General responses from the authors:

We greatly appreciate the time and effort spent by both reviewers in providing their helpful comments. One comment shared by both reviewers is that some parts of the text need to be shortened. We have taken this comment seriously and have shortened the paper accordingly. We are also very grateful for other general and specific comments from the reviewers, and we believe that by addressing these comments the manuscript has been substantially improved. In addition, revisions were also made to address comments received externally from Canada's Oil Sands Innovation Alliance (COSIA) and internally from two other divisions within our organization (Environment and Climate Change Canada), who are responsible for compiling Canada's national emissions inventories of criteria air contaminants and greenhouse gases.

Below are our point-by-point responses (in blue font) to all of the review comments:

Anonymous Referee #1:

General comments:

The authors describe the anthropogenic and biogenic emissions datasets developed for the global air quality model GEM-MATCH to simulate air quality (AQ) in summer 2013 over the Athabasca Oil Sands Region (AOSR) of Canada. The paper provides a detailed description of the number of datasets and emission inventories that are used to generate a new hybrid emissions inventory for high-resolution AQ modeling over the AOSR. I recommend the manuscript for final publication in ACP after addressing these questions and comments

We appreciate your comments very much. See below for our responses to your specific comments.

Specific Comments:

1) The text needs to be shortened, e.g. some parts (Abstract, Summary) are too long and repetitive.

   Following your comment and the other reviewer's comments, we have shortened the Abstract and the Summary and Future Work sections, moved Sections 2.1.1 and 2.1.2 to the Supplemental Material, and revised some other sections of the paper.

2) The paper uses too many acronyms that are hard to follow. I suggest adding a table to introduce all the acronyms that are used in the paper.

   When we were preparing this manuscript, we were also concerned about this problem and therefore we included a table (Table S1) in the Supplemental Material to define all of the acronyms. We have now moved that table to the main paper as an Appendix to make it more accessible to the reader.

3) One of key improvements of the new emission dataset is the improved biogenic VOC emissions due to the use of the new land use map (forest clearing and water and ponds). The regional and global AQ models typically use the outdated static vegetation and LAI maps, hence introducing large uncertainties to the biogenic VOC simulations. Was the land-use map modified to improve the meteorological simulations as well?

Yes, we also modified the land-use map to improve the meteorological simulations. To address this point we have added one more plot in the Supplemental Material (Figure S3) to show the impact of the modified land use on model-predicted PBL height. The impact can be a factor of two or more.

4) The paper reports that the aircraft did measure high isoprene from the Suncor Millenium/ Steepbank and the CNRL Horizon facilities. What are the sources emitting the high amount of isoprene?

Based on aircraft observations and preliminary lab test results, the isoprene seems to come from bitumen vapor emissions. We are curious about the isoprene source as well and are conducting further field studies this year (2018). We have made two text additions: "likely originated from bitumen vapor emissions from" and "Further studies are needed to confirm the source of non-biogenic isoprene emissions" to Page 15, Lines 22 and 24-25 (note that page and line numbers have changed after the revisions to the paper).

5) The paper mentions Stroud et al., 2017 study to model SOA over the AOSR by using the emissions inventory developed in this study. There a number of uncertainties in the emission inventories that affect the modeled SOA levels. First, does this emission dataset include intermediate VOCs (IVOCs) emissions from the anthropogenic sources in the AOSR? How various long-chain alkane and other species are lumped in the developed inventory, which can affect the SOA production in the model? Did the emission dataset characterize the semi-volatile organic species (SVOCs)? This also depends on the volatility distribution of the primary OA emissions. Not sure if the POA is assumed to be non-volatile in this dataset. Are the improvements for such SOA precursors (S- and I-VOCs) in this new emission development over the existing inventories used for the regulatory purposes?

IVOC and SVOC species were not measured by the aircraft. Canister samples of VOCs up to C12 and the aerosol mass spectrometer (AMS) PM_1 organic aerosol were measured. The box flights encircling individual facilities were able to calculate net fluxes of VOCs and PM_1 organic aerosol from entire facilities based on the measured concentrations and wind speeds. These measurement-derived VOC emissions were used as input to the GEM-MACH sensitivity simulation performed by Stroud et al. and lumped to a long-chain alkane (ALKA), two aromatic species (AROM, TOLU), a long-chain alkene (ALKE) and non-volatile POA. In GEM-MACH, the long-chain alkane has an SOA yield representative of a C10 species, but only a small fraction of the ALKA lumped species is assumed to be C10 or greater, so GEM-

MACH does not account for the IVOC SOA formation by lumping the IVOCs into the ALKA species.

The aircraft observations suggest POA emissions and/or rapid SOA formation over and just downwind of the open pit mines. It is not clear the relative proportion of each. The O/C ratio of the organic aerosol over or just downwind of the open pit mines suggests it is already partially aged, as a ratio greater than 0.4 was already observed. This favours the rapid SOA formation mechanism from reactive precursors such as anthropogenic VOCs and/or from the SVOCs that only need one oxidation step to reach an O/C ratio of 0.4 (based on a box model study). The IVOCs likely need more time than one hour after emission to start dominating the SOA production. The previous box model study, by Liggio et al. (2015), was initialized at the first transect of a downwind Lagrangian flight pattern and a large concentration of organic aerosol was already measured at this point. The box modelling suggested that high concentrations in the IVOC range were needed thereafter to account for the near-steady organic aerosol concentration, where continued SOA production was needed to balance dilution loss in the plume.

It is likely that the model emissions are missing the IVOCs and that the aircraft-measured organic aerosol already contains some SOA from the reactive anthropogenic VOC precursors and from the SVOCs emitted that only need one oxidation step to form low-volatile material. This rapid SOA mass production would be, at least partially, accounted for in the new model POA emissions measured by aircraft box method. The current GEM-MACH version does not include the volatility basis set (VBS) approach to account for POA volatility, but rather assumes the POA to be non-volatile. A research version of GEM-MACH has recently been coded with the VBS approach; however, the IVOC emission factors remain highly uncertain as does the IVOC/SVOC aging scheme. A new aircraft field study in the Oil Sands region is planned for this year (2018) that will measure IVOC gaseous species over the open pit mines, as well as employ an electrospray-ionization AMS to measure organic aerosol composition. This will provide additional information to better address these uncertainties.

6) The new emission dataset also includes some emission estimates based on the aircraft measurements and mass balance approach. I think the authors need to put more emphasis on the use of the top-down emission estimates in the paper. As the Summary section discusses, there are some uncertainties associated with the top-down emission datasets. However, in the text it isn't clear the distinction between the top-down and bottom-up emission datasets and their use in the AQ models.

   This is a very good point. We have added the modifier "top-down" to the titles of Sections 4.2 and 4.3 and revised the contents of these sections to differentiate between the top-down and bottom-up emissions estimates and to emphasize the top-down emissions.

7) There were studies in the US to improve the emission inventories for the oil and gas sector and simulate air quality by taking advantage of the top-down emission estimates for NOx,

CH4 and VOCs from the oil and gas sector. Unfortunately, the findings of those studies aren't discussed in this paper. Below are some references:

Thank you very much for pointing out these studies in the U.S. We found three of them to be very relevant to our study and we have cited them in Sections 4.2 and 4.3 when aircraft-measurement-based VOC and PM emissions estimates are discussed.

Anonymous Referee #2:

General comments:

As it stands, this paper is largely a straightforward emission development application and its format resembles a report more than a scientific paper. The abstract is very lengthy similar to a report executive summary. Same information is provided repetitively. The paper needs to be a lot more concise especially when the same information is available elsewhere (references). If qualitative arguments like "significantly impacted" are used to characterize the results, they need to be supported by facts and figures. While subject to a variety of weaknesses and not developing any new approaches or containing extremely unique analyses, it is a useful paper and fits well in this special issue. Essentially, it serves as a basis for other papers in the same special issue. The paper could be improved by presenting a linkage to uncover what lessons are learned from the air quality applications that have used this inventory.

Thank you very much for these comments. As you point out, this emissions paper serves as a foundation for several of the modelling papers in this special issue and hence it makes a useful contribution to this special issue. Following your specific comments, we have shortened the Abstract and the Summary and Future Work sections, moved Sections 2.1.1 and 2.1.2 to the Supplemental Material, and revised some other sections of the paper. We have replaced some descriptions like "significantly impacted" by more fact-based statements. We also linked this paper more closely with the air quality modelling papers in the special issue to summarize what emissions-based lessons were learned from this research activity. More details are provided in the responses to your specific comments.

Specific Comments:

1) Abstract - The current form is too long and is read like a 2-page executive report summary rather than a research abstract. Focus on key messages and make the abstract more concise.

   We have shortened the abstract by more than one-third.

2) Page 4 – The summary on the first two phases is repetitive. Much of the text in the first paragraph can be excluded. Page 6-8 already give more than sufficient background information (which can still be further condensed in my opinion).

We agree that much of the text in the first paragraph is repetitive. We have condensed this paragraph (from 407 words to 280 words). We have also moved most part of Section 2.1 ("Review of emissions inventories used for JOSM Phases 1 and 2 AQ modelling", Pages 6-8, now Page 5) and two corresponding tables (Tables 1 and 3) to the Supplemental Material.

3) Page 5 (from Line 14-25) – This section is not needed.

This section briefly describes the following sections of the paper. As a common practice, it serves as a transitional paragraph from the Introduction section to the main body of the paper. We did not remove this section completely, but we did shorten it considerably.

4) Page 6 (Line 28) If a setup for processing 2006 APEI already exists, it is not clear why 2010 APEI could not be formatted to leverage that. Information in the APEI is simple, e.g., province, SCC, pollutant, emissions.

Canada's Air Pollutant Emission Inventory (APEI) is a comprehensive anthropogenic emissions inventory that is prepared to fulfill various national and international reporting obligations and to provide data for air quality modelling. However, additional preparation steps are needed to transfer the emissions database underlying the published APEI data to a format that is suitable for processing emissions for AQ modelling, such as a further breakdown to more detailed source types for SCC assignment. More detailed emissions data are also needed than are stored in the annual APEI, such as (a) monthly on-road and off-road emissions and (b) process-level reporting such as on-road evaporative emissions vs. exhaust emissions. Other countries have similar gaps between their national reported inventories and their emissions-processing-ready inventories. For example, the U.S. EPA compiles emissions for their policy modelling platform (https://www.epa.gov/air-emissions-modeling/emissions-modeling-platforms) based on their NEI (National Emissions Inventory, https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei). We have added the following explanation to this line: ", which requires the published APEI data to be transferred to a format that is suitable for processing emissions for AQ modelling as well as the addition of more detailed emissions data, such as monthly on-road and off-road emissions and process-based separation of emissions from some sectors (e.g., evaporative vs. exhaust emissions from on-road vehicles)" (now in Section S1 of the Supplemental Material, Page 1, Lines 22-25).

5) Page 7 (Line 12) Why didn't each facility level scale up by facility-specific scaling factor as supposed to a uniform constant of 2.6? For example, VOC emissions from the CNRL facility should have scaled up by about a factor of 10. Would not this offer better spatial representation? Also, were there particular VOC sources missing in the CEMA inventory? The VOC discrepancy is surprising given that the CEMA inventory is a bottom-up inventory. I suspect that the 2010 NPRI values were too high considering much lower estimates in the 2013 NPRIv2. This point should be clarified because this updated inventory relied heavily on both CEMA and NPRI. The referenced report does not address this point.

This is a good question. First of all, we do not think that the CEMA inventory misses any significant sources of VOC emissions, but it is a static, "one-off" inventory. For the ongoing NPRI inventory, whose preparation is a requirement under the authority of the Canadian Environmental Protection Act, owners or operators of facilities that meet published reporting requirements are required to report to the NPRI annually. However, various estimation methods with different uncertainties may be used. (See https://www.canada.ca/en/environment-climate-change/services/national-pollutant-release-inventory/publications/guide/exemptions-exclusions.html#a3_8 for the list of methods that the facility can use to estimate emissions.) For the same activity data, different estimation methods can result in different emission estimates for a bottom-up inventory. Facilities are also allowed to update their emissions after the official NPRI inventory is published if they deem it necessary due to the availability of new information or new estimation methodologies. As a result, the emissions reported by a facility for a given year can vary from inventory version to inventory version. In fact, as you commented below, you found the "difference between version 1 and 2 of the 2013 NPRI to be disturbing", but we view this aspect of the NPRI to be positive since it allows better emissions estimates to be included after the initial submission.

It is therefore difficult to choose the most suitable inventory. Before Phase 1, our understanding of the emissions from the Oil Sands facilities was limited. Our decision in Phase 1 to scale the CEMA VOC emissions to the 2010 NPRI level for the OS facilities using a uniform factor was based mainly on three considerations: (1) emissions in the NPRI inventory were required by law to be reported by each facility, and we assumed that each facility is best placed to know its own emissions; (2) the CEMA inventory had the lowest total VOC emissions for these five facilities compared to four other inventories (ECCC & AEP, 2016); and (3) large uncertainties may be present in the reported NPRI inventory (and also in the CEMA inventory) and the use of a uniform scaling factor will not affect the impact of VOC emissions from the OS facilities as a whole. We had already explained the first and second reasons in the paper and we have now added the following text to clarify the third reason: "as large uncertainties may exist in both inventories and the use of a uniform scaling factor should not affect the impact of VOC emissions from the OS facilities as a whole" (now in Section S1 of the Supplemental Material, Page 2, Lines 7-8). Note that after we gained more knowledge of the emissions from this region through Phases 1 and 2 of the study, we treated VOC emissions separately for each facility in Phase 3.

6) Page 8 (Line 1) Treating industrial non-stack sources as area sources (e.g., distributing within the facility boundary and specific to source type) is appropriate. However, this is quite a standard approach, NOT a new approach as claimed.

To the contrary, our understanding is that treating non-stack sources in an individual industrial facility as area sources is not a standard approach. Such emissions are usually allocated to a single grid cell based on the single pair of latitude–longitude values provided for the non-stack sources of that facility, which is clearly not optimal when a facility is very large and spans multiple grid cells. Moreover, it is also not a common practice to develop a

set of facility-specific, process-specific spatial surrogate fields for each facility to allocate non-stack source emissions. We believe our approach to be unique; at least we are not aware of any other groups that have applied a similar approach, which may not be that surprising given the unusually large spatial scale of the oil sands facilities considered in this study and the fine horizontal grid spacing (2.5 km) that was used by the model.

7) Page 10 (first paragraph) The US EPA's trend suggests a reduction of NOx by 8% and SO2 by 23% between 2011 and 2013 [https://www.epa.gov/air-emissionsinventories/air-pollutant-emissions-trends-data]. This should be mentioned in the paper.

Thank you very much for pointing out this. We have added the following sentence to the end of this paragraph:
"Note, however, that the U.S. EPA's emissions trend data set suggests a reduction of NO_x emissions by 8% and SO_2 emissions by 23% between 2011 and 2013 (https://www.epa.gov/air-emissionsinventories/air-pollutant-emissions-trends-data)" (now Page 6, Lines 24-26).

8) Page 10 (Line 20) This comment is related to my comment made above on the 2010 NPRI inventory. I find the difference between version 1 and 2 of the 2013 NPRI to be disturbing. What is the level of confidence when using NPRI inventory? Would you recommend extra procedures to ensure that the NPRI values are representative?

As we mentioned in the paper and explained above, NPRI is the Canada's legislated inventory of large point sources and is based on emissions reported by facilities. Various methods with different inherent uncertainties can be used for estimation of emissions. We are confident with emissions derived from CEMS measurement, such as SO_2 and NO_x emissions, although even for that method, large uncertainty still remains when a facility experiences upset conditions and the CEMS instruments are bypassed. Emissions derived from Engineering Judgement or general Emission Factors, for example, are less reliable. We must trust that facilities have done their best to report their emissions, but we must also always keep in mind that the emissions reported may not be representative. Other inventories, such as the U.S. NEI, have similar uncertainties. This is one of the reasons for the 2013 OS field study: to allow independent emissions estimates to be produced based on aircraft observations, thus facilitating evaluation of reported NPRI facility emissions. However, such field studies are very difficult and expensive to undertake and are only possible under special circumstances for a very small number of facilities.

9) Page 11 (Line 16) The argument is that VOC emissions in the CEMA inventory are underestimated with no reasons given. What are your justifications of using the CEMA inventory to represent source profile (to allocate fugitive VOC emissions among mine faces, tailing ponds, etc.)? If the underlying assumptions of VOC estimates are vastly different between NPRI and CEMA, this approach would not be appropriate especially if VOC speciation profiles differ significantly among these sources.

We explained in Section 2.1.1 that the VOC emissions reported in the CEMA inventory might be underestimated (Page 7, Line 7-11, now in Supplemental Material, Section S1, Page 2, Lines 1-5). The relevant text reads:
"The 2010 NPRI was also used to scale the CEMA facility-total VOC emissions for the five AOSR surface mines active at that time (Figure 1), since it was found that the CEMA inventory had the lowest total VOC emissions for these five facilities compared to four other inventories (ECCC & AEP, 2016) and the NPRI is Canada's legislated inventory of large point sources based on emissions reported by facilities."

As mentioned in this same paragraph, fugitive VOC emissions are reported to NPRI as facility-total emissions without differentiation between source type (i.e., mine faces, tailings ponds, and extraction/upgrading plants), whereas VOC area-source emissions in the CEMA inventory are reported by process, including fugitive VOC emissions from tailings ponds, plants, and mine faces. We therefore used split factors from the CEMA inventory to separate NPRI VOC emissions by source type because there is no better information than the CEMA inventory available to estimate the relative contributions of sub-facility, process-level emissions. We explained this reasoning in the paper as well for Phase 3 (now Page 8, Lines 1-6), where the text reads:
"Surface-level fugitive VOC emissions are reported to NPRI as facility-total emissions without differentiation between source type (i.e., mine faces, tailings ponds, and extraction/upgrading plants). To distribute 2013 NPRI fugitive VOC emissions spatially within an OS mining facility, process allocation factors calculated from the process-specific fugitive VOC emissions in the 2009/10 CEMA inventory for each AOSR mining facility were used to allocate fugitive VOC emissions between mine faces, tailings ponds, and plants (similar to the procedure used in Phase 2; see ECCC & AEP, 2016)."

Aircraft measurements conducted during the 2013 field study are not helpful here because they can only be used to calculate emissions estimates at the facility level. We understand this limitation and we recommended this as an area for improvement in the Summary and Future Work section (Page 23, Lines 21-28). We have modified the text further for clarity. The text now reads (new text is shown in black font):
"Moreover, these aircraft measurements were carried out at the facility level, but within these very large facilities the individual VOC species emitted from mine faces, tailings ponds, and plants can be very different. More aircraft measurements, especially at other times of year, and additional measurements of emissions at the sub-facility level, from mine faces, tailings ponds, and plants for multiple AOSR facilities are needed to confirm and augment the findings of the 2013 field study and to further improve emissions factors, temporal profiles, and chemical speciation profiles used for OS emissions inventories and emissions processing(e.g., Small et al., 2015; Stantec Consulting Ltd. et al., 2016)."

10) Page 11 (Line 26) How was the 2011 UOG emissions projected to 2013? Page 12 (Line 5) From what is shown in Figure S2, the spatial distribution of UOG emissions does not change very much with a few exceptions. Perhaps, include UOG emission spatial plots to support your statement.

The projection from 2011 UOG emissions to 2013 is based on activity data and a methodology described in a Clearstone Engineering Ltd. letter report entitled "Documentation for UOG emission inventory extrapolation database (ExtrapolateR.accdb)". We have changed the sentence to read (new text shown in black font):
"This new subinventory was then projected by ECCC to 2013 for inclusion in the 2013 APEI based on activity data and a methodology described in a letter report from Clearstone Engineering Ltd. (2014d)." (now Page 8, Lines 17-19).
We have also added this report to the References section.

The majority of the facilities in the UOG inventory are conventional oil & gas wells and batteries, which are not abundant in the immediate vicinity of the AOSR. That is the reason why the spatial distribution of UOG emissions does not change very much in the original Figure S2. As suggested, we prepared some new UOG emissions spatial plots to support our statement that "some UOG facilities that existed in 2000 have been closed while many new facilities have opened since 2000". However, it is difficult to see in these new plots whether the differences are due to changes in the numbers and locations of facilities or to changes of emissions magnitude. We then plotted the spatial distribution of UOG facilities over a larger area near the AOSR as shown below. We believe these new plots, which are now both superpositions of the two UOG versions but in different order, support our statement well. We have replaced the old Figure S2 with these two new plots.

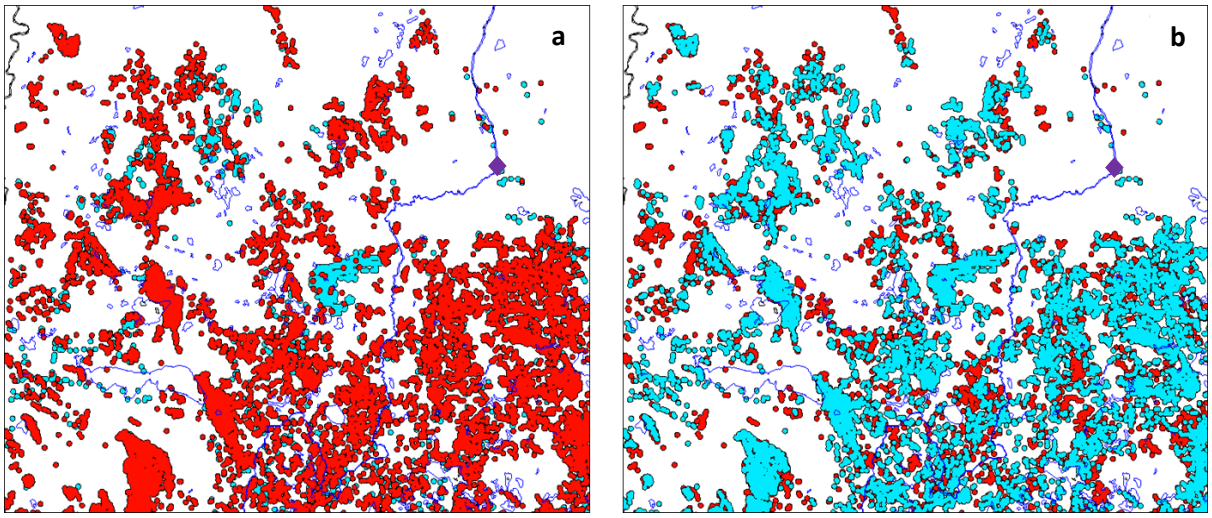

Figure S2: Location of UOG facilities in the vicinity of the Fort McMurray AOSR area from (a) the 2011-based projected 2013 inventory (red dots) superposed on the 2000-based projected 2010 inventory (cyan dots) and (b) the 2000-based projected 2010 inventory (cyan dots) superposed on the 2011-based projected 2013 inventory (red dots). In panel (a) the cyan dots not covered by the red dots are facilities that were in the projected 2010 inventory, but not in the projected 2013 inventory, whereas in panel (b) the red dots not covered by the

cyan dots are facilities that were in the projected 2013 inventory, but not in the projected 2010 inventory. The location of Fort McMurray is marked by the purple diamond symbol

11) Page 12 (Line 20) SMOKE reference has been mentioned – no need to repeat here.

SMOKE reference removed as suggested.

12) Page 13 (Line 14) I agree that the temporal profiles for fugitive VOC are dependent of temperature, but that's not the only factor. Wind speed (and snow cover) can also affect emission flux. How did you address wind speed effects?

This is a very good point. We did not take wind speed (and snow cover) into account for the current study. However we did point out in the Summary and Future Work section that this is an area for future improvement. The text reads:
"In future, model-predicted or locally measured hourly temperature and wind speed may be used to estimate hourly fugitive VOC emissions if the dependence of fugitive VOC emission rates on temperature and wind speed can be parameterized (Li et al., 2017)" (now Page 25, Lines 6-9).
We have now added one more sentence to the paper:
"Snow cover over the mining areas and ice cover over the ponds during wintertime also affect fugitive VOC emissions and need to be considered." (Page 25, Lines 9-10).

13) Page 14 (Line 29 and also Page 15 Line 5) Need a reference to those tests that show 'significant (as the authors claimed)' effects of the artificial water bodies. Be specific about what effects are.

Thank you for the comment. We have added one plot (Figure S3) to show that the change of land use can affect the predicted planetary boundary layer height by a few hundred meters, and we added the following text to the end of the paragraph:
"Meteorological fields are also affected. For example, Figure S3 shows that the predicted planetary boundary layer height over the OS facilities can be a few hundred meters lower than the surrounding areas, similar to the effect of natural lakes." (Page 12, Lines 7-9)

14) Page 15 (2nd paragraph) The authors' effort in updating landuse/land cover is to be commended. This is rarely done in air quality applications. Since BELD4 is already available, would future work require this step?

Thank you very much for recognizing our efforts in this regard. BELD4 is currently available with updated data for the U.S., but MODIS land-use data are still used in BELD4 for Canada with much less detailed land-cover data. We are currently updating the Canadian biogenic emissions land-use database to be consistent with the U.S. BELD4. Once the update is finished, this step will not be needed, at least for the near future. However, it may still be needed for areas that undergo fast development, such as the AOSR.

15) Page 15 (Line 9) Instead of using qualitative description such as 'significantly impacted', present facts. , e.g. change in biogenic VOCs (%).

Thank you for the comment. We have changed the statement as follows:
"By applying these masks to update vegetation and land-cover data, GEM-MACH-calculated biogenic emissions can be reduced by as much as 100% for the cleared areas related to mining activities." (Page 12, Lines 5-7).
This impact can be seen in Figure 7.

