Late-Spring and Summertime Tropospheric Ozone and NO$_2$ in Western Siberia and the Russian Arctic: Regional Model Evaluation and Sensitivities.

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Abstract

15   We use a regional chemistry transport model (WRF-Chem) in conjunction with surface observations of tropospheric ozone
and Ozone Monitoring Instrument (OMI) satellite retrievals of tropospheric column NO$_2$ to evaluate processes controlling the
regional distribution of tropospheric ozone over Western Siberia for late-spring and summer in 2011. This region hosts a range
of anthropogenic and natural ozone precursor sources, and serves as a gateway for near-surface transport of Eurasian pollution
to the Arctic. However, there is a severe lack of in-situ observations to constrain tropospheric ozone sources and sinks in the

20   region. We show widespread negative bias in WRF-Chem tropospheric column NO$_2$ when compared to OMI satellite
observations from May – August, which is reduced when using ECLIPSE v5a emissions (FMB= -0.82 to -0.73) compared
with the EDGAR-HTAP-2 emissions data (FMB= -0.80 to -0.70). Despite the large negative bias, the spatial correlations
between model and observed NO$_2$ columns suggest that the spatial pattern of NO$_X$ sources in the region is well represented.
Scaling transport and energy emissions in the ECLIPSE v5a inventory by a factor of 2 reduces column NO$_2$ bias (FMB=-0.66

25   to -0.35), but with overestimates in some urban regions, and little change to a persistent underestimate in background regions.
Based on the scaled ECLIPSE v5a emissions, we assess the influence of the two dominant anthropogenic emission sectors
(transport and energy) and vegetation fires on surface NO$_X$ and ozone over Siberia and the Russian Arctic. Our results suggest
regional ozone is more sensitive to anthropogenic emissions, particularly from the transport sector, and the contribution from
fire emissions maximises in June and is largely confined to latitudes south of 60°N. Ozone dry deposition fluxes from the

30   model simulations show that the dominant ozone dry deposition sink in the region is to forest vegetation, averaging 8.0 Tg of
ozone per month, peaking at 10.3 Tg of ozone deposition during June. The impact of fires on ozone dry deposition within the

domain is small compared to anthropogenic emissions, and is negligible north of 60°N. Overall, our results suggest that surface ozone in the region is controlled by an interplay between seasonality in atmospheric transport patterns, vegetation dry deposition, and a dominance of transport and energy sector emissions.

1. Introduction

In recent decades, the high latitudes have warmed disproportionately relative to global mean temperature increases, resulting in rapid environmental changes in the Arctic region, most notably substantial loss of summer sea ice (IPCC, 2014). This disproportionate warming is termed Arctic Amplification, and results from efficient Arctic feedback processes, such as surface albedo and temperature feedbacks (Pithan and Mauritsen, 2014). Although Arctic warming has been predominantly controlled by radiative forcing from well mixed greenhouse gases, such as carbon dioxide (CO_2), warming from changes in the abundances and distributions of short-lived climate pollutants (SLCPs) such as tropospheric ozone and aerosol particles may have contributed substantially (Sand et al., 2016). Targeting such SLCPs, through short-term emission controls could have a substantial benefit in mitigating Arctic and global warming, particularly in the near-term (Shindell et al., 2012).

Tropospheric ozone (O_3) is a secondary pollutant, and an SLCP, being a greenhouse gas with an atmospheric lifetime of several weeks (Stevenson et al., 2006). Tropospheric ozone is formed through photochemical oxidation of volatile organic compounds (VOCs), in the presence of nitrogen oxides (NO_X = $NO + NO_2$) and sunlight (Crutzen et al., 1999). Enhancements in near-surface ozone degrade air quality, and are linked with premature mortality in humans (Atkinson et al., 2016; Jerrett et al., 2009; Lelieveld et al., 2015; Turner et al., 2016). Ozone is also detrimental to natural vegetation and crops (Fuhrer, 2009; Hollaway et al., 2012; Rydsaa et al., 2016), and can indirectly impact climate and hydrology through its impacts on vegetation carbon sequestration (Sitch et al., 2007) and transpiration (Arnold et al., 2018). Sources of tropospheric ozone and its precursors are poorly characterised in the Arctic region, resulting in poor understanding of sensitivity of ozone and its impacts to potential changes in Arctic atmospheric processes, and remote and local emission sources (Law et al., 2017). Local Arctic sources of ozone

precursors may increase in the future with northward migration of population, an expanding tourism industry, and increased industrial activity and shipping traffic (Arnold et al., 2016; Schmale et al., 2018).

Western Siberia is an important region in the context of high latitude tropospheric ozone concentrations, as it possesses an array of potential precursor sources. During winter and spring, the region acts as a "gateway" for poleward near-surface advection of Eurasian pollution into the Arctic (Stohl, 2007), which contributes to the well-characterised "Arctic haze" (Shaw, 1995; Quinn et al., 2008). However, a severe paucity of in-situ observations limits our understanding of sources, sinks and processing of pollution over Western Siberia, including ozone and its precursors. Current emission inventories have large uncertainties for high latitude emissions, including those from Western Siberia (Schmale et al., 2018).

The Western Siberia region is impacted by both anthropogenic and natural ozone precursor sources, many of which are poorly quantified. Anthropogenic sources in the region include those associated with large urban regions such as transport, domestic heating and power generation (Stohl et al., 2013), as well as sources specific to industrial and commercial activities in the region, such as gas flaring (Huang et al., 2014; Huang et al., 2015; Marelle et al., 2018) and shipping (Corbett et al., 2010). Moreover, future emission increases are likely, meaning a better understanding of these sources is important in the context of future Arctic SLCP budgets (Arnold et al., 2016). The Ob Valley region (approx. 45-65°N, 60-95°E) in particular is home to multiple populous cities, such as Novosibirsk (1.5 million people), Yekaterinburg (1.4 million), Novokuznetsk (550,000) and Tomsk (550,000). Emissions from these urban regions are uncertain, and poorly constrained by in-situ monitoring, except for some long-term datasets reported for Tomsk by Davydov et al., (2019). Vivchar et al., (2009) used a back-trajectory model to quantify local source regions of NO_x emissions from Siberia to the Zotino observation tower (Zotino location shown in Fig. 1). Their results suggest a significant contribution to NO_x pollution levels found in the background Siberian atmosphere originating from sources to the south of Zotino, which includes regions of intense pollution, such as the Ob Valley area.

Past studies have attempted to improve quantification of several ozone precursor emission sources in Western Siberia. Low light imaging data from the Defence Meteorological Satellite Program (DMSP) suggest that the volume of gas flared decreased between 2005 and 2008 (Elvidge et al., 2009), however a study examining tropospheric column NO_2 specifically from gas flaring locations in Western Siberia found no significant trends between 2004-2015 (Li et al., 2016). Improvements in flaring efficiency is most likely the reason for the observed decrease. High latitude residential combustion (Stohl et al., 2013) and transport emissions (Huang et al., 2015) are often underestimated, or overlooked entirely in this region. Residential combustion at the latitudes relevant to the Ob Valley (55°N - 65°N), can result in emissions all year round which relates to indoor heating and cooking, due to prolonged wintertime low outdoor temperatures, and frequent summer cold spells. The use of diesel generators to provide the energy for this heating is frequently understated, which may be used both domestically and commercially as space heaters for up to 12 hours a day (Evans et al., 2015). Attempts to better quantify Russian transport sector emissions suggest major flaws in current emissions. In particular a severe lack of regional activity data, a problem shared across all major anthropogenic sectors, leads to missing contributions from major sources. This is highlighted by Evans et al., (2015), where a detailed inventory is provided for Murmansk in northern European Russia, which is the largest city within the Arctic Circle. Murmansk is recognised as a particular region of poor emission quantification due to high levels of industrial mining, which is often overlooked (Stohl et al., 2013). A coherent evaluation of anthropogenic ozone precursor sources across the region is lacking.

In addition to fossil combustion sources, during summertime, large wild and agricultural fires emit substantial amounts of ozone precursor species (AMAP, 2015), and are the largest natural source of pollutants from within the Arctic region (Schmale et al., 2018). The intensity and locations of these fires vary annually, but the frequency of high impact Siberian fire events is increasing (Kukavskaya et al., 2016). Over the past 20 years, severe Siberian fire-events occurred in 2003 (Jeong et al., 2008), 2010 (Konovalov et al., 2011), 2012 (Antokhin et al., et) 2014 (Jung et al., 2016) and 2016 (Sitnov et al., 2017). These fires can have severe impacts on regional air quality, and lead to increases in aerosol and ozone concentrations, which can perturb the radiative budget, affecting regional climate. During a severe heat

wave in 2010, which led to severe wildfires to the east of Moscow, 11,000 nonaccidental deaths were associated with increased levels of pollutants and degradations in air quality attributed to wildfires (Shaposhnikov et al., 2014). Understanding the controls on tropospheric ozone concentrations in a region of wildfires can be further complicated, due to high levels of aerosols associated with fires (Jaffe and Wigder, 2012). In Siberia, this has been found to limit photochemical ozone production, and also act as an ozone sink in some cases (Antokhin et al., 2018).

Siberia is also characterised by extensive vegetation cover, which may act as an important dry deposition sink for pollution in the region. Previous studies have demonstrated observations of suppressed high latitude ozone concentrations in air masses that have had extensive surface contact with Siberian forests (Hirdman et al., 2010; Engvall et al., 2012). This implies a key role for Siberian vegetation as a sink for ozone pollution in the region, potentially reducing the abundance of ozone within air masses transported polewards from ozone precursor source regions. An understanding of the extent to which this ozone sink mediates anthropogenic ozone influence in high latitude Siberia requires detailed quantification.
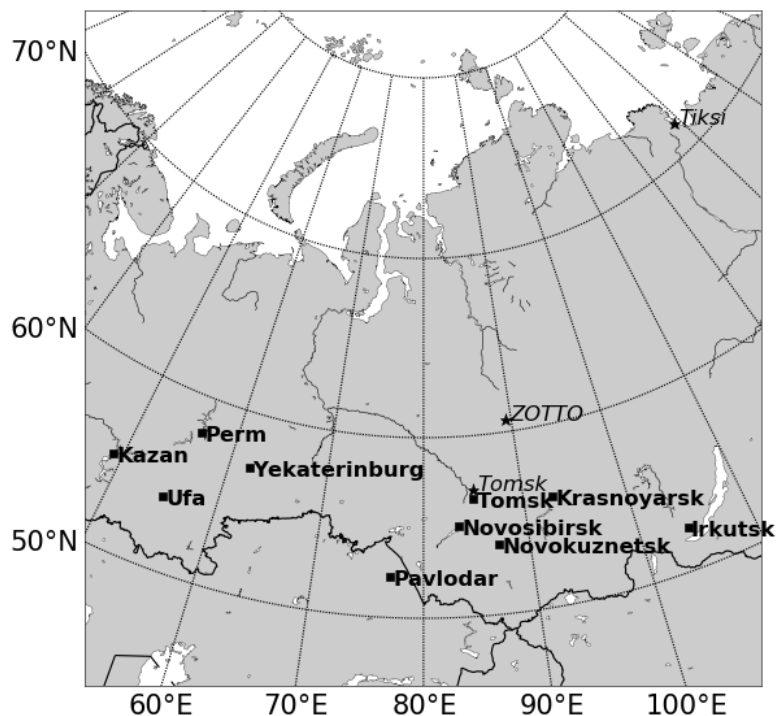
Figure 1 - Map of domain used for model simulations. Centred on Western Siberia region, major cities (squares) (population > 100,000) shown in bold. Observation sites (star symbols) are given in italics.

