size, (ii) their initiation after the last available satellite overpass, or (iii) the presence of thick cloud or heavy smoke above the fire. Second, detected fires are assumed to continue burning but to remain the same size over the duration of the forecast. Third, fire emissions are treated as point sources and hence are assigned to a single grid cell. And fourth, wildfire combustion heat and emitted smoke are assumed not to affect meteorological fields, which may not be a good assumption for large, intense fires.

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