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Supporting Information for

High resolution mapping of nitrogen dioxide with TROPOMI:

First results and validation over the Canadian oil sands

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Introduction

Supplementary material for "High resolution mapping of nitrogen dioxide with TROPOMI: First results and validation over the Canadian oil sands" by D. Griffin et al. This document contains further details about the methodology used in this study to determine the alternative air mass factors (AMFs) as well as details about the collection of the ground-based in-situ measurements. Figures that help with the interpretation of the results, but could not be included in the main manuscript (due to size limitations) are also included here.

Text S1.

Alternative Air Mass Factors

The information on the NO_2 profile shape is taken from ECCC's air quality forecast model; the Global Environmental Multiscale - Modelling Air-quality and Chemistry (GEM-MACH). The operational version of the model (Moran et al, 2010; Pendlebury et al, 2018) has a 10x10km2 grid cell size for North American domain, a 2-size bin aerosol size distribution and 8 chemical species (sulphate, nitrate, ammonium, primary organic aerosol, secondary organic aerosol, sea-salt, black carbon and crustal material). The operational forecast makes use of 2013 emissions information (Zhang et al., 2018). Here, we use the daily model output for the closest hour of the measurements and the closest grid-box of the 10km resolution version of GEM-MACH. The TM5-MP model used for the standard TROPOMI product has global coverage but with coarser resolution (1x10 , or about 111 x 111 km2) and thus will be unable to capture the NO_2 profile distribution, due to very localized enhancements. This performance can be improved by using input from regional models. The meteorological component of GEM-MACH is within the physics module of the Global Environmental Multiscale (GEM) weather forecast model (Coté et al., 1998; Girard et al., 2014). The emissions used in the model are processed using the Sparse Matrix Operator Kernel Emissions (SMOKE; Coats et al., 1996). Further details on GEM-MACH can be found in, e.g., Makar et al. (2015a,b) and Akingunola et al. (2018). To generate an improved a priori NO_2 profiles, we use the NO_2 concentrations from 0-1.5 km from the GEM-MACH model for the closest hour of the TROPOMI overpass. Between 1.5-12 km we use the concentrations from a monthly GEOS-Chem model run at the approximate time of the TROPOMI overpass on a 0.5x0.67^0 resolution version v8-03-01 (http://www.geos-chem.org) (Bey et al., 2001; McLinden et al., 2014), as the GEM-MACH model currently does not include NOx sources in the free troposphere, such as lightning and aircraft emissions.

MODIS provides white-sky albedo (WSA) and black-sky albedo (BSA), based on 16-day averages available every 8 days, at a resolution of 0.05x0.05^0

(collection 6.1 MCD43C3; Schaaf et al., 2002). From this, a monthly-mean albedo is computed considering only 100 % snow-free pixels. For surfaces with snow-cover, a climatology of the MODIS surface reflectance is used that only includes pixels with full snow-cover. To determine whether the TROPOMI pixel is snow covered, we use the daily IMS snow flag (http://www.natice.noaa.gov/ims/) on a on a 4x4 km resolution. It has been shown that the IMS product is better suited than other snow-products in differentiating between snow and snow-free scenes (Cooper et al., 2018), including the NISE snow flag used for the standard TROPOMI product that has a tendency of missing thin snow layers (McLinden et al., 2014). The MODIS snow albedo (see supplementary material) shows that the value over snow and ice is not necessarily 0.6 as assumed for the original TROPOMI product. For many areas in North America this can be as high as 0.9, however, over the boreal forest the reflectance is relatively low (0.2-0.3) even with snow cover.

Text S2.

Wood Buffalo Environmental Association

The Wood Buffalo Environmental Association (WBEA; www.wbea.org) operates 26 continuous air monitoring stations in and around the oil sands region (Percy et al., 2012). Some of these stations are equipped with in-situ NO2 detectors (Percy, 2013; Hsu et al., 2010). A summary of the stations used in this study is given in Table S1 and the location of the stations is shown in Fig. S1.

The WBEA data protocols, standard operating procedures, and quality control/quality assurance procedures are all compliant with the regulations for routine monitoring. This includes daily zero/span calibration and monitoring of instrument performance, monthly multi-point calibrations, annual independent third-party audits, and independent system evaluations conducted every three years (Phillips, 2010).

Different types of NO2 detectors are deployed but they are all Chemiluminsescent gas analyzers and possess a similar performance with a precision of approximately 0.4 ppb according to manufacturer product specifications (see https://www.thermofisher.com/order/catalog/product/42I for the Thermo instruments and http://eservices.teledyne-api.com/products/T200.asp for the Teledyne instruments). Accuracy is more difficult to gauge since this NO2 measurements method is also sensitive to other reactive non-NOx compounds such as nitric acid (e.g., U.S. Environmental Protection Agency [1975]). The magnitude of this high bias depends on many factors (including the time since NOx emission). However, systematic errors are not as relevant in this study since these data are only used to evaluate correlation with satellite observations.