In this study we use satellite observations, surface measurements, and a regional air quality model to evaluate late spring and summer tropospheric NO_2 and ozone in Western Siberia. Our model domain encompasses both the major Western Siberian cities to the south and the Arctic Ocean coast to the north, whilst also enabling us to capture potential shifts in patterns of spring and summertime transport of pollutants (Fig. 1). Our overall aim is to exploit satellite NO_2 observations to better understand sources of ozone precursors in a region of sparse in-situ measurements, and to investigate major processes controlling surface ozone in this region. We evaluate the performance of two different commonly used anthropogenic emission inventories in the region, and use the model to quantify contributions to surface ozone from anthropogenic and vegetation fire precursor emissions. Finally, we use the model to estimate the contributions from different types and regions of vegetation in Western Siberia to dry deposition loss of ozone produced from anthropogenic and fire emissions from the region and ozone originating upstream. Section 2 introduces the methodology used, Section 3 presents the results, Section 4 provides a discussion and Section 5 presents the main conclusions from the study.

2. Data & Methodology

2.1 Anthropogenic Emission Inventories

We use and compare two different anthropogenic emission inventories: the EDGAR (Emissions Database for Global Atmospheric Research)-HTAP (Hemispheric Transport of Air Pollution) v2.2 inventory, and the ECLIPSE (Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants) V5a inventory. We carry out several model simulations to compare the impacts of these different emission datasets on ozone and its precursors in the Western Siberia region.

2.1.1 EDGAR-HTAP V2.2

The EDGAR-HTAP v2.2 (hereafter "EH2") (Janssens-Maenhout et al., 2015) anthropogenic emissions used are for the year 2010 and acquired in a monthly 0.1° x 0.1° gridmap format and split into anthropogenic sectors (aircraft, shipping, energy, industry, transportation, residential and agriculture). 2010 EH2 emission species include: carbon monoxide (CO), sulphur dioxide (SO_2), NO_x, NMVOC, ammonia (NH_3), particulate matter smaller than 10 µm (PM_{10}), particulate matter smaller than 2.5 µm ($PM_{2.5}$), black carbon (BC), organic carbon (OC) and methane (CH_4). The EH2 emissions are created through supplementing globally reported emissions with regional inventories, with the aim of producing an inventory for hemispheric transport of air pollution. These data are readily available online in NetCDF format (http://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=123).

2.1.2 ECLIPSE V5a

ECLIPSE v5a (hereafter "ECL") anthropogenic emissions data are created by the Greenhouse gas-Air pollution Interactions and Synergies (GAINS) model, which contains information on the sources of emissions, environmental policies and mitigation efforts and opportunities for approximately 160 countries (Stohl, et al., 2015). The emission data have been rigorously evaluated through comparisons with multiple ground-based and satellite observational data sets from Europe, Asia and the Arctic, with improvements for Arctic aerosols, when compared to previous studies (Stohl, et al., 2015). The emissions used are for the year 2010 at a resolution of 0.5° x 0.5°. Shipping emissions are available at a 1° x 1°

resolution. ECL provides emissions for SO_2, NO_x, NH_3, NMVOC, BC, OC, $PM_{2.5}$, PM_{10}, CO and CH_4. split into different anthropogenic sectors (agricultural waste burning, residential, energy, industry, transport, waste, and shipping).

2.1.3   Anthropogenic Emission Dataset Comparison for Western Siberia

Comparisons between the two anthropogenic emission inventories for NO_x show larger emissions in the ECL inventory for Western Siberia (Fig. 2). NO_x emissions within the domain are dominated by the Transport and Energy sectors, which together contribute 75% of emissions for EH2, and 82% for ECL respectively (Table 1). For both emission inventories the largest sector contribution is from transport, which accounts for 41% of total EH2 emissions and 48% of total ECL emissions. Figure 2 shows that despite larger magnitude of emissions in ECL, with the largest difference seen over the urban regions
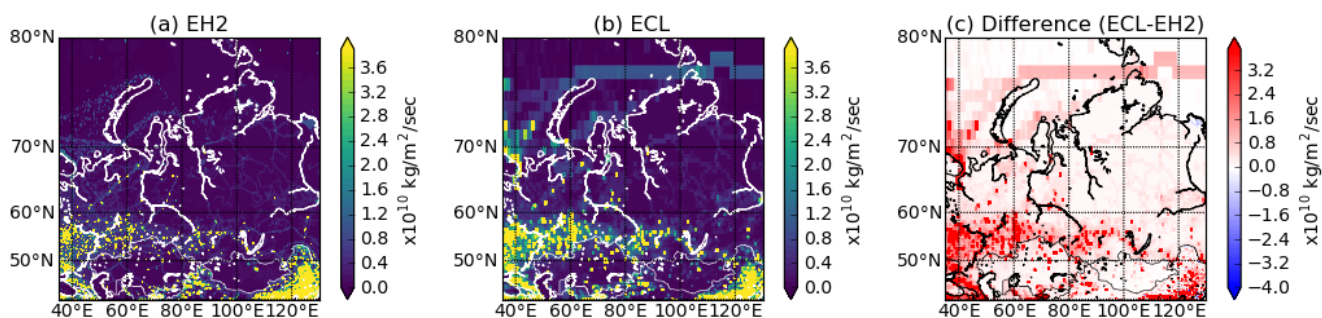
Figure 2 –Spatial distribution of anthropogenic emissions according to EDGAR HTAP v2.2 (panel (a)) and ECLIPSE v5a (panel (b)) inventories. Difference between the 2 inventories is shown in panel (c) (ECLIPSE v5a – EDGAR HTAP v2.2).

within the domain, the spatial patterns of total emissions are similar in both inventories. Differences are also seen in the shipping emissions, with large emissions north of Murmansk in the ECL inventory, which are not seen to the same extent in EH2 emissions. ECL attempts to better account for point source emissions associated with gas flaring north of 60°N, which can be seen between 60°E – 80°E (Fig. 2b).

Table 1 - Total NO$_x$ emissions (kilotons per month) for the study domain from EH2 and ECL anthropogenic emission inventories, and soil NO$_x$ contribution from GEIA. Contributions from energy and transport sectors shown for each inventory.

	EH2 Total	EH2 Energy	EH2 Transport	ECL Total	ECL Energy	ECL Transport	GEIA Soil NO$_x$
May	915.5	306.0	375.2	989.8	324.4	481.7	51.4
June	911.3	307.8	374.1	985.3	326.3	480.3	71.9
July	870.4	297.1	367.8	941.1	315.0	472.2	84.1
August	864.4	294.0	368.7	934.6	311.7	473.3	88.9
Total	**3561.6**	**1204.9**	**1485.8**	**3850.8**	**1277.4**	**1907.5**	**296.3**

2.2 Anthropogenic Soil NO$_x$ Emissions

Past studies have highlighted potential missing sources of anthropogenic soil NO$_x$ emissions in current inventories, associated with fertilized agricultural soils (Ganzeveld et al., 2010; Jaeglé et al., 2005; Visser et al., 2019). In particular it is suggested that during summer in northern mid-latitude regions, soil NO$_x$ emissions can contribute up to half those from fossil fuel combustion, which could have important impacts upon background ozone concentrations (Jaeglé et al., 2005). The missing source is attributed to large areas of fertilized agricultural soils, which are not well represented in current global or regional models. Estimates of global soil NO$_x$ emissions have been undertaken through different methodologies which include using top-down emission estimates (Vinken et al., 2014); scaling based upon multiple field measurement campaigns (Davidson and Kingerlee, 1997); and using an empirical model (Steinkamp and Lawrence, 2011; Yienger and Levy, 1995). Despite this, global soil NO$_x$ estimates vary significantly (9-27 Tg per year) (Oikawa et al., 2015). Agricultural NO$_x$ emissions are available within the EH2 inventory but missing in ECL, therefore we supplement all ECL simulations with additional anthropogenic soil NO$_x$. These are from the GEIA global soil NO$_x$ anthropogenic emissions, and distributed spatially according to the Yienger and Levy (1995) empirical model. The contributions per month from these anthropogenic soil NO$_x$ emissions to Western Siberia emissions are shown in Table 1.

2.3 Observational Data

2.3.1 Surface Sites

Tomsk observations are taken from the Fonovaya Observatory, which is an Institute of Atmospheric Optics (IAO) observational site, part of the Russian Academy of Sciences (RAS) Siberian Branch (Antonovich et al., 2018). This is located 60 km to the west of Tomsk (approx. 57°N, 85°E) in a rural, boreal location. Hourly ozone measurements are available at the surface from 2010 – 2011 (Davydov et al., 2018). These measurements are taken using an OPTEC 3.02-P chemiluminescence analyser at 10 m on an observational mast outside of the Observatory. Near real-time graphical representation of the data is available at http://lop.iao.ru/EN/.

The Zotino Tall Tower Observatory (ZOTTO) is situated in central Siberia (61°N, 89°E). The tower is 304 m in height, with 6 measurement platforms at 4, 52, 92, 158, 227 and 301m for meteorological variables, and 2 air sampling inlets positioned at 30 and 301m for ozone measurements carried out by Dasibi 1008AH-type and Thermo Electron Model 42C-TL gas analysers, respectively (Moiseenko et al., 2019). At present, human impacts on the local air quality are minimal due to the low population density of the area. These observations are therefore useful in evaluating the background atmospheric composition in the central Siberian region. In this study we use hourly ozone measurements taken from 30 m.

