Estimates of atmospheric deposition of polycyclic aromatic compounds at local and regional scales in the Athabasca oil sands region

Leiming Zhang[1], Zhiyong Wu[1], Irene Cheng[1], Tom Harner[1], Xin Qiu[2], Fuquan Yang[2], Erin Horb[2],

[1]Air Quality Research Division, Science and Technology Branch, Environment and Climate Change Canada, 4905 Dufferin Street, Toronto, Ontario, M3H 5T4, Canada (leiming.zhang@canada.ca)
[2]Novus Environmental Inc., 150 Research Lane, Suite 105, Guelph, Ontario, N1G 4T2, Canada

To assess ecosystem health and cumulative environmental effects associated with mining activities in the Athabasca oil sands region, western Canada, a monitoring program was established in 2011 to measure polycyclic aromatic compounds (PACs), including 17 polycyclic aromatic hydrocarbons (PAHs), 21 alkylated PAHs, and 5 parent and alkylated dibenzothiophenes (DBTs) at 17 passive and three active sampling sites. One of the primary deliverables of this project is to provide estimated total deposition of various pollutants at local and regional scales.

A dry deposition framework was developed to estimate dry deposition of PACs, and was first applied to the three active sampling sites. Modelled dry deposition velocities for various gaseous PACs and over various land covers surrounding the three sites were mostly in the range of 0.01-0.5 cm s^{-1} with median and annual mean values between 0.08 and 0.24 cm s^{-1}, comparable with literature values obtained from field studies. Annual dry deposition of the sum of PAHs (including both gaseous and particulate forms) was estimated to range from 330 to560 μg m^{-2} over forested canopies surrounding the three sites and from 270 to 490 μg m^{-2} over grass and shrubs. The corresponding values are 3920 to 5380 μg m^{-2} and 2850 to 4920 μg m^{-2} for the sum of 21 alkylated PAHs, and 230 to 1120 μg m^{-2} and 450 to 930 μg m^{-2} for the sum of 5 DBTs. The three monitoring sites are situated nearby the Athabasca River, and the direct annual atmospheric dry deposition to water surface was estimated to range from 350 to 500, 3170 to 4530, and 170 to 840 μg m^{-2} for PAHs, alkylated PAHs, and DBTs, respectively. Alkylated PAHs contributed 80% of the total dry and 60% of the total wet deposition budget, suggesting the importance of including this group of PAHs in the atmospheric deposition budget estimation for subsequent ecosystem impact studies.

In order to produce dry deposition estimates at the regional scale, gridded air concentrations of individual PACs are needed. These are obtained through model simulations using the U.S. EPA's CALPUFF model and based on available monitoring data. The model horizontal grid resolution is chosen as 4 km by 4 km with the domain covering most of the oil sands area. Emissions of PACs were based on the Cumulative Environmental Management Association (CEMA) 2010 emissions inventory and U.S. EPA's speciation profiles (Speciate 4.0). Model results were compared at the regional scale with the 17 sites passive data, then compared at the three sites with both active and passive data at the time resolution of the passive samples (bimonthly), and finally compared at the three sites with the active data at high time resolution (daily). On an annual basis and at the regional scale, modelled total PAH concentrations in air are within a factor of 1.2 of measurements and within a factor of 2 of the total of measured alkylated PAHs. Modelled DBTs concentrations are much lower than monitored levels due to the lack of speciated emissions data. Bias correction techniques were then applied to the modelled air concentrations based on available measurement data to generate spatial distributions of air

concentrations of individual PACs species. This approach which combines emissions, measurements and a model shows promise for the assessment of PAC concentrations in air and deposition across the entire oil sands region. Further development of alkylated PAH and DBT emissions is needed to improve modelling results.

Wet deposition monitoring data were only available at the three active air sampling sites. Gridded wet deposition for the whole domain will be estimated in future work using modelled air concentrations, forecasted precipitation amounts, and PAC species-dependent scavenge ratio values. The scavenging ratio was generated from the concurrent monitored precipitation chemistry and air concentration data at the three sites, which showed median values of 6100-$1.1x10^6$ for snow scavenging and 350-$2.3x10^5$ for rain scavenging depending on the PAC species.