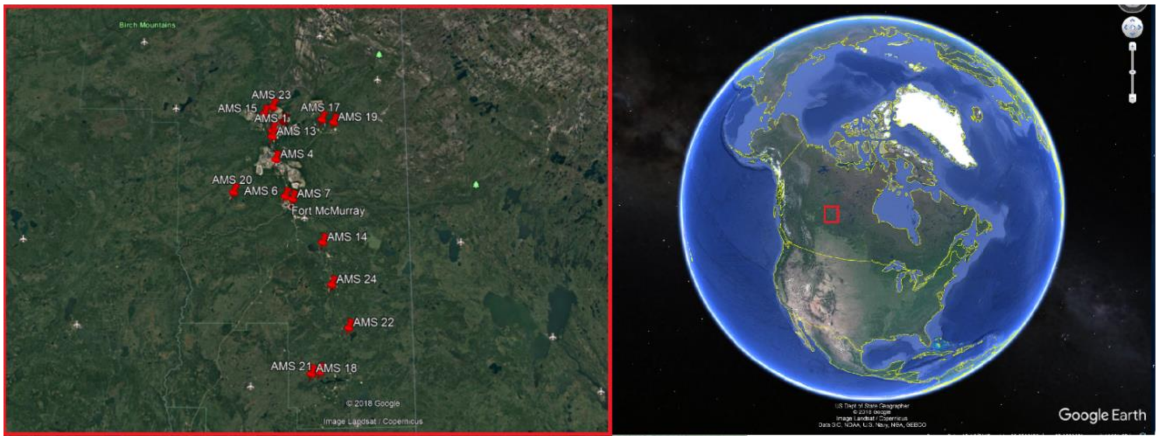

Figure S1. Location of the current WBEA monitoring stations measuring NO_2.