The Tiksi Observatory (71.36°N, 128.53°E) is located at the mouth of the Lena River, on the remote northern Russia coast. It is situated in a region far from any major sources of anthropogenic pollution, other than the town of Tiksi (5000 population) which is 5 km northeast of the observatory. This location offers an opportunity to gain observations at high latitudes in a near pristine environment. At present, the observatory is run in collaboration with NOAA (National Oceanic and Atmospheric Administration), the Tiksi Data Centre at the Arctic and Antarctic Research Institute in St Petersburg, Russia, which is responsible for the collection and distribution of the data, the Yakutian Service for Hydrometeorology and Environmental Monitoring, and the FMI (Finnish Meteorological Institute) (Asmi et al., 2016; Uttal

et al., 2016). For this study, we use hourly O_3 concentrations measured with a Thermo Scientific Model 19*i* analyser, which are available for 2011 (https://www.esrl.noaa.gov/gmd/dv/iadv/).

2.3.2   Ozone Monitoring Instrument satellite data

We make use of Dutch OMI (Ozone Monitoring Instrument) satellite data for tropospheric NO_2 (DOMINO v2.0), on-board NASA's polar orbiting Aura satellite, launched in 2004 (Boersma et al., 2011; Vinken et al., 2014). OMI retrievals of trace gases are through the Differential Optical Absorption Spectroscopy (DOAS) method, which involves using the on-board spectrometer to make UV-visible measurements. This provides tropospheric column NO_2 by first calculating the slant columns, which is the quantity of NO_2 along the whole photon path through the atmosphere to the instrument (Vinken et al., 2014). Using a tropospheric air mass factor (AMF), tropospheric vertical column density (VCD) of NO_2 can be retrieved, which is mapped onto a 0.25° x 0.25° grid. This data was provided on a daily temporal scale and has been averaged into monthly means (May-August) to provide reliable spatial coverage at high latitudes. To allow for a direct comparison of OMI with modelled column NO_2, averaging kernels are applied to daily model fields (Pope et al., 2015), to account for OMI vertical sensitivity varying through the tropospheric profile. The averaging kernel provides a relative sensitivity of the satellite instrument to the abundance of species of interest at different vertical points within the column (Herron-Thorpe et al., 2010). We apply averaging kernels to WRF-Chem that are provided as a column vector alongside the total column retrieval for NO_2 from the DOMINO product. Retrievals with geometric cloud cover greater than 20% and poor-quality data flags (where flag =-1) were removed. We compare model output and satellite observations on days where OMI data was available at the satellite overpass time (1330 local time).

25   All column comparisons presented in this work are undertaken at 0.5° x 0.5° resolution and limited to south of 65°N latitude. This latitude is chosen as a cut-off for the comparisons, since satellite retrieval uncertainty increases at higher latitudes, for solar zenith angle greater than 70°. Furthermore, ± 65° is the

latitudinal range used to map global NO_2 VCD when using the DOAS retrieval method (Bucsela et al., 2006).

2.4. Model Simulations

We use the Weather Research and Forecasting model coupled with chemistry (WRF-Chem) version 3.7.1 (Grell et al., 2005) to simulate tropospheric chemical and aerosol composition over Western Siberia. WRF-Chem is a fully coupled online model, in which atmospheric chemistry and meteorological components are fully consistent, using the same transport scheme, time step, advection, and physics

10 schemes. The model domain (Fig. 1) has a 30 km x 30 km horizontal resolution, in a 140 x 140 grid. There are 32 vertical levels, with the model top at 10 hPa, and the model uses terrain following hydrostatic pressure coordinates. Model gas phase chemistry is simulated using the Model of Ozone and Related Chemical Tracers v4 (MOZART-4; Emmons et al., 2010), whilst the model aerosol scheme is the 4-bin Model for Simulating Aerosol Interactions and Chemistry (MOSAIC; Zaveri et al., 2008), using

15 chemistry option 201, but with updates to aromatic photochemistry, biogenic hydrocarbons, and further species which are important for regional air quality (Hodzic and Jimenez, 2011; Knote et al., 2014). Biogenic emissions are calculated online using the Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2006). Biomass Burning emissions are from the Fire Inventory from NCAR (FINN) for 2011 (Wiedinmyer et al., 2010). The dry deposition scheme used in this model setup

20 is the Wesley Scheme (Wesley, 1989), and we use the modified International Geosphere-Biosphere Programme (IGBP) Moderate Resolution Imaging Spectroradiometer (MODIS) Noah land surface scheme (Ek, 2003), which has 20 land surface types. For more information on model setup please refer to Supplementary Table 1.

25 Model simulations are conducted between May and August 2011, with a spin-up period of 2 weeks preceding this. This simulation length is chosen at it represents the optimum period of time for valid OMI satellite comparisons at the latitudes of interest, and the year 2011 has good surface observation data availability within the domain. WRF-Chem has successfully been used at high latitudes previously to

investigate air quality issues (Marelle et al., 2015; Marelle et al., 2017 Raut et., 2017; Stohl, 2007; Thomas et al., 2013), with model output being compared to both flight campaigns and ground observations. Three separate sensitivity simulations are conducted, within each of which emissions of all species from one of three different emission sectors are removed: biomass burning emissions (fires_off simulation), anthropogenic transport emissions (trans_off simulation), and anthropogenic energy emissions (ene_off simulation). Model evaluation with observations is presented in Sections 3.1 and 3.2, for two control simulations using the two anthropogenic emissions inventories, and an additional simulation to test sensitivity to scaling the baseline emissions. The subsequent sensitivity simulations in Section 3.3 use the optimal inventory for the domain based on this evaluation.

3   Results

3.1. OMI – model Comparisons

For both anthropogenic emission inventories (EH2 and ECL), an overall negative bias is seen in WRF-Chem tropospheric column NO_2 when compared with OMI satellite observations (Fig. 3). The greatest negative bias is during June for both anthropogenic emission inventories (Fig. 3f & 3j). During June, July and August there is a statistically significant negative bias in the southwestern section of the domain using

10   both anthropogenic emission inventories (highlighted by hatching in Fig. S1). This significance is most prominent during June and July, particularly over urban regions in the EH2 simulation (Fig. 3f). For large parts of the domain that are located further from large anthropogenic sources, there is better agreement between the observed and modelled column NO_2 values.

15   Over urban regions with large emission sources south of 60°N, OMI tropospheric column NO_2 distributions show values exceeding 2×10^{15} molec cm^{-2} (Fig. 3a-d), with some variability across the 4-month period. Tropospheric column NO_2 biases greater than 1×10^{15} molec cm^{-2} are seen over the major cities within the north-western section of the domain, for example in Kazan (56°N, 49°E), Perm (58°N, 56°E), Yekaterinburg (57°N, 61°E), and Ufa (55°N, 56°E), whilst also showing positive biases over the

20   cities more centrally located, such as Tomsk (56°N, 85°E) and Novosibirsk (55°N, 83°E).
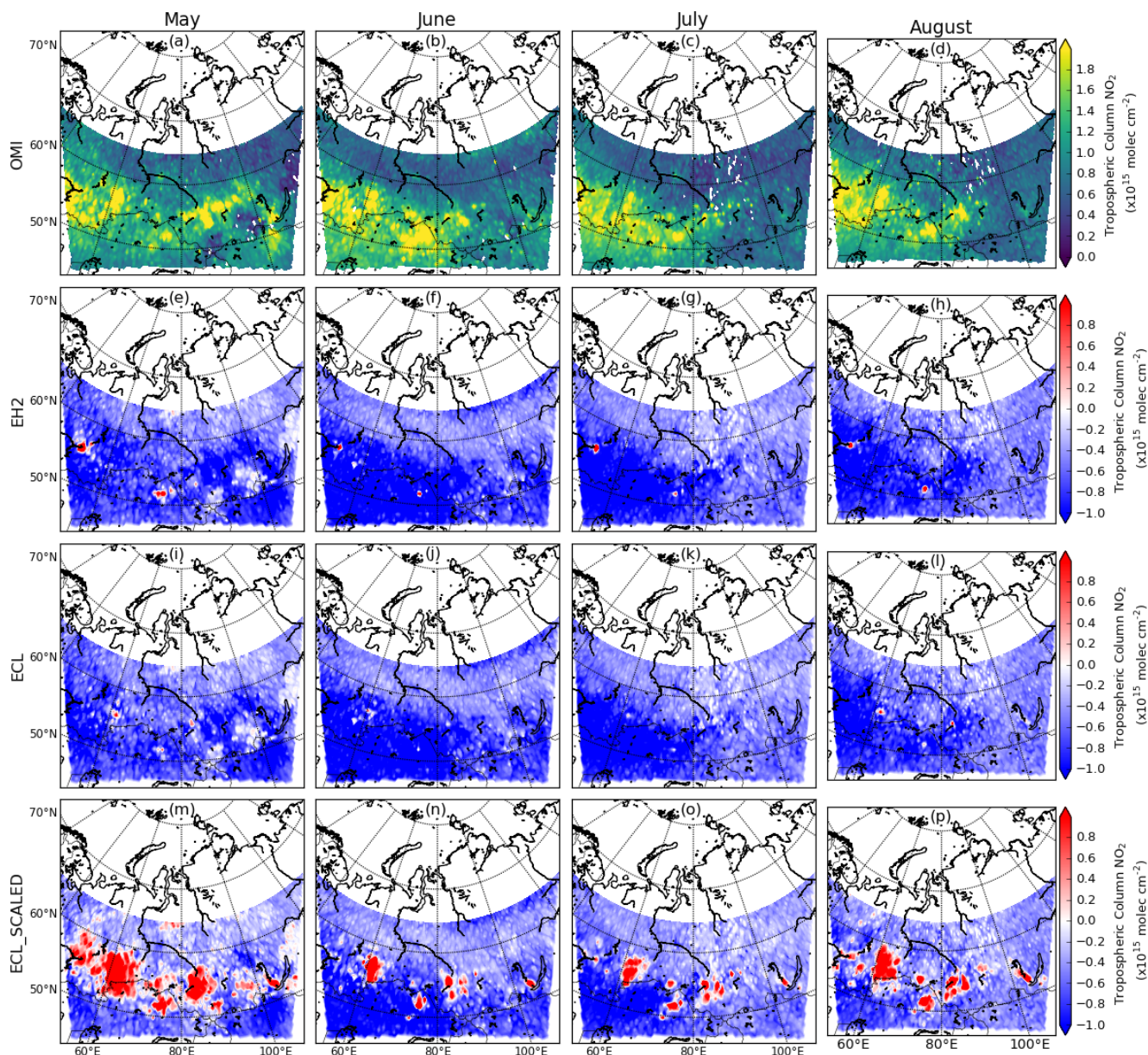
Figure 3 – Observed and model-observed tropospheric column NO$_2$. Panels a-d show mean OMI tropospheric column NO$_2$ for May-August. Panels e-h show WRF-Chem bias (model – satellite) using the EDGAR HTAP v2.2 anthropogenic emission inventory for May-August. Panels i-l show WRF-Chem bias using the ECLIPSE v5a anthropogenic emission inventory for May-August. Panels m-p show WRF-Chem bias using the ECLIPSE v5a anthropogenic emission inventory for May-August, with the energy and transport sectors scaled by a factor of 2. Results not shown <65°, due to satellite retrieval uncertainty increasing at high latitudes, associated with a large solar zenith angle.