16) Page 16 (Line 25; Figure S5, S6) Why not comparing 'annual' CEMS to the NPRI inventory? As the authors noted, there could be large variation in the CEM measurements. Using two-month worth of data does not give me confidence in the comparison shown in Figure S5 and S6. Alternatively, you can report NPRI emissions that are temporally allocated to the same two months.

This is a very good comment. Detailed CEMS data are not required to be reported to the national inventory. We obtained the two months of CEMS data directly from the Alberta provincial government (their assistance is acknowledged in the Acknowledgements section). NPRI annual emissions are reported annually. Facilities are also required to report monthly emissions profiles. We checked the NPRI monthly profiles for SO_2, VOC, and NO_x for all six of OS mining facilities for years 2013 to 2016. All but one facility reported constant monthly profiles for all three species for all four years, which is very surprising in light of the major wildfire in May 2016 that ravaged the AOSR area and caused mining operations to be shut down for most of May and June. The monthly profiles reported by the one facility that did not report constant monthly profiles are shown in the table below. We can see that the monthly profiles for this facility also seem questionable. For example, SO_2 monthly profiles did not change at all for the first three years. Although the SO_2 monthly profile was changed for the fourth year, it only varies by season. VOC profiles are constant for the first three years, but the fourth year has the same seasonal variation as the SO_2 and NO_x emissions.

Alternatively it is possible to allocate the NPRI annual emissions to monthly emissions using the production-based derived monthly profiles. However, this approach will not account for SO_2 and NO_x emissions during upset events, when large amount of emissions may be released, as indicated from the CNRL daily reports during one upset event. Since these two plots are mainly for "sanity-check" purposes, we decided to annualize the CEMS data with an understanding that there is large uncertainty associated with the comparison by showing the CEMS standard deviation as error bars.

	SO2					VOC					NOX			
	2013	2014	2015	2016		2013	2014	2015	2016		2013	2014	2015	2016
January	10.1%	10.1%	10.1%	10.3%		8.33%	8.33%	8.33%	10.30%		11.40%	11.40%	9.20%	10.30%
February	10.1%	10.1%	10.1%	10.3%		8.33%	8.33%	8.33%	10.30%		8.90%	8.90%	9.60%	10.30%
March	7.5%	7.5%	7.5%	10.3%		8.34%	8.34%	8.34%	10.30%		9.70%	9.70%	9.20%	10.30%
April	3.6%	3.6%	3.6%	2.7%		8.33%	8.33%	8.33%	2.70%		7.40%	7.40%	9.80%	2.70%
May	2.6%	2.6%	2.6%	2.6%		8.33%	8.33%	8.33%	2.70%		3.20%	3.20%	8.20%	2.70%

June	7.6%	7.6%	7.6%	2.7%		8.34%	8.34%	8.34%	2.70%		8.30%	8.30%	7.60%	2.70%
July	7.1%	7.1%	7.1%	9.7%		8.33%	8.33%	8.33%	9.60%		7.50%	7.50%	8.20%	9.60%
August	9.3%	9.3%	9.3%	9.7%		8.33%	8.33%	8.33%	9.60%		6.70%	6.70%	6.50%	9.60%
September	13.5%	13.5%	13.5%	9.7%		8.34%	8.34%	8.34%	9.70%		7.60%	7.60%	7%	9.70%
October	9.7%	9.7%	9.7%	10.6%		8.33%	8.33%	8.33%	10.70%		8.30%	8.30%	8.20%	10.70%
November	8.8%	8.8%	8.8%	10.7%		8.33%	8.33%	8.33%	10.70%		10.40%	10.40%	7.90%	10.70%
December	10.1%	10.1%	10.1%	10.7%		8.34%	8.34%	8.34%	10.70%		10.60%	10.60%	8.60%	10.70%

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17) Page 17 (Line 14) How are the aircraft-estimated VOC emissions spatially allocated within each facility for modeling? The numbers shown in Table 6 are in tonnes/year, but aircraft observations are only available during the field campaign. Hourly aircraft data was scaled to daily rate based on temperature profile during the flight day (Li et al). Were the aircraft-derived estimated annualized by scaling daily values with temperature (as was done in the base case)?

In order to obtain gridded model emissions, the aircraft-estimated facility-total VOC emissions were first split by process based on the process-specific VOC emissions compiled for the base case. The process-specific emissions for each facility were then spatially allocated within each facility based on the facility-specific and process-specific gridded spatial surrogate fields.

To respond to your question about the annualization calculation for the aircraft-measurement-based VOC estimates, we have added the following sentence to Page 15, Lines 6-7 (note that Table 6 is now Table 4):
"The aircraft-derived VOC emissions estimates shown in Table 4 were annualized by scaling daily values with seasonal variation factors as discussed in Li et al. (2017)."

18) Page 18 (Line 1) The observation-based VOC profiles show lower higher-aromatic fractions. Could this be a result of flight altitude (150-1470 m) which could be more influenced by stack emissions rather than surface sources like tailing ponds?

Total stack VOC emissions are estimated to be very low compared to fugitive emissions from tailings pond, mine faces, and plants. Therefore, we think it unlikely that the composition of VOC emissions from stacks had a material impact on the observation-based VOC speciation profiles.

19) Page 18 (Line 11) My understanding is that the aircraft-derived emissions are at facility level so any scaling would be the same across all grid cells within individual facilities. Why are the ratios shown in Figure S7 varying within individual facilities?

This is a very good question. It is due to the different emission rates and different VOC speciation profiles for plants, mine faces, and tailings ponds. Aircraft-derived VOC profiles

are for the whole facility, but the VOC speciation profiles used for the base case are process-specific, i.e., profiles are different between plants, mine faces, and tailings ponds. Therefore, the ratio of aircraft-observation-based ADOM-2 higher-alkane emissions to the base-case higher-alkane emissions for the GEM-MACH 2.5-km grid varies slightly within the facility due to spatial variations in the base-case emissions field associated with the geographic locations of different processes. We had explained this in the paper on Page 18, Line 11 (now Page 16, Lines 4-5) as "The variations seen within individual facilities are due to different emission rates for plants, mine faces, and tailings ponds". However, in formulating our response here we noticed that the phrase "and different VOC speciation profiles" was missing from that sentence. We have now added it.

20) Page 19 (Line 29) I understand that the aircraft-derived emissions are only used for summertime modeling. The emission comparison needs to be limited to just summer months (so summing basecase emissions for these months), instead of assuming constant daily emissions throughout the year to get annualized value. Reporting 61,500 tpy of PM2.5 is totally misleading and overemphasizing the role of wind-blown dust.

This is a very good point. We agree that the figure of 61,500 t/year is misleading, although we had stated near the beginning of the paragraph that "Note that the latter were annualized for this comparison simply by assuming constant daily emissions throughout the year, which does not account for modulation by snow cover, frozen ground, or precipitation, but the aircraft-observation-based estimates were only used in GEM-MACH for summertime modelling."

Instead of annualizing the aircraft-derived emissions, we have now estimated two-month emissions during the field study period (August and September) based on a similar simple assumption that daily emissions were constant throughout these two warm-season months, which is a more reasonable assumption than assuming constant daily emissions throughout the year. We found that even the estimated aircraft-derived $PM_{2.5}$ emissions for two months are larger than the annual $PM_{2.5}$ emissions from the base-case inventory for all of the facilities except for the Imperial Oil Kearl facility. We think comparing the estimated two-month aircraft-derived emissions with the annual base-case emissions serves the same purpose of showing that the observation-based $PM_{2.5}$ emissions can be much larger than the base-case inventory $PM_{2.5}$ emissions while avoiding the larger uncertainty associated with the annualization calculation. Therefore, we have modified Figure 9. We now compare facility-level $PM_{2.5}$ emissions between the annual, base-case, inventory-based values and the aircraft-observation-based estimates for two summer months for the six AOSR facilities. The wording in the last paragraph of Page 19 (now the second paragraph of Page 17) was also revised accordingly.

21) Page 20 (Line 29) It is interesting to see a high fraction of OC from soil in this area. Is there any literature that reports similar finding?

As far as we can determine, Wang et al. (2015) is the only published report studying the chemical composition of dust over the AOSR area. However, there are a number of "Unpaved Road" dust speciation profiles archived in the U.S. EPA SPECIATE4.5 database that are based on measurements from various locations (see https://www.epa.gov/air-emissions-modeling/speciate-version-45-through-40). The Organic Carbon percentage varies from less than 1% at a California site to more than 20% at a Mexico City site.

22) Summary Section – Information on Phase I and Phase II is repetitive (page 23 and 24). The authors should focus on Phase III and some new insights that this work has revealed. Some recommendations made in this paper are reasonable. Since the basecase and sensitivity emissions were already applied in GEM-MACH, what are the lessons learned? On page 27 (last paragraph), the authors suggest that fugitive dust emissions are underestimated based on air quality modeling (by how much?). The authors should distill some scientific findings to share with readers for other pollutants (such as NOx, VOC, or SO2) based on other air quality modeling activities referred in the paper.

Thank you for this "arms-length" perspective. As suggested, we have shortened the Phase 1 and Phase 2-related discussion by two-thirds. Now the main focus of the paper is on Phase 3 and the lessons learned from this study.

On page 27 (last paragraph, now Page 25, second paragraph), we referred to some GEM-MACH modelling results (Makar et al., 2018) that suggested the fugitive dust emissions, or the fraction of their mass that is composed of base cations, might be underestimated by even more than suggested by the aircraft measurements. However, we were not certain whether this larger underestimate is in fact true and we stated later in the same paragraph that "Further aircraft-based measurements of fugitive dust emissions and their speciation are needed to improve the emissions inventories used here" (now Page 25, Lines 20-21). Nevertheless, we agree that the original phrase "are underestimated" is too strong and therefore we changed it to "might be underestimated" (Page 25, Line 19).

We also agree with the suggestion to summarize scientific findings based on other air quality modelling activities using emissions discussed in this paper. We have revised the paper accordingly. Specifically, we have expanded/added the following statements:

"Akingunola et al. (2018) showed that model-predicted SO_2 concentration could be changed by as much as 50% and the NO_x concentration by about 10% using the CEMS-measured hourly stack flow rate and temperature. On the other hand, the use of the more realistic CEMS-measured volume flow rates and temperatures resulted in a slight degradation of model performance with a new, improved plume-rise algorithm." (Page 14, Line 3-7)

"Similar to Ahmadov et al. (2015), Stroud et al. (2018) demonstrated that the measurement-derived top-down emissions improved the modelled VOC and organic aerosol (OA) concentration maxima in plumes. Bias was also improved for OA predictions. Their study

suggested that intermediate volatile organic compound (IVOC) emissions needs to be included as precursors to SOA for further improvement of SOA predictions." (Page 19, Lines 15-18)

"In their examination of acidifying deposition in the region, Makar et al. (2018) found that the new aircraft-based emissions improved the model fit to observations, increasing correlation coefficients (R from 0.47 to 0.54) and improving slopes of the model-to-observation best-fit line (slope changed from 0.051 to 0.73, correcting most of the large underestimate in predicted base cation deposition). The revised fugitive dust estimates from the aircraft study, while resulting in greatly improved model performance relative to the reported emissions, still resulted in an underestimate of base cations relative to observations, implying the need for further improvements to these emissions data." (Page 19, Lines 18-25)

23) Figure 1. This figure needs improvement on print quality/resolution. Table S3. Why was the 2010 CEMA off-road inventory selected? Did you compare these emissions with other inventories? Figure S3. Resolution issue. Color scales are not readable. And why is ethane shown instead of total VOC?

We have replotted Figure 1 with better quality and resolution.

Concerning Table S3 (now Table S4), there were two main reasons for using the 2010 CEMA off-road inventory for Phase 3. First, based the inventory analysis conducted during Phase 1 and Phase 2, we deemed the 2010 CEMA off-road inventory to be the best available inventory for the study area. Second, for Phase 3, the 2013 Canadian APEI emissions were available for all sectors, except for on-road and off-road emissions. Therefore, we had to continue to use the 2010 CEMA off-road emissions. We documented this fact on Page 6, Lines 2-4, as follows:
"2013 Canadian APEI Version 1 from ECCC for all sectors, including the first version of reviewed, publicly-available 2013 NPRI (released December 2014), except for on-road and off-road mobile source emissions (Sassi et al., 2016)."

We also identified the issue with off-road emissions as an area for further improvement in the "Summary and Future Work" section (see the first two paragraphs on Page 24).

Concerning Figure S3 (now Figure S4), we have replotted this figure with better quality and resolution. Ethene (or ethylene, C_2H_4) was shown in this figure because it is one of the model speciated VOC species and we used it as a representative for total VOC emissions. However, for the new plot, we have replaced ethene with total VOC as suggested.

Emissions Preparation and Analysis for Multiscale Air Quality Modelling over the Athabasca Oil Sands Region of Alberta, Canada

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Abstract. The oil sands of Alberta, Canada, which are classified as unconventional oil, ~~, but and they~~ are ~~also~~ the third-largest oil reserves in the world~~, behind only Venezuela and Saudi Arabia~~. We describe here a six-year effort to improve the emissions data used for air quality (AQ) modelling of the roughly 100 km x 100 km oil extraction and processing industrial complex operating in the Athabasca Oil Sands Region (AOSR) of north-eastern Alberta. ~~The objective of this work was to~~This paper reviews the ~~available~~ national, provincial, and sub-provincial emissions inventories~~data~~ that were available during the three phases of the ~~field~~ study, supplemented by hourly SO_2 and NO_x emissions and stack characteristics for larger point sources measured by Continuous Emission Monitoring Systems (CEMS), and daily reports of SO_2, from one AOSR facility for a one-week period during ~~the~~a 2013 fie~~l~~ed campaign when the facility experienced upset conditions. ~~,~~ Next it describes ~~, provide information for comparesison reported bottom-up emissions with aircraft-observation-based top-down emissions estimates, provides information for~~the creation of several~~a~~ detailed hybrid emissions inventories~~y,~~ and the generation of~~e~~ model-ready emissions input files for the Global Environmental Multiscale–Modelling Air-quality and CHemistry (GEM-MACH) AQ modelling system ~~for~~ that were used during ~~application to the AOSR~~the 2013 field study and ~~-~~for various post-campaign GEM-MACH sensitivity studies, in particular for a high-resolution model domain with 2.5-km grid spacing covering much of western Canada and centred over the AOSR. Lastly, it compares inventory-based bottom-up emissions with aircraft-observation-based top-down emissions estimates.~~,~~ ~~GEM-MACH was used to produce nested AQ forecasts during an AQ field study carried out in the AOSR in summer 2013 as well as ongoing experimental forecasts since then and retrospective model~~

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~~simulations and analyses for the field-study period. This paper discusses the generation of GEM-MACH emissions input files, in particular for a high-resolution model domain with 2.5 km grid spacing covering much of western Canada and centred over the AOSR. Prior to the field study, ten pre-2013 national, provincial, or sub-provincial emissions inventories for up to seven criteria air contaminant species (NO_x, VOC, SO_2, NH_3, CO, $PM_{2.5}$, and PM_{10}) that covered the AOSR study area and that had been compiled for various purposes were reviewed, and then a detailed hybrid emissions inventory was created by combining the best available emissions data from some of these ten inventories. After the field study, additional sources of emissions-related data became available, including 2013 hourly SO_2 and NO_x emissions and stack characteristics for large point sources measured by Continuous Emission Monitoring Systems, 2013 specific national inventories, daily reports of SO_2 emissions from one AOSR facility for a one-week period during the field campaign when that facility experienced upset conditions, aircraft measurements of VOC and PM2.5 concentrations from the 2013 field campaign and derived estimates of their emissions, and measurements of chemical composition of dust collected from various AOSR sites. These new data were used to generate updated emissions input files for various post-campaign GEM-MACH sensitivity studies.~~ Results show that emissions values obtained from different data sources can ~~be~~differ significantly ~~different~~, such as a possible ~~possible~~ ten-fold differences in $PM_{2.5}$ emissions and ~~about~~approximately 40% and 20% differences for total VOC and SO_2 emissions~~,~~ ~~respectively~~. A novel emissions-processing approach was also employed to allocate emissions spatially within six large AOSR mining facilities in order to address the urban-scale spatial extent of the facilities and the high-resolution 2.5-km model grid. ~~Their inclusion resulted in some significant emissions revisions, including a reduction in total VOC and SO2 emissions from surface mining facilities of about 40% and 20%, respectively, and a ten-fold increase in $PM_{2.5}$ emissions based on aircraft observations. In addition, standard emissions processing approaches could not provide an accurate representation of emissions from such large, unconventional emissions sources as AOSR surface mines. In order to generate more accurate high-resolution, model-ready emissions files, AOSR-specific improvements were made to the emissions processing methodology. To account for the urban-scale spatial extent of the AOSR mining facilities and the high-resolution 2.5-km model grid, novel~~ Gridded f~~F~~facility- and process-specific ~~gridded~~ spatial surrogate fields that were generated using spatial information from GIS (geographic information system) shapefiles and satellite images ~~to allocate emissions spatially within each mining facility to account for the urban-scale spatial extent of the AOSR mining facilities and the high-resolution 2.5-km model grid~~ were used to allocate non-smokestack emissions for each facility~~,~~ to multiple grid cells instead of treating these emissions ~~from each facility~~ as point sources and allocating them to a single grid cell as is normally done. Facility- and process-specific temporal

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profiles and VOC speciation profiles were also developed. The pre-2013 vegetation and land-use data-bases normally used to estimate biogenic emissions and meteorological surface properties were modified to account for the rapid change of land use in the study area due to marked, year-by-year changes in surface mining activities, including the 2013 opening of a new mine. Lastly, mercury emissions data were also processed in addition to the seven criteria-air-contaminant species (NO_x, VOC, SO_2, NH_3, CO, $PM_{2.5}$, and PM_{10}). ~~mercury emissions data were also processed~~ to support AOSR mercury modelling activities. ~~The combination of emissions inventory updates and methodological improvements to emissions processing has resulted in a more representative and more accurate set of emissions input files to support AQ modelling to predict the ecosystem impacts of AOSR air pollutant emissions. Seven~~ Six ~~other~~ GEM-MACH modelling papers in this special issue used some of these new sets of emissions and land-use input files.

1 Introduction

Alberta's oil sands (OS: see ~~Table S1~~Appendix for a list of acronyms), which consist of a mixture of bitumen, sand, clay, and water, are found in the Athabasca, Cold Lake, and Peace River areas of northern Alberta. Together these three areas cover 142,200 km^2, about 21% of the area of the province of Alberta (Alberta Energy, 2017) or about the same area as Greece. The Athabasca Oil Sands Region (AOSR) contributes the largest share of OS bitumen production: 82% in 2015 (Alberta Energy Regulator, 2017a). There are two main methods used to produce oil from the bitumen, each of which has associated atmospheric emissions. For bitumen deposits buried less than ~~200 feet~~60 m or so below the surface, the oil sands are mined by open-pit mining methods, in which large excavators dig up oil sand ore and transfer it to heavy-hauler trucks for transport to crushers, where large ore lumps are broken up. The crushed ore is then mixed with hot water and transported to an extraction plant, where the bitumen is separated from the other components and then transferred to either an on-site or a remote upgrader to create synthetic crude oil. About 3% of the OS area, mainly within the AOSR, can be surface-mined but it accounts for about 20 ~~percent~~ % of the recoverable OS oil reserves. Oil sands in the remaining 97% of the OS area are situated too deep for surface mining and can only be recovered by *in situ* extraction methods such as steam-assisted gravity drainage (Alberta Energy Regulator, 2017b). As of 2015, about 46% of Alberta oil production from oil sands comes from surface mines in the AOSR (Alberta Energy Regulator, 2017a).

According to the 2013 National Pollutant Release Inventory (NPRI; Canada's legislated inventory of pollutant releases reported by industrial, commercial, and institutional facilities that meet certain reporting requirements), emissions from Alberta's OS sector account for 61%, 34%, and 14% of the total reported VOC (volatile organic compound), SO_2, and NOx emissions, respectively, for the province, whose NPRI total VOC, SO_2, and NO_x provincial emissions are the highest of the Canadian provinces (https://www.canada.ca/en/environment-climate-change/services/national-pollutant-release-inventory.html). The OS industrial sector is also a significant source of PM (particulate matter) and CO emissions. Due to the complex nature of open-pit mining and the OS oil extraction processes, pollutants are mainly emitted from the following five processes: (1) exhaust emissions from off-road vehicles used for removal of the surface overburden and for excavation and transportation of the OS ore to an extraction plant; (2) pollutants emitted from processing taking place at the extraction and upgrading plants; (3) fugitive VOC emissions from mine faces, tailings ponds, and extraction plants; (4) fugitive dust emissions from surface disturbances such as the passage of the large vehicles belonging to the off-road mine fleets; and (5) wind-blown dust emissions from open surfaces such as mine faces and tailings-pond "beaches". The emissions of criteria-air-contaminant (CAC) pollutants (NO_x, VOC, SO_2, NH_3, CO, $PM_{2.5}$, and PM_{10}) from *in situ* OS extraction activities are ~~currently~~ believed to be lower currently than those of open-pit ~~mines~~ mining facilities based on the emissions reported to NPRI by facilities (https://www.canada.ca/en/environment-climate-change/services/national-pollutant-release-inventory.html).

Comment [JZ1]: Response to COSIA

To support air quality (AQ) modelling activities that are part of the Governments of Canada and Alberta Joint Oil Sands Monitoring (JOSM) Plan (see JOSM, 2011), emissions input files were created over the past six years ~~through~~in three successive phases for Environment and Climate Change Canada's~~—~~ (ECCC) Global Environmental Multiscale – Modelling Air-quality and CHemistry (GEM-MACH) AQ modelling system, which was set up to conduct nested AQ forecasts at model horizontal grid spacings of 10 km and 2.5 km (see Figure S1). The generation of emission input files was particularly challenging for the inner 2.5-km grid because the AOSR surface mining and processing facilities at the centre of the grid are large, complex, and unconventional industrial facilities that cannot be well represented by standard emissions processing approaches for point sources. At the beginning of emissions-related work for the JOSM plan in 2012 (referred to as Phase 1, 2012 - 2013), considerable effort was invested in reviewing ~~various~~a number of available emissions inventories, compiling~~ation of~~ a hybrid emissions inventory, and prepar~~ation of~~ GEM-MACH emissions input files for multiple model grids to support AQ forecasting for an Aug.–Sept. 2013 AQ field campaign in the AOSR. P~~;~~ ~~p~~articular attention was paid to the emissions input files for the inner (2.5-km) model domain centred over the

AOSR, since the model forecasts for this grid were the primary numerical guidance used during the field campaign period. ~~three areas: (i) the review of different available emissions inventories covering the AOSR compiled for different agencies with various geographic and temporal coverages (Alberta Environment and Sustainable Resource Development (AESRD [now AEP (Alberta Environment and Parks)], 2013; Marson, 2013); (ii) the compilation and synthesis of best-available emissions data into a hybrid JOSM emissions inventory (ECCC & AEP, 2016); and (iii) the preparation of GEM-MACH emissions input files for multiple model grids to support AQ forecasting for an Aug.–Sept. 2013 AQ field campaign in the AOSR (Gordon et al., 2015; Liggio et al., 2016; Li et al., 2017). Particular attention was paid to the emissions input files for the inner (2.5 km) model domain centred over the AOSR, which comprised the largest share of OS bitumen production during the 2013 field study period.~~ Additional emissions input files were then developed for JOSM Plan post-campaign AQ modelling activities in the second phase (2014-15) based on new emissions-related information available after the field study and in the third phase (2016-17) with updated emissions inventories, as well as new emissions estimates obtained from analysis of the 2013 field-study measurements.

~~two more phases as 2013-specific emissions data became available. The three phases to support GEM-MACH development, testing, evaluation, and application can thus be summarized as follows: (1) in the first phase (2012-13), emissions input files based on older inventories were created prior to and in support of AQ forecasting for the 2013 field study and post-study analysis shortly thereafter; (2) in the second phase (2014-15), updated sets of emissions files were created for post-study analyses based on new emissions-related information available after the field study; and (3) in the third phase (2016-17), more sets of emissions input files were created based on updated emissions inventories, as well as new emissions estimates from analysis of the 2013 field-study measurements.~~

GEM-MACH emissions input files developed during the first two phases using the SMOKE (Sparse Matrix Operator Kernel Emissions) emissions processing system (https://www.cmascenter.org/smoke) have been discussed in Zhang et al. (2015) and in a joint report by ECCC and AEP (Alberta Environment and Parks, ~~(~~formerly AESRD (Alberta Environment and Sustainable Resource Development)) for the JOSM project (ECCC & AEP, 2016: hereinafter referred to as the JOSM report). This paper briefly summarizes the work of the first two phases but focuses on the development of new emissions input files during the third phase for the following GEM-MACH AQ modelling applications:

1) Base-case study for AQ forecasting and a long-term deposition study for the region (Makar et al., 2018~~7~~, this issue) and for improvements for NH_3 predictions (Whaley et al., 2018~~7~~, this issue);
2) Model sensitivity study on the use of CEMS (Continuous Emission Monitoring System) measurements of SO_2, NO_x, exit temperature, and flow rate (Akingunola et al., 2018~~7b~~ and Gordon et al., 2018~~7~~ this issue);
3) Model sensitivity study on the impact of updated VOC and $PM_{2.5}$ emissions and speciation derived from surface measurements and from airborne measurements made during the 2013 field campaign (Stroud et al., 2018~~7~~, this issue);
~~4) Sensitivity study on the impact of increased model horizontal resolution down to 1 km to model predictions (Russell et al., 2017, this issue)~~
~~5)~~4) Mercury modelling over North America and the OS area using updated emissions (Fraser et al., 2018~~7~~, this issue).