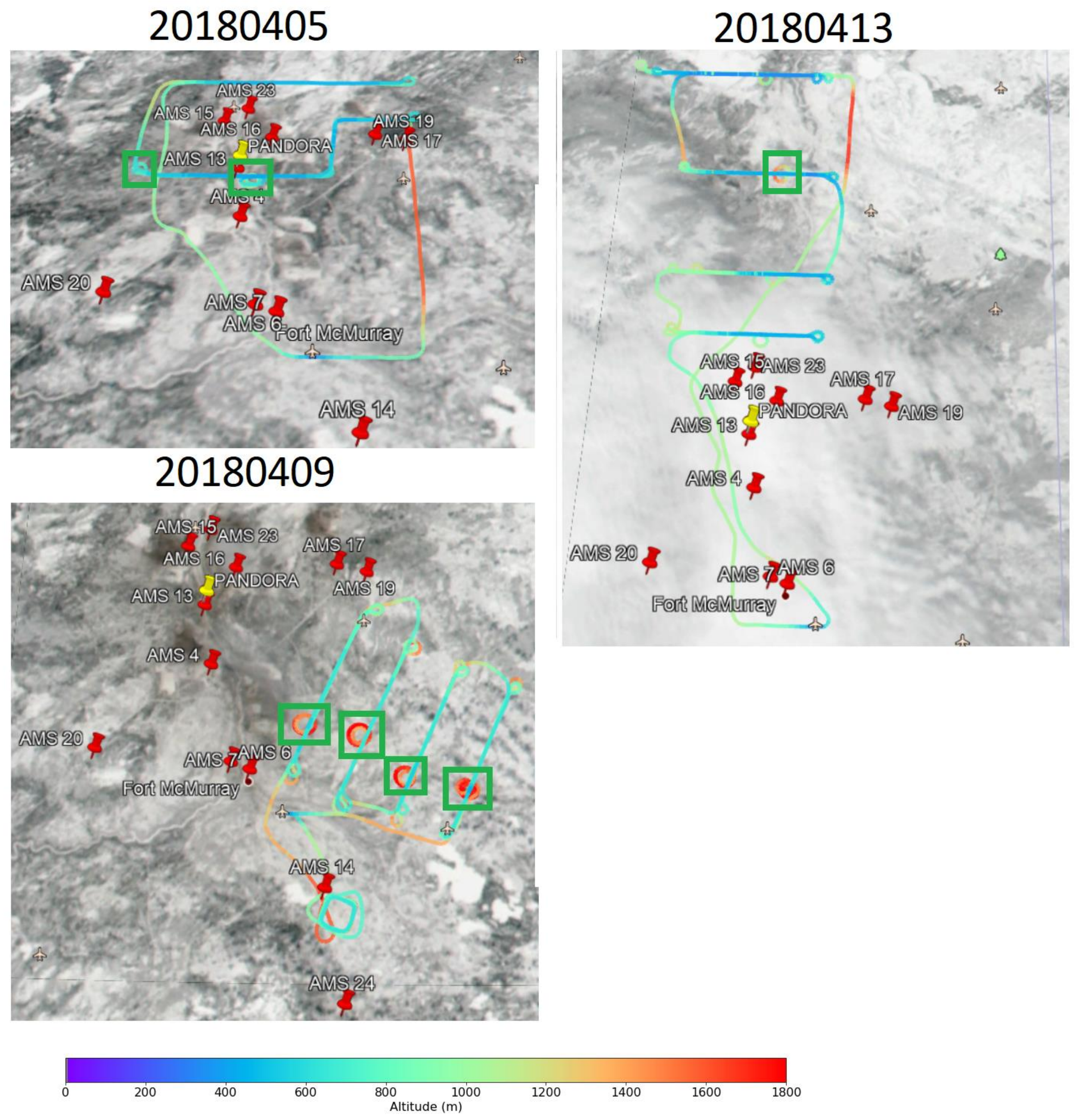

Figure S2. Showing the three trajectories from the flight on April 5, 9, and 13, 2018. Overlaid is the Visible Infrared Imaging Radiometer Suite (VIIRS) image of the corresponding day showing that the surface was covered in snow and ice, and sky was cloud free in the area. The altitudes are in units of m asl; the surface elevation is approximately 300 m asl in this area. The red pins indicate the location of the WBEA measurement sites, and the yellow pin shows the location of the Pandora site. The green boxes highlight the location of the spirals.