For all four months, the WRF-Chem simulations using ECL anthropogenic emissions provide better agreement with observations for tropospheric column NO₂ over Western Siberia (Fig. 4). Despite this, negative biases persist across the whole simulated period with both anthropogenic emission inventories. In particular, negative biases are marked during June and July using either anthropogenic emission inventory, reflected in the regression slope values for ECL during June (slope = 0.2), and EH2 during July (slope = 0.1). However, better correlation coefficients are produced using ECL anthropogenic
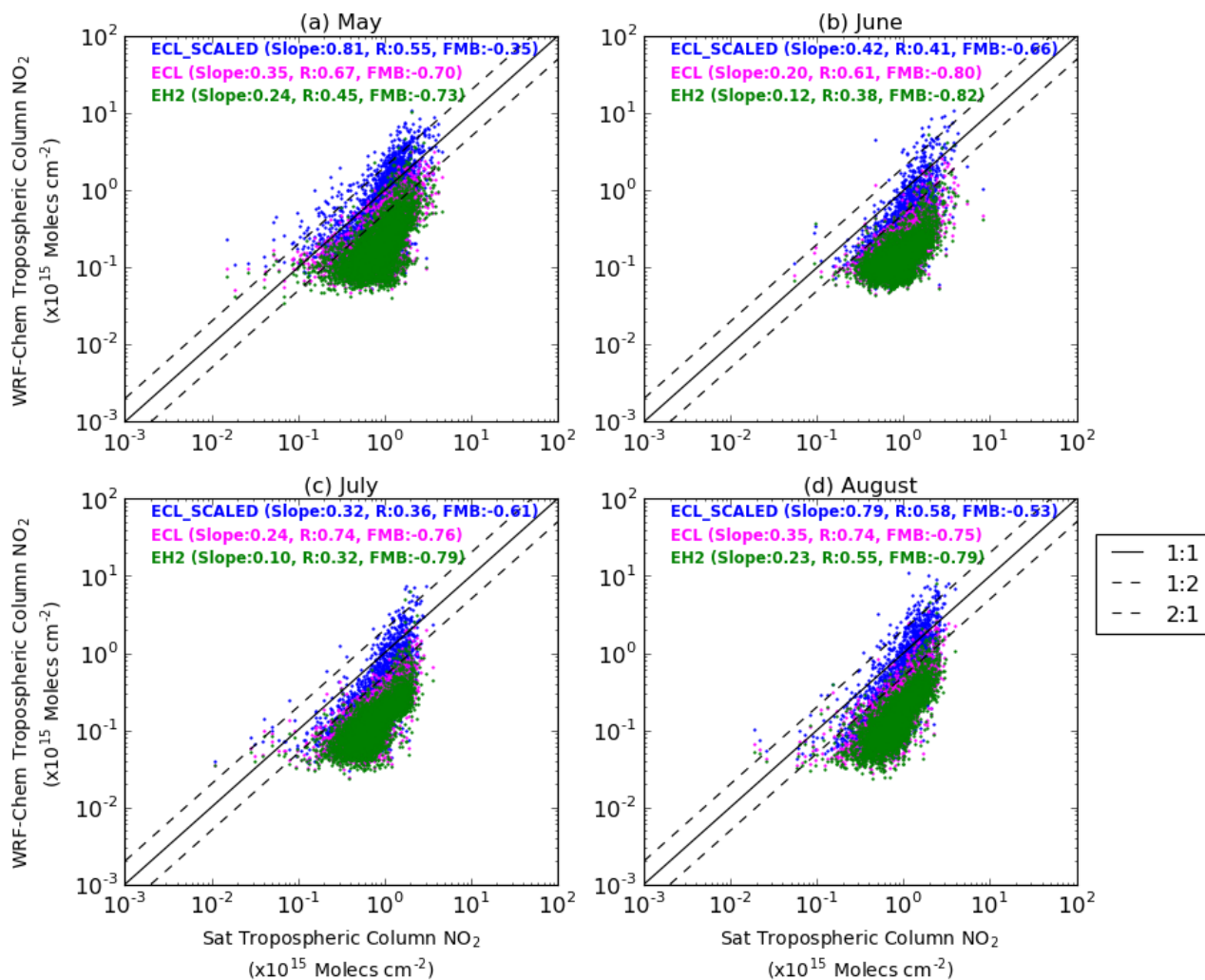
Figure 4 – WRF-Chem simulated versus OMI tropospheric column NO₂ using ECL (magenta), EH2 (blue), and ECL_SCALED anthropogenic emissions, for May (a), June (b), July (c) and August (d).All plots show total domain below 65°N. Slope, correlation coefficient (R) and fractional mean bias (FMB) are shown in each panel.

emissions during July (r = 0.74) and August (r = 0.74), and with EH2 anthropogenic emissions during August (r = 0.55), which suggests spatial patterns in NO_2 sources are well simulated, especially in ECL, but may be underestimated.

5 Table 2 – Root mean squared error (RMSE) values for monthly simulated tropospheric column NO_2 in urban and background regions within Western Siberia when compared with OMI values, for each of the model simulations using EH2, ECL and ECL_SCALED emissions.

Month	EH2 (x10^{15} Molecs cm^{-2})		ECL (x10^{15} Molecs cm^{-2})		ECL_SCALED (x10^{15} Molecs cm^{-2})	
	Urban	Background	Urban	Background	Urban	Background
May	1.41	0.34	1.26	0.39	1.25	0.22
June	1.44	0.63	1.61	0.64	0.84	0.59
July	1.33	0.47	1.14	0.48	1.09	0.45
August	1.07	0.43	1.03	0.44	1.03	0.40

10 Background values for tropospheric column NO_2 across Western Siberia show little sensitivity to changing anthropogenic emission inventories across the entire 4-month period (Table 2). We see an improved RMSE value in urban regions within the domain when using the ECL anthropogenic emissions compared to EH2 in all months but June.

15 Major cities located within western Siberia generally show smaller fractional mean biases for tropospheric column NO_2 when using the ECL anthropogenic emission inventory across the whole study period (Fig. 5). This is especially the case over Novosibirsk, Novokuznetsk and Tomsk in the centre of the domain, where the mean fractional bias is larger for almost all months when using the EH2 anthropogenic emissions, Novosibirsk in August being a notable exception. Examination of sector totals in the ECL and 20 EH2 emissions datasets shows that the transport sector is the dominant source for NO_x in ECL and EH2 over Novosibirisk and Tomsk, whilst in Novokuznetsk it is the industrial sector (EH2) and energy sector (ECL). Despite the different dominant sector over Novokuznetsk, a mean negative bias is seen across all 4 months using both inventories.

The same overall pattern is replicated at the other major cities within the western section of the domain, where we see predominantly lower fractional mean biases using the ECL anthropogenic emissions at nearly all cities (Kazan and Pavlodar being the only exceptions). In both of the anthropogenic emission inventories the cities in this western section of the domain are dominated by NO_x emissions from the transport sector, with Yekaterinburg in the EH2 inventory the only city with a different anthropogenic sector as its main NO_x source sector (Industry). The model bias could therefore suggest a potential underestimation of NO_x emissions in the transport sectors of both anthropogenic inventories over urban areas.
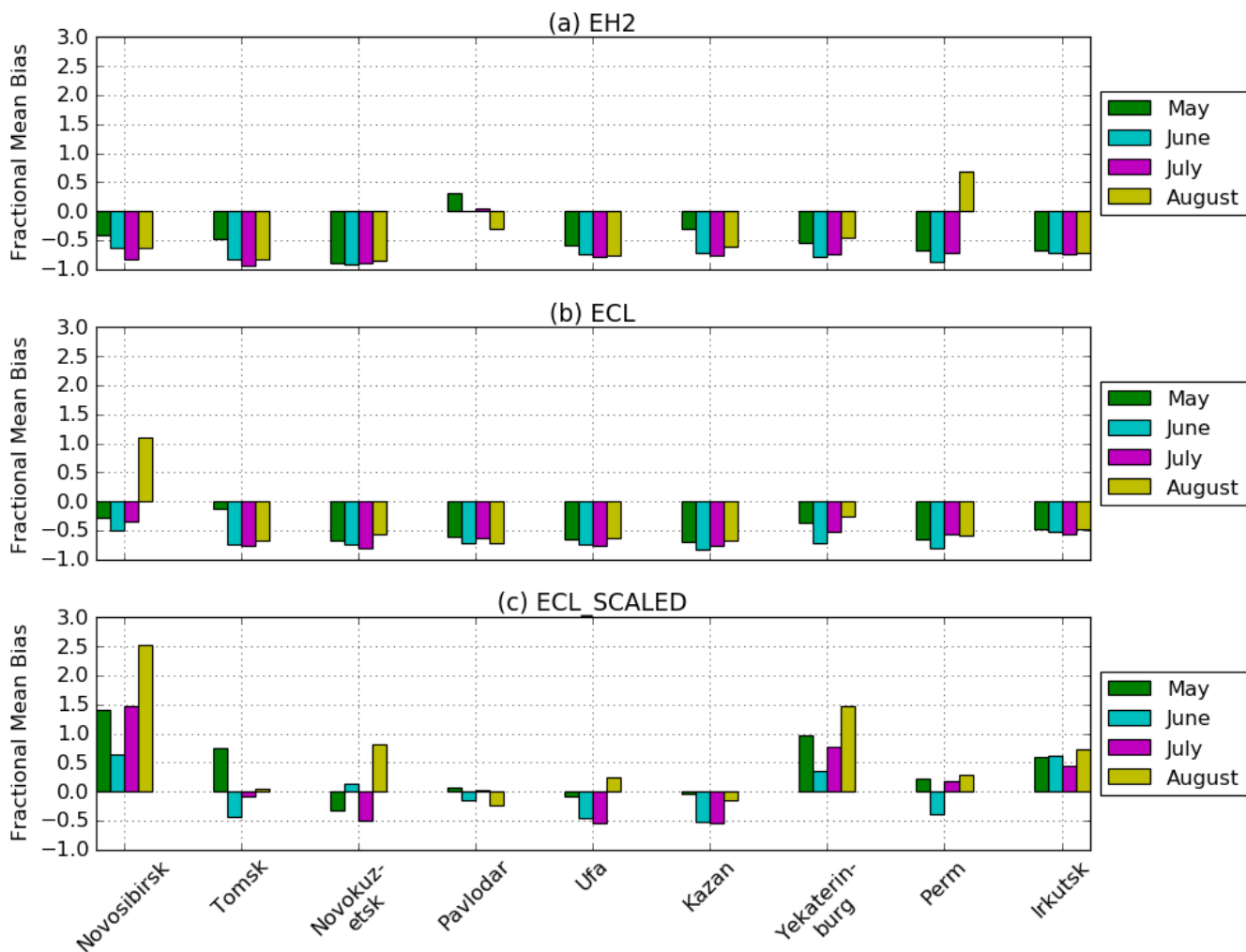
Figure 5 –Fractional mean bias of monthly simulated tropospheric column NO_2 for major cities (population >100,000) within Western Siberia when compared with OMI values. Panel (a) shows results using the EH2 anthropogenic emission inventory. Panel (b) shows results using the ECL anthropogenic emission inventory. Panel (c) shows results using the ECL_SCALED anthropogenic emission inventory.

