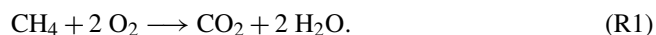


of the water emitted due to combustion. Wren et al. (2023) calculated the average ratios of CO_2 to NO_x emission rates from OSM 2018 aircraft campaign data for individual OS facilities and source types (e.g. stack, area). For this work, the CO_2 : NO_x ratios estimated by Wren et al. (2023) for the stack sources were used in turn to estimate CO_2 emission rates from NO_x reported in NPRI and CEMS. CO_2 and H_2O are primarily generated from combustion of natural gas, with methane (CH_4) as its main component, in OS production operations:



Therefore, for every mole of CO_2 , 2 moles of H_2O is emitted due to combustion. Accordingly, a stoichiometric ratio of 1 : 2 of CO_2 to H_2O can be used to estimate H_2O emissions levels, as was done for this work. H_2O emissions were then calculated from NPRI- and/or CEMS-reported NO_x emission rates based on source-specific CO_2 to NO_x ratios. For the period corresponding to the aircraft study, the continuous emissions monitoring system (CEMS) hourly data were available for SO_2 and NO_x for only two of the OS Suncor stack sources and for SO_2 for the other facilities/stacks. Canadian emissions reporting requirements for NPRI reporting for large stacks are for annual totals. Therefore, the hourly NO_x and consequently hourly H_2O for the rest of the facilities were estimated from NPRI annual emissions data. CEMS hourly data for stack parameters (e.g. exit temperature, flow rate) and SO_2 emission rates were available for April to July 2018, partially overlapping with the period of our 6-month run simulations from February to July 2018, and were used in the simulations for the same period. We note that the estimation of stack water emissions is a required input for our algorithm – the methodology demonstrated here is easily expandable to other combustion stack sources. Knowledge of the fuel type is required, with different fuels having different amounts of water produced per carbon atom combusted – i.e. Reaction (R1) depends on the fuel used for generating heat for stack emissions. As we will discuss below, the accuracy of the stack emissions and the consequent estimates of water emissions have a key impact on the accuracy of our plume rise algorithm. Note that we used the estimates of combustion-generated water as described above in our simulations (both standalone and GEM-MACH simulations, with PRISM) for the specific stack sources for which the following information was available: (a) reported NO_x and SO_2 emission rates (CEMS or NPRI) and (b) facility-specific estimates (aircraft-based) of CO_2 to NO_x emission ratios. Such source emission information was not available for the majority of the stack sources within our large-scale GEM-MACH modelling domain (10 km resolution domain over North America, 2.5 km resolution domain over Alberta and Saskatchewan). Nevertheless, in our GEM-MACH simulations with PRISM (GM-PRISM), the plume rise from major point sources, including those without combustion-

generated water data, was also impacted by the moist thermodynamics of the entrained water from ambient air.

2.5 Aircraft campaign and WBEA surface monitoring network

During the OSM 2018 campaign (April to July), aircraft-based measurements of environmental variables (meteorology, pollutant concentrations) were conducted over the Canadian oil sands (OS) (ECCC, 2018). Figure 1b shows the flight tracks taken by the aircraft during the OSM 2018 campaign over the OS region. The aircraft conducted several flights during different days and times from April to July 2018, including single screen flights tens of kilometres downwind of OS facilities and box flights around the facilities at near range. The designation box flight refers to a flight pattern during which the aircraft would fly along closed loops around a specific emitting facility at several consecutive altitudes while making measurements of environmental variables. The box flights were specifically designed to capture emissions from individual facilities. Aircraft-measured data during box flights were converted into source emission rates through flux estimations and mass-balance calculations, utilizing the Top-down Emission Rate Retrieval Algorithm (TERRA) algorithm described in Gordon et al. (2015). For further discussion on the application of TERRA and the uncertainties in emission rate retrievals based on aircraft measurements, see Fathi et al. (2021). This was done for several emitted species such as SO_2 , NO_x , and CO_2 . As discussed in Sect. 2.4, aircraft-based estimates, emission inventory data, and continuous emissions monitoring system (CEMS) data for NO_x were used to derive the NO_x to CO_2 emission rate ratio, which in turn was used to estimate the water emissions rate.

Here, we also used aircraft measurements of SO_2 concentrations downwind of several oil sands facilities (CNRL, Syncrude, and Suncor) to determine observed plume heights and evaluate our model-predicted plume rise (using both GM-orig and GM-PRISM) vs. these observations. For our analysis, we considered aircraft data from box flights where measurements were made just a few kilometres downwind or upwind of emission sources. This was done to avoid flights that included a large long-range transport path/time of emitted pollutant to the point of measurement so that the observed plumes would be a better representation of emission and plume rise conditions at the stack locations. We focused on SO_2 as the emitted pollutant, since it is a primary emitted pollutant (i.e. not generally generated due to photo-chemical reactions in the atmosphere) and due to the availability of CEMS-based direct observations of SO_2 within emitting stacks. SO_2 in oil sands (OS) regions is mainly emitted from large high-temperature stack sources (over 90 % of the emitted SO_2 in the region originates in the large stacks, unlike NO_2 , only about 40 % of which is emitted from large stacks; Zhang et al., 2018), with low background levels from other