In the rest of this paper, Section 2 provides an overview of the various ~~comprehensive national, provincial, and subprovincial~~ emissions inventories considered to build the base-case model emissions for all three phases. Challenges faced and approaches taken to compile a best-available hybrid emissions inventory for each of the three phases are discussed. Section 3 describes the emissions processing methodology applied in Phase 3 ~~(2016-17)~~ to generate base-case ~~hourly, gridded emissions input files, including both anthropogenic and biogenic~~ emissions. ~~Unique facility-specific and process-specific spatial surrogate fields were created for six AOSR surface mines to allocate emission from these large and unconventional sources.~~ A land-cover database was also updated for biogenic emissions and for land-surface characterization to account for the rapid change of land use over this region. Next, Section 4 describes the emissions data and emissions processing used for several post-campaign emissions sensitivity studies~~, including those based on new hourly CEMS measurement data, on aircraft and surface observation based estimates of VOC and $PM_{2.5}$ emissions and chemical speciation, and on updated mercury emissions~~. Lastly, Section 5 provides a summary of this work and give~~present~~s ~~plans~~ recommendations for future updates and improvements of emissions for AOSR AQ modelling.

2 Emissions Inventories Used for the Base-Case Emissions

2.1 Review of emissions inventories used for JOSM Phases 1 and 2 AQ modelling

In 2012, prior to the summer 2013 AOSR field study (Gordon et al., 2015; Liggio et al., 2016; Li et al., 2017), the national emissions inventories used to generate the emissions input files for ECCC's operational GEM-MACH AQ forecast model consisted of the AQ modelling version of the 2006 Canadian national Air Pollutant Emission Inventory (APEI) from ECCC, a projected 2012 U.S. National Emissions Inventory (NEI) from the U.S. Environmental Protection Agency (EPA) based on version 4 of the 2005 U.S. NEI, and the 1999 Mexican inventory (Moran et al., 2013a, 2014). The 2006 Canadian APEI represented a base year seven years earlier than the field study period, an important consideration for the AOSR due to its rapid development. ~~–~~For example, one of the five AOSR surface mining facilities in operation in 2012, the Canadian Natural Resources Limited (CNRL) Horizon mine (see Figure 1), only began production in 2009. Hence, pollutant emissions from that mine were not available in the 2006 APEI. Thus, while the 2006 APEI was being used as the basis for national-scale operational AQ forecasting for Canada, it was not an ideal choice for high-resolution AQ modelling for the AOSR field study.

A number of newer emissions inventories, however, had been developed for the AOSR area or for the province of Alberta, albeit not always for the purpose of supporting AQ modelling.

After an intense review of 10 available national, provincial, and sub-provincial emissions inventories in 2012 (AESRD, 2013; Marson, 2013), a hybrid inventory was compiled for Phase 1 and was used to pr~~ocess~~epare GEM-MACH-ready emissions input files for near-real-time GEM-MACH forecasts during the 2013 field study. Section S1 of the Supplemental Material ~~recapped the~~provides details about~~of~~ the creation of the Phase 1 emissions files. After the field study, emissions were updated during the 2014-2015 period (~~for~~ Phase 2) to incorporat~~clud~~e newly available emissions information, including new versions of national inventories, measurements from ~~s~~CEMS ~~for~~ attached to 17 smokestacks at four AOSR mining facilities for the field-study months of August and September 2013, and daily reports of SO_2 emissions during a one-week period in August 2013 when the CNRL Horizon facility experienced abnormal operating conditions. Details of the creation of the Phase 2 emissions files are summarized in Section S2 of the Supplemental Material. ~~Prior to the 2013 AOSR field study, the ten inventories listed in Table S2 by name, target region, and base year were reviewed to choose the most suitable emissions inventory data for AQ modelling for the OS area (AESRD, 2013; Marson, 2013).~~

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After an intensive review of these newer inventories, it became clear that no one inventory was the "best" choice in all respects, but three inventories contained emissions data that were either unique (i.e., not reported elsewhere), more detailed, and/or the most recent. The 2009/10 Cumulative Environmental Management Association (CEMA) inventory (Davies et al., 2012) had the most detailed stack- and process-level emissions for the AOSR surface mining facilities shown in Figure 1 (including separate emissions from mine faces, tailings ponds, and off-road mine hauler fleets, except for fugitive dust emissions from the off-road fleet); the 2010 Canadian NPRI included emissions for some species (NH_3 and PM_{10}) and source types (fugitive dust emissions from OS mine fleets) missing from the CEMA inventory; and the 2010 Canadian APEI from ECCC was the most comprehensive and had the largest spatial coverage (national) for area sources. Note, however, that at this time the 2010 Canadian APEI was not yet available in the detailed format required for emissions processing (referred to as the AQ modelling version).

2.1.1 Phase 1 hybrid emissions inventory and ancillary files

The solution adopted in the first phase for the best base case inventory to use to generate GEM-MACH emissions input files for the 2013 field study was to create a synthesized or hybrid AQ modelling emissions inventory, as summarized in Table 1, that combined the best available information from the above inventories. The 2009/10 CEMA inventory, supplemented by the 2010 NPRI for NH_3 and PM_{10} emissions, was mainly used to provide emissions for the AOSR field study area while the 2006 APEI was used outside the AOSR where the CEMA inventory's coverage ended. The 2010 NPRI was also used to scale the CEMA facility total VOC emissions for the five AOSR surface mines active at that time (Figure 1), since it was found that the CEMA inventory had the lowest total VOC emissions for these five facilities compared to four other inventories (ECCC & AEP, 2016) and the NPRI is Canada's legislated inventory of large point sources based on emissions reported by facilities. The ratio of 2010 NPRI total VOC emissions for the five mining facilities vs. the CEMA total yielded a scaling factor of 2.6, which was applied to the CEMA facility total VOC emissions for the individual facilities (Table 2). One reason to focus on the VOC emissions from these five facilities was that for 2010 they were estimated by NPRI to have contributed 75% of VOC emissions from all Alberta facilities. The 2009/10 CEMA inventory was also used to specify the process-specific allocation of these facility-total emissions between mine faces, tailing ponds, plants, and smoke stacks, which then dictated the spatial and temporal allocation and chemical speciation of these process-level emissions (ECCC & AEP, 2016).

~~The focus of the OS field study was a roughly 100 km by 100 km subregion of the AOSR located north of Fort McMurray, Alberta (Figure 1). This study area contains a complex of six large surface bitumen mining and processing facilities situated on both sides of the Athabasca River. As shown in Figure 1, each mining facility covers a very large area, ranging from 66 to 275 km^2, and each facility contains various area sources within their boundaries, including NOx, CO, VOC, and $PM_{2.5}$ emissions from each mine's off-road heavy-hauler fleet, evaporative VOC emissions from tailings ponds and mine faces, and point sources of SO_2, NOx, CO, VOC, $PM_{2.5}$ and fugitive VOC emissions from extraction and upgrading plants (Zhang et al., 2015). Although emissions from industrial facilities are normally treated as point sources by emissions processing systems and AQ models (e.g., Houyoux et al., 2000), each of these six facilities spans more than 10 GEM-MACH 2.5 km grid cells (area of 6.25 km^2 each), and many of the emissions are distributed over large areas within the facility boundaries. Treating such large facilities as point sources that can be assigned to a single grid cell is thus not realistic.~~

~~To address this concern, a new approach was taken in which these nominal point sources were treated as area sources. First, a GIS shapefile based on data collected by AESRD was obtained for the year 2010 with detailed locations of mine faces, extraction plants (and, for three facilities, upgrading plants), and tailings ponds for the five AOSR mines that were active within the study area at that time: Suncor Millenium and Steepbank mines; Syncrude Mildred Lake mine; Syncrude Aurora North mine; Shell Canada Muskeg River mine and Jackpine mine (known collectively as the Shell Canada Albian Sands mine); and CNRL Horizon mine (Figure 1). This shapefile was then used to develop three spatial surrogates for each facility to be used for spatial allocation of mine face, tailings pond, and extraction/upgrading plant emissions, respectively, including emissions from the off-road mining fleet and evaporative VOC emissions from mine faces, extraction plants, and tailing ponds (Zhang et al., 2015). It was assumed that the off-road fleets operated mainly in the mine face areas, so the mine face spatial surrogate field was used to allocate CAC emissions from the off-road fleet as well as evaporative VOC emissions from the mine faces. Note that emissions from the main smokestacks of the facilities were still treated as point sources. Finally, once all of the above development work was completed, the hybrid Phase 1 emissions inventory was input to the SMOKE (Sparse Matrix Operator Kernel Emissions) emissions processing system (https://www.cmascenter.org/smoke) together with the new AOSR facility specific spatial surrogate fields to generate Phase 1 model-ready emissions input files for use by GEM-MACH during the 2013 summer field study (Zhang et al., 2015).~~

~~**2.1.2 Phase 2 hybrid emissions inventory and ancillary files**~~

~~In Phase 2, after the field study, emissions updates were made during the 2014-2015 period to include newly available emissions information, including (i) an AQ modelling version of the 2010 Canadian APEI, (ii) a preliminary version of the 2013 NPRI point-source inventory, (iii) stack-level continuous emission monitoring system (CEMS) measurements for 17 smokestacks at four AOSR mining facilities for the field-study months of August and September 2013, and (iv) daily reports of SO_2 emissions during abnormal operating conditions from one AOSR mining facility (CNRL Horizon) during a one-week period in August 2013 when up to 20 times normal daily SO_2 emissions were released to the air during several upset events (ECCC & AEP, 2016). The six inventories and other emissions data sources that were used to create a second hybrid Canadian AQ modelling emissions inventory for 2013 are listed in Table 3.~~

~~The GIS shapefile describing the OS mines was also updated using 2013 satellite imagery (Zhang et al., 2015). These shapefile updates captured growth in the boundaries of existing mine faces and tailings ponds as well as new mine faces and tailings ponds that had been opened post-2010, and they were used to update the facility-specific spatial surrogate fields. In addition, a sixth mine, the Imperial Oil Kearl mine, entered production in 2013 (see Figure 1). Annual emissions estimates for this facility were obtained from the preliminary 2013 NPRI and three new spatial surrogates were developed to allocate emissions from this facility (Zhang et al., 2015). As well, monthly facility specific bitumen production data reported to the province of Alberta for 2013 for the six OS mining facilities were used to create facility-specific monthly temporal profiles (Alberta Energy Regulator (AER), 2014; Zhang et al., 2015). Note that a more comprehensive and detailed description of the Phase 2 hybrid inventory, the updated ancillary data sets for emissions processing, and the emissions processing procedure that was followed with the SMOKE system to generate model-ready emissions input files using the Phase 2 inventory is available in the JOSM report (ECCC & AEP, 2016).~~

2.2 Inventory updates for the Phase 3 hybrid emissions inventory

After the generation of the Phase 2 emissions input files for GEM-MACH, five important new sources of 2013-related emissions data became available:

1) 2011 U.S. NEI Version 1 from U.S. EPA (Eyth et al., 2013);
2) a) 2013 Canadian APEI Version 1 from ECCC for all sectors, including the first version of reviewed, publicly-available 2013 NPRI (released December 2014), except for on-road and off-road mobile source emissions (Sassi et al., 2016);

b) Second version of reviewed, publicly-available 2013 NPRI (released December 2015)

3) 2011 Canadian upstream oil and gas (UOG) point-source inventory for small and medium UOG facilities (Clearstone Engineering Ltd., 2014-a,b,c) and a projected 2013 Canadian UOG inventory (created by ECCC as part of the 2013 APEI Version 1);
4) CEMS measurements for all CEMS stacks with relatively large SO_2 and/or ~~/~~NO_X emissions in the province of Alberta for August and September~~,~~ 2013 (from AEP);
5) Top-down a~~A~~ircraft-measurement-based estimates of VOC emissions during the 2013 field study period for four of the six AOSR mining facilities (Li et al., 2017) and aircraft-measurement-based size-resolved PM emissions for all six facilities.

There were large differences noted between the 2011 U.S. NEI and the older projected 2012 U.S. NEI (projected from the 2005 U.S. NEI) used in Phases 1 and 2, despite the one-year difference in base year. For example, the projected 2012 NEI SO_2 emissions from all sectors were reduced by 48% in the 2011 NEI, but NO_2 emissions increased in the latter by 8%, due mainly to a 40% increase of on-road NO_x emissions (Moran et al., 2015). Among the many reasons that may have contributed to these large differences between the two inventories, one is the change in on-road emissions estimation tool used by the U.S. EPA from MOBILE6.2 and ~~+~~MOVES2010 (U.S. EPA, 2010) to SMOKE-MOVES2014 (U.S. EPA, 2015; Choi, 2016). Given that the 2011 U.S. NEI is a retrospective inventory based on actual activity data and CEMS data for base-year 2011, it was chosen to replace the projected 2012 U.S. NEI used in Phases 1 and 2 for the creation of the Phase 3 emissions input files for base year 2013. Note, however, that the U.S. EPA's emissions trend data set suggests a reduction of NO_x emissions by 8% and SO_2 emissions by 23% between 2011 and 2013 (https://www.epa.gov/air-emissionsinventories/air-pollutant-emissions-trends-data).

The first AQ modelling version (i.e., SMOKE-ready version) of the 2013 Canadian APEI (v1), which included point-source emissions from the first version (v1) of the reviewed, publicly-available 2013 NPRI (released in late 2014), became available in early 2016 for most sectors, with the exception of the on-road and off-road mobile source sectors. There are significant differences for some sectors between the modified 2010 APEI used in Phase 2 (Table S3) and the 2013 APEI. Figure 2 shows a comparison of fugitive-dust $PM_{2.5}$ emissions from four sectors for the province of Alberta. $PM_{2.5}$ emissions from construction more than doubled from 2010 to 2013 due to a combination of increased construction activities and changes in the methodology used to estimate PM emissions for this sector (Environment Canada, 2014). ~~Table 2~~Table 1 provides a comparison of facility-

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total VOC emissions for the six surface OS mining facilities used for Phases 1/2 vs. Phase 3. For Phases 1 and 2 these emissions were 2010-NPRI-scaled CEMA VOC emissions (Tables S2~~1~~ and S3), whereas for Phase 3, version 2 (v2) of the 2013 NPRI, which became available in late 2015, was used (~~Table 4~~Table 2). VOC emissions from the Suncor Millenium/Steepbank facility were reduced from about 28,000 tons/year in Phase 2 to 9,500 tons/year in Phase 3, a 64% reduction; the Shell Canada Muskeg River/Jackpine mine had a similar percentage reduction. One additional complication is that facilities may submit modified reports to NPRI for past reporting years based on updated information, as can be seen by comparing the last two columns of ~~Table 2~~Table 1, where reported total VOC emissions increased for Suncor Millenium/Steepbank, Syncrude Mildred Lake, and Syncrude Aurora North in the 2013 NPRI v2 (see also Li et al., 2017). One other important change evident in ~~Table 2~~Table 1 is the inclusion of emissions from the Imperial Oil Kearl surface mine, which began production in 2013, in the two 2013 emission inventory versions.

Emissions from smokestacks that are released at high ~~-~~volume flow rates and high temperatures may rise much higher into the atmosphere than stack releases with lower volume flow rates and temperatures. As a consequence, AQ models such as GEM-MACH include specialized parameterizations to calculate this plume rise (see Akingunola et al., 2018~~7b~~; Gordon et al., 2018~~7~~, this special issue). However, the extent to which this information is reported depends on the regulatory environment. One limitation of the 2013 NPRI is that only emissions from stacks higher than 50 m must be reported separately. Emissions from all other shorter stacks are aggregated together with surface-level fugitive emissions and are treated as surface releases (ECCC, 2016). On the other hand, the 2009/10 CEMA inventory has separate emissions information for all individual stacks. To allow plume rise to be calculated for stacks both above and below the NPRI reporting threshold, facility-total NPRI aggregate stack emissions were allocated proportionately to each stack in the CEMA inventory based on the 2009/10 CEMA stack emissions.

There are a variety of activities with pollutant releases to air within any given facility's boundaries, and the type of activity may influence the type and amount of VOCs being emitted at the facility. The extent to which these activities can be identified to allow spatial allocation within a facility once again depends on the regulatory environment and the reporting requirements. Surface-level fugitive VOC emissions are reported to NPRI as facility-total emissions without differentiation between source type (~~i.~~e.g., mine faces, tailings ponds, and extraction/upgrading plants). To distribute 2013 NPRI fugitive VOC emissions spatially within an OS mining facility, process allocation factors calculated from the process-specific fugitive VOC emissions in the 2009/10

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CEMA inventory for each AOSR mining facility were used to allocate fugitive VOC emissions between mine faces, tailings ponds, and plants (similar to the procedure used in Phase 2; see ECCC & AEP, 2016). For the Imperial Oil Kearl mine, which was not operating in 2010, 2013 fugitive VOC emissions were differentiated based on process allocation factors from the Shell Muskeg River/Jackpine facility given that both facilities use Paraffinic Froth Treatment (PFT) technology to produce diluted bitumen, which is then transported through pipelines to off-site refineries for further processing (http://www.oilsandsmagazine.com/technical/mining/froth-treatment/paraffinic; Li et al., 2017). However, due to the fact that the operation of a new mine during ~~the~~its first months ~~of its operation~~ may be quite different than a mine that has been operating for years, this was at best a necessary assumption with considerable uncertainty.

The UOG emissions input files generated for Phase 2 were based in part on a year-2000 Canadian UOG inventory projected to 2010 (Table S3). After Phase 2, a 2011 Canadian UOG inventory that was compiled for ECCC became available (Clearstone Engineering Ltd., 2014a,-b,-c). This new subinventory was then projected by ECCC to 2013 for inclusion in the 2013 APEI based on activity data and a methodology described in a letter report from Clearstone Engineering Ltd. (2014d). Figure 3 shows the national-level differences between the year-2000-based projected 2010 UOG inventory and the year-2011-based projected 2013 UOG inventory for the seven CAC pollutants, where about 95% of the UOG facilities are located within the high-resolution OS modelling domain. VOC, CO, and NOx emissions are higher for the new subinventory by 27%, 23%, and 11%, respectively, while SO_2 emissions are 11% lower. Thus, the projection of total UOG emissions from 2000 to 2010 that was used for Phase 2 seems to have been reasonable in total. However, the number of UOG facilities with CAC emissions increased from about 207,000 in the 2000 UOG inventory to 334,000 in the 2011 UOG inventory, a 61% increase. Figure S2 shows the locations of UOG facilities in the Ft. McMurray AOSR area for the 2000 and 2011 UOG inventories. We can see that some UOG facilities that existed in 2000 have been closed while many new facilities have opened since 2000. Updating the UOG inventory to the 2011-based 2013 projected inventory might thus be expected to have a significant impact on the spatial distribution of UOG emissions.

Given the availability of these new emissions data sets, ~~the~~ a synthesized Phase 3 hybrid emissions inventory was created from the inventories listed in ~~Table 4~~Table 2. As a complement to ~~Table 2~~Table 1, which compared the VOC emissions from the AOSR mines used for the three phases, Tables S4~~3~~ to S6~~5~~ compare the facility-total emissions of other CAC species compiled for the three phases from three main source types: CEMA off-

road mobile emissions; facility smokestack and area-source emissions; and road-dust emissions. As described in the next section, further improvements were also made to the emissions processing methodology before new Phase 3 model-ready 2013 base-case emissions files were generated from the Phase 3 hybrid inventory. Additional Phase 3 emissions input files that were generated for GEM-MACH emissions sensitivity runs using an expanded set of CEMS measurements and aircraft-observation-based emissions estimates are then discussed in Section 4.

3 Phase 3 Emissions Processing for GEM-MACH 2013 Base-Case Simulations

The same overall emissions-processing methodology described in Zhang et al. (2015) and the JOSM report (ECCC & AEP, 2016) was used in Phase 3 to generate ~~model-ready,~~ gridded, hourly, model-ready emissions fields for GEM-MACH using the SMOKE emissions processing system ~~(https://www.cmascenter.org/smoke/)~~. The three main steps required to process a typical emissions inventory that contains monthly or annual CAC emissions reported by jurisdiction for a small number of pollutants into ~~hourly,~~ gridded, hourly, model-ready emissions input files are (a) spatial disaggregation, (b) temporal disaggregation, and (c) chemical speciation (e.g., Dickson and Oliver, 1991; Houyoux et al., 2000; Moran et al., 2013b). ~~–~~ Note that before spatial disaggregation (i.e., spatial allocation) can be performed, a set of spatial surrogate fields must first be generated on the model grid of interest for such proxy or surrogate fields as population, road density, and agricultural land-use. Different inventories are then processed separately, often subinventory by subinventory (e.g., point sources, area sources, off-road sources, on-road sources), and as a last step some of the resulting gridded output fields may be merged.

Key aspects of the emissions-processing methodology for Phase 3 that were specific to the AOSR emissions included the following:

1) Updated f~~F~~acility-specific and process-specific spatial surrogate fields were ~~again~~ used (~~same as~~similar to Phase 2) for the 10-km North American grid and 2.5-km western Canada grid based on GIS polygons of mine faces, tailings ponds, and plants for the six AOSR mining facilities (Figure 1) in order to spatially allocate the surface area emissions from off-road fleet and fugitive sources~~,~~ between mine faces, tailings ponds, and plants. Emissions from individual smokestacks within these facilities, on the other hand, were treated as point-source emissions and assigned to the specific grid cells in which the stacks are located.

2) Facility-specific monthly temporal profiles for production-related emissions, such as emissions from off-road mine fleets and extraction plants, were generated based on facility-specific monthly production statistics for 2013 (Alberta Energy Regulator, 2014). Weekly and diurnal temporal profiles were treated as constant (i.e., "flat") as a default because the AOSR mining facilities usually operate around-the-clock throughout the year (note, however, the discussion on CEMS emissions in Section 4.1). Temperature-based monthly temporal profiles were created for fugitive VOC emissions from mine faces and tailing ponds, similar to the methodology that has been used in past AOSR environmental impact assessment (EIA) submissions (e.g., Cenovus, 2010; Imperial Oil, 2005).

3) Facility-specific and process-specific VOC speciation profiles were created based on VOC speciation profiles compiled in the CEMA inventory (Davies et al., 2012; Zhang et al., 2015).

4) PM speciation profiles from version 4.3 of the U.S. EPA SPECIATE database (https://www.epa.gov/air-emissions-modeling/speciate-version-45-through-40; Reff et al., 2009) were used to split PM emissions into six model chemical components: sulfate; nitrate; ammonium; elemental carbon; primary organic matter; and crustal material. Process-specific PM profiles were used for stack emissions based on the Source Classification Code (SCC) assigned to the stacks in the CEMA inventory (Davies et al., 2012). The "Unpaved Road Dust - composite~~Unpaved Road~~" PM speciation profile from SPECIATE v4.3 was used to speciate fugitive dust emissions from unpaved roads within each facility in the base-case emissions.

Another required emissions processing step was to perform PM size disaggregation. As discussed in Makar et al. (2018~~7~~, this issue) GEM-MACH may be configured to represent the PM size distribution with either two or 12 size bins. Accordingly, the PM emissions were processed twice, once for each representation of the PM size distribution ~~representation~~. The two-bin version separates PM_{10} emissions into two size bins, $PM_{2.5}$ (fine bin) and PMC (coarse bin, equal to PM_{10} - $PM_{2.5}$), whereas the 12-bin version separates PM_{10} emissions into the 10 size bins listed in ~~Table 5~~Table 3, plus two larger size bins for diameters greater than 10 µm (note that the base-case emissions thus assumed no primary particulate emissions for sizes greater than 10 µm diameter). For the 12-bin PM emissions, generic PM size distribution profiles were applied for three broad source types (area, mobile, and point) based on 10 source-specific particle size distributions ~~as~~ discussed in Eldering and Cass (1996). Figure 4 shows the distribution of the eight $PM_{2.5}$ bins for these three source types. Mobile-source $PM_{2.5}$ emissions have a normal size distribution centred around 0.16 micron in diameter, but point-source and area-source $PM_{2.5}$ emissions are skewed to the smaller and larger size bins, respectively.

In addition to anthropogenic emissions, GEM-MACH must also consider natural emissions, including biogenic VOC emissions, which depend on local vegetation type and light and/or temperature conditions. GEM-MACH calculates biogenic emissions dynamically (that is, making use of the GEM meteorological model's predictions of temperatures and light levels during a simulation combined with vegetation-type-dependant biogenic emissions formulas from BEIS (Biogenic Emission Inventory System) v3.06). Vegetation type is described using the BELD3 (Biogenic Emissions Landuse Database, Version 3) database, which contains 230 vegetation classes at 1-km resolution (Pierce et al., 2000). However, by 2013 the vegetation fields in the BELD3 database, which is based on early 1990's satellite imagery (Kinnee et al., 1997), were outdated over the AOSR mining area – much of the area ~~which~~ that was forested in the 1990's ~~but subsequently~~was later cleared of forest cover during the construction of the AOSR mining facilities. This is illustrated in Figure 5, which shows mean Leaf Area Index (LAI) for the gridded vegetation and corresponding summer peak isoprene emissions computed from the original BELD3 database. Except for some areas within the two oldest AOSR mining facilities, Suncor Millenium/Steepbank and Syncrude Mildred Lake, LAI values and isoprene emissions over the other mining facilities as computed with the BELD3 database are erroneously high, due to the fact that these areas, which by 2013 had been cleared for surface mining, were still characterized in the database as forested. Furthermore, the only water bodies contained in the land cover database over this area are natural lakes. The large artificial tailings ponds present in the mining facilities are not characterized as water-covered in the database (Figure 6a) even though in 2013, the tailings ponds in the AOSR covered an area of about 180 km^2 (http://www.energy.alberta.ca/OilSands/pdfs/FSTailings.pdf), the equivalent of 29 grid cells on the OS 2.5-km grid. Tests of the GEM-MACH model's meteorology for plume-rise algorithm analysis have shown that these artificial water bodies can have a significant influence on local meteorology and atmospheric vertical stability. In addition, an examination of the default water-body field portion of the grid cells overlapping the Athabasca River (centre of Figure 6a, flowing from south to north) showed that the river was also not being treated as a body of water in the default meteorological model database. The accuracy of the land-use database thus influences both meteorological and biogenic emissions estimation accuracy.