Despite simulations using the ECL anthropogenic emissions providing better agreement with observations than those using EH2 emissions, large underestimates remain, particularly in background regions (Fig. 3). Therefore, to further investigate the sensitivity of the model NO_2 bias to anthropogenic emissions, we scale the dominant anthropogenic sectors (energy and transport) in the ECLIPSE data by a

factor of 2 (hereafter called "ECL_SCALED"). Based on analysis of the ECL emission inventory, these sectors combined account for 82% of total anthropogenic NO_x emissions within the domain (Table 1).

Across Western Siberia, we see overall improved model tropospheric column NO_2 when evaluated against OMI observations using these ECL_SCALED emissions, compared to the standard ECL simulations, with a reduction in overall FMB for each month (Fig.4). In particular we see an improvement over anthropogenic source regions where RMSE values for tropospheric column NO_2 improve for May, June and July, whilst remaining similar in August (Table 2). FMB is reduced for most major cities within the domain, however bias increases over both Novosibirsk and Yekaterinburg. These cities showed relatively small bias in the ECL simulation (Fig. 5), meaning that the simple emission scaling applied leads to an NO_2 overestimate. Despite improvement in simulated NO_2 over anthropogenic source regions when using ECL_SCALED, there is little change in the model error in background regions during June, July, August, although there is a marked reduction in background model error in May (Table 2). This suggests that background tropospheric column NO_2 is much less sensitive to anthropogenic emission increases. We note that model bias in much of this background domain is lower than the OMI observational error (Fig. S1), meaning that it is difficult to draw conclusions regarding the bias in these regions.

3.2 Surface Observation – model Comparisons

We evaluate surface ozone in WRF-Chem over the same May-August 2011 period using observations from Tomsk, ZOTTO, and Tiksi, described in Section 2.3.1 (Fig. 6). Our model simulations suggest that tropospheric ozone in these locations is relatively insensitive to the choice of anthropogenic emissions inventory. This is particularly the case at the 2 sites of Tomsk (EH2 FMB: 0.43; ECL FMB: 0.43) and Tiksi (EH2 FMB: 1.26; ECL FMB: 1.27). The greatest sensitivity to the differences between the anthropogenic emissions datasets is at ZOTTO, where the bias is improved using the EH2 emissions (EH2 FMB: 0.08; ECL FMB: 0.15).
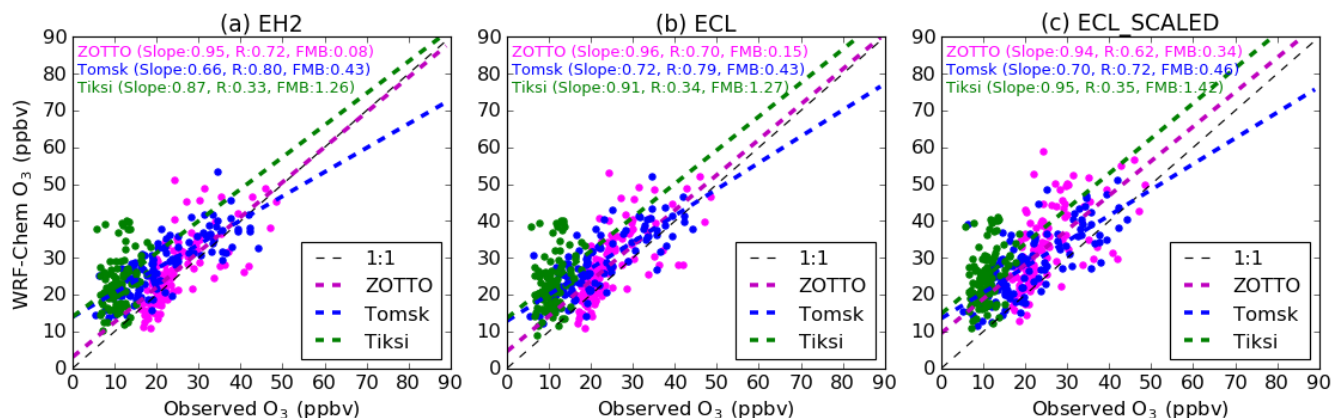
Figure 6 – Daily mean surface ozone comparisons for three ground observation sites within the study domain: ZOTTO (magenta), Tomsk (blue) and Tiksi (green) for 01/04/11 – 31/08/11. Panel (a) shows WRF-Chem surface ozone using EH2 anthropogenic emissions; panel (b) shows WRF-Chem surface ozone using ECL anthropogenic emissions; panel (c) shows WRF-Chem surface ozone using ECL_SCALED anthropogenic emissions.

There is a consistent positive bias in modelled surface ozone values at all 3 observation sites, with the largest bias seen at Tiksi for both simulations (EH2 FMB: 1.26; ECL FMB: 1.27). Both model simulations also show substantially lower fractional mean bias values for ZOTTO when compared to the other observation sites, EH2 (FMB=0.08) and ECL (FMB=0.15). Due to the largely boreal forest land surface cover at the ZOTTO observation site, this could suggest improved model performance over boreal forest regions, when compared to the Arctic tundra at Tiksi.

We note that the ECL_SCALED emissions produce an increase in ozone FMB at all sites compared to both ECL and EH2 simulations (Fig 6c). However, given the limited spatial coverage of the ozone observations, we base the optimal model choice to use in the subsequent analysis on the more extensive NO$_2$ evaluation.

3.3 Sensitivity Studies

Based upon the results in Sections 3.1 and 3.2, from this point onwards we use model simulations with the improved ECL_SCALED anthropogenic emissions to perform sensitivity simulations, since for tropospheric column NO_2, these emissions produced a smaller model bias against observations averaged across the domain. Three sensitivity simulations are used to gain a better understanding of the impacts of transport (trans_off), energy (ene_off) and fire (fires_off) emissions. Transport and energy are chosen as the two dominant anthropogenic NO_x emission sectors. From here forward, the simulation titled "control" will use the ECL_SCALED anthropogenic emissions and standard fire emissions.

3.3.1 NO_2 Source Contributions

Simulated surface NO_2 concentrations show enhancements in regions close to major anthropogenic emission sources, mainly urban regions south of 60°N, throughout the 4-month study period (Fig. 7 a-d). Hotspots in OMI-observed NO_2 north of 60°N in the central and western portions of the domain are associated with the influence of high latitude gas flaring emissions and are evident as substantial reductions in the ene_off simulation (Fig.7m-p). In the fires_off sensitivity simulation (Fig. 7 e-h) there is a small reduction in NO_2 concentrations, including background regions not in close proximity to fire source regions, across all 4 monthly periods.

As expected, given their relative source sizes, NO_2 concentrations in Western Siberia are most sensitive to anthropogenic emissions relating to transport and energy activities (Fig. 7i-p), rather than those associated with fires. Transport sector emissions are the largest source of surface NO_2 during the 4-month simulation (Fig. 7i-l). A widespread reduction of surface NO_2 is simulated in the trans_off simulation south of 60°N, both close to the urban source regions and in between cities, associated with on-road transport emissions. We also see reductions in surface NO_2 in the ene_off simulation confined to the major urban regions (Fig. 7p-t), which is likely due to emissions being point sources of high emissions associated with energy production facilities. Reductions in the abundance of peroxyacetyl nitrate (PAN) sourced from lower latitude NO_2 likely also play a role in reducing high latitude NO_2 abundance in the emission perturbation simulations. NO_2 source contributions based on perturbation simulations with