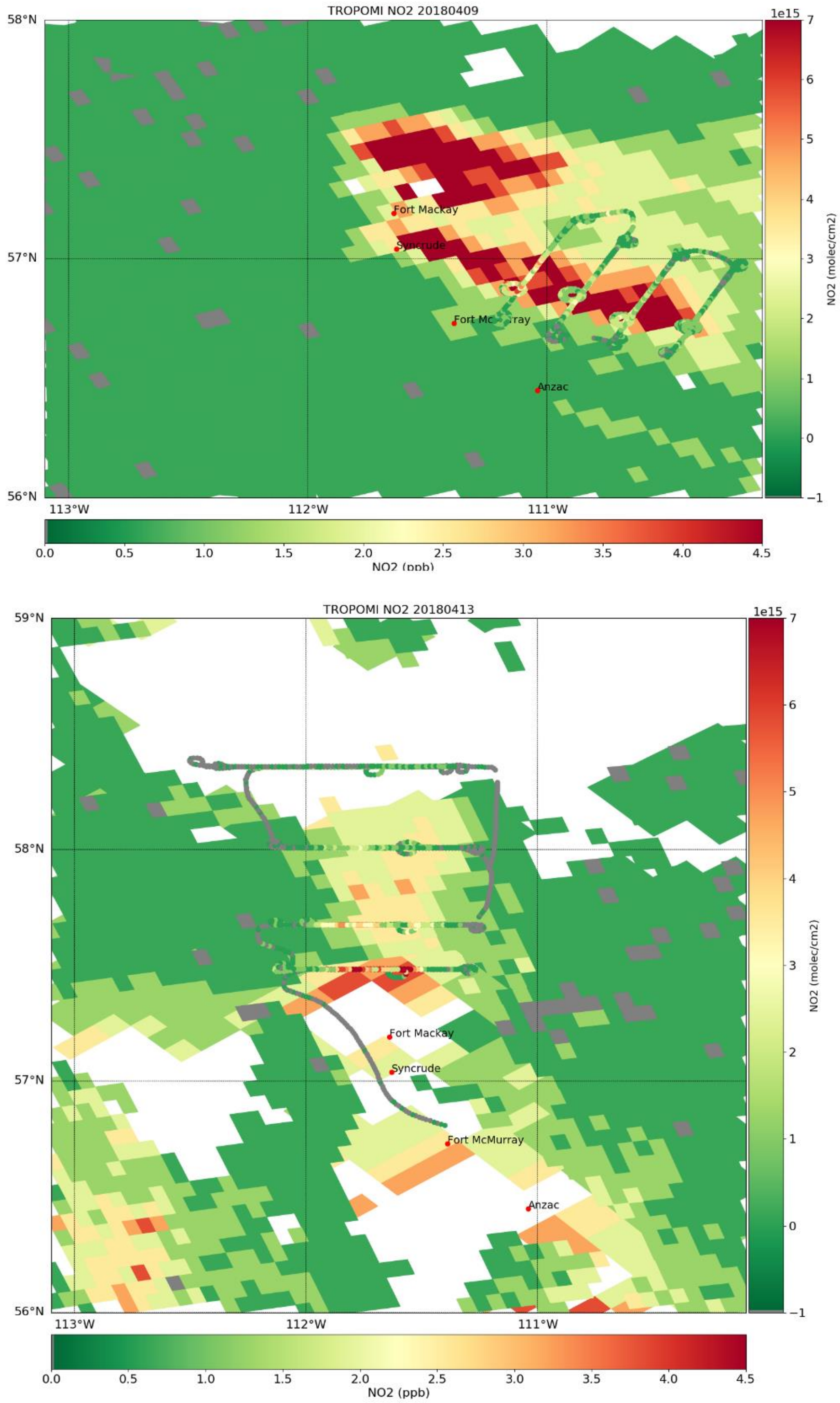

Figure S3. Map around the Oil Sand region showing the concentrations as sampled by the aircraft and the pixels show the tropospheric NO2 VCD as measured by TROPOMI for April 9 and 13, 2018.

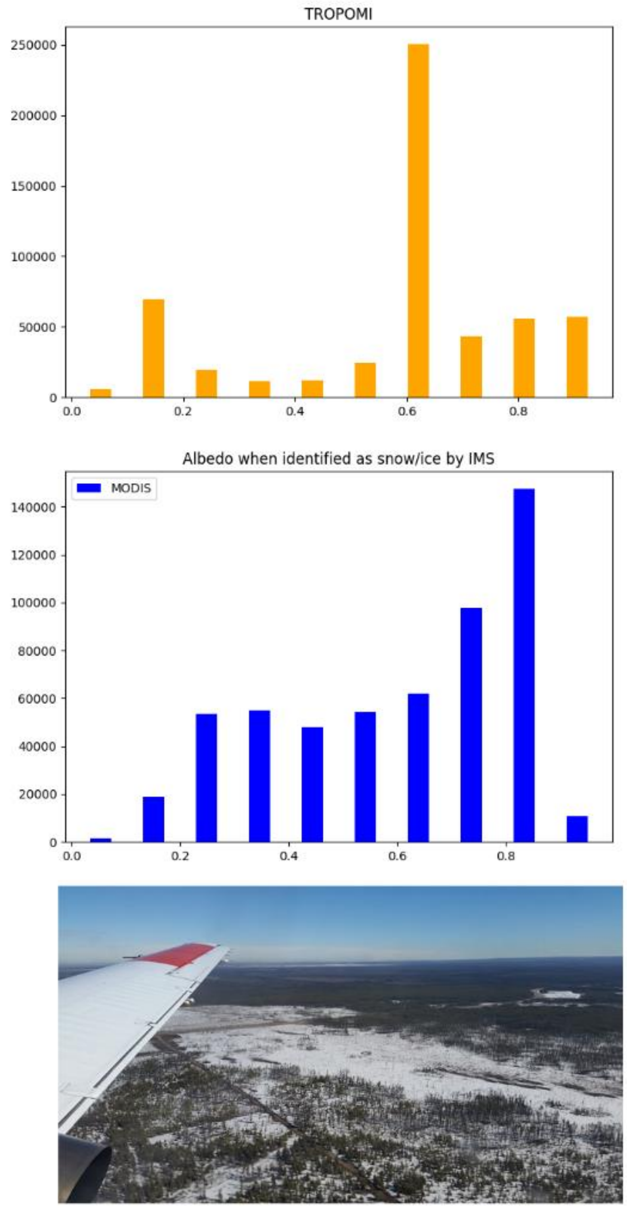

Figure S4. Example (April 9, 2018) of the albedo over snow covered surfaces over North America (as identified by the daily Interactive Multisensor Snow and Ice Mapping System (IMS; http://www.natice.noaa.gov/ims/). The albedo used for the standard TROPOMI product is snow in orange, it is set to 0.6 for snow cover, the values that are not 0.6 occur if the snow cover has not been identified by the TROPOMI snow flag. The orange bars show the surface reflectance using a MODIS climatology for snow-covered surfaces. Over the boreal forest (near the Athabascan Oil Sand Region) the albedo can be as low as 0.1-0.2 even if there is snow. The photograph on the right was taken out of the aircraft flight during the campaign showing that the surface reflectance is quite low due to the dark trees of the boreal forest.