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The outdated land-cover characteristics over the AOSR area would thus have an impact on GEM-MACH predictions, particularly at high spatial resolutions. To improve the land-use and vegetation characterization of this area, masks for cleared land and artificial water bodies were generated as GIS polygons based on 2013

satellite images. Rivers were added using more detailed GIS water-body data. ~~By applying these masks to update vegetation and land-cover data, GEM-MACH-calculated biogenic emissions were significantly impacted.~~ Figure 7a shows the biogenic isoprene emissions over the AOSR surface mining area after the modification (cf. Figure 5b) and Figure 7b shows the difference between the original and modified isoprene emissions. The modified inland water coverage is shown in Figure 6b. By applying these masks to update vegetation and land-cover data, GEM-MACH-calculated biogenic emissions can be reduced by as much as 100% for the cleared areas related to mining activities. Meteorological fields are also affected. For example, Figure S3 shows that the predicted planetary boundary layer height over the OS facilities can be a few hundred meters lower than the surrounding areas, similar to the effect of natural lakes.

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As an example of the emissions input files generated with the SMOKE emissions processing system from the Phase 3 inventory, Figure S4~~3~~ shows gridded August mean monthly emissions of six ~~model~~ pollutant species for a portion of the 2.5-km OS grid centred on the AOSR study area. Similar to Figure 7b, the locations of the six AOSR mining facilities can be seen clearly, but other emissions sources are also evident such as on-road vehicle emissions and emissions from the city of Fort McMurray. GEM-MACH results from the use of the new Phase 3 base-case emissions input files generated using these updated emissions inventories (~~Table 4~~Table 2), updated AOSR facility- and process-specific spatial surrogate fields, new AOSR facility-specific monthly temporal profiles~~,~~ and ~~new AOSR facility-specific~~ VOC speciation profiles, and updated BELD3 vegetation and land-use data sets are described in Makar et al. (2018~~7~~).

4 Additional Phase 3 Emissions Processing for GEM-MACH Sensitivity and Scenario Studies

In addition to the Phase 3 base-case emissions input files described in Section 3, additional GEM-MACH emissions input files were generated using four special emissions data sets in order to examine the effects of specific changes to the emissions data on model predictions. These four data sets were (1~~a~~) an expanded 2013 CEMS emissions data set, (2~~b~~) 2013 OS field campaign aircraft-measurement-based top-down VOC emissions estimates, (3~~c~~) 2013 OS field campaign aircraft-measurement-based top-down $PM_{2.5}$ emissions estimates, and (4~~d~~) updated mercury emissions. These additional GEM-MACH emissions input files were used for a number of Phase 3 GEM-MACH sensitivity studies that are referenced in this section and described in detail elsewhere in this special issue.

4.1 Expanded CEMS emissions data set

As noted in Section S2~~.1.2~~, CEMS-measured hourly SO_2 and NO_x emissions from 17 stacks within four AOSR mining facilities were used in Phase 2 emissions processing for a GEM-MACH sensitivity test (ECCC & AEP, 2016; Makar et al., 2015; Zhang et al., 2015). This earlier work showed a relatively large impact of the better time-resolved CEMS data on model results. Recall that in Canada, regulatory reporting at the national level requires only annual total emissions from large stacks; hence, details on specific time periods within the year are lost and calculations to reconstruct this time variation using each facility's operating schedule for the emitting activities can only be approximate at best. However, detailed CEMS records are reported to the Alberta provincial government. For Phase 3, CEMS measurements from about 100 stacks ~~from~~ at 33 facilities with relatively large SO_2 or NOx emissions were obtained for the province of Alberta for August and September~~,~~ 2013. A sensitivity study was designed to investigate the impacts of both CEMS-measured hourly SO_2 and NO_x emissions, and CEMS-measured stack volume flow rates and exit temperatures on GEM-MACH predictions (Akingunola et al., 2018~~7b~~, this issue). For this study, the Phase 3 base-case stack emissions (based on 2013 NPRI annual reporting of stack emissions) were replaced with the corresponding CEMS hourly measurements. For the Phase 3 base-case emissions, the stack flow rate and exit temperature, which are used to calculate plume rise, were assumed to be static at the annual reported values. However, CEMS-measured stack exit temperature and flow rate often display significant temporal variation as shown in Figure S5~~4~~ for one example; hence, these measured~~ir~~ values were ~~generated~~ saved in model-ready form for the two-month period to evaluate their impact on model predictions.

Due to the NPRI reporting threshold that facility operators are not required to report stack-specific emission from smokestacks shorter than 50 meters (Section 2.2), not all CEMS stacks could be matched to NPRI stacks. Overall, 38 of the 100 stacks in the expanded CEMS data set were matched with NPRI stacks at 20 facilities. However, since the 38 matched stacks were *de facto* all tall stacks with generally large emissions, emissions from the matched stacks account for 77% and 43% of total SO_2 and NO_x emissions, respectively, from all NPRI point sources in Alberta. Figures S6~~5~~ and S7~~6~~ show comparisons by facility of SO_2 and NO_x emissions between the NPRI annual inventory and the two-month CEMS measurements for SO_2 and NO_x, scaled up to annual values. Overall, these scaled CEMS-based estimates agree well with NPRI annual totals, in spite of the large short-term temporal variation shown in the CEMS measurements. This is reasonable since facilities are expected to base their reported annual stack emissions on CEMS measurements. ~~However,~~ O~~o~~ver shorter time intervals, however, the stack emissions levels may vary by up to several orders of magnitude, thus having a

significant influence on model predictions. As well, the differences between CEMS volume flow rates and exit temperatures and the annual reported values may also have a significant influence on ~~model~~ plume dispersion and transformation of ~~emitted~~ SO_2 and NO_x emitted from tall stacks. ~~(~~Akingunola et al. ~~,~~ (201~~7~~8~~a,b~~) ~~,~~showed that model~~-~~predicted SO_2 concentration could be changed by as much as 50% and the NO_x concentration by about 10% using the CEMS~~-~~measured hourly stack flow rate and temperature. ~~However~~On the other hand, the use of the more realistic CEMS-measur~~observ~~ed ~~more realistic~~ volume flow rates and temperatures resulted in a slight degradation of model performance with a new, improved plume~~-~~rise algorithm.

4.2 Aircraft-measurement-based top-down VOC emissions estimates for AOSR mining facilities

~~Recently~~ A~~a~~irborne measurements have recently been used to quantify emissions from various oil and gas fields. For example, Karion et al. (2013) ~~measur~~estimated methane emissions over a western U.S. natural gas field, Peischl et al. (2015) quantified methane emissions as well from three U.S. shale production regions~~,~~ and Li et al. (2017) estimated VOC emissions for ~~the~~four AOSR ~~region~~facilities during the 2013 OS field campaign. As described in Li et al. (2017), aircraft observations of VOC species concentrations ~~made during the 2013 AOSR field campaign have been~~were used to estimate facility-total emissions of individual VOC species using a mass-balance approach (Gordon et al., 2015) for ~~four AOSR mining facilities:~~the Suncor Millenium/Steepbank~~,~~ Syncrude Mildred Lake~~,~~ Shell Canada Muskeg River/Jackpine~~,~~ and CNRL Horizon mining facilities (see Figure 1). Comparisons between the aircraft-observation-based top-down estimates of individual VOC species emissions and the corresponding bottom-up emissions reported to NPRI by these four facilities showed differences in terms of the magnitude of both VOC species emissions and total VOC emissions (Li et al., 2017).

Some p~~P~~revious studies have shown~~ed~~ that the use of air~~-~~craft~~-~~derived top-down emissions improved model performance. For example, in an attempt to understanding high O_3 events during winter time in a western U.S. oil and gas region, Ahmadov et al. (2015) compared AQ model performance using emissions from two different sources: (1) the U.S. EPA NEI (bottom-up) and (2) emissions derived from aircraft observations (top-down). They found that the top-down emissions improved model prediction of methane, other VOCs, and NOx. ~~Model with~~The use of these top-down emissions also captured the O_3 episode better than using the bottom-up emissions. To assess the impact of the suggested uncertainty of VOC emissions for these four OS facilities on GEM-MACH predictions, emissions of the individual VOC species estimated from the aircraft observations (top-down) were mapped to the model VOC species used by GEM-MACH's ADOM-2 (Acid Deposition and

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Oxidant Model, version 2) gas-phase chemistry mechanism (Makar et al., 2003; Stroud et al., 2008) to replace the corresponding Phase 3 base-case model VOC species emissions (bottom-up) for the four facilities.

~~Table 6~~Table 4 shows a comparison of facility-total emissions of ADOM-2 model VOC species between the Phase 3 base-case emissions input files (bottom-up) and the aircraft-observation-based emissions input files (top-down). The aircraft-derived VOC emissions estimates shown in Table 4 wee~~a~~re annualized by scaling daily values with seasonal variation factors as discussed in Li et al. (2017). Except for Syncrude Mildred Lake, the totals of the aircraft-observation-based top-down VOC emissions for these facilities are higher than the corresponding bottom-up base-case totals, ranging from a factor of 2.5 for Suncor Millenium/Steepbank to 6.7 for Shell Canada Muskeg River/Jackpine and 7.2 for CNRL Horizon. The relative rankings of the emissions by model VOC species also differ for the two data sources. Figure 8 compares the process-specific VOC speciation profiles for these four facilities that were used for the Phase 3 base-case study based on the CEMA inventory (Davies et al., 2012; Zhang et al., 2015). Figure 8 also compares the inventory-based VOC speciation profiles (bottom-up) with the aircraft-observation-based VOC speciation profiles (top-down) by facility. As the emissions estimate~~d~~s from the aircraft observations corresponded to facility-total emissions, an emissions-weighted, base-case "composite" VOC speciation profile was created for each facility by combining the plant, mine-face, and tailings-pond VOC speciation profiles based on the emissions of each ADOM-2 model VOC species. Both the aircraft-observation-based VOC speciation profiles and the "composite" VOC profiles vary from facility to facility, but there are some differences between the two profile types. Consistent with Li et al. (2017), for example, the aircraft-observation-based VOC profiles have a higher propane emissions fraction and a much lower higher-aromatic emissions fraction than the composite profiles for all four facilities. The aircraft also measured significant amounts of isoprene emissions likely originated from bitumen vapour emissions from~~rom~~ the Suncor Millenium/Steepbank and the CNRL Horizon facilities, which are not present in the corresponding bottom-up base-case profiles. Further studies are needed to confirm the source of non-biogenic isoprene emissions.

To generate~~t~~ model-ready emissions files, the aircraft-estimated top-down VOC emissions were first split by process based on the process-specific VOC emissions compiled for the base ~~-~~case and then spatially allocated within each facility based on the process-specific and facility-specific surrogates. Figure S8~~7~~ shows spatial variations in the ratio of the gridded, model-ready, aircraft-observation-based higher-alkane emissions (top-

down) to corresponding base-case emissions (bottom-up) for the GEM-MACH 2.5-km grid over the AOSR study area. Consistent with ~~Table 6~~Table 4, the ADOM-2 higher-alkane emissions estimated from ~~aircraft observations~~the top-down estimation are about eight times higher for the Shell Canada Muskeg River/Jackpine and CNRL Horizon facilities than corresponding bottom-up emissions from the 2013 NPRI but are closer for the Suncor Millenium/Steepbank and Syncrude Mildred Lake facilities. The variations seen within individual facilities are due to different emission rates and different VOC speciation profiles for plants, mine faces, and tailings ponds. As expected there is no difference for areas outside of these four facilities. The new GEM-MACH emissions input files generated using the aircraft-observation-based VOC emissions have been used for a GEM-MACH sensitivity test (see Stroud et al., 2018~~7~~, this issue).

4.3 Aircraft-measurement-based top-down PM emissions estimates for AOSR mining facilities

PM emissions from the AOSR mining facilities originate mainly from four major source categories: (1) emissions from plant stacks; (2) tail-pipe emissions from the off-road mining fleet; (3) fugitive dust originating from various activities, such as excavation of oil-sand ore, loading and unloading trucks, and wheel abrasion of surfaces by off-road vehicles; and (4) wind-blown dust. As summarized in ~~Table 4~~Table 2, PM emissions from plant stacks and fugitive dust source categories were obtained from the 2013 NPRI wh~~il~~ereas emissions from tail-pipe emissions were provided by the 2009/10 CEMA inventory. However, none of the inventories included wind-blown dust emissions, and the estimates of anthropogenic fugitive dust emissions are highly uncertain. In addition, emissions of construction dust from one facility, the Imperial Oil Kearl mine, a portion of which was still under construction during the aircraft monitoring campaign, were expected to be large. In order to evaluate and potentially to improve these emissions estimates, top-down estimates of size-resolved PM emissions were also calculated based on aircraft measurements of size-resolved PM concentrations made during the 2013 AOSR field campaign for all six AOSR mining facilities.

The 2013 aircraft campaign used a top-down mass–balance approach (Gordon et al., 2015) to determine PM emissions from all six AOSR surface mining facilities. For particles with a diameter in the range of 0.3 to 20 μm, a Forward Scattering Spectrometer Probe (FSSP) model 300 was deployed from a wing-mounted pod (Baumgardner et al., 1989) to measure the particle number–concentration size distribution in 30 size bins. An Ultra-High Sensitivity Aerosol Spectrometer (UHSAS) was used inboard to determine the number-concentration size distribution of particles with diameter from 0.06 to 1.00 μm in 99 size bins. Volume-concentration size distributions of particles were derived from these number–concentration size distributions

from 0.06 to 20 μm by combining both sets of measurements from the two instruments. Size-dependent particle densities, varying from 1.5 to 2.5 g/cm^3, were used to convert the volume ~~-~~ concentration size distributions to mass ~~-~~ concentration size distributions, based on the known mineralogy for the supermicron particles for the top soil in the region and the known chemical composition for submicron particles from concurrent aerosol mass spectrometer measurements (Liggio et al., 2016). The resulting particle mass concentration size distributions were combined to match the 12-bin version of the GEM-MACH model particle size distribution. The mass balance emission algorithm TERRA (Top-Down Emission Rate Retrieval Algorithm) (Gordon et al., 2015) was then applied to these particle size bins to determine the particle mass emission rates for each bin. Uncertainties in the particle mass emission rate from each facility determined this way were estimated at approximately 36% for supermicron particles, and 32% for submicron particles. Based on the aircraft observations, 68% of the PM_{10} emissions are in the coarse mode (2.5 to 10 μm).

Figure 9 shows a comparison of ~~annual~~ facility-level $PM_{2.5}$ emissions between the ~~annual~~ base-case inventory-based annual values (bottom-up) and the aircraft-observation-based estimates (top-down) for the two summer months (August and September) for the six AOSR facilities. Note that the latter were ~~annualized~~ calculated for this comparison simply by assuming constant daily emissions throughout the two summer months. This avoided an annualization calculation, for which~~year because~~ it is difficult to ~~, which does not~~ account for modulation by snow cover, frozen ground, or precipitation during winter~~-~~time. Moreover,~~, but~~ the aircraft-observation-based top-down estimates were only used in GEM-MACH for summertime modelling. Except for the Imperial Oil Kearl facility, the $PM_{2.5}$ emissions estimated from the top-down aircraft observations, even for ~~only~~just two summer months, were ~~at least~~ a factor of 1.5 to 5 ~~one order of magnitude~~ larger than the ~~annual~~ bottom-up 2013 APEI $PM_{2.5}$ annual emissions used for the Phase 3 base-case emissions processing. One reason for the difference is that wind-blown dust is not included in the ~~-~~ APEI inventory, which ~~i~~was compiled for anthropogenic emissions only. For the base~~-~~case bottom-up inventory, total $PM_{2.5}$ emissions from off-road vehicle tail-pipe emissions and stacks are 2,272 tonnes/year (Tables ~~S3~~ S4 and ~~S4~~S5) while road dust emissions are 4,134 tonnes/year (Table ~~S5~~S6). Thus, anthropogenic fugitive dust emissions account for 65% of total $PM_{2.5}$ emissions from the AOSR mines. Aircraft-observation-based estimated total $PM_{2.5}$ emissions from all six facilities are about ~~61,500~~10,300 tonnes for the two summer months ~~tonnes/year~~. If we assume that all of the unreported $PM_{2.5}$ emissions come from natural wind-blown dust, then fugitive dust emissions ~~accounts for 96% of~~will dominate total $PM_{2.5}$ emissions from those facilities even more.

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Figure 10 shows the observed size distribution of the first eight GEM-MACH size bins, which correspond to the $PM_{2.5}$ size range (cf. ~~Table 5~~Table 3). Although the size distribution of the $PM_{2.5}$ emissions varies from facility to facility, 65%–95% of $PM_{2.5}$ emissions are in Bin 8 (diameter range from 1.28 to 2.56 µm), implying that the majority of the $PM_{2.5}$ emissions are from fugitive-dust area sources (Eldering and Cass, 1996), either from dust kicked up by off-road mining vehicles or from wind-blown dust. Compared to the area-source PM size distribution profile used by SMOKE to process the bottom-up base-case emissions (Figure 4), a much larger Bin 8 mass fraction and smaller Bin 1 to 7 (i.e., <1.28 µm) mass fractions were observed by the aircraft for the AOSR mining facilities.

An AOSR-specific PM chemical speciation profile consisting of six chemical components was also constructed for fugitive dust emissions from these facilities to replace the ~~standard~~ "Unpaved Road Dust - Composite~~Fugitive Dust~~" profile from the U.S. EPA SPECIATE v4.3 database (see Section 3). Wang et al. (2015) analysed soil samples collected from 17 AOSR facility sites and 10 forest sites. The samples were further characterized as paved road dust, unpaved road dust, tailings sands, and overburden soil. Their analysis showed that PM speciation is clearly different between the dust collected from the facility sites and from the forest sites. For this study, the new AOSR-specific fugitive-dust PM speciation profile was compiled by averaging the site-specific profiles from all 17 facility sites from Wang et al. (2015) to represent surface PM speciation with the following three exceptions:

1) For the unpaved-road site S16, the elemental-carbon percentage seemed to be too large, which might be an artefact due to dry deposition from heavy-duty diesel exhaust (Wang et al., 2015). This site was excluded from the facility profile average in their study and was excluded in this study too.
2) The organic-carbon percentage for site S10 was much smaller and the elemental-carbon percentage was larger than those of other facility sites. That site was excluded from the organic-carbon range discussion in Wang et al. (2015) and was excluded here as well.
3) S17 is located on Highway 63, so it was also excluded from the facility average.

Figure 11 shows a comparison of the fugitive-dust PM speciation profile used for the Phase 3 base-case emissions processing, which is the standard "Unpaved Road Dust - Composite~~Unpaved Road~~" profile from the U.S. EPA SPECIATE v4.3 database, and the new profile described above. The organic-matter (OM = organic carbon + particulate non-carbon organic matter) percentage in the AOSR-specific PM speciation profile (21.8%) is about three times larger than the fraction in the standard "Unpaved Road Dust - Composite~~Unpaved Road~~"

profile (7.6%), suggesting that soils in the AOSR facilities contain more organic matter than soils in other areas. The crustal-material percentage decreases correspondingly, from over 91% to 76%. The AOSR-specific PM speciation profile also has more sulfate and elemental carbon, but the fractions are relatively small.

Figure S98 shows spatial variations in the ratio of the gridded aircraft-observation-based Bin 8 OM emissions (top-down) to the corresponding base-case emissions (bottom-up) for the GEM-MACH 2.5-km grid over the AOSR study area. Except for the Imperial Oil Kearl facility, the top-down OM emissions ~~estimated from the aircraft observations~~ are more than two orders of magnitude larger than those for base-case study (bottom-up) due to the combination of higher PM emissions (Figure 9), larger Bin 8 mass fraction (Figures 4 and 10), and the larger OM mass fraction (Figure 11).

The new estimates of total fugitive dust emissions and the new PM size-distribution and speciation profiles were used for two GEM-MACH sensitivity simulations. One of these simulations focussed on the impact of the increases of VOC and primary OM emissions on total organic aerosol and the formation of secondary organic aerosol (Stroud et al., 2018~~7~~, this special issue). The second examined the impact of the increased crustal-material emissions on regional acid deposition by making use of the Wang et al. (2015) PM speciation profile to further speciate the model's crustal material into a base-cation fraction (Makar et al., 2018~~7~~, this special issue). Similar to Ahmadov et al. (2015), Stroud et al. (2018) demonstrated that the measurement-derived top-down emissions improved the modelled VOC and organic aerosol (OA) concentration maxima in plumes. Bias was also improved for OA predictions. Their study suggested that intermediate volatile organic compound (IVOC) emissions needs to be included as precursors to SOA for further improvement of SOA predictions. ~~Both studies suggest that these improvements to emissions have a significant impact on model performance.~~ In their examination of acidifying deposition in the region, Makar et al. (2018) found that the new aircraft-based top-down emissions improved the model fit to observations, increasing correlation coefficients (R from 0.47 to 0.54) and improving slopes of the model-to-observation best-fit line (slope changed from 0.051 to 0.73, correcting most of the large underestimate in predicted base cation deposition). The revised fugitive dust estimates from the aircraft study, while resulting in greatly improved model performance relative to the reported emissions, still resulted in an underestimate of base cations relative to observations, implying the need for further improvements to these emissions data.

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~~The acid deposition study (Makar et al., 2017) also found that the fugitive dust estimates from the 2013 aircraft field study, while larger than the reported inventory values, may themselves underestimate the total fugitive dust emissions when compared to deposition observations in the immediate vicinity of the oil sands.~~

4.4 Mercury emissions

Mercury emissions from the SMOKE-ready versions of the 2010 Canadian APEI and version 1 of the 2011 U.S. NEI (NEIv1) were used in Phase 2 for creating gridded GEM-MACH-ready mercury emissions. In Phase 3 these emissions input files were updated with two AOSR-specific adjustments. First, annual total mercury emissions to air from all NPRI facilities in the 2010 Canadian APEI, including the six AOSR mining facilities, were 3,429 kg/year. In comparison, the annual total mercury emissions to air reported by all NPRI facilities for 2013 were 2,529 kg/year, of which, only 61 kg were emitted from the surface mining facilities. Thus for the 2013 field study, the 2013 NPRI reported values were used for the model Hg emissions. Second, the U.S., mercury emissions from off-road vehicles were only available for the state of California in the SMOKE-ready version of the 2011 NEIv1 (https://www.epa.gov/air-emissions-modeling/2011-version-6-air-emissions-modeling-platforms), whereas the original 2011 NEIv1 (https://www.epa.gov/air-emissions-inventories/2011-national-emissions-inventory-nei-data) included off-road-mobile mercury emissions for other states as well. The amount of off-road-mobile mercury emissions for California was the same in the two inventory versions. Based on the original 2011 NEIv1 inventory, total annual off-road-mobile mercury emissions for the entire U.S. were 40.9 kg/year, of which 26.1 kg/year was from California. Although these off-road-mobile mercury emissions were relatively small compared with other emissions sources (see ~~Table 7~~Table 5) and more than 60% of the off-road-mobile mercury emissions were from California, the second adjustment was to use off-road-mobile mercury emissions from the original 2011 NEIv1 to add in mercury emissions for the missing states in the off-road-mobile subinventory of the SMOKE-ready version of the 2011 U.S. NEIv1.

~~Table 7~~Table 5 presents a summary of source-specific anthropogenic mercury emissions used for Phase 3 for both the U.S. and Canada. Total 2011 U.S. annual mercury emissions from all four broad categories were 46,992 kg, of which nearly 90% was from point sources and the rest were mainly from area sources (9%). Mercury emissions from on-road and off-road vehicles accounted for less than 1% of total mercury emissions, and most of these vehicular emissions (90%) came from on-road vehicles. The summary of 2010/2013 Canadian mercury emissions shows that point sources were the largest anthropogenic source of mercury emissions in Canada (58%), followed by area sources (42%), and on-road and off-road vehicle emissions

contributed little. Total mercury emissions from Canada for 2010/2013 were about 9% of those emitted in the U.S. for 2011. The two adjustments made for Phase 3 reduced U.S. and Canadian anthropogenic mercury emissions by 885 kg/year or less than 2%. However, emissions of mercury from forest fires were also recognized as a major source (Fraser et al., 2018~~7~~, this special issue).

Three mercury species (elemental, divalent gas, and particulate) are considered in the mercury version of the GEM-MACH model (Fraser et al, 2018~~7~~). Mercury emissions for the Canadian 2013 NPRI point source emissions were pre-speciated based on the 2006 Canadian point-source emissions inventory used for the 2008 mercury assessment (UNEP, 2008). For other inventories, mercury emissions were reported as unspeciated totals in the 2010 Canadian APEI and the 2011 U.S. NEIv1. For these other inventories, mercury speciation was carried out using speciation profiles for nine broad source categories following the same methodology used in the U.S. EPA 2005 NEIv4.1 platform. The same profiles had also been used in the U.S. EPA 2002v3 platform (see Table 3-14 in U.S. EPA, 2011).