standard ECLIPSE emissions are shown in Supplementary Figure S2. Patterns and relative contributions are similar to those shown in Figure 7, and consistent with the larger magnitude transport and energy emission perturbations.
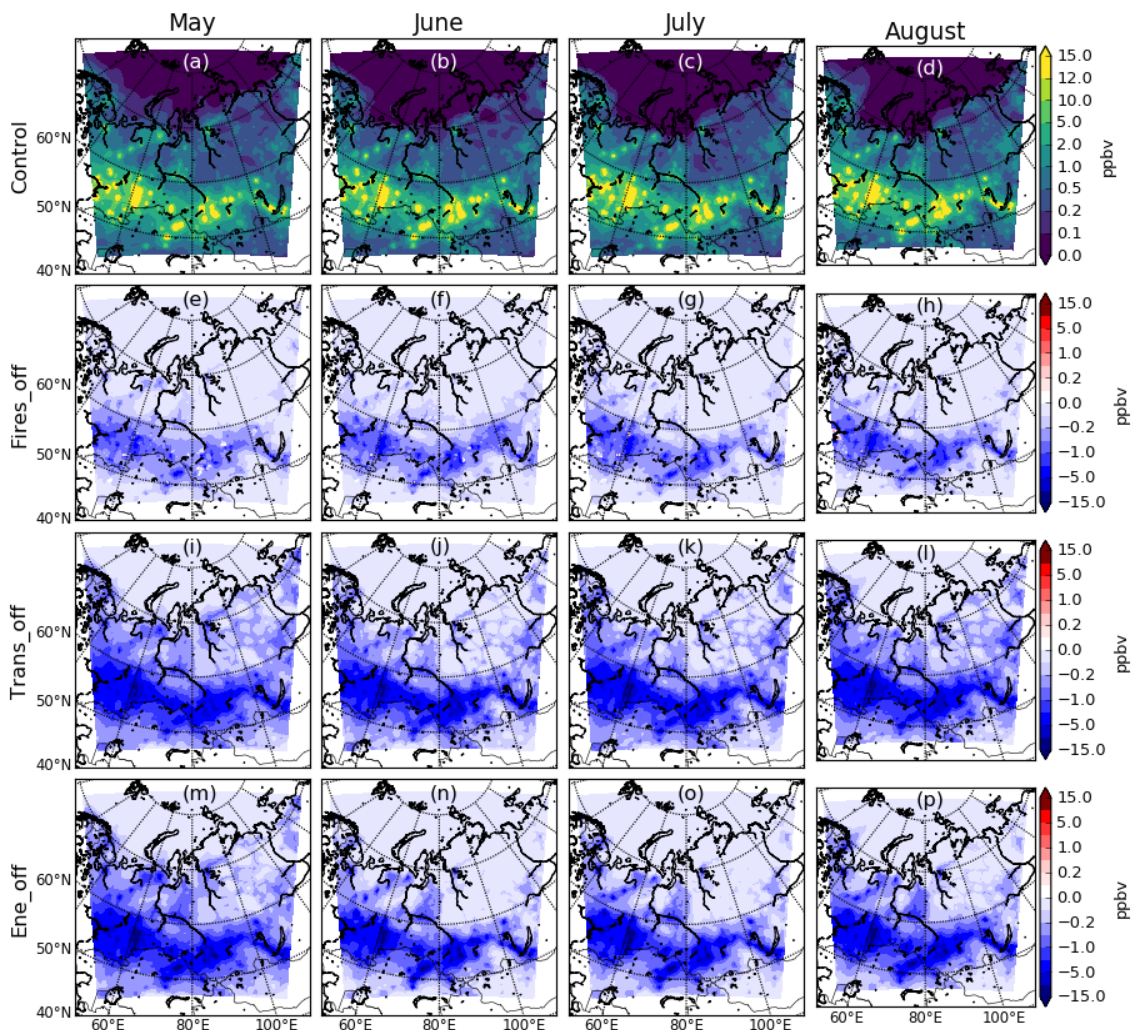
Figure 7– Simulated control and sensitivity run changes in surface NO₂ concentrations. Panels (a)–(d) show monthly means of WRF-Chem surface NO₂ for May-August using ECL_SCALED emissions. Panels (e)-(h) show monthly means of WRF-Chem Surface NO₂ with all fire emissions switched off in domain (fires_off simulation) minus control simulation for May-August. Panels (i)-(l) show monthly means of WRF-Chem Surface NO₂ with all transport emissions switched off in domain (trans_off) minus control simulation for May-August. Panels (m)-(p) show monthly means of WRF-Chem Surface NO₂ with all energy emissions switched off in domain (ene_off) minus control simulation for May-August.

3.3.2 Ozone Source Contributions

Surface ozone concentrations in Western Siberia are largest during May (mean=36.9 ppbv) (Fig. 8a), coinciding with the well-characterised springtime peak in Arctic surface ozone (Quinn et al., 2008; Stohl et al., 2007). This has been attributed to poleward import of ozone precursors, or an increase in stratospheric downwelling, which is more frequent during late spring at high latitudes (Berchet et al., 2013). In July and August (Fig. 8c-d) a surface ozone gradient from north to south begins to emerge and is strongest during August (Fig. 8d). This results from smaller simulated ozone concentrations over the Arctic, where the mean surface ozone concentration north of 60°N is 21.9 ppbv, whilst south of 60°N it is 36.0 ppbv. These low concentrations of modelled surface ozone at high latitudes occur as wind directions change to a northerly direction during summer, limiting the import of ozone precursors from lower latitudes into the Arctic. During June-August (Fig. 8b-d), largest surface ozone concentrations occur over the areas of significant precursor emissions, where monthly surface ozone averages across the summer exceed 35 ppbv. During June (Fig. 8c) concentrations exceeding 45 ppbv are simulated in the region to the east of the Ob valley, which is associated with a major fire event during this month.

Overall, surface ozone shows greatest sensitivity to anthropogenic emissions (Fig. 9). In June, July and August, transport sector emissions produce the largest ozone sensitivity, while energy emissions dominate during May. This is the case for both north and south of 60°N, where we see the maximum differences relative to the control simulation for the transport off simulation occur in August north of 60°N (-2.2 ppbv) and in July (-5.5 ppbv) south of 60°N. Across the 4-month period for the total domain, widespread reductions in surface ozone concentrations are seen in both the trans_off (Fig. 8i-l) and ene_off (Fig. 8m-p) simulations. However, within the ene_off simulations an increase in ozone is simulated over urban regions with high anthropogenic emissions, due to a decrease in the loss of ozone via $NO + O_3$ where NO_x emissions are reduced. In the fires_off simulation there is a small reduction over a large area in surface ozone south of 60°N in May (Fig. 8e), whereas in June (Fig. 8f) we see a significant reduction of surface ozone to the east of the Ob valley, due to the large biomass burning event.

Anthropogenic emissions from the energy and transport sectors sourced from within the domain contribute more to surface ozone north of 60°N than fire emissions for all months (Fig. 9), with surface ozone sourced from fires predominantly confined to south of 60°N. In the high fire month of June, we see the greatest influence of fires on surface ozone north of 60°N for the entire study period, but the difference compared to the control simulation is less than 1 ppbv. During May north of 60°N large contributions to surface ozone from energy emissions are seen, likely due to emissions associated with high latitude oil and gas extraction within the domain. This contribution is enhanced due to poleward movement of air which occurs during late springtime (Stohl, 2007) (Fig. S4). There is a shift in wind direction north of 60°N during May/June to a more northerly flow bringing in cleaner Arctic air, which leads to efficient southward export of the energy sourced ozone at high latitudes, evident as an increase in surface ozone energy sector sensitivity south of 60°N from May to June (Fig. 9).
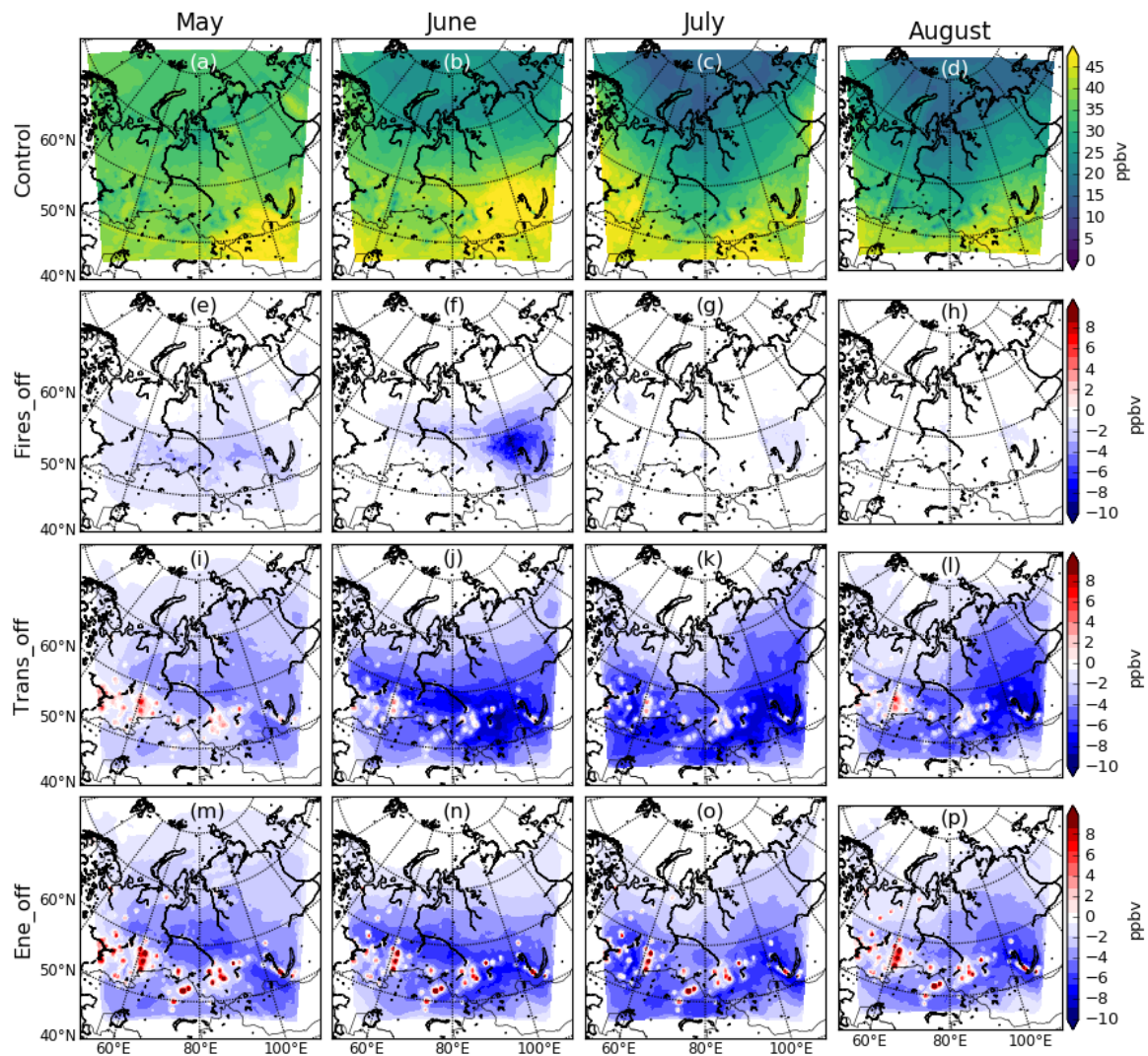
Figure 8 – As Figure 7, but for changes in surface ozone concentrations.
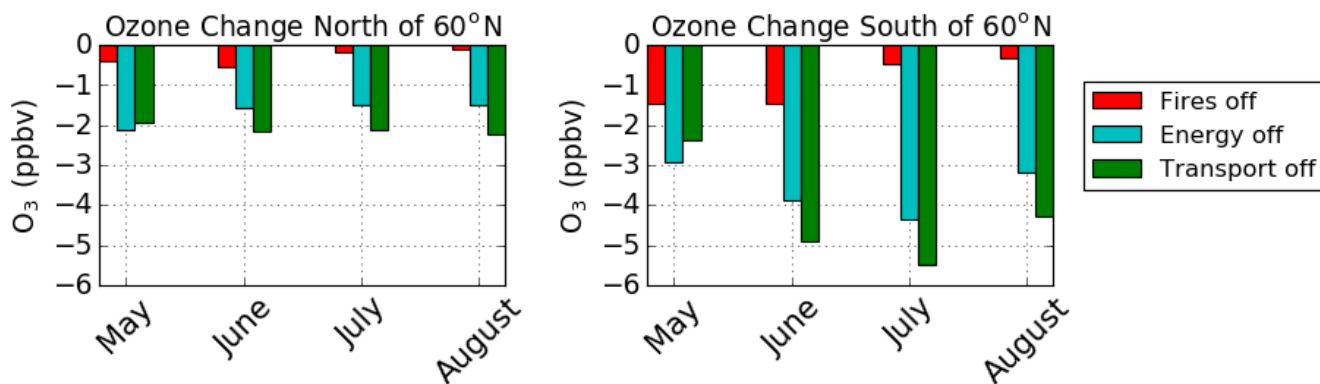
Figure 9 – Surface ozone change relative to control simulation for the section of the domain north of 60°N (left panel) and south of 60°N (right panel) for the 3 sensitivity simulations, fires_off (red), ene_off (blue) and trans_off (green).