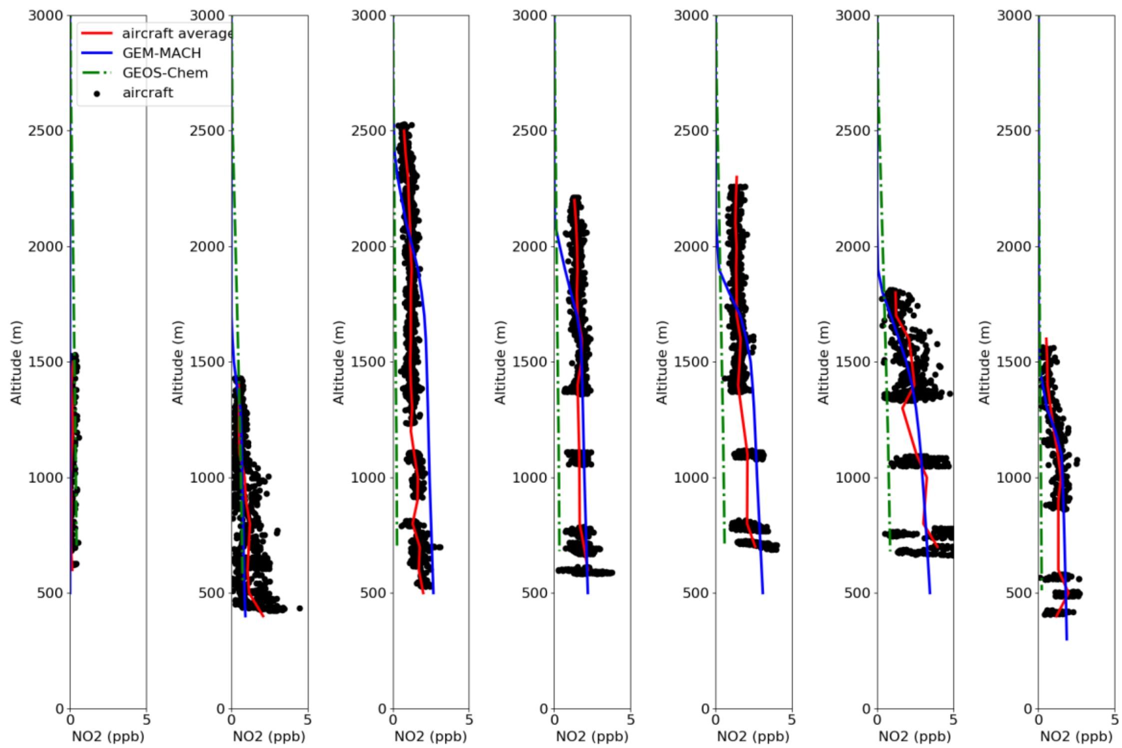

Figure S5. In-situ aircraft measurements and model output of the seven spiral flights. The full vertical extend of all seven spirals taken during the three days (April 5,9 and 13, 2018) of flights is shown.