Figure 12 shows the spatial distribution of Phase 3 elemental mercury emissions for both Canada and the U.S. on the 10-km GEM-MACH continental grid for one afternoon hour in August. Most of the mercury emissions are from populated and industrial areas. Figure S10~~9~~ shows the domain-average percentages of the three mercury species based on total emissions summed over the nine source categories. About 50%, 30%, and 20% of the total mercury emissions are in the elemental, divalent gas, and particulate states, respectively. Fraser et al. (2018~~7~~, this issue) present some results from the use of these Phase 3 mercury emissions input files.

5 Summary and Future Work

A number of sets of model-ready emissions input files have been prepared over the past six years in three successive phases for the GEM-MACH air quality modelling system in support of the Governments of Canada and Alberta Joint Oil Sands Monitoring (JOSM) plan. These emissions files were used by GEM-MACH to conduct nested AQ forecasts in support of an Oil Sands field campaign carried out in summer 2013 as well as ongoing experimental forecasts since then and retrospective model simulations and analyses for the field-study period. Two GEM-MACH grids were considered: a North American continental grid with 10-km grid spacing and a high-resolution western Canada grid with 2.5-km grid spacing centred over the Athabasca Oil Sands Region (AOSR) of north-eastern Alberta, Canada.

~~Successful preparation of emissions input files for AQ modelling requires accurate and representative emissions inventories and emissions processing. The JOSM Phase 1 emissions processing undertaken from 2012 to 2013 was a particular challenge because the base years of all available emissions inventories were three years or more out of date compared to 2013, that is, 2010 or earlier. Moreover, the six large AOSR mining facilities that were the focus of the 2013 field campaign were changing year by year, which made emissions representativeness an important issue. These facilities are also complex and unconventional industrial sources that cannot be well represented by standard emissions processing approaches.~~

~~The approach adopted in Phase 1 was to review all available emissions inventories covering the study area and to extract the best available information from the 10 inventories considered in order to construct a detailed synthesized or hybrid AQ modellers' emissions inventory for this specific project. One important change in Phase 1 to the emissions processing methodology that was used with the new hybrid modellers' inventory was to treat three types of major emissions sources within each AOSR mining facility – mine faces, tailings ponds, and extraction plants – as area sources rather than point sources due to their large spatial extent. This required three spatial surrogate fields to be developed for each individual facility based on a 2010 GIS shapefile describing the AOSR mines. Chemical speciation profiles were also chosen to be as source-specific as possible~~ Ten available emissions inventories covering the study area were reviewed in Phase1 (2012-2013) and a detailed synthesized or hybrid AQ modellers' emissions inventory was constructed. An important approach developed in Phase 1 was to treat three types of major emissions sources within each AOSR mining facility – mine faces, tailings ponds, and extraction plants – as area sources rather than point sources due to their large spatial extent by developing and using three sets of facility-specific and process-specific spatial surrogate fields based on a 2010 GIS shapefile describing the AOSR mines.

For Phase 2 emissions processing from 2014 to 2015, more up-to-date emissions inventories and other relevant emissions information became available, including ~~a modellers' version of a newer (2010) Canadian national comprehensive emissions inventory (APEI), a preliminary version of the 2013 Canadian large point source inventory (from NPRI), monthly bitumen production statistics for 2013 that included a new AOSR mining facility (Imperial Oil Kearl),~~ continuous emissions monitoring system (CEMS) data sets for 2013 for 17 smokestacks in four AOSR mining facilities, and updated, 2013-specific AOSR shapefiles. ~~A more comprehensive and detailed description of the modellers' inventory compilation and emissions processing for~~

~~Phases 1 and 2 to prepare GEM-MACH emissions input files is contained in a JOSM report (ECCC & AEP, 2016), which also identified gaps and recommended areas for future improvements.~~

This paper focused on the Phase 3 emissions processing that was carried out from 2016 to 2017. Some of the gaps and recommendations raised in the JOSM report (ECCC & AEP, 2016) were addressed during this phase~~.~~. Newer Canadian and US inventory compiled for, or close to, 2013 were used. ~~Canadian area and point source emissions were updated to a 2013 criteria air contaminant (CAC) inventory (the AQ modellers' version of the 2013 APEI), which included a new upstream oil and gas subinventory, and U.S. emissions were updated to version 1 of the 2011 NEI to replace an earlier projected 2012 NEI.~~ An expanded CEMS data set of hourly SO_2 and NO_x emissions and smokestack operating characteristics for August–September 2013 was obtained for the entire province of Alberta, increasing the provincial total coverage of point~~-~~source SO_2 and NO_x emissions by CEMS measurements from 31% and 3% to 77% and 43%, respectively. New VOC and PM emissions estimates and chemical speciation profiles for the AOSR mining facilities that had been derived from on-site surface observations and aircraft observations made during the 2013 field campaign were processed for several GEM-MACH sensitivity studies. The aircraft-observation-based top-down VOC emissions were about two times larger than the bottom-up base-case emissions from the 2013 NPRI (Li et al., 2017). For PM emissions, two-month PM emissions estimated from the top-down aircraft-observation-based emissions were even larger than ~~a comparison between the annualized aircraft-observation-based emissions and~~ the bottom-up NPRI annual emissions ~~shows a factor of 10 difference~~ for five of the six facilities (Figure 9). The VOC and PM chemical speciation profiles used to speciate emissions from the AOSR mines were also noticeably different than those used to process the Phase 3 base-case emissions. A vegetation data~~-~~base used to estimate biogenic emissions and a land-cover data~~-~~base used in the parameterizations of land-surface processes and dry deposition were also modified to account for the rapid change of vegetation cover and land use in the AOSR region due to year-by-year changes in surface mining activities. In addition to CAC emissions, mercury emissions were also processed to support mercury modelling activities using newly available data sets.

~~The various Phase 3 emissions input data sets have been used to drive a number of GEM-MACH simulations as well as to evaluate plume rise algorithms, results from which are discussed in a number of companion papers in this special issue: see Akingunola et al., 2017b; Fraser et al., 2017; Gordon et al., 2017, Makar et al., 2017; Russell et al., 2017; Stroud et al., 2017, and Whaley et al., 2017.~~

This study also provides specific examples of some common issues related to the preparation of emissions input files for AQ models. First, there is always a time lag between a year of interest and the year in which an emissions inventory becomes available for that year of interest. Second, inventories are always subject to change due to reported corrections or to changes in estimation methodology. Third, if multiple inventories are available for the same region and the same base year, they are unlikely to be in perfect agreement. Fourth, a synthesized or hybrid inventory can provide a more accurate representation of emissions than any of its component inventories. ~~And f~~Fifth, extra effort and investigation related to the specific year and region of interest can yield significant improvements over standard emissions processing methodologies. And sixth, top-down emissions, such as those from aircraft observations, ~~are important~~can be used to verify bottom-up emissions and to improve ~~air quality~~AQ modelling performance, as demonstrated by the ~~ac~~companion~~ed~~ ~~air quality~~AQ modelling papers in this special issue (Stroud et al., 2018; Makar et al., 2018).

Nevertheless, although improved sets of emissions input files were generated during Phase 3 after a considerable effort to acquire and apply new sources of emissions data representative of the 2013 AOSR field-study period, there are still large uncertainties associated with these emissions. Here are six areas that still need further improvement:

1) Top-down emissions estimates from ~~A~~aircraft measurements made in late summer 2013 during the AOSR field study show that VOC and PM emissions reported to the NPRI using currently accepted estimation methods ~~are~~might be underestimated for the AOSR facilities (Li et al., 2017). However, these measurements were made during a limited time period (four weeks) and the mass-balance calculations used to estimate emissions were only applied to a relatively large area (Gordon et al., 2015; Li et al., 2017). Large variations in PM emissions results were also seen from flight to flight for the same facilities, probably related at least in part to the variation of mined volume of oil sands from day to day or recent precipitation. There are thus still issues with the spatial and temporal allocation of emissions to the right location at the right time. ~~More aircraft measurements, especially at other times of year, and further attempts to spatially locate emissions on a sub-facility level, are needed to confirm and augment the findings of the 2013 field study.~~

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The aircraft measurements also indicated that the VOC speciation reported to NPRI by individual AOSR mining facilities may need~~s~~ to be improved (Li et al., 2017), and additional VOC speciation data should be collected to improve speciation profiles. Moreover, these aircraft measurements were carried out at the facility level, but within these very large facilities the individual VOC species emitted from mine faces, tailings ponds, and plants can be very different. More aircraft measurements, especially at other times of year, and a~~A~~dditional measurements of emissions at the sub-facility level, from mine faces, tailings ponds, and plants for multiple AOSR facilities, are needed to confirm and augment the findings of the 2013 field study and to further improve emissions factors, temporal profiles, and chemical speciation profiles ~~that can improve the~~used for OS emissions inventories~~y~~ and emissions processing (e.g., Small et al., 2015; Stantec Consulting Ltd. et al., 2016). Given the above differences between field study measurements and reports, the AOSR mining facilities should also review the methodologies that they employ to estimate and report VOC emissions to NPRI.

2) The off-road mining fleets in the six AOSR mining facilities are a large source of NO_x emissions, but large differences are seen in the emissions estimates for this source sector between different inventories. For example, the 2010 CEMA inventory lists 38,362 tonnes of NOx emissions for this sector, but the 2010 APEI for the same year lists 27,786 tonnes. The 2013 APEI then reduced NOx emissions from the OS off-road mining fleets to 12,370 tonnes. Since mined oil sands increased by 17% between 2010 to 2013, the significant drop of NOx emissions is probably due to different emissions factors being used for these two inventory years (possibly due in part to the introduction of cleaner heavy-hauler trucks: e.g., M.J. Bradley & Associates, 2008).

Additional sources of information are needed to reconcile the differences amongst existing inventories. One possible data source is satellite remote sensing. For example, a methodology has been developed recently to use repeated satellite measurements of NO_2 vertical column density over the AOSR to estimate NO_x emissions (McLinden et al., 2014, 2016). Preliminary top-down results from satellite remote sensing show that area ~~-~~source NO_x emissions in the OS area, which are mainly from the off-road fleets, are about 38 kt per year for 2013, comparable to the bottom-up 2010 CEMA inventory. The 2010 CEMA inventory was also deemed to have the best estimation of off-road emissions for the AOSR facilities (ECCC & AEP, 2016). Satellite remote sensing (e.g., McLinden et al., 2014; Shephard

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et al., 2015; Sioris et al., 2017) and ground-based remote sensing (e.g., Fioletov et al., 2016), should thus be considered in future for emissions estimation and verification.

3) There have been ongoing efforts to improve the spatial allocation of emissions within the huge AOSR mining facilities using spatial surrogate fields generated from the locations of mine faces, tailings ponds, and extraction/upgrading plants. For example, the 2010 version of the shapefile used for generating these surrogates was updated in Phase 2 based on 2013 satellite images (Zhang et al., 2015). Further improvements, however, are possible. As one example, the spatial surrogate used to allocate emissions from the off-road mining fleet currently allocates all of the emissions to the mine-face locations and does not account for the movement of the heavy-hauler trucks between the mine faces and the extraction plants. Year-specific shapefiles with locations of active mining areas and current boundaries of tailing ponds as well as activity data sets for the actual or average movement of mining vehicles and time spent at locations throughout the mine should be obtained to improve the spatial allocation of off-road emissions for the AOSR mining operations (ECCC & AEP, 2016)

4) Fugitive VOC emissions from tailing ponds and mine faces are currently provided as annual totals in the inventory. A temperature-based monthly temporal profile was used to allocate the annual emissions to each month while weekly and diurnal temporal profiles were assumed to be constant, which is likely not realistic. For example, night-time emission rates over the ~~mining~~ mine faces are likely lower than daytime rates due to lower surface temperatures. In future, model-predicted or locally measured hourly temperature and wind speed may be used to estimate hourly fugitive VOC emissions if the dependence of fugitive VOC emission rates on temperature and wind speed can be parameterized (Li et al., 2017). Snow cover over the mining areas and ice cover over the ponds during winter-time also affect fugitive VOC emissions and need~~s~~ to be considered. A related issue is ~~that~~that the tailings ponds are of different ages; some are receiving fresh tailings while others have been inactive for years, which may mean lower emission rates due to past off-gassing of more volatile components. Consideration should thus be given to tailings-pond age when allocating VOC emissions between different tailings ponds. A recently completed study (summer 2017) of tailings pond emissions conducted by ECCC is expected to lead to improved estimates of emissions from these sources.

5) Top-down ~~F~~fugitive dust emissions estimates based on aircraft observations suggest large underestimates in the reported inventory totals, and GEM-MACH modelling suggests that even these revised estimates, or the fraction of their mass which is composed of base cations, ~~are~~ might be underestimated (Makar et al., 201~~8~~7, this issue). Further aircraft-based measurements of fugitive dust emissions and their speciation are needed to improve the emissions inventories used here. A parameterization of wind-blown dust emissions should also be added to GEM-MACH.

6) For mercury emissions, although unspeciated mercury emissions were obtained from inventories with base years close to 2013, chemical speciation was done crudely using speciation profiles for nine broad source categories. This methodology needs to be updated as more detailed speciation information becomes available in the future.

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REFERENCES

Ahmadov, R., McKeen, S., Trainer, M., Banta, R., Brewer, A., Brown, S., Edwards, P. M., de Gouw, J. A., Frost, G. J., Gilman, J., Helmig, D., Johnson, B., Karion, A., Koss, A., Langford, A., Lerner, B., Olson, J., Oltmans, S., Peischl, J., Petron, G., Pichugina, Y., Roberts, J. M., Ryerson, T., Schnell, R., Senff, C., Sweeney, C., Thompson, C., Veres, P. R., Warneke, C., Wild, R., Williams, E. J., Yuan, B., Zamora, R.: Understanding high wintertime ozone pollution events in an oil- and natural gas-producing region of the western US, Atmos. Chem. Phys., 15, (1), 411-429, 2015.

Akingunola, A., Makar, P.A., Zhang, J., Baratzadeh, P., Moran, M. D., Melick, R., Przybylo-Komar, E., Carmody, T.-L., and Mackay, K.: High resolution simulations of Alberta and Saskatchewan using GEM-MACH: Comparing continuous emissions monitoring versus inventory estimates, 8th International Workshop on Air Quality Forecasting Research, 10-12 Jan., Toronto, Ontario, Canada, https://cpaess.ucar.edu/sites/default/files/meetings/2017/iwaqfr/presentations/3.%20Ayodeji_Akingunola%2C%20350%20pm%20-%20IWAQFR_2017_Presentation_final.pptx, 2017~~a~~.

Akingunola, A., Makar, P.A., Zhang, J., Darlington, A., Li, S.-M., M. Gordon, Moran, M. D., and Zheng, Q.: A chemical transport model study of plume rise and particle size distribution for the Athabasca oil sands~~Impact of continuous monitoring emissions stack parameters on model simulations using 2.5km resolution GEM-MACH~~, ~~submitted, Atmospheric Chemistry and Physics Discussions~~ Atmos. Chem. Phys. Disc., https://www.atmos-chem-phys-discuss.net/acp-2018-155, 2018 (this special issue)~~, 2017b~~.

Alberta Energy: http://www.energy.alberta.ca/OilSands/791.asp, accessed on Aug. 8, 2017.

Alberta Energy Regulator: ST98 Alberta's Energy Reserves and Supply/Demand Outlook, Feb., available at http://aer.ca/data-and-publications/statistical-reports/crude-bitumen-production, accessed July 20, 2017, 2017a.

Alberta Energy Regulator: In situ impacts, available at http://aer.ca/about-aer/spotlight-on/oil-sands/in-situ-impacts, accessed on July 20, 2017b.

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Formatted: Font: 11 pt

Alberta Energy Regulator: ST39 Alberta Mineable Oil Sands Plant Statistics (Updated monthly, 3 months in arrears), Data in MS Excel., February, https://www.aer.ca/documents/sts/ST39-2013.xls (accessed on May 18, 2017), 2014.

Alberta Environment & Sustainable Resource Development: Report on the Inventory of Oil Sands Inventories, internal report, Apr., 88 pp., 2013.

Baumgardner, D., Dye, J.E., and Gandrud, B. W.: Calibration of the forward scattering spectrometer probe used on the ER-2 during the Airborne Antarctic Ozone Experiment. J. Geophys. Res., 94, 16,475-16,480, 1989. DOI: 10.1029/JD094iD14p16475.

Cenovus FCCL Ltd.: Environmental Impact Assessment - Cenovus FCCL Ltd. Narrows Lake Project, Volume 3 - Air quality, noise and health, https://open.alberta.ca/dataset/e41cfccf-1ab3-40ca-8efd-4079633216ac/resource/9f1ab94d-379c-4c7d-8544-ca709b3cacc4/download/Volume-3.pdf, 1607pp, June 2010 (last accessed on May 19, 2017).

Choi D.: MOVES2014 Overview and Plans for the Future, report for FACA MOVES Review Work Group, https://www.epa.gov/sites/production/files/2016-10/documents/moves2014-overview-and-plans-for-the-future.pdf, September 14, 2016 ((last accessed on November 1, 2017)

Clearstone Engineering Ltd.: Technical Report, Volume 2, Overview of CAC and Other Priority Substance Emissions Inventory, prepared for Environment Canada, June, 354 pp., 2014a.

Clearstone Engineering Ltd.: Technical Report, Volume 3, UOG Emissions Inventory Methodology Manual, prepared for Environment Canada, March, 217 pp., 2014b.

Clearstone Engineering Ltd.: Technical Report, Volume 4, Compendium of Factors and Terminology, prepared for Environment Canada, June, 180 pp., 2014c.

Clearstone Engineering Ltd.: Letter Report, Documentation for UOG emission inventory extrapolation database (ExtrapolateR.accdb), prepared for Environment Canada, March, 17 pp., 2014d.

Davies, M., Person, R., Nopmongcol, U., Shah T., Vijayaraghavan, K., Morris, R., and Picard, D.: Lower Athabasca Region Source and Emission Inventory, report prepared by Stantec Consulting Ltd. and ENVIRON International Corporation for Cumulative Environmental Management Association - Air Working Group, http://library.cemaonline.ca/ckan/dataset/0cfaa447-410a-4339-b51f-e64871390efe/resource/fba8a3b0-72df-45ed-bf12-8ca254fdd5b1/download/larsourceandemissionsinventory.pdf, 274 pp., 2012 (last accessed on October 24, 2017).

Dickson R. J. and Oliver W. R.: Emissions models for regional air quality studies. Environ. Sci. Technol., 25, 1533-1535, 1991.

Eldering, A. and Cass, G. R.: Source-oriented model for air pollutant effects on visibility. J. Geophys. Res., 101, 19343-19369, 1996.

Environment Canada: 2012 Air Pollutant Emissions Inventory & Trends: Methodology, Summary & Highlights, Internal report, 133 pp., March 2014.

Environment and Climate Change Canada: Guide for Reporting to the National Pollutant Release Inventory (NPRI), 2016 and 2017, http://www.ec.gc.ca/inrp-npri/AFC98B81-A734-4E91-BD16-C5998F0DDE6B/2016-17%20Guide%20for%20Reporting%20-%20EN.pdf, 2016 (last accessed May 19, 2017).

Environment and Climate Change Canada & Alberta Environment and Parks: Joint Oil Sands Monitoring Program Emissions Inventory Compilation Report, http://aep.alberta.ca/air/reports-data/documents/JOSM-EmissionsInventoryReport-Jun2016.pdf, 146 pp, 2016.

Eyth, A., Mason, R., and Zubrow, A.: Development and Status of EPA's 2011 Modeling Platform, 12th CMAS Conference, 28-30 Oct., Chapel Hill, North Carolina, https://www.cmascenter.org/conference//2013/slides/eyth_development_status_2013.pptx, 2013.

Formatted: English (U.K.)

Fioletov, V.E., McLinden, C.A., Cede, A., Davies, J., Mihele, C., Netcheva, S., Li, S.-M., O'Brien, J.: Sulfur dioxide (SO_2) vertical column density measurements by Pandora spectrometer over the Canadian oil sands. Atmos. Meas. Tech, 9, 2961-2976, 2016.

Fraser, A., Dastoor, A., and Ryjkov, A.: How important is biomass burning in Canada to mercury contamination? ~~submitted,~~ Atmos. Chem. Phys~~. Discuss.~~, 18, 7263-7286, https://doi.org/10.5194/acp-18-7263-2018, 2018 ~~https://doi.org/10.5194/acp-2017-736, 2017~~ (this special issue)~~, revised manuscript accepted for ACP~~.

Gordon, M., Li, S.-M., Staebler, R., Darlington, A., Hayden, K., O'Brien, J., and M. Wolde: Determining air pollutant emission rates based on mass balance using airborne measurement data over the Alberta oil sands operations. Atmos. Meas. Tech., 8, 3745–3765. doi:10.5194/amt-8-3745-2015, 2015.

Gordon, M., Makar, P. A., Staebler, R., Zhang, J., Akingunola, A., Gong, W., Li, S.-M.: A comparison of plume rise algorithms to stack plume measurements in the Athabasca Oil Sands, Atmos. Chem. Phys. Disc., https://doi.org/10.5194/acp-2017-1093, 2017 (this special issue).~~submitted, Atmospheric Chemistry and Physics Discussions (this special issue), 2017.~~

Houyoux, M. R., Vukovich, J. M., Coats, Jr., C. J., Wheeler, N. J. M., and Kasibhatla, P. S.: Emission inventory development and processing for the Seasonal Model for Regional Air Quality (SMRAQ) project. J. Geophys. Res., 105, 9079-9090, 1999JD900975, 2000.

Imperial Oil Ltd.: Environmental Impact Assessment – Kearl Oil Sands Project, Volume 5 - Air quality & noise, http://www.acee.gc.ca/050/documents_staticpost/cearref_16237/KR-0007-5.pdf, 706 pp, July 2005 (last accessed on May 19, 2017).

Joint Oil Sands Monitoring Plan: Integrated Monitoring Plan for the Oil Sands: Air Quality Component, ISBN 978-1-100-18813-3, 72 pp., 2011. Available at http://publications.gc.ca/site/eng/394253/publication.html (last accessed Nov. 6, 2017).

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Formatted: Default Paragraph Font

Karion, A., Sweeney, C., Petron, G., Frost, G., Hardesty, R. M., Kofler, J., Miller, B. R., Newberger, T., Wolter, S., Banta, R., Brewer, A., Dlugokencky, E., Lang, P., Montzka, S. A., Schnell, R., Tans, P., Trainer, M., Zamora, R., Conley, S.: Methane emissions estimate from airborne measurements over a western United States natural gas field. Geophys. Res. Lett., 40, (16), 4393-4397, 2013.

Kinnee, E., Geron, C., and T. Pierce, United States land use inventory for estimating biogenic ozone precursor emissions, Ecological Applications, 7, 46-58, 1997.

Li, S.-M., Leithead, A., Moussa, S. G., Liggio, J., Moran, M. D., Wang, D., Hayden, K., Darlington, A., Gordon, M., Staebler, R., Makar, P. A., Stroud, C. A., McLaren, R., Liu, P. S. K., O'Brien, J., Mittermeier, R. L., Zhang, J., Marson, G., Cober, S. G., Wolde, M., and Wentzell, J. J. B.: Differences between measured and reported volatile organic compound emissions from oil sands facilities in Alberta, Canada, PNAS, 114, E3756–E3765, doi:10.1073/pnas.1617862114, 2017.

Liggio, J., Li, S.-M., Hayden, K., Taha, Y. M., Stroud., C., Darlington, A., Drollette, B. D., Gordon, M., Lee, P., Liu, P., Leithead, A., Moussa, S. G., Wang, D., O'Brien, J., Mittermeier, R. L., Brook, J. R., Lu, G., Staebler, R. M., Han, Y., Tokarek, T. W., Osthoff, H. D., Makar, P. A., Zhang, J., Plata, D. L., and Gentner, D. R.: Oil sands operations as a large source of secondary organic aerosols. Nature, 534, 91–94, doi:10.1038/nature17646, 2016.

Makar, P. A., Moran, M. D., Scholtz, M. T., and Taylor, A.: Speciation of volatile organic compound emissions for regional air quality modelling of particulate matter and ozone. J. Geophys. Res., **108** (D2), 4041, doi:10.1029/2001JD000797, 51 pp, 2003.

Makar, P.A., Zhang, J., Akingunola, A., Moran, M. D., Hayden, K., Li, S.-M., Staebler, R., and Aliabadi, A.: The Sensitivity of Model Plume Rise to Emissions Inputs, 7th International Workshop on Air Quality Forecasting Research (IWAQFR), September 1–3, 2015, College Park, MD, http://www.arl.noaa.gov/documents/IWAQFR/Posters2015/S4_Makar_Poster_IWAQFR_2015.pdf, 2015.

Makar, P.A., Akingunola, A., Aherne, J., Cole, A. S., Aklilu, Y., Zhang, J., Wong, I., Hayden, K., Li, S.-M., Kirk, J., Scott, K., Moran, M. D., Robichaud, A., Cathcart, H., Baratzedah, P., Pabla, B., Cheung, P., ~~and~~ Zheng, Q., and Jeffries, D. S., Estimates of Exceedances of Critical Loads for Acidifying Deposition in Alberta and

Formatted: Font: English (U.S.)

Formatted: Font: (Default) Times New Roman, 10 pt, Font color: Auto, English (U.K.)