Ozone source contributions based on perturbation simulations with standard ECLIPSE emissions are shown in Supplementary Figure S3. Spatial patterns in ozone changes are consistent with those using the ECL_SCALED emissions (Fig. 8), with lower relative magnitude contributions from energy and transport sector emissions in the south of the domain, consistent with smaller NOx perturbations. However, the relative importances of the different emission source contributions to ozone are consistent between the two sets of simulations. In regions remote from terrestrial transport and energy sources, in the north-east of the model domain and over the Arctic ozone, ozone changes are more similar in magnitude between the ECL and ECL_SCALED perturbations.

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3.4 Ozone Dry Deposition

To investigate the impact of vegetation as a surface sink of ozone in Western Siberia, we analyse ozone dry deposition fluxes from the WRF-Chem simulations (Fig. 10). These fluxes are partitioned across each of the 20 land surface types from the IGBP MODIS Noah land surface scheme used in the model. We group similar land surface types together to provide total fluxes over 8 broad land cover categories (Fig. 11). Maximum ozone dry deposition to the surface occurs during the summer months of June, July & August (Fig. 10b-d), which coincides with the summer peak in photosynthesis in vegetation (Karlsson et al., 2007; Stjernberg et al., 2012). Dry deposition fluxes are lower during May (Fig. 10a), which is coincident with the period of highest concentrations for modelled surface ozone at high latitudes (Fig. 8a).

Ozone dry deposition flux is most sensitive to anthropogenic ozone precursor emissions, especially during June, July and August (Fig 10). The reduction in dry deposition flux in the anthropogenic perturbation simulations is greater south of 60°N during this period but extends north of 60°N in July and August, for both the trans_off and ene_off simulations. This is despite relatively low concentrations of surface ozone at high latitudes during these months (Fig. 8c-d). This is likely due to enhanced photosynthesis activity and stomatal conductance during the summertime, leading to enhanced ozone deposition flux.
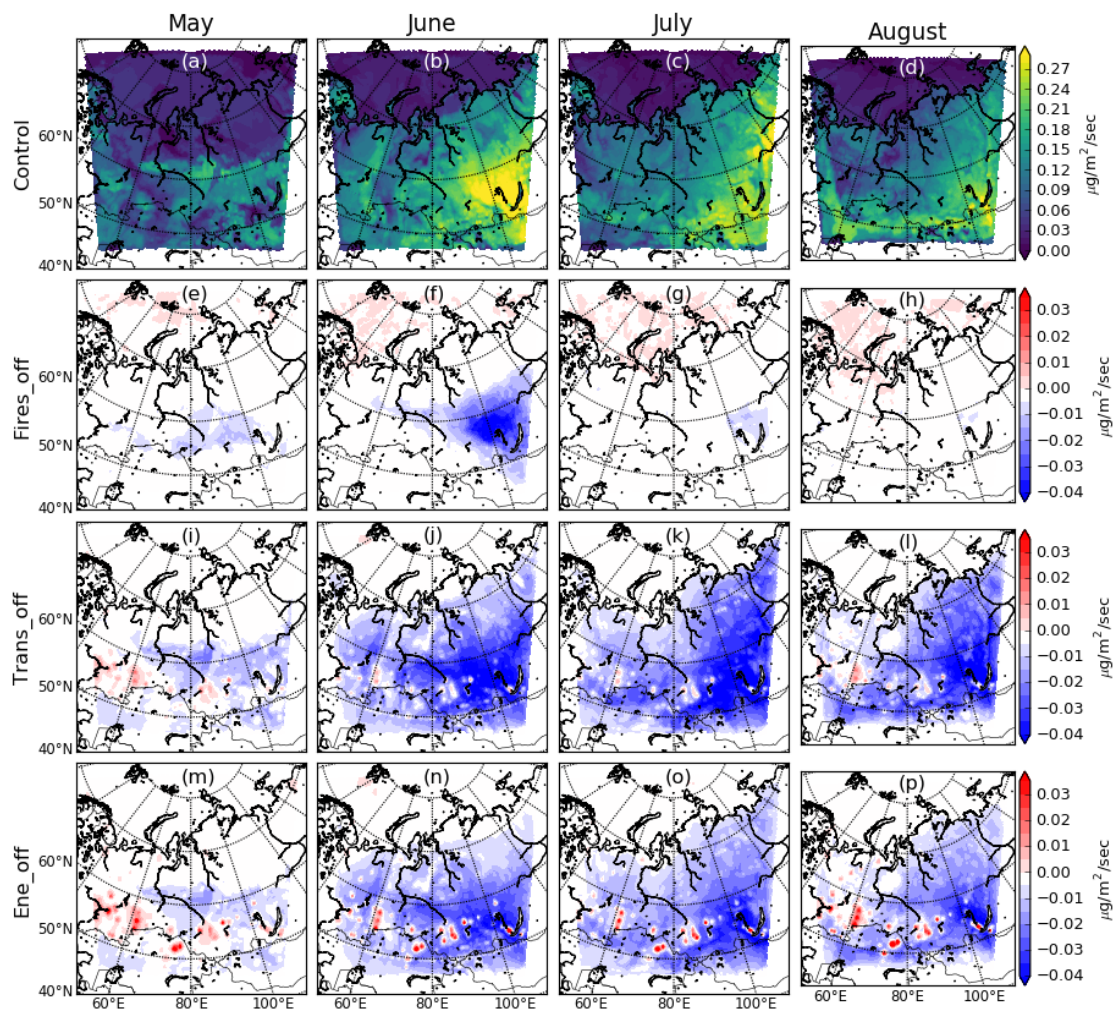
Figure 10 – As Figure 7, but for model simulated ozone dry deposition flux.
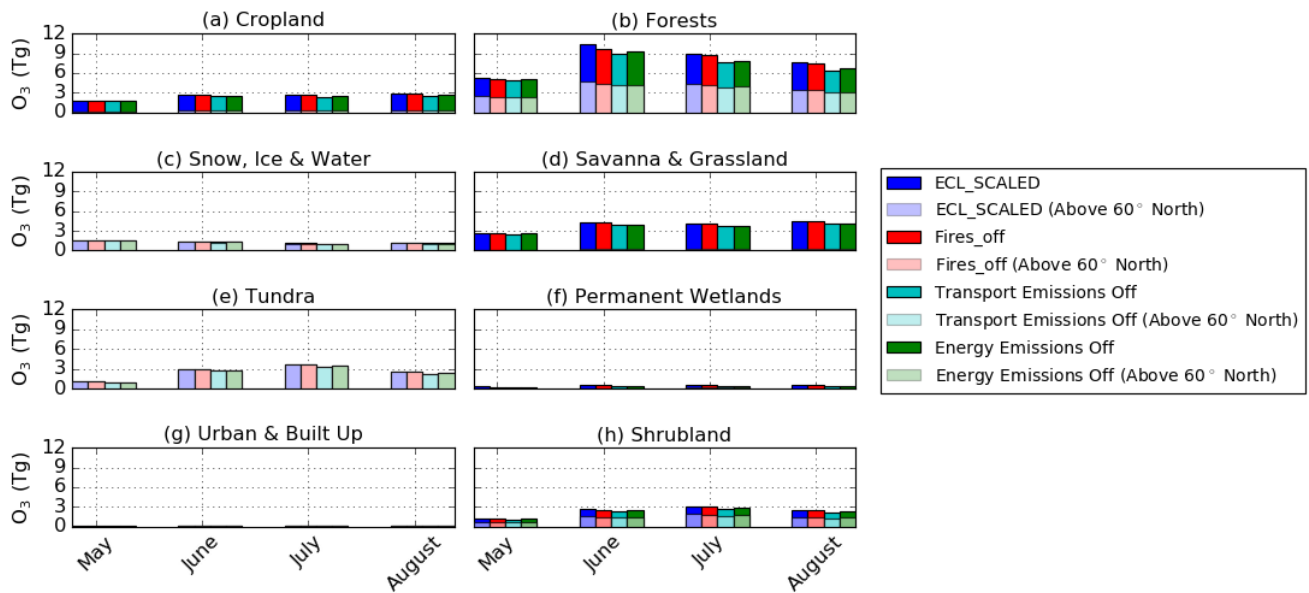
Figure 11 – Quantity of ozone deposited to modified IGBP MODIS NOAH land surface cover categories per month for total domain (solid bars) and for the section of the domain north of 60°N (pale bars).

The largest deposition sink for ozone in the model domain is to forest vegetation, averaging 8.0 Tg of ozone deposition per month in the control simulation, peaking at 10.3 Tg of ozone deposition during June (Fig. 11b). Forest covers 29% of the domain, spanning large areas both north and south 60°N. For the total domain, "cropland & vegetation" and "savanna & grassland" surface types account for an average of 2.5 and 3.9 Tg/month of ozone loss, respectively. North of 60°N, forest and tundra are the dominant sinks, which account for 65% of dry deposition flux, and 77% of the terrestrial surface cover at these latitudes.

Ozone deposition flux responds most in the trans_off and ene_off simulations, in particular during July and August. Deposited ozone reduces by 12% over forests when anthropogenic energy emissions are removed during August, and by 16% when anthropogenic transport emissions are removed during August. The impact of fires on ozone dry deposition within the domain is small compared to anthropogenic emissions, and is negligible north of 60°N. The greatest impact of the fires_off simulation

on ozone deposition occurs during May and June, with the largest percentage change for forest land cover (May: 4%, June: 6%).