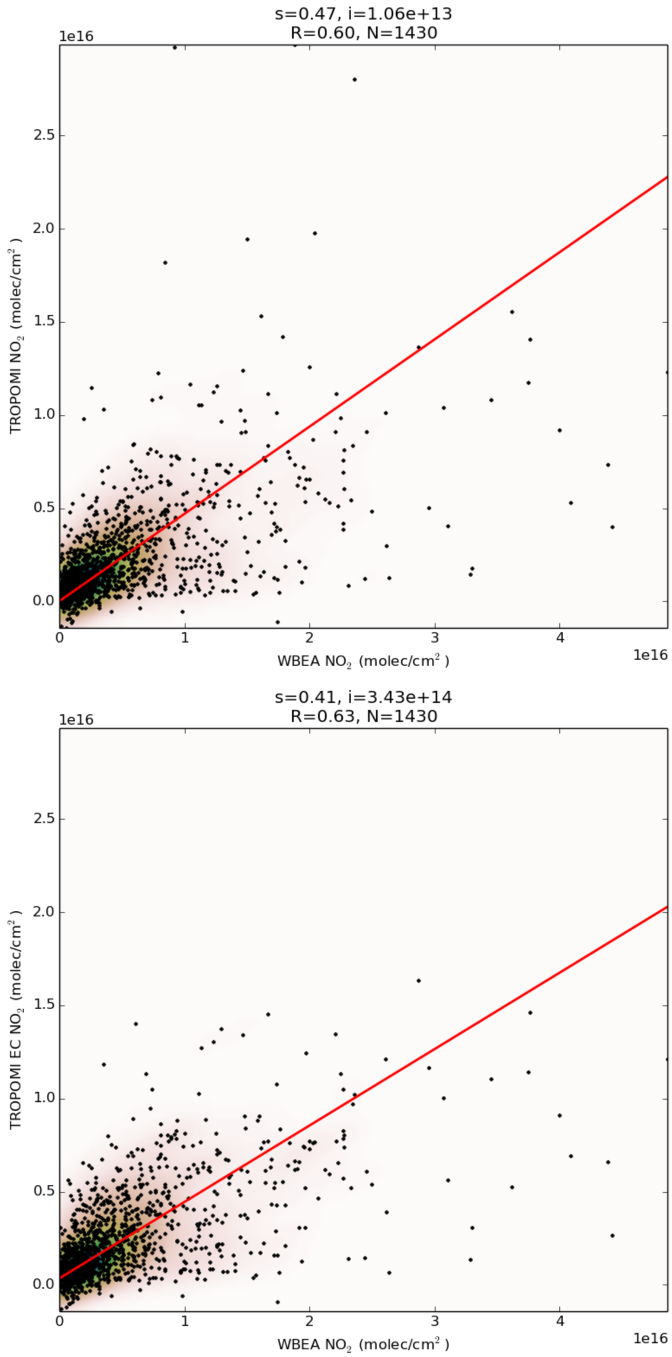

Figure S6. (a) TROPOMI tropospheric NO2 columns and (b) the alternative TROPOMI columns versus aircraft WBEA VCDs (converted from the surface concentrations to VCDs; Eq. 1). For the regression analysis, we use a geometric mean analysis with y = sx + i (s and i values are indicated in the plots, as well as the correlation coefficient, R and the number of points, N). The surface measurements are expected to have a larger spread as they only measure a single point inside the TROPOMI pixels. The correlation improves slightly with the alternative TROPOMI NO2 columns.

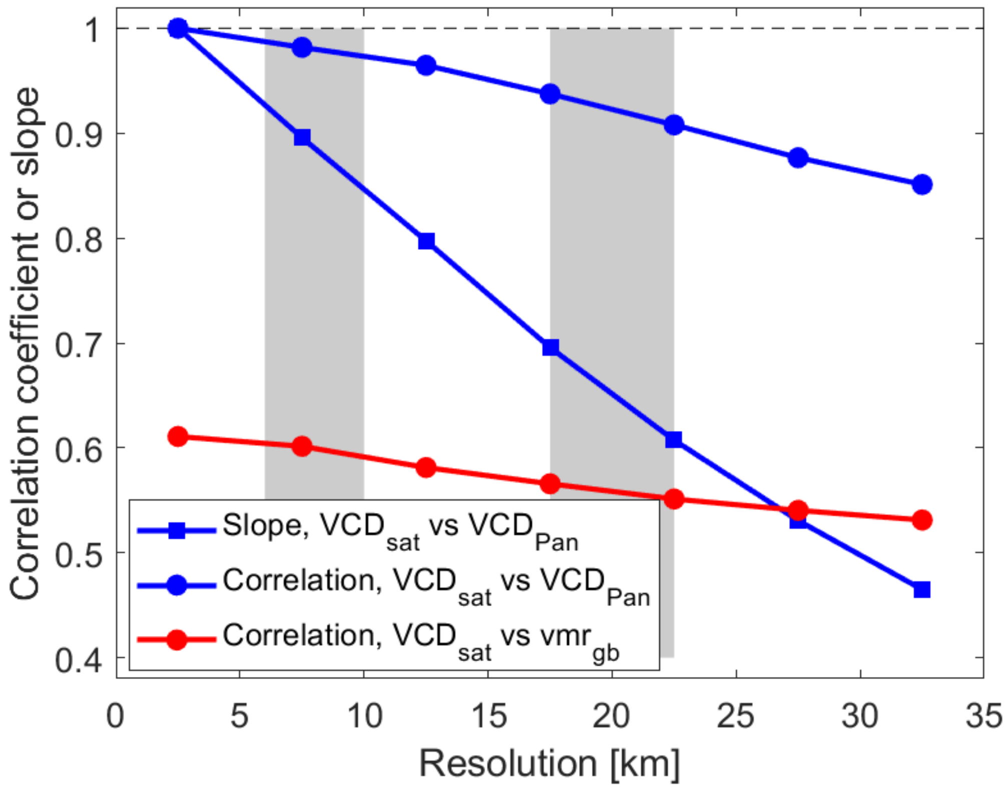

Figure S7. Effect of the resolution on the regression slope or correlation coefficient as determined using high-resolution (2.5 km) output from the GEM-MACH model. Model output spanning one year and sampled at monitoring station locations and 1-2 pm local time over the oil sands was used. The blue lines show the slope and correlation coefficient when model VCD output, smoothed to the indicated resolution as might be the case for a satellite, is compared with the original 2.5 km output for the Fort McKay station location, simulating a satellite-Pandora comparison. The red line shows the correlation coefficient considering smoothed satellite VCD and ground-based volume mixing ratio considering all WBEA stations given in Table S1. The shaded areas show the approximate (effective) resolutions of TROPOMI (left) and OMI (right).