Saskatchewan, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-1094, 2018 (this special issue).(~~Estimates of acid deposition critical load exceedances for Alberta and Saskatchewan using GEM-MACH, submitted, Atmospheric Chemistry and Physics Discussions (this special issue), 2017.~~

Marson G.: Oil Sands Emission Inventories Review, draft internal report, Environment and Climate Change Canada, 56 pp, 2013.

McLinden, C. A., Fioletov, V., Boersma, K. F., Kharol, S. K., Krotkov, N., Lamsal, L., Makar, P. A., Martin, R.V., Veefkind, J. P., and Yang, K.: Improved satellite retrievals of NO_2 and SO_2 over the Canadian oil sands and comparisons with surface measurements. Atmos. Chem. Phys., 14, 3637-3656, 2014.

McLinden, C. A., Fioletov, V., Krotkov, N. A., Li, C., Boersma, K. F., and Adams, C.: A decade of change in NO_2 and SO_2 over the Canadian oil sands as seen from space. Environ. Sci. Technol., 50, 331-337, 2016.

M.J. Bradley & Associates: Evaluation of vehicle emissions reduction options for the oil sand mining fleet, final report prepared for Environment Canada, http://www.mjbradley.com/sites/default/files/Oil%20Sands%20Retrofit%20Feasibility%20Study%20Mar08_0.pdf, 40 pp., March 2008 (last accessed on September 8, 2017).

Moran, M. D., Ménard, S., Gravel, S., Pavlovic, R., and Anselmo, D.: RAQDPS Versions 1.5.0 and 1.5.1: Upgrades to the CMC operational Regional Air Quality Deterministic Prediction System released in October 2012 and February 2013, Technical note, Canadian Meteorological Centre, Montreal, April, 31 pp., 2013a [see http://collaboration.cmc.ec.gc.ca/cmc/cmoi/product_guide/docs/lib/op_systems/doc_opchanges/technote_rdaqps_20121003_e.pdf]

Moran, M. D., Dastoor, A., and Morneau, G.: Long-range transport of air pollutants and regional and global AQ modelling, In: Air Quality Management -- Canadian Perspectives on a Global Issue, E. Taylor and A. McMillan, Editors, ISBN 978-94-007-7556-5, Springer, Dordrecht, pp. 69-98, 2013b.

Moran, M. D., Ménard, S., Pavlovic, R., Anselmo, D., Antonopoulos, S., Makar, P. A., Gong, W., Stroud, C., Zhang, J., Zheng, Q., Robichaud, A., Landry, H., Beaulieu, P.-A., Gilbert, S., Chen, J., and Kallaur, A.: Recent

advances in Canada's national operational AQ forecasting system. Air Pollution Modeling and its Application XXII, NATO Science for Peace and Security Series C: Environmental Security, D.G. Steyn, P.J.H. Builtjes, and R.M.A. Timmermans, Editors, Springer, Dordrecht, pp. 215-220, DOI 10.1007/978-94-007-5577-2_4, 2014.

Moran, M. D., Zheng, Q., Zhang, J., and Pavlovic, R.: RAQDPS Version 013: Upgrades to the CMC Operational Regional Air Quality Deterministic Prediction System Released in June 2015. Technical note, November, Canadian Meteorological Centre, Montreal, 54 pp., 2015 [see http://collaboration.cmc.ec.gc.ca/cmc/cmoi/product_guide/docs/lib/op_systems/doc_opchanges/Technical_Note_GEM-MACH10_v1.5.3+SET2.1.1_Emissions_9Nov2015.pdf].

Peischl, J., Ryerson, T. B., Aikin, K. C., de Gouw, J. A., Gilman, J. B., Holloway, J. S., Lerner, B. M., Nadkarni, R., Neuman, J. A., Nowak, J. B., Trainer, M., Warneke, C., Parrish, D. D.: Quantifying atmospheric methane emissions from the Haynesville, Fayetteville, and northeastern Marcellus shale gas production regions. J. Geophys. Res.-Atmos., 120, (5), 2119-2139, 2015.

Pierce Jr., T. E., Kinnee, E. J., and Geron, C. D.: Development of a 1-km vegetation database for modeling biogenic fluxes of hydrocarbons and nitric oxide, Sixth International Conference on Air Surface Exchange of Gases and Particles, July 3-7, Edinburgh, England, https://www.epa.gov/sites/production/files/2015-08/beld3_web.ppsx, 2000.

Reff, A., Bhave, P. V., Simon, H., Pace, T. G., Pouliot, G. A., Mobley, J. D., and Houyoux, M.: Emissions inventory of $PM_{2.5}$ trace elements across the United States, Environmental Science and Technology, 43: 5790–5796, 2009.

~~Russell, M., Hakami, A., Makar, P.A., Akingunola, A., Moran, M. D., Zhang, J., and Zheng, Q.: submitted, Atmospheric Chemistry and Physics Discussions (this special issue), 2017.~~

Sassi, M., Cousineau, S., Duhamel, A., Ménard, S., Racine, J., Zaganescu, C., and Mashayekhi, R.: The 2013 Canadian air quality modelling platform and the base and future cases used for policy regulations, 15th Annual CMAS Conference, October 24-26, Chapel Hill, NC, https://www.cmascenter.org/conference//2016/abstracts/sassi_2013_canadian_2016.pdf, 2016.

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Formatted: Font: English (U.S.)

Shephard, M.W., McLinden, C. A., Cady-Pereira, K. E., Luo, M., Moussa, S. G., Leithead, A., Liggio, J., Staebler, R. M., Akingunola, A., Makar, P., Lehr, P., Zhang, J., Henze, D. K., Millet, D. B., Bash, J. O., Zhu, L., Wells, K. C., Capps, S. L., Chaliyakunnel, S., Gordon, M., Hayden, K., Brook, J. R., Wolde, M., and Li, S.-M.: Tropospheric Emission Spectrometer (TES) satellite observations of ammonia, methanol, formic acid, and carbon monoxide over the Canadian oil sands: Validation and model evaluation. Atmos. Meas. Tech,, 8, 5189-5211, 2015.

Sioris, C. E., McLinden, C. A., Shephard, M. W., Fioletov, V. E., and Abboud, I.: Assessment of the aerosol optical depths measured by satellite-based passive remote sensors in the Alberta oil sands region. Atmos. Chem. Phys., 17, 1931-1943, 2017.

Small, C. C., Cho, S., Hashishoa, Z., and Ulricha, A. C.: Emissions from oil sands tailings ponds: Review of tailings pond parameters and emission estimates. , J. Petrol. Sci. Eng., 127:490–501, 2015

Stantec Consulting Ltd., Clearstone Engineering Ltd., and Intrinsik Environmental Sciences Inc.: Air Pollutant and GHG Emissions from Mine Faces and Tailings Ponds, prepared for Alberta Environmental Monitoring, Evaluation and Reporting Agency, 413 pp., September 2016.

Stroud, C. A., Morneau, G., Makar, P. A., Moran, M. D., Gong, W., Pabla, B., Zhang, J., Bouchet, V. S., Fox, D., Venkatesh, S., Wang, D., and Dann, T.: OH-reactivity of volatile organic compounds at urban and rural sites across Canada: Evaluation of air quality model predictions using speciated VOC measurements. Atmos. Environ., **42**, 7746-7756, 2008.

Stroud, C. A., Makar, P. A., ~~Akinqunola , A.,~~ Zhang, J., Moran M. D., Akinqunola , A., Li, S.-M., Leithead, A., Hayden, K., Siu~~Wang~~, M~~D~~.: Air Quality Predictions using Measurement-1 Derived Organic Gaseous and Particle Emissions in a Petrochemical-Dominated Region, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-93, 2018 (this special issue).~~Impact of Updated Organic Gaseous and Particle Emissions on Air Quality Predictions in the Athabasca Oil Sands Region, submitted, Atmospheric Chemistry and Physics Discussions (this special issue), 2017.~~

Formatted: English (U.K.)

UNEP Chemical Branch: The Global Atmospheric Mercury Assessment: Sources, Emissions and Transport, UNEP-Chemicals, Geneva, https://wedocs.unep.org/bitstream/handle/20.500.11822/11517/UNEP_GlobalAtmosphericMercuryAssessment_May2009.pdf?sequence=1&isAllowed=y, 42 pp, 2008 (last accessed on November 15, 2017).

U.S. Environmental Protection Agency: Technical Support Document: Preparation of Emissions Inventories for the Version 4, 2005-based Platform, ftp://ftp.epa.gov/EmisInventory/2005v4/2005_emissions_tsd_07jul2010.pdf, 73 pp, July 2010 (last accessed on August 30, 2017).

U.S. Environmental Protection Agency: Technical Support Document: Preparation of Emissions Inventories for the Version 4.1, 2005-based Platform, https://www.epa.gov/sites/production/files/2015-11/documents/2005v4.1_main.pdf, 76 pp, March 2011(last accessed on November 15, 2017).

U.S. Environmental Protection Agency: Technical Support Document: Preparation of Emissions Inventories for the Version 6.2, 2011 Emissions Modeling Platform, https://www.epa.gov/sites/production/files/2015-10/documents/2011v6_2_2017_2025_emismod_tsd_aug2015.pdf, 204 pp, August 2015 (last accessed on August 8, 2017).

Wang, X., Chow, J. C., Kohl, S. D., Percy, K. E., Legge, A. H., and Watson, J. G.: Characterization of PM2.5 and PM10 fugitive dust source profiles in the Athabasca Oil Sands Region, Journal of the Air & Waste Management Association, 65:12, 1421-1433, DOI: 10.1080/10962247.2015.1100693, 2015.

Whaley, C. H., Makar, P. A., Shephard, M. W., Zhang, L., Zhang, J., Zheng, Q., Akingunola, A., Wentworth, G. R., Murphy, J. G., Kharol, S. K., and Cady-Pereira,K. E.: Contributions of natural and anthropogenic sources to ambient ammonia in the Athabasca Oil Sands and north-western Canada, Atmos. Chem. Phys., 18, 2011-2034, https://doi.org/10.5194/acp-18-2011-2018, 2018~~Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-627, 2017~~ (this special issue).

Zhang, J., Zheng, Q., Moran, M. D., Makar, P. A., Akingunola, A., Li, S.-M., Marson, G., Gordon, M., Melick, R., and Cho, S.: Emissions preparation for high-resolution air quality modelling over the Athabasca oil sands

region of Alberta, Canada. 21st Intern. Emissions Inventory Conference, 13-17 April, San Diego, http://www.epa.gov/ttn/chief/conference/ei21/session1/zhang_emissions.pdf, 18 pp, 2015.

~~**Table 1: Summary of Canadian emission sources used for generating JOSM Phase 1 emissions input files.**~~

~~Data Category~~	~~Data Sources~~
~~Point/Facility Sources~~	~~• 2009/10 CEMA Inventory for AOSR study area (except VOC, NH_3, PM_{10})~~
	~~• 2010 NPRI for rest of the domain~~
~~OS Off-road Fleet~~	~~• 2009/10 CEMA Inventory~~
~~Fugitive Dust from Major Facility~~	~~• 2010 NPRI~~
~~Tailings Ponds, Mines and Plant Fugitives~~	~~• 2010 facility-total VOC emissions from CEMA scaled by NPRI:CEMA~~
	~~• Splitting factors for fugitive VOC emissions for tailings ponds, mines and plants based on 2009/10 CEMA Inventory~~
~~Small & Medium Upstream Oil and Gas (UOG) Sources~~	~~• 2006 APEI (projected to 2006 from the 2000 Canadian upstream oil and gas emissions inventory)~~
~~Non-Mobile Area Sources~~	~~• 2006 APEI~~
~~Mobile Sources~~	~~• 2006 APEI~~

~~**Table 2**~~**Table 1: Comparison of annual facility-total VOC emissions (tonnes) between 2010 NPRI, 2010 CEMA, and versions 1 and 2 of the 2013 NPRI for the OS mining facilities within the AOSR study area.**

Emissions Processing Phase			1/2		3
Facility Name	**2010 APEI/NPRI**	**Original 2010 CEMA**	**2010-NPRI-Scaled CEMA**	**2013 APEI/NPRI**	**2013 NPRI version 2**
Suncor Millenium/Steepbank	28,940	10,808	28,013	6,768	9,529
Syncrude Mildred Lake	8,591	7,663	19,861	8,291	20,732
Syncrude Aurora North	5,182	3,319	8,602	2,572	8,268
Shell Muskeg River/Jackpine	1,460	2,813	7,291	2,614	2,614
CNRL Horizon	27,853	2,623	6,798	4,328	4,560
Imperial Oil Kearl				2,546	2,546

Total	**72,026**	**27,226**	**70,566**	**27,119**	**48,249**

~~**Table 3: Summary of Canadian emission sources used for generating JOSM Phase 2 emissions input files.**~~

~~Data Category~~	~~Data Sources~~
~~Point/Facility Sources~~	~~• 2009/10 CEMA Inventory for AOSR study area (except VOC, NH_3, PM_{10})~~
	~~• 2010 NPRI for rest of the domain~~
	~~• 2013 preliminary NPRI for AOSR Imperial Kearl facility and for NH_3 emissions~~
	~~• SO_2 and NO_x from CEMS measurements for stacks of the OS facilities during study period~~
	~~• SO_2 from CNRL daily reports during one-week period in August 2013~~
~~OS Off-road Fleet~~	~~• 2009/10 CEMA Inventory~~
~~Fugitive Dust from Major Facility~~	~~• 2013 preliminary NPRI~~
~~Tailings Ponds, Mines and Plant Fugitives~~	~~• 2010 facility-total VOC emissions from CEMA scaled by NPRI:CEMA~~
	~~• Splitting factors for fugitive VOC emissions for tailings ponds, mines and plants based on 2009/10 CEMA Inventory~~
~~Small & Medium UOG Sources~~	~~• 2010 APEI (projected to 2010 from the 2000 Canadian UOG emissions inventory)~~
~~Non-Mobile Area Sources~~	~~• 2010 APEI~~
~~Mobile Sources~~	~~• 2010 APEI~~

~~**Table 4**~~**Table 2: Summary of Canadian data sources used for generating JOSM Phase 3 base-case emissions input files.**

Data Category	Data Sources
Point/Facility Sources	• 2013 NPRI v1 for the whole domain except for the OS facilities
	• 2013 NPRI v2 for the OS facilities, but 2009/2010 CEMA stack information used
OS Off-road Fleet	• 2009/10 CEMA Inventory
Fugitive Dust from Major Facility	• 2013 NPRI v1
Tailings Ponds, Mines and Plant Fugitives	• Facility-total VOC emissions from 2013 NPRI v2
	• Splitting factors for fugitive VOC emissions from tailings ponds, mines and plants based on 2009/10 CEMA Inventory

Small & Medium UOG Sources	• 2013 APEI (projected from the 2011 Canadian UOG inventory)
Non-Mobile Area Sources	• 2013 APEI
On-road and Off-road Mobile Sources	• 2010 APEI

~~Table 5~~Table 3: PM10 size-bin ranges as Stokes diameter (μm) for the 12-bin version of GEM-MACH.

Bin 1	Bin 2	Bin 3	Bin 4	Bin 5	Bin 6	Bin 7	Bin 8	Bin 9	Bin 10
0.01-	0.02-	0.04-	0.08-	0.16-	0.32-	0.64-	1.28-	2.56-	5.12-
0.02	0.04	0.08	0.16	0.32	0.64	1.28	2.56	5.12	10.24

~~Table 6~~Table 4: Comparison of speciated annual ADOM-2 model VOC species emissions (tonnes/year) between base-case emissions from the 2013 NPRI version 2 (bottom-up) and the aircraft-observation-based estimates (top-down). Note that unknown or unreactive VOC species are not included.

	Suncor – M/S		Syncrude - ML		Shell – MR/J		CNRL - Horizon	
SPECIES	**Base Case**	**Aircraft**	**Base Case**	**Aircraft**	**Base Case**	**Aircraft**	**Base Case**	**Aircraft**
Higher Alkenes	601	1,038	863	513	34	1,219	177	1,657
Higher Alkanes	5,636	13,488	12,348	10,022	1,690	14,384	2,651	23,779
Higher Aldehydes	15	0.0	40	301	64	28	10	0.0
Higher Aromatics	1,457	1,569	5,273	1,696	746	88	1,125	500
Propane	0.5	953	0.0	1,592	3.1	955	0.0	1,928
Ethene	8.0	0.0	15	77	0.2	290	3.5	0.0
Formaldehyde	3.8	235	4.5	647	0.7	0.0	0.7	0.0
Isoprene	0.3	2,230	0.5	0.0	0.3	143	0.1	1,346
Toluene	486	1,112	806	1,539	6.8	72	135	393
Methyl ethyl ketone	0.0	0.0	0.0	212	0.0	0.0	0.0	0.0
Total VOC	8,208	20,625	19,350	16,600	2,545	17,180	4,102	29,603

~~Table 7~~Table 5: Sum of source-sector-specific mercury emissions (kg) for the 2011 U.S. inventory (version 1) and the 2010/2013 Canadian inventory.

Source Category	2011 U.S.	2010/13 Canada
Point	42,202	2,529

Area	4,321	1,803
On-road	358	2.3
Off-road	41	0.0
Total	**46,922**	**4,334**

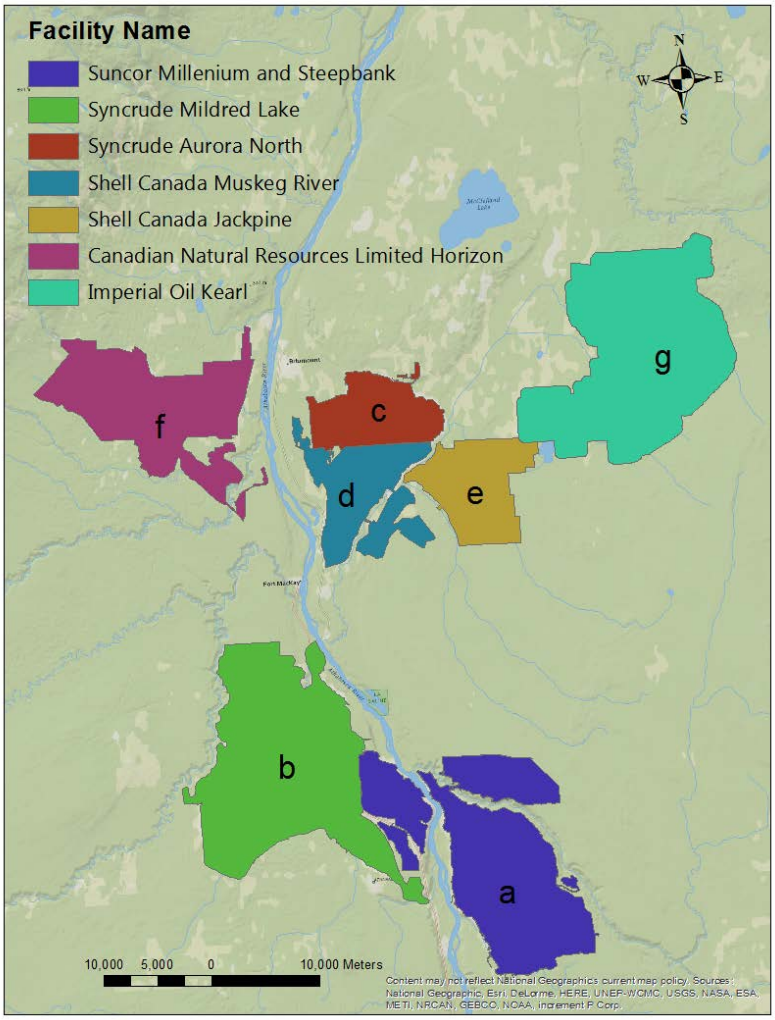

Figure 1: Location of six AOSR surface mining and processing facilities: (a) Suncor Millenium and Steepbank; (b) Syncrude Mildred Lake; (c) Syncrude Aurora North; (d) Shell Canada Muskeg River and (e) Shell Canada Jackpine (reported to NPRI as one facility); (f) Canadian Natural Resources Limited Horizon; and (g) Imperial Oil Kearl (only started production in 2013, not considered in earlier inventories). The city of Fort McMurray is located about 10 km to the south.

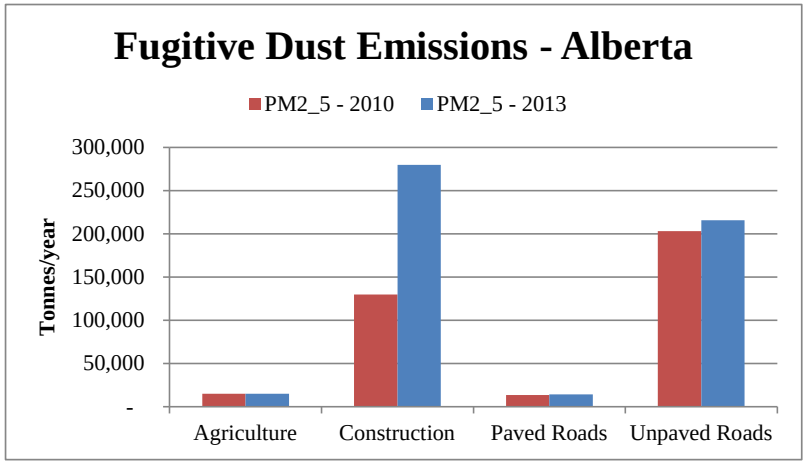

Figure 2: Comparison of fugitive $PM_{2.5}$ emissions for four sectors between 2010 APEI (used for Phase 2) and 2013 APEI (used for Phase 3) for the province of Alberta.

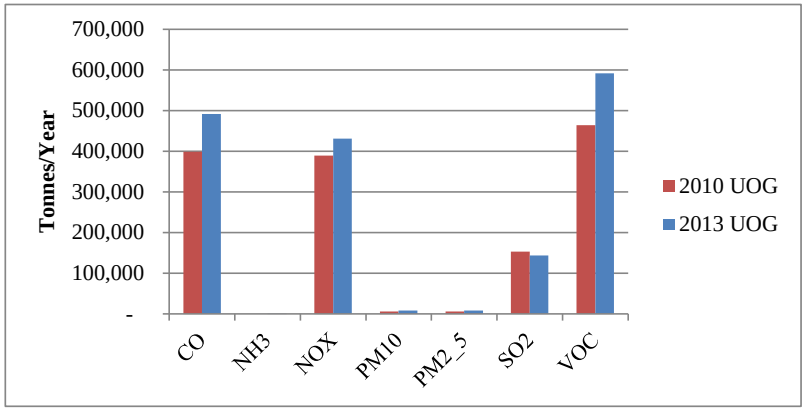

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Figure 3: Comparison of national CAC emissions between the year-2000-based projected 2010 UOG inventory and the year-2011-based projected 2013 UOG inventory.

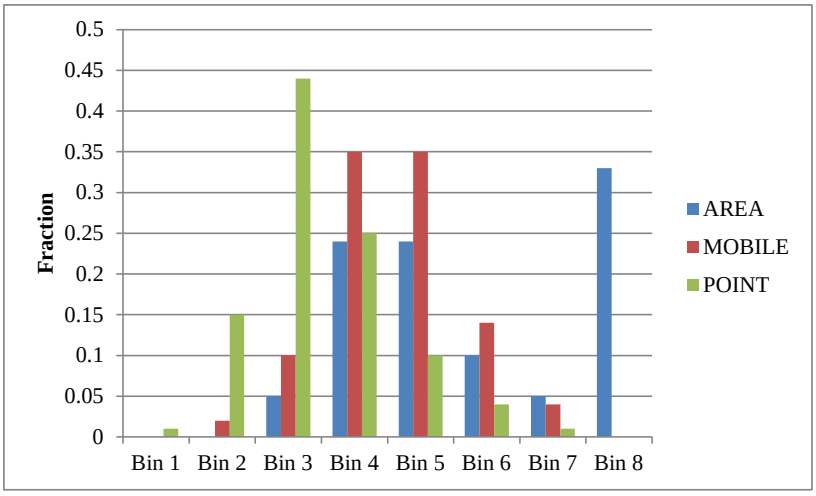

Figure 4: Fractional distribution of the eight $PM_{2.5}$ size bins for the 12-bin version of GEM-MACH modelling for three broad types of emissions sources.

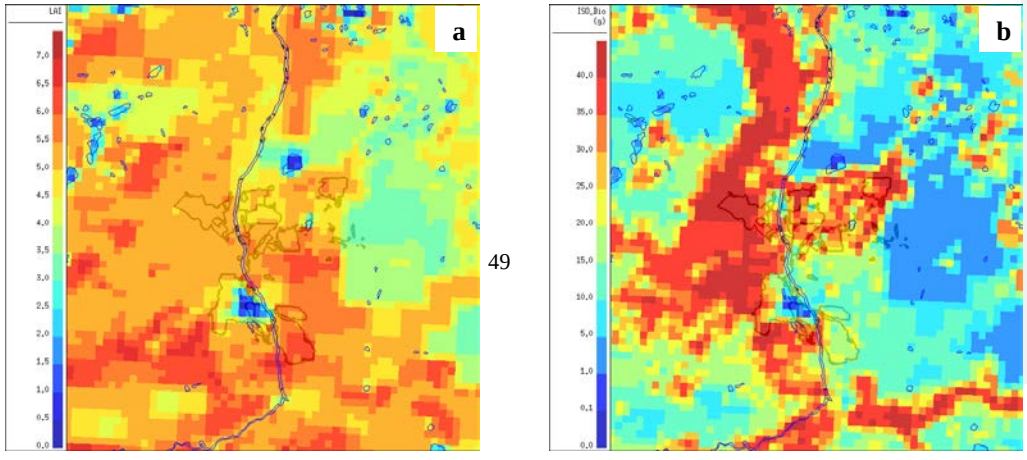

Figure 5: (a) Leaf Area Index and (b) peak summer isoprene emissions computed on the 2.5-km for a portion of the 2.5-km OS grid centred on the AOSR study area from the original BELD3 database. The gray lines indicate the cleared areas within the boundaries of the six AOSR mining and processing facilities (cf. Figure 1).

