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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Emissions databases for polycyclic aromatic compounds in the Canadian Athabasca Oil Sands Region – development using current knowledge and evaluation with passive sampling and air dispersion modelling data" *by* Xin Qiu et al.

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Response to Referee #3

We appreciate the comments by the reviewer to help us improve the paper. Our responses to the specific comments are shown below.

This research work compared CALPUFF modelling results applying the two air emis-

sions databases of CEMA and JOSM programs. The modelling results are then compared with observations to evaluate accuracy of the air emissions values. This research makes significant contribution to the work of PAHs air emission estimation in the oil sands region. While dispersion models could have systematic error existing inherently in the model, particularly and usually lead to underestimation at low pollutant concentrations, this research presents a progressive approach to compare the modelling results relatively for the original emissions data and the improved one. I suggest to publish it to make colleagues working in this field be aware of the work progress.

It would be clearer if the author could add more information on meteorology and emission summary.

Response: A summary of the meteorological model that drives the CALPUFF model will be provided in sect. 2.2 of the revised paper.

CALPUFF takes three-dimensionally varying wind, temperature and turbulence fields from the CALMET model. The 3-D winds and temperature fields from CALMET are reconstructed using meteorological measurements, orography and land use data. Besides wind and temperature fields, CALMET determines the 2-D fields of micrometeorological variables needed to carry out dispersion simulations (mixing height, Monin Obukhov length, friction velocity, convective velocity, etc.). A two-step approach is typically used to compute the wind fields in CALMET. In the first step, an initial guess wind field is adjusted for kinematic effects of terrain, slope flows, and terrain blocking effects to produce a Step 1 wind field. The second step applies an objective analysis procedure to introduce observational data into the Step 1 wind fields to produce the final wind fields. In this study, CALMET used the Weather Research and Forecasting (WRF) model due to its capability of simulating regional flows and certain aspects of local meteorological conditions such as complex terrain. It replaces the two-step approach because of the higher spatial resolution of the WRF output compared to observational data. The output of the CALMET model is directly interfaced with the CALPUFF dispersion model for further air quality modelling.

A paragraph summarizing the PAC emissions will be included in the beginning of the Results and Discussion (sect. 3.1) and is provided below:

3.1 PAC emissions estimates

Unsubstituted and alkylated PAHs and DBTs emissions from oil sands development and non-industrial sources were estimated over the model domain (Table 1). Total unsubstituted PAH emissions (2009-2014) are estimated to be 56 to 58 tonnes yr-1 based on emissions from tailings ponds, mine face, mine fleet, residential, commercial, local traffic, airport, point, and transportation sources. Point sources accounted for most of the total unsubstituted PAH emissions (75-77%). The major difference in the total unsubstituted PAH emissions between the CEMA-derived and JOSM-derived emissions databases is the higher evaporative PAH emissions from tailings ponds and mine face in the JOSM-derived emissions database. Alkylated PAH and DBT emissions (2011-2014) are estimated to be 17 tonnes yr-1 and 0.26 tonnes yr-1 respectively; however, they consisted of fewer emission sources (tailings ponds, mine fleet and transportation sources) due to a lack of PAC speciation data to estimate emissions from other sources. Nevertheless, the PAC emissions estimates in this study may still be underestimated, e.g. tailings pond and fugitive dust emissions. A common technique for measuring tailings pond emissions is a flux chamber. However, recent studies suggest that this technique underestimates organic compound emission fluxes (Tran et al., 2018). Windblown petcoke dust observed recently over surface mining areas in the AOSR (Zhang et al., 2016) also have not been accounted for in the PAC emissions databases.

Additionally, PAHs have a wide spectrum including compounds in gaseous phase and particulate phase, which can exhibit different characteristics during transport and deposition. Although the research is focused on relative comparison of two emissions databases with only considering dispersion, the author may analyze qualitatively the resultant impact of turning off deposition modelling on modelling results in general.

Response: We decided to present the model results without deposition processes after running a few model scenarios. One of the impacts of turning off deposition modeling is that the modeled air concentrations are higher compared to those with deposition modeling turned on. However, we found that the modeled concentrations from simulating emissions, transport and dispersion processes, but without deposition processes, are already lower than measurements, demonstrating that the emissions inputs are conservative or underestimated. If deposition processes were included, modeled concentrations would be even lower than measurements; however, in this model scenario it would be hard to say if this was caused by too low emissions input or too high deposition rates, knowing that large uncertainties exist in treating dry and wet deposition processes. For example, there are large uncertainties in the PAC dry deposition velocities (Zhang et al., 2015a), PAC scavenging ratios for snow and rain scavenging of gas-phase and particulate-phase PACs (Zhang et al., 2015b), and scavenging coefficients of aerosols in general by snow and rain scavenging processes (Zhang et al., 2013; Wang et al., 2014). Our next study related to this project is to compare the deposition output using various approaches.

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