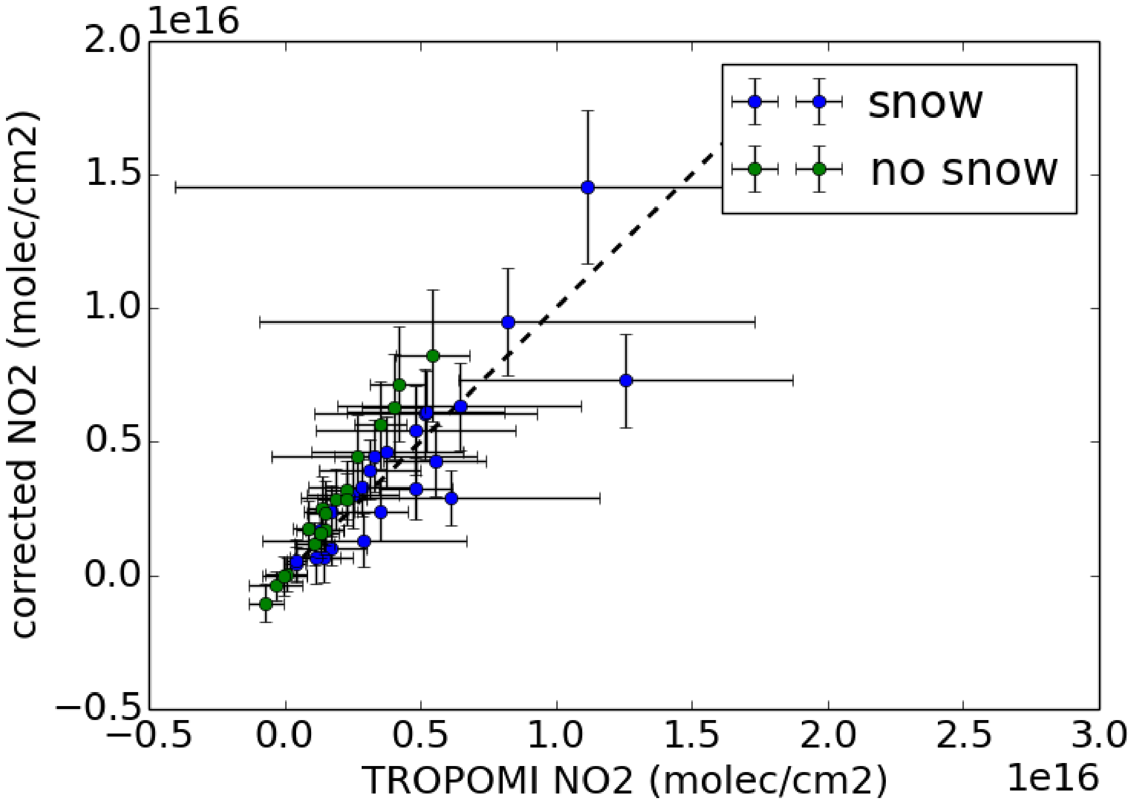

Figure S8. Comparison between the original TROPOMI columns and the corrected TROPMI columns (the same data as in Fig. 4 are used). For reference the 1:1-line is shown here as a dashed line. Overall, the corrected and the original NO_2 VCDs agree within the estimated uncertainties, however, the uncertainty of the original TROPMI columns can be as big as 100% over snow. Using higher resolution input can reduce the uncertainties significantly.