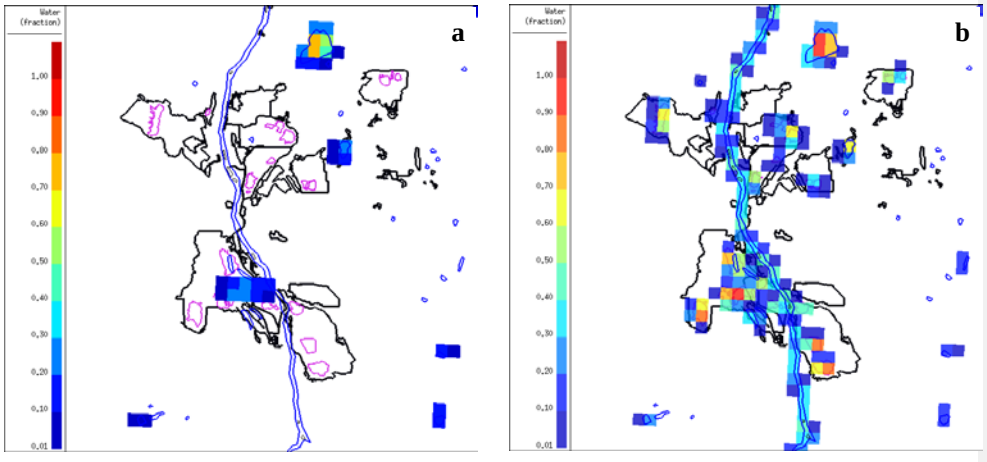

Figure 6. (a) Inland water coverage ~~on the 2.5-km~~ for a portion of the 2.5-km OS grid centred on the six AOSR mining and processing facilities generated from the original land-cover database (only natural lakes); and (b) modified inland water coverage including tailings ponds and rivers. The black and pink lines in panel (a) indicate the cleared-land areas and the tailings ponds within the boundaries of the six AOSR mining and processing facilities, whereas the blues lines in panel (a) mark the boundaries of natural lakes and rivers.

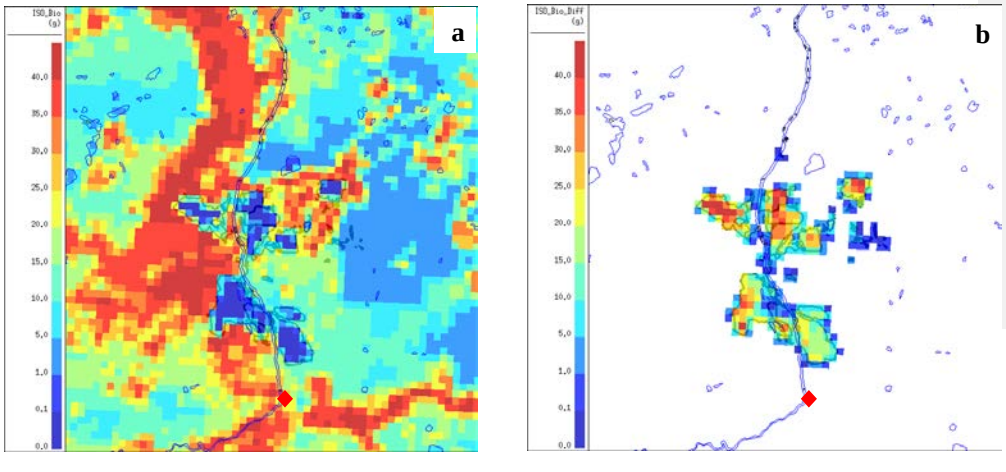

Figure 7: (a) Modified biogenic isoprene emissions for a portion of the 2.5-km OS grid centred on the AOSR study area and (b) difference between the original and the modified isoprene emissions (original – modified). The gray lines indicate the cleared-land areas within the boundaries of the OS mining facilities. The location of Fort McMurray is indicated by the diamond symbol.

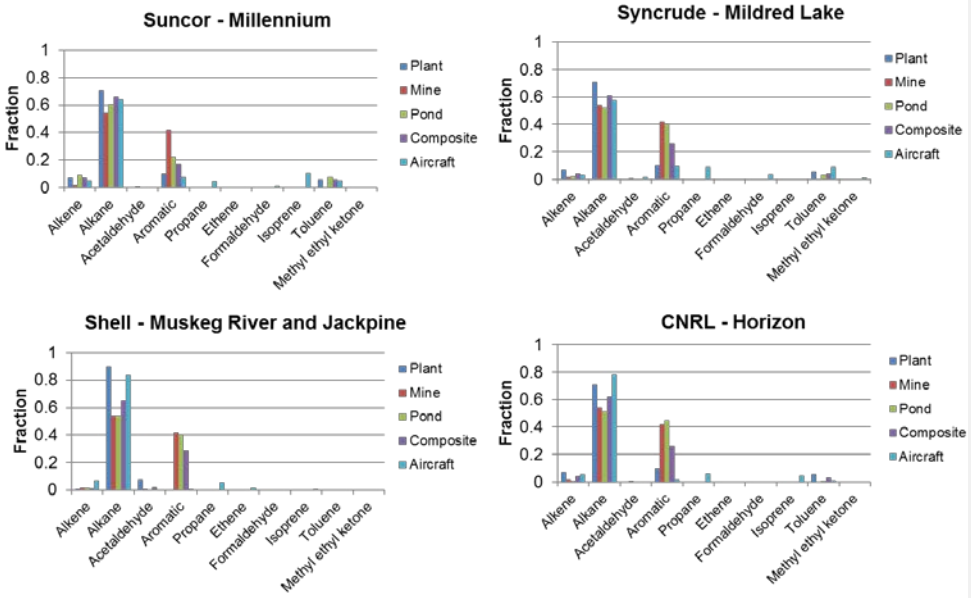

Figure 8: Comparisons of facility-specific VOC speciation profiles for ADOM-2-mechanism for four AOSR mining facilities used for the base-case study with facility-specific profiles derived from aircraft observations. Different VOC speciation profiles for plants, mine faces, and tailings ponds were used for the base-case study. The "composite"

VOC speciation profile for the base case is an emissions-weighted combination of the plant, mine-face, and tailings-pond profiles for each facility to allow comparison with the aircraft-based facility-specific VOC speciation profiles.

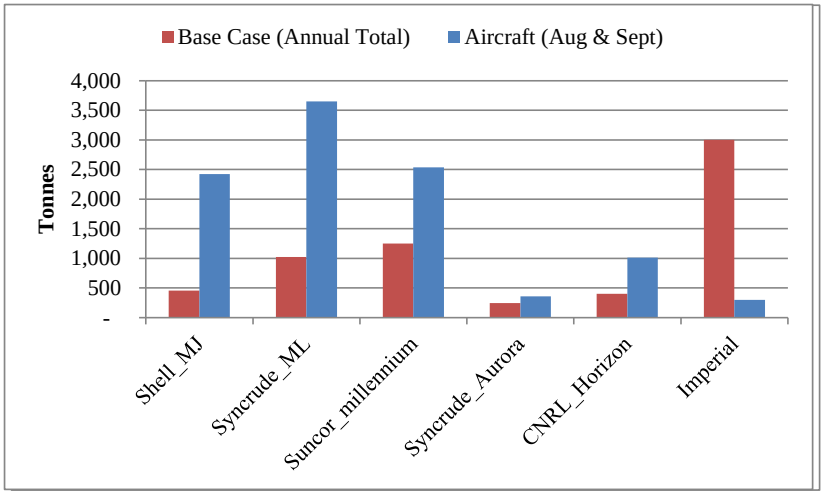

Figure 9: Comparison of ~~annual~~ $PM_{2.5}$ emissions between ~~annual~~ base-case annual emissions and aircraft-observation-based estimates for the two summer months (August and September) for the six AOSR mining facilities.

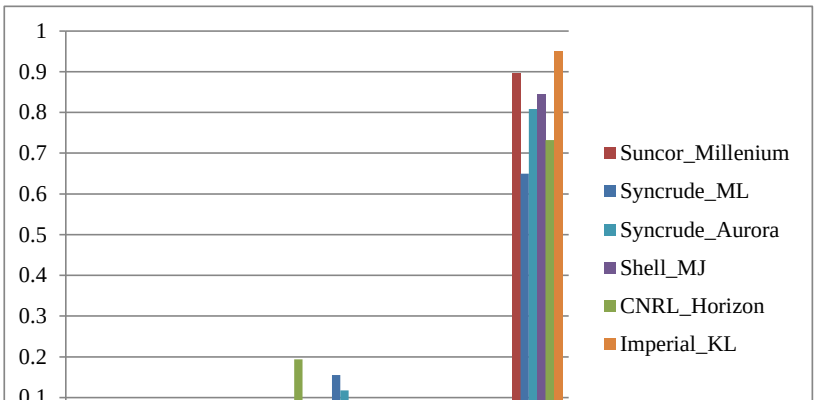

Figure 10: $PM_{2.5}$ size distribution derived from the aircraft observations for the six AOSR mining facilities.

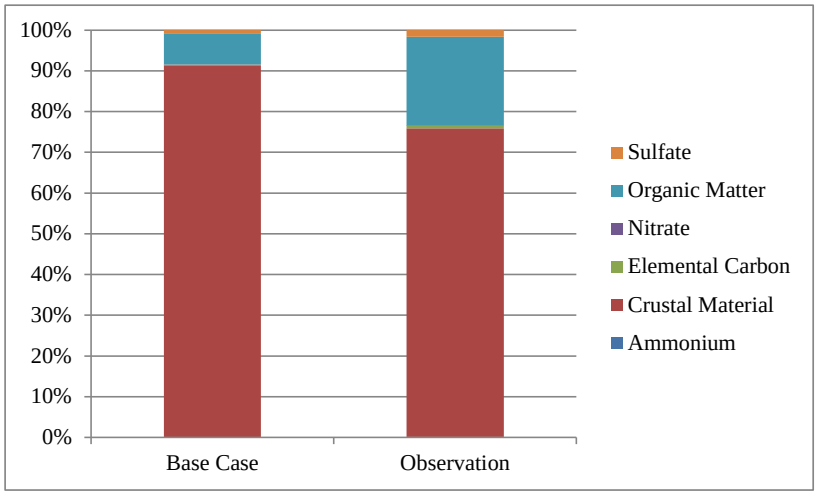

Figure 11: Comparison of the fugitive-dust PM speciation profile used for the base-case study and the one compiled from soil analyses from Wang et al. (2015) for the AOSR mining facilities.

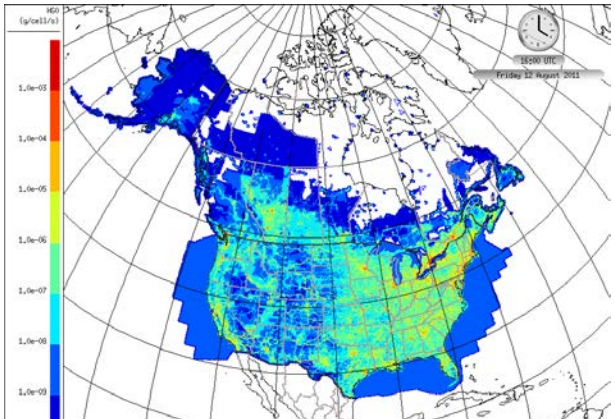

Figure 12: Spatial distribution of Phase 3 elemental mercury emissions for Canada and the U.S. for the 10-km continental model grid for one afternoon hour in August. Note logarithmic spacing of the emissions contour intervals; white areas have emissions less than 10^{-10} g/cell/s.

Appendix: List of acronyms used in the paper.

~~AAEI~~Acronym	~~Alberta Air Emissions Inventory~~Expansion
AAEI	Alberta Air Emissions Inventory
ADOM-2	Acid Deposition and Oxidant Model, version 2
AEP	Alberta Environment and Parks (formerly AESRD)
AER	Alberta Energy Regulator
AESRD	Alberta Environment and Sustainable Resource Development (now AEP)
AOSR	Athabasca Oil Sands Region
APEI	Air Pollutant Emission Inventory
AQ	air quality
BEIS	Biogenic Emission Inventory System
BELD	Biogenic Emissions Landuse Database
CAC	criteria air contaminants
CEMA	Cumulative Environmental Management Association
CEMS	Continuous Emission Monitoring System
CNRL	Canadian Natural Resources Limited
ECCC	Environment and Climate Change Canada
EIA	environmental impact assessment

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EPA	Environmental Protection Agency (U.S.)
EPEA	Environmental Protection and Enhancement Act (Alberta)
FSSP	Forward Scattering Spectrometer Probe
GEM-MACH	Global Environmental Multiscale model – Modelling Air-quality and CHhemistry
GIS	geographic information system
JOSM	Joint Oil Sands Monitoring plan
LAI	Leaf Area Index
LARP	Lower Athabasca Regional Plan
NEI	National Emissions Inventory
NPRI	National Pollutant Release Inventory
OS	oil sands
PFT	Paraffinic Froth Treatment
PM	particulate matter
SCC	Source Classification Code
SMOKE	Sparse Matrix Operator Kernel Emissions
TERRA	Top-Down Emission Rate Retrieval Algorithm
UHASP	Ultra-High Sensitivity Aerosol Spectrometer
UOG	upstream oil and gas
VOC	volatile organic compound
WBEA	Wood Buffalo Environmental Association

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Supplemental -Material for:

Emissions Preparation and Analysis for Multiscale Air Quality Modelling over the Athabasca Oil Sands Region of Alberta, Canada

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S1. Phase 1 hybrid emissions inventory and ancillary files

Prior to the 2013 AOSR field study, the ~~ten~~10 inventories listed in Table S1 by name, target region, and base year were reviewed to choose the most suitable emissions inventory data for AQ modelling for the OS area (AESRD, 2013; Marson, 2013). After an intensive review of these newer inventories, it became clear that no one inventory was the "best" choice in all respects, but three inventories contained emissions data that were either unique (i.e., not reported elsewhere), more detailed, and/or the most recent. The 2009/10 Cumulative Environmental Management Association (CEMA) inventory (Davies et al., 2012) had the most detailed stack- and process-level emissions for the AOSR surface mining facilities shown in Figure 1 (including separate emissions from mine faces, tailings ponds, and off-road mine hauler fleets, except for fugitive dust emissions from the off-road fleet); the 2010 Canadian NPRI included emissions for some species (NH_3 and PM_{10}) and source types (fugitive dust emissions from OS mine fleets) missing from the CEMA inventory; and the 2010 Canadian APEI from ECCC was the most comprehensive and had the largest spatial coverage (national) for area sources. Note, however, that at this time the 2010 Canadian APEI was not yet available in the detailed format required for emissions processing (referred to as the AQ modelling version), which requires the published APEI data to be transferred to a format that is suitable for processing emissions for AQ modelling as well as the addition of more detailed emissions data, such as monthly on-road and off-road emissions and process-based separation of emissions from some sectors (e.g., evaporative vs. exhaust emissions from on-road vehicles).

The solution adopted in the first phase for the best base-case inventory to use to generate GEM-MACH emissions input files for the 2013 field study was to create a synthesized or hybrid AQ modelling emissions inventory, as summarized in Table S2, that combined the best available information from the above inventories. The 2009/10 CEMA inventory, supplemented by the 2010 NPRI for NH_3 and PM_{10} emissions, was mainly used to provide emissions for the AOSR field-

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study area while the 2006 APEI was used outside the AOSR where the CEMA inventory's coverage ended. The 2010 NPRI was also used to scale the CEMA facility-total VOC emissions for the five AOSR surface mines active at that time (Figure 1), since it was found that the CEMA inventory had the lowest total VOC emissions for these five facilities compared to four other inventories (ECCC & AEP, 2016) and the NPRI is Canada's legislated inventory of large point sources based on emissions reported by facilities. The ratio of 2010 NPRI total VOC emissions for the five mining facilities vs. the CEMA total yielded a scaling factor of 2.6, which was applied to the CEMA facility-total VOC emissions for the individual facilities (Table 1), as large uncertainties may exist in both inventories and the use of a uniform scaling factor should not affect the impact of VOC emissions from the OS facilities as a whole. One reason to focus on the VOC emissions from these five facilities was that for 2010 they were estimated by the NPRI to have contributed 75% of VOC emissions from all Alberta facilities. The 2009/10 CEMA inventory was also used to specify the process-specific allocation of these facility-total emissions between mine faces, tailing ponds, plants, and smoke stacks, which then dictated the spatial and temporal allocation and chemical speciation of these process-level emissions (ECCC & AEP, 2016).

The focus of the OS field study was a roughly 100 km by 100 km subregion of the AOSR located north of Fort McMurray, Alberta (Figure 1). This study area contains a complex of six large surface bitumen mining and processing facilities situated on both sides of the Athabasca River. As shown in Figure 1, each mining facility covers a very large area, ranging from 66 to 275 km^2, and each facility contains various area sources within their boundaries, including NOx, CO, VOC, and $PM_{2.5}$ emissions from each mine's off-road heavy-hauler fleet, evaporative VOC emissions from tailings ponds and mine faces, and point sources of SO_2, NO_x, CO, VOC, $PM_{2.5}$ and fugitive VOC emissions from extraction and upgrading plants (Zhang et al., 2015). Although emissions from industrial facilities are normally treated as point sources by emissions processing systems and AQ models (e.g., Houyoux et al., 2000), each of these six facilities spans more than 10 GEM-MACH 2.5-km grid cells (area of 6.25 km^2 each), and many of the emissions are distributed over large areas within the facility boundaries. Treating such large facilities as point sources that can be assigned to a single grid cell is thus not realistic.

To address this concern, a new approach was taken in which these nominal point sources were treated as area sources. First, a GIS shapefile based on data collected by AESRD was obtained for the year 2010 with detailed locations of mine faces, extraction plants (and, for three facilities, upgrading plants), and tailings ponds for the five AOSR mines that were active within the study area at that time: Suncor Millenium and Steepbank mines; Syncrude Mildred Lake mine; Syncrude Aurora North mine; Shell Canada Muskeg River mine and Jackpine mine (known collectively as the Shell Canada Albian Sands mine); and CNRL Horizon mine (Figure 1). This shapefile was then used to develop three spatial surrogates for each facility to be used for spatial allocation of mine-face, ~~tailings-pond, and~~ extraction/upgrading plant ~~emissions~~, and tailings-pond emissions, respectively, including emissions from the off-road mining fleet and evaporative VOC emissions from mine faces, extraction plants, and tailing ponds (Zhang et al., 2015). It was assumed that the off-road fleets operated mainly in the mine-face areas, so the mine-face spatial surrogate field was used to allocate CAC emissions from the off-road fleet as well

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as evaporative VOC emissions from the mine faces. Note that emissions from the main smokestacks of the facilities were still treated as point sources. Finally, once all of the above development work was completed, the hybrid Phase 1 emissions inventory was input to the SMOKE (Sparse Matrix Operator Kernel Emissions) emissions processing system (https://www.cmascenter.org/smoke) together with the new AOSR facility-specific spatial surrogate fields to generate Phase 1 model-ready emissions input files for use by GEM-MACH during the 2013 summer field study (Zhang et al., 2015).

S2. Phase 2 hybrid emissions inventory and ancillary files

In Phase 2, after the field study, emissions updates were made during the 2014-2015 period to include newly available emissions information, including (i) an AQ modelling version of the 2010 Canadian APEI, (ii) a preliminary version of the 2013 NPRI point-source inventory, (iii) stack-level continuous emission monitoring system (CEMS) measurements for 17 smokestacks at four AOSR mining facilities for the field-study months of August and September 2013, and (iv) daily reports of SO_2 emissions during abnormal operating conditions from one AOSR mining facility (CNRL Horizon) during a one-week period in August 2013 when up to 20 times normal daily SO_2 emissions were released to the air during several upset events (ECCC & AEP, 2016). The six inventories and other emissions data sources that were used to create a second hybrid Canadian AQ modelling emissions inventory for 2013 are listed in Table S3.

The GIS shapefile describing the OS mines was also updated using 2013 satellite imagery (Zhang et al., 2015). These shapefile updates captured growth in the boundaries of existing mine faces and tailings ponds as well as new mine faces and tailings ponds that had been opened post-2010, and they were used to update the facility-specific spatial surrogate fields. In addition, a sixth mine, the Imperial Oil Kearl mine, had entered production in 2013 (see Figure 1). Annual emissions estimates for this facility were obtained from the preliminary 2013 NPRI and three new spatial surrogates were developed to allocate emissions from this facility (Zhang et al., 2015). As well, monthly facility-specific bitumen production data reported to the province of Alberta for 2013 for the six OS mining facilities were used to create facility-specific monthly temporal profiles (Alberta Energy Regulator (AER), 2014; Zhang et al., 2015). Note that a more comprehensive and detailed description of the Phase 2 hybrid inventory, the updated ancillary data sets for emissions processing, and the emissions processing procedure that was followed with the SMOKE system to generate model-ready emissions input files using the Phase 2 inventory is available in the JOSM report (ECCC & AEP, 2016).

~~**Table S1: List of acronyms used in paper.**~~

~~AAEI~~	~~Alberta Air Emissions Inventory~~
~~ADOM-2~~	~~Acid Deposition and Oxidant Model, version 2~~
~~AEP~~	~~Alberta Environment and Parks (formerly AESRD)~~

~~AER~~	~~Alberta Energy Regulator~~
~~AESRD~~	~~Alberta Environment and Sustainable Resource Development (now AEP)~~
~~AOSR~~	~~Athabasca Oil Sands Region~~
~~APEI~~	~~Air Pollutant Emission Inventory~~
~~AQ~~	~~air quality~~
~~BEIS~~	~~Biogenic Emission Inventory System~~
~~BELD~~	~~Biogenic Emissions Landuse Database~~
~~CAC~~	~~criteria air contaminants~~
~~CEMA~~	~~Cumulative Environmental Management Association~~
~~CEMS~~	~~Continuous Emission Monitoring System~~
~~CNRL~~	~~Canadian Natural Resources Limited~~
~~ECCC~~	~~Environment and Climate Change Canada~~
~~EIA~~	~~environmental impact assessment~~
~~EPA~~	~~Environmental Protection Agency (U.S.)~~
~~EPEA~~	~~Environmental Protection and Enhancement Act (Alberta)~~
~~GEM-MACH~~	~~Global Environmental Multiscale model – Modelling Air-quality and Chemistry~~
~~GIS~~	~~geographic information system~~
~~JOSM~~	~~Joint Oil Sands Monitoring plan~~
~~LAI~~	~~Leaf Area Index~~
~~LARP~~	~~Lower Athabasca Regional Plan~~
~~NEI~~	~~National Emissions Inventory~~
~~NPRI~~	~~National Pollutant Release Inventory~~
~~OS~~	~~oil sands~~
~~PM~~	~~particulate matter~~
~~SCC~~	~~Source Classification Code~~
~~SMOKE~~	~~Sparse Matrix Operator Kernel Emissions~~
~~TERRA~~	~~Top-Down Emission Rate Retrieval Algorithm~~
~~UOG~~	~~upstream oil and gas~~
~~VOC~~	~~volatile organic compound~~
~~WBEA~~	~~Wood Buffalo Environmental Association~~

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References

Alberta Energy Regulator: ST39 Alberta Mineable Oil Sands Plant Statistics (Updated monthly, 3 months in arrears), Data in MS Excel., February, https://www.aer.ca/documents/sts/ST39-2013.xls (accessed on May 18, 2017), 2014.

Alberta Environment & Sustainable Resource Development: Report on the Inventory of Oil Sands Inventories, internal report, Apr., 88 pp., 2013.

Davies, M., Person, R., Nopmongcol, U., Shah T., Vijayaraghavan, K., Morris, R., and Picard, D.: Lower Athabasca Region Source and Emission Inventory, report prepared by Stantec Consulting Ltd. and ENVIRON International Corporation for Cumulative Environmental Management Association - Air Working Group, http://library.cemaonline.ca/ckan/dataset/0cfaa447-410a-4339-b51f-e64871390efe/resource/fba8a3b0-72df-45ed-bf12-8ca254fdd5b1/download/larsourceandemissionsinventory.pdf, 274 pp., 2012 (last accessed on October 24, 2017).

Environment and Climate Change Canada & Alberta Environment and Parks: Joint Oil Sands Monitoring Program Emissions Inventory Compilation Report, http://aep.alberta.ca/air/reports-data/documents/JOSM-EmissionsInventoryReport-Jun2016.pdf, 146 pp, 2016.

Houyoux, M. R., Vukovich, J. M., Coats, Jr., C. J., Wheeler, N. J. M., and Kasibhatla, P. S.: Emission inventory development and processing for the Seasonal Model for Regional Air Quality (SMRAQ) project. J. Geophys. Res., 105, 9079-9090, 1999JD900975, 2000.

Marson G.: Oil Sands Emission Inventories Review, draft internal report, Environment and Climate Change Canada, 56 pp, 2013.

Zhang, J., Zheng, Q., Moran, M. D., Makar, P. A., Akingunola, A., Li, S.-M., Marson, G., Gordon, M., Melick, R., and Cho, S.: Emissions preparation for high-resolution air quality modelling over the Athabasca oil sands region of Alberta, Canada. 21st Intern. Emissions Inventory Conference, 13-17 April, San Diego, http://www.epa.gov/ttn/chief/conference/ei21/session1/zhang_emissions.pdf, 18 pp, 2015.

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Table S12: Emissions inventories reviewed in Phase 1 prior to the 2013 AOSR field study.

Inventory Name	Geographic Coverage	Base Year
Cumulative Environmental Management Association (CEMA) Air Working Group Emission Inventory	Lower Athabasca Region of Alberta	2009/2010
Lower Athabasca Regional Plan (LARP) Emissions Inventory	Lower Athabasca Region of Alberta	2006
Environmental Protection and Enhancement Act (EPEA) Approvals Emissions Data	entire province of Alberta	2010
Alberta Industrial Air Emissions Survey	entire province of Alberta	2010
Alberta Air Emissions Inventory (AAEI)	entire province of Alberta	2006-2008
Canadian National Pollutant Release Inventory (NPRI)	all of Canada	2010
Canadian Air Pollutant Emission Inventory (APEI, (not an AQ-modelling-ready version, NPRI is a subset of APEI)	all of Canada	2010
Wood Buffalo Emissions Inventory	WBEA Airshed Zone of Alberta	2005/2006
Two EPEA Approval Applications / Environmental Impact Assessments (EIA) emissions inventories (Frontier and Voyageur South)	project-specific area coverage (Alberta)	various years

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Table S2: Summary of Canadian emission sources used for generating JOSM Phase 1 emissions input files.

Data Category	Data Sources
Point/Facility Sources	• 2009/10 CEMA Inventory for AOSR study area (except VOC, NH_3, PM_{10})
	• 2010 NPRI for rest of the domain
OS Off-road Fleet	• 2009/10 CEMA Inventory
Fugitive Dust from Major Facility	• 2010 NPRI
Tailings Ponds, Mines and Plant Fugitives	• 2010 facility-total VOC emissions from CEMA scaled by NPRI:CEMA
	• Splitting factors for fugitive VOC emissions for tailings ponds, mines and plants based on 2009/10 CEMA Inventory
Small & Medium Upstream Oil and Gas (UOG) Sources	• 2006 APEI (projected to 2006 from the 2000 Canadian upstream oil and gas emissions inventory)

Non-Mobile Area Sources	• 2006 APEI
Mobile Sources	• 2006 APEI

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Table S3: Summary of Canadian emission sources used for generating JOSM Phase 2 emissions input files.

Data Category	Data Sources
Point/Facility Sources	• 2009/10 CEMA Inventory for AOSR study area (except VOC, NH_3, PM_{10})
	• 2010 NPRI for rest of the domain
	• 2013 preliminary NPRI for AOSR Imperial Kearl facility and for NH_3 emissions
	• SO_2 and NO_x from CEMS measurements for stacks of the OS facilities during study period
	• SO_2 from CNRL daily reports during one-week period in August 2013
OS Off-road Fleet	• 2009/10 CEMA Inventory
Fugitive Dust from Major Facility	• 2013 preliminary NPRI
Tailings Ponds, Mines and Plant Fugitives	• 2010 facility-total VOC emissions from CEMA scaled by NPRI:CEMA
	• Splitting factors for fugitive VOC emissions for tailings ponds, mines and plants based on 2009/10 CEMA Inventory
Small & Medium UOG Sources	• 2010 APEI (projected to 2010 from the 2000 Canadian UOG emissions inventory)
Non-Mobile Area Sources	• 2010 APEI
Mobile Sources	• 2010 APEI

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Table S43: Annual facility-total CO, NO_x, PM_{10}, $PM_{2.5}$, and SO_2 off-road vehicle tail-pipe emissions (tonnes) from 2010 CEMA inventory. These emissions were used for all three phases. (Note: a) PM_{10} emissions were estimated based on the $PM_{2.5}$ emissions and the typical PM_{10} to $PM_{2.5}$ ratio for similar type of off-road vehicles; b) emissions for the Imperial Oil Kearl facility were estimated based on mined oil sands statistics for 2013; and c) off-road VOC emissions are included in this table because they are not included in Table 12 as they are not required for NPRI reporting.)

Facility Name	CO	NO_X	PM_{10}	$PM_{2.5}$	SO_2	VOC

Suncor Millenium/Steepbank	9,087	10,768	484	444	62	1,173
Syncrude Mildred Lake	1,931	8,030	194	178	365	332
Syncrude Aurora North	1,341	7,045	159	146	117	219
Shell Muskeg River/Jackpine	4,577	6,935	225	206	128	653
CNRL Horizon	602	5,585	38	35	66	134
Imperial Oil Kearl	505	1,258	33	30	26	75
Total	**18,042**	**39,620**	**1,132**	**1,039**	**763**	**2,585**

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Table S54: Annual facility-total CO, NH_3, NO_x, PM_{10}, $PM_{2.5}$, and SO_2 emissions (tonnes) from stacks and area sources, except for road dust emissions from the off-road mining fleet, by phase for the three phases (P1, P2, P3).

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Species & Phase / Facility Name		Suncor Millenium/ Steepbank	Syncrude Mildred Lake	Syncrude Aurora North	Shell Muskeg River/Jackpine	CNRL Horizon	Imperial Oil Kearl	TOTAL
CO	P1	3,080	5,196	883	107	2,785	0	**12,051**
	P2	3,080	5,196	883	107	2,785	484	**12,535**
	P3	6,096	5,184	343	57	1,353	484	**13,517**
NH_3	P1	0	0	0	0	0	0	**0**
	P2	1	1,436	0	0	174	0	**1,612**
	P3	1	1,436	0	0	174	0	**1,612**
NO_X	P1	11,526	14,003	561	696	1,841	0	**28,629**
	P2	11,526	14,003	561	696	1,841	256	**28,884**
	P3	7,848	13,900	519	1,064	1,472	256	**25,059**
PM_{10}	P1	804	4,166	69	26	243	0	**5,308**
	P2	804	4,166	69	26	243	75	**5,382**
	P3	564	2,221	8	35	195	75	**3,098**
$PM_{2.5}$	P1	460	1,538	69	23	243	0	**2,332**
	P2	460	1,538	69	23	243	73	**2,406**
	P3	340	635	7	19	158	73	**1,233**
SO_2	P1	20,619	77,120	0	0	6,512	0	**104,251**
	P2	20,619	77,120	0	0	6,512	0	**104,251**
	P3	13,868	63,321	0	0	4,005	0	**81,194**

Table S65: Annual facility-total PM_{10} and $PM_{2.5}$ fugitive dust emissions from the off-road mining fleet (tonnes) for the three phases.

Facility Name		Suncor Millenium/ Steepbank	Syncrude Mildred Lake	Syncrude Aurora North	Shell Muskeg River/Jackpine	CNRL Horizon	Imperial Oil Kearl	TOTAL
PM_{10}	P1	4,086	1,958	1,702	972	1,642	0	**10,361**
	P2	4,667	2,091	954	2,330	2,099	8,236	**20,377**
	P3	4,667	2,091	954	2,330	2,099	8,236	**20,377**
$PM_{2.5}$	P1	409	196	170	97	164	0	**1,037**
	P2	467	209	95	233	210	2,921	**4,134**
	P3	467	209	95	233	210	2,921	**4,134**

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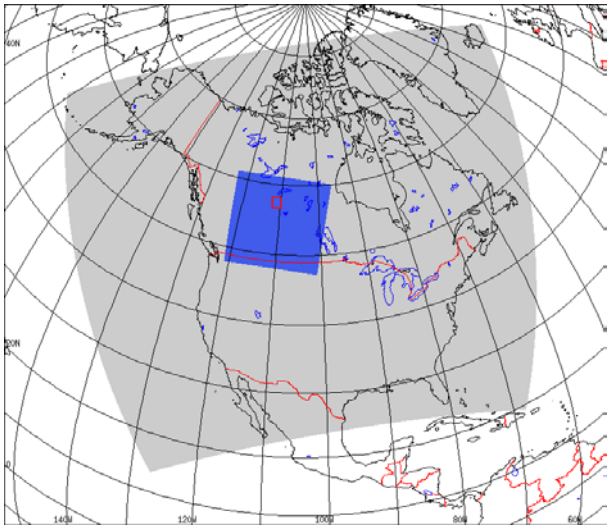

Figure S1: Locations of the North American continental model grid with 10-km grid spacing (grey), the OS western Canada grid with 2.5-km grid spacing (blue), and the subgrid shown in Figures 5 and 7 (red).

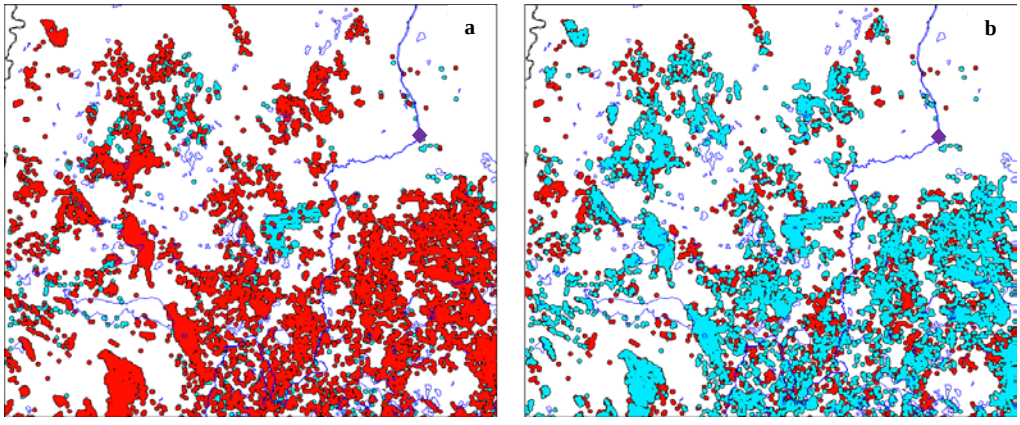

Figure S2: Location of UOG facilities in the vicinity of the Fort McMurray AOSR area from (a) the 2011-based projected 2013 inventory (red dots) superposed on the 2000-based projected 2010 inventory (cyan dots) and (b) the 2000-based projected 2010

inventory (cyan dots) superposed on the 2011-based projected 2013 inventory (red dots). In panel (a) the cyan dots not covered by the red dots are facilities that were in the projected 2010 inventory, but not in the projected 2013 inventory, whereas in panel (b) the red dots not covered by the cyan dots are facilities that were in the projected 2013 inventory, but not in the projected 2010 inventory. The location of Fort McMurray is marked by the purple diamond symbol.~~Location of UOG facilities in the Fort McMurray AOSR area from (a) the 2000-based projected 2010 inventory and (b) the 2011-based projected 2013 inventory (red dots) superposed on the 2000-based projected 2010 inventory (cyan dots). The location of Fort McMurray is marked by the diamond symbol.~~

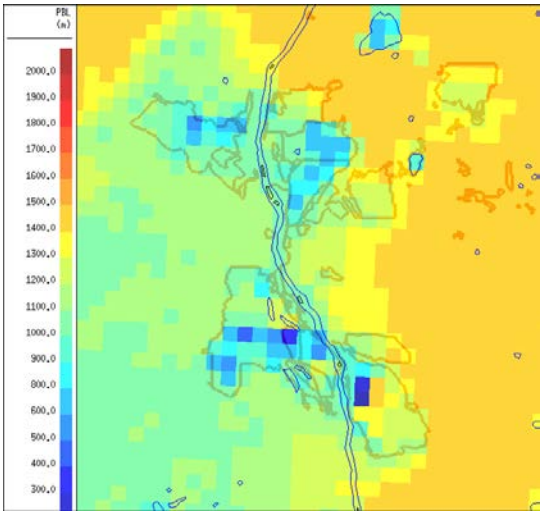

Figure S3: Planetary b~~B~~oundary l~~L~~ayer (PBL) height predicted by ~~the modeling~~GEM-MACH using the modified land cover data. The thick grey lines mark~~e~~ the boundaries of the ~~oil sand~~OS facilities and the thin blue lines mark ~~thee~~ boundary~~y~~ of natural lakes.

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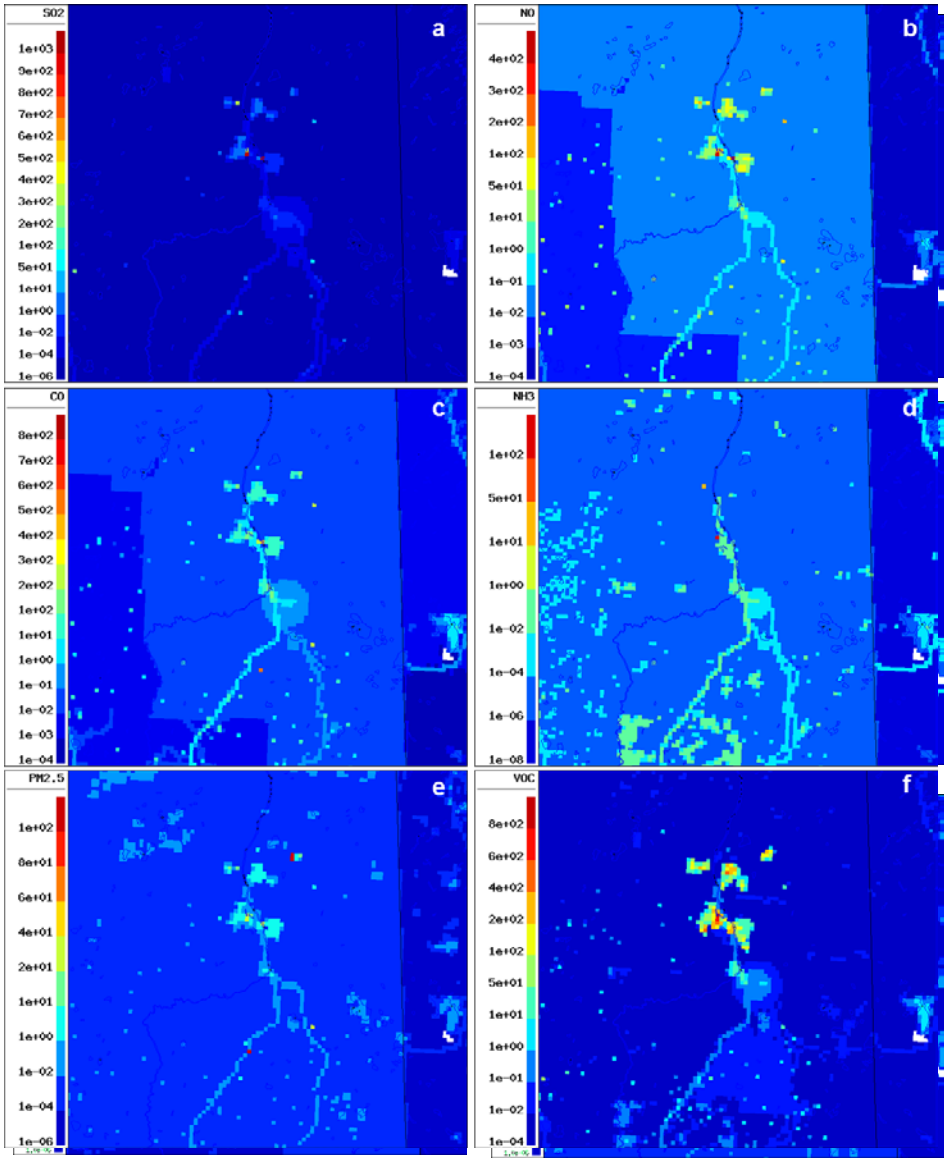
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9e+02
8e+02
7e+02
6e+02
5e+02
4e+02
3e+02
2e+02
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5e+01
1e+01
1e+00
1e-02
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5e+01
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7e+02
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5e+02
4e+02
3e+02
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6e+02
4e+02
2e+02
1e+02
5e+01
1e+01
1e+00
1e-01
1e-02
1e-04

Figure S43: Phase 3 August monthly emissions fluxes (tonnes month^{-1} grid-cell^{-1}) on an interior portion of the OS 2.5-km grid centred on the AOSR study area of the following GEM-MACH species: (a) SO_2; (b) NO; (c) CO; (d) NH_3; (e) $PM_{2.5}$; and (f) ~~C_2H_4 (ethene or ethylene)~~VOC.

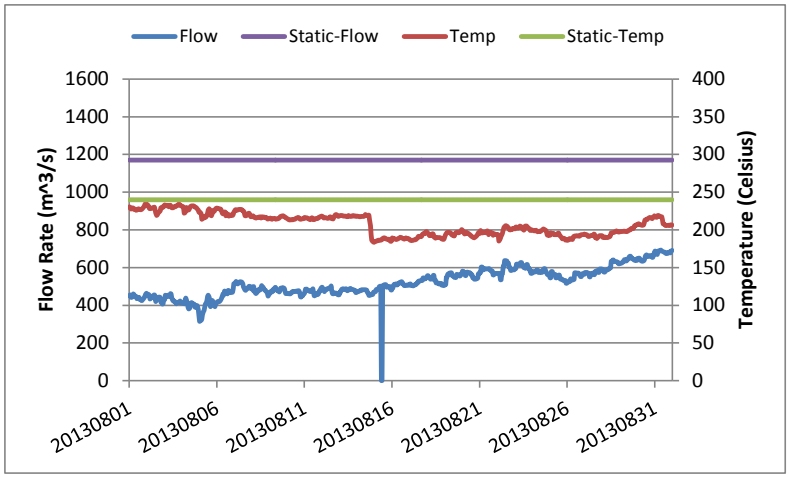

Figure S54: CEMS-measured hourly and NPRI static exit temperature (°C) and volume flow rate (m^3s^{-1}) for the Main stack of the Syncrude Mildred Lake facility for August 2013. Note that CEMS measurements are not available for some hours due to instrument calibration or other interruptions (e.g., flow rate on August 15). Those missing data have been filled by averaging the available data before and after the missing hours.

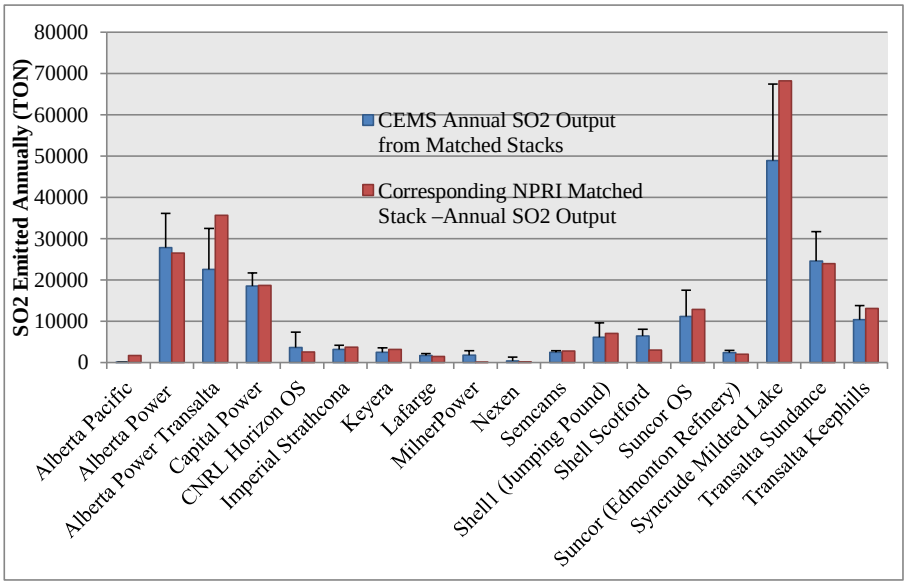

Figure S65: Comparison of annual SO_2 emissions estimated from CEMS measurements at 18 facilities and reported to NPRI for 2013. The error bars shown on the CEMS data represent the standard deviation of the hourly CEMS data measured for August and September, 2013.

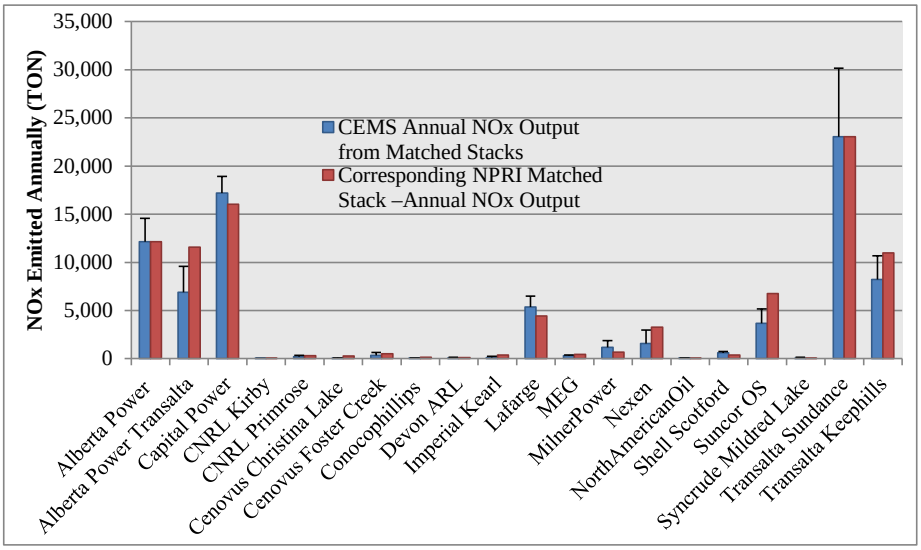

Figure S76: Same as Figure S5, but for NOx comparison and 20 facilities.

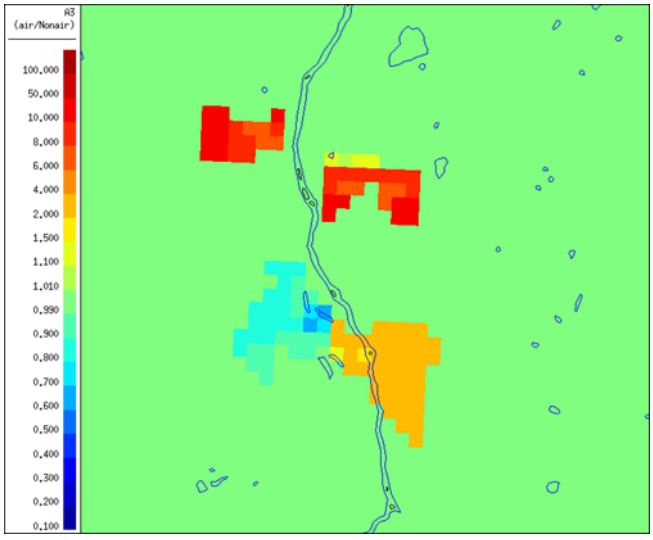

Figure S8~~7~~: Ratio of gridded model-ready aircraft-observation-based ADOM-2 higher-alkane emissions (top-down) to the base-case emissions (bottom-up) for the GEM-MACH 2.5-km grid over the AOSR area.

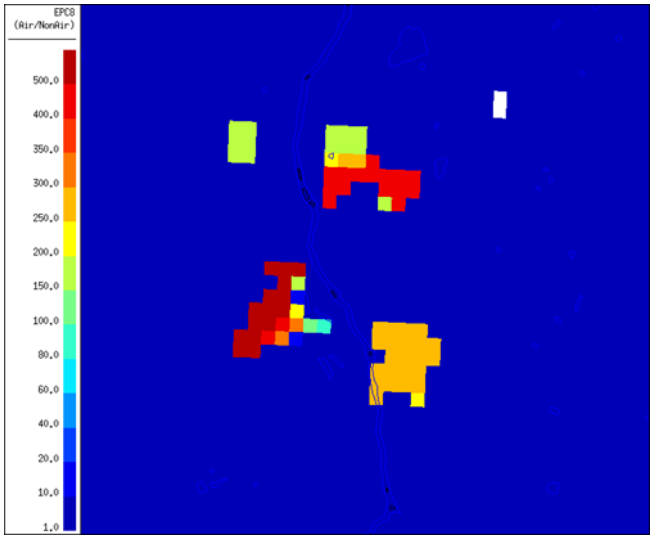

Figure S9~~8~~: Ratio of gridded, model-ready aircraft-observation-based (size) Bin 8 OM emissions to the base-case emissions for the GEM-MACH 2.5-km grid over the AOSR study area.

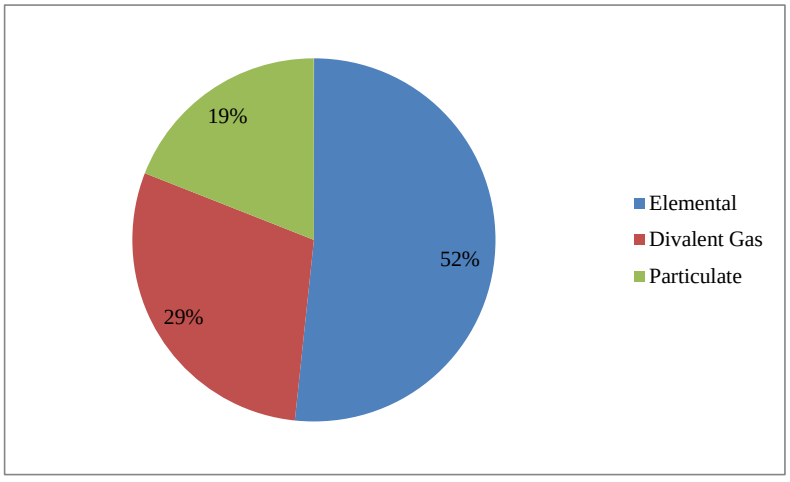

Figure S10~~9~~: Domain-average percentages of the three speciated mercury species for Phase 3 emissions.