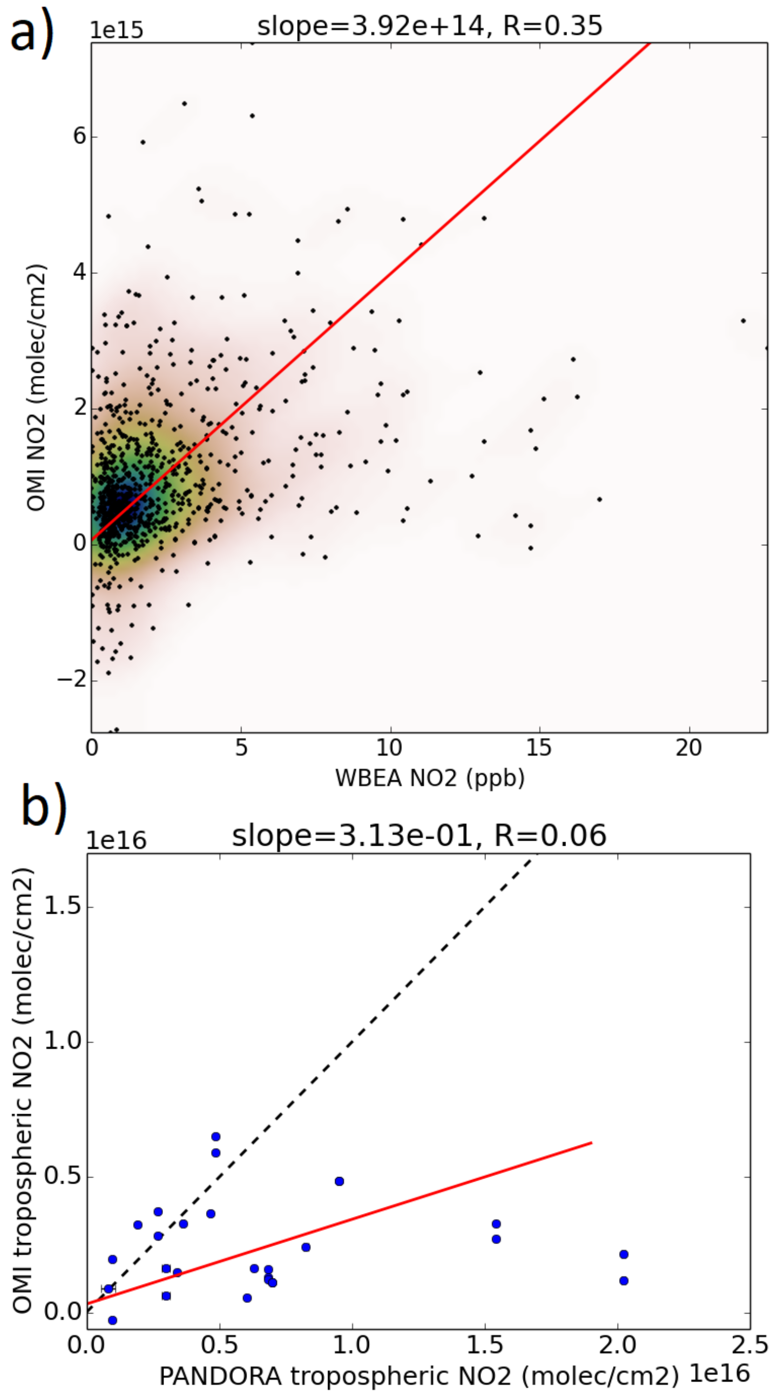

Figure S9. Comparison between the OMI tropospheric NO2 columns and the ground-based in-situ (a) and remote-sensing (b) measurements between March and May 2018. The solid and the dashed lines represent the line of best fit and the 1:1 line, respectively. The same measurements are used and the same method is applied for this comparison with OMI measurements as described in the text of the paper for TROPOMI.

Table S1. List of WBEA monitoring stations used in this study.

station	latitude	longitude	Instrument
AMS 1 Ft McKay	57.18943	-111.64058	Thermo 17C
AMS 4 Buffalo Viewpoint	56.99627	-111.5941	
AMS 6 Patricia McInnes	56.75138	-111.47669	Thermo 17C
AMS 7 Athabasca Valley	56.73339	-111.3905	Thermo 42CTL
AMS 8 Ft. Chipewyan	58.70924	-111.17499	Teledyne API T200U
AMS 13 Syncrude	57.14918	-111.64234	Thermo 42C
AMS 14 Anzac	56.4489	-111.03798	Thermo 42i
AMS 15 CNRL Horizon	57.30369	-111.73949	Teledyne API 200A
AMS 16 Albian Muskeg River	57.24909	-111.50865	Teledyne API, 200A
AMS 17 Wapasu	57.2592	-111.03858	
AMS 18 Stony Mountain	55.62141	-111.17269	Thermo 42i
AMS 19 Firebag	57.23958	-110.89799	
AMS 20 Brion MacKay River	56.77972	-112.08917	Thermo 42i
AMS 21 Conklin Community	55.63233	-111.07887	Thermo 42i
AMS 22 Janvier Community	55.90324	-110.74974	Thermo 42i
AMS 23 Fort Hills	57.3489	-111.63969	
AMS 24 Surmont	56.17799	-110.93561	