Ozone deposition fluxes and their contributions based on perturbation simulations with standard ECLIPSE emissions are shown in Supplementary Figures S4 and S5. Spatial patterns and relative contributions are consistent with those shown in Figure 10, with increased contributions from energy and transport emissions resulting from the larger surface ozone contributions simulated with ECL_SCALED emissions. Small increases in forest ozone uptake using the ECL_SCALED emissions are evident in Jun-Aug. The relative importance of different land cover categories for ozone deposition loss in the domain across different months are consistent between the ECL_SCALED and ECL simulations.

Discussion

There are limited studies comparing WRF-Chem and OMI tropospheric column NO_2, especially at high latitudes, but this technique has been shown in previous studies to be an effective regional model evaluation tool (Han et al., 2011; Herron-Thorpe et al., 2010; Pope et al., 2015). Our results are consistent with previous work using OMI tropospheric NO_2 columns to evaluate an ensemble of regional models at similar latitudes over Europe (Huijnen et al., 2010). This showed negative model biases for tropospheric column NO_2, which were greatest in magnitude during summertime in background regions, with ensemble mean column NO_2 values up to 50% lower than OMI. Furthermore, it was shown that greatest spread between models occurred during the summer, with model underestimation ranging from 40-60% depending on the region. A positive bias in the DOMINO v1 product has been identified in previous studies of up to 40% in summer (Hains et al., 2010; Huijnen et al., 2010; Lamsal et al., 2010; Zhou et al., 2009), attributed to errors in the a priori NO_2 profile, air mass factors and albedo. These errors were improved in the DOMINO v2 product (Boersma et al., 2011), which we use here. This improved product is shown to lower summertime satellite positive biases of tropospheric column NO_2 relative to retrievals using the previous version of the DOMINO product, and we therefore expect these retrieval issues to play less of a role in explaining our negative model bias during summer. We see widespread negative bias in

WRF-Chem tropospheric column NO_2 when compared to the satellite measurements, especially during June (ECL FMB = -0.82; ECL_SCALED FMB = -0.66) across the background regions of western Siberia, despite improvements in the DOMINO v2.0 retrieval algorithm. These biases could also result from an underestimation of emissions and/or model deficiencies in NO_x chemistry, leading to underestimation of the NO_2 lifetime. Kanaya et al., (2014) compared 2007 - 2012 OMI tropospheric column NO_2 retrievals using the DOMINO v2 product with Multi-Axis Differential optical absorption spectroscopy (MAX-DOAS) observations from multiple sites in Asia and one in Russia. The Russian site was located in Zvenigorod (55°N, 37°E) approximately 50 km to the west of Moscow, where they found very good agreement between ground observations and satellite observations of tropospheric column NO_2, especially during the summer period of 2011 and 2012. Although limited in spatial scope, this comparison lends some limited confidence to the reliability of the OMI observations for this region during summer and may further support our model-observation differences being a result of poor representation of NO_2 sources or sinks in the model. However, it is important to note that it is difficult to completely rule out errors in the DOMINO v2 retrieval, since it has not been extensively evaluated for this region. Despite an overall reduction in NO_2 bias when scaling the ECLIPSE transport and energy sector emissions by a factor of 2, simulated negative NO_2 bias largely persists in background regions.

We include anthropogenic soil NO_x emissions in our model, which have been shown in previous studies to be a potentially overlooked source of NO_x (Oikawa et al., 2015; Visser et al., 2019). Visser et al., (2019) highlighted potential underestimates in anthropogenic soil NO_x emissions from the MEGAN emissions model, which resulted in negative model biases against surface observations of NO_2 across eastern Europe. Implementation of satellite-constrained surface NO_x emissions inferred from OMI tropospheric column NO_2, subsequently improved low model bias in their analysis. In our study we supplement our ECL simulations with anthropogenic soil NO_x emissions from the GEIA emission inventory. However, we find little impact from including these emissions on our model bias.

Cities within the domain demonstrate varying model tropospheric column NO_2 biases, with generally improved bias when using scaled surface NO_2 emissions. Other studies have shown that despite significant model underestimations of background tropospheric column NO_2 when compared to satellite observations, model performance is better over cities (Huijnen et al., 2010). NO_2 underestimation persists in the model over the majority of major urban regions in the domain, particularly outside of spring. Our results show that for all cities in the Ob valley where the dominant anthropogenic NO_x sector is transport, an underestimation is simulated for almost every month, when using both EH2 and ECL standard anthropogenic emission inventories. Evans et al., (2017) suggest that the transport sector has grown dramatically between 2000-2013 in Russia, with a doubling of passenger vehicles, and a 40% increase in truck ownership. This rise of on-road vehicles may not be well represented within Western Siberian transport emissions in ECL and EH2, as global inventories often do not have access to up-to-date country wide data (Kholod et al., 2016). Our simulations in which transport and energy emissions are scaled by a factor of 2 show improved comparison with OMI NO_2 over urban regions in our domain, although there are overestimates in some regions.

Simulated tropospheric column NO_2 in the region is sensitive to the anthropogenic emission inventory used, with the ECL inventory providing an improved NO_2 simulation when compared to EH2 and OMI. ECL has been extensively used in previous high latitude, regional modelling studies (Marelle et al., 2018; Sand et al., 2015; Stohl et al., 2013). Our results support the view that the ECL dataset is more suitable over the Western Siberia region compared with EH2. The ECL anthropogenic emission inventory attempts to add detail in the Arctic compared to EH2, accounting for better quantification of direct and associated emissions from gas flaring, and also a better understanding of emission seasonality (Stohl et al., 2015). Despite this we still see a significant widespread low bias over the region, especially from May-August. Good spatial correlation (R=0.61-0.74) between model and OMI observations despite the low bias during this period further supports the possibility of an underestimation in sources. We find that scaling anthropogenic transport and energy sector emissions by a factor of 2 leads to a reduction in average RMSE model error over urban regions in our domain from 1.26 to 1.05×10^{15} molec cm^{-2}. Huang et al., (2014) discuss potential unreliable representation of Russian anthropogenic emissions within global

inventories due to difficulties in accurate quantification of local emission factors, and incorrect locations of point sources.

The sensitivity of modelled surface ozone concentrations to the differences in the two anthropogenic emission inventories is small. This may be due to 2 of the ground observation sites being located far from precursor source regions (Tiksi & ZOTTO). In Fig. 6a & b we see a positive bias in modelled surface ozone at all sites for both anthropogenic emission inventories. Pankratova et al., (2011) suggest that during the night-time in Siberia ozone destruction can occur under intense temperature inversions through surface deposition to snowless surfaces. However, WRF-Chem does not perform well when recreating high latitude temperature inversions with strongly stable stratification periods, which are often of high importance for high latitude surface air quality (Mölders et al., 2011; Schmale et al., 2018).

At Tiksi we see a particularly marked positive bias in modelled surface ozone across the study period, which could be associated with missing halogen chemistry at high latitudes during the early part of our simulation. During ice melt the release of bromine can lead to ozone depletion events, causing ozone concentrations to go from background concentrations (~30ppbv) to concentrations lower than 5ppbv within days (Cao et al., 2016; Zhao et al., 2016). Therefore, gaining a better understanding of bromine behaviour at high latitudes is important due to the impact it can potentially have on near surface ozone concentrations. The impact of such ozone depletion on continental surface ozone across Western Siberia may be limited however, due to predominantly southerly winds in the north of the domain over the Siberian coast, during spring. A summertime low ozone bias at Tiksi may also be indicative of an underestimation in the ozone dry deposition sink, which has been shown to be a key ozone loss process for high latitude Siberia (Stjernberg et al., 2012).

The dominant ozone dry deposition sink within the domain is to forest, with approximately half of this deposition occurring north of 60°N to Arctic forest vegetation. This agrees well with the findings of Stjernberg et al., (2012) who suggest that the Siberian forest is an important ozone surface sink through dry deposition. Our results show that summer (JJA) is the most active time for surface ozone deposition,

correlating with peak photosynthetic activity and longer periods of stomatal opening, leading to more stomatal gas exchange. Stjernberg et al., (2012) also suggest that both tundra and wetlands are significant surface sinks for ozone. Our findings support the importance of tundra, which is the second largest sink above 60°N behind forest land cover type. We find wetlands to have a small contribution to ozone deposition in our domain. However, we note that our domain is different and substantially smaller than the region considered by the Stjernberg et al., (2012) study.

4   Conclusions

We have used in-situ observations for surface ozone evaluation and OMI satellite observations of tropospheric column NO_2 for large spatial scale evaluation of ozone precursor distributions in the regional chemistry model WRF-Chem over Western Siberian during late-spring and summer. Gaining a better understanding of controls on tropospheric ozone concentrations in western Siberia is important due to the role it plays as a direct pathway to the Arctic. The region provides substantial surface sinks via efficient dry deposition to vegetation, and important sources for polluted air travelling polewards to the Arctic. We attempt to better quantify major sinks and sources of ozone and its precursors within this key region for high latitude and Arctic atmospheric composition, which is vastly understudied with limited in-situ observations.

WRF-Chem shows an underestimation of tropospheric column NO_2 when compared with OMI, despite the use of a more recent OMI retrieval product (DOMINO v2), which has reduced a previously characterised high bias in OMI NO_2 columns in earlier product versions (Boersma et al., 2011). We suggest that the low model bias could be a result of lacking or underestimated emissions within the region in current emissions datasets, or due to model errors in the NO_2 atmospheric lifetime. Our results suggest that from May – August the simulated spatial pattern in NO_2 produced by the ECL anthropogenic emissions is consistent with observed NO_2 from OMI (R=0.61-0.74), but a persistent low bias continues throughout. Both EH2 (monthly FMB= -0.82 to -0.73) and ECL (monthly FMB= -0.80 to -0.70) produce simulated atmospheric distributions that underestimate the magnitude of satellite observed NO_2 during this period. Scaling transport and energy emissions in the model by a factor of 2 results in an overall improvement in the underestimation of OMI column NO_2 (monthly FMB=-0.66 to -0.35), but with overestimates in some urban regions, and little change to a persistent underestimate in background regions. Deficiencies in model tropospheric NO_y chemistry have been identified in previous studies as a contributor to bias in the simulated NO_x lifetime (Huijnen et al., 2010). These include removal through wet and dry deposition, and NO_x too readily converted to reservoir species such as nitric acid and peroxyacetyl nitrate (PAN). Our model biases during summer could be an indication of errors in the conversion of NO_2 to nitric acid, when OH concentrations are enhanced and the NO_2+OH reaction is

more important. Future work is needed to better understand drivers of the model NO_2 bias relative to OMI.

Our results suggest that surface ozone in the region studied is influenced by an interplay between seasonality in atmospheric transport patterns, vegetation dry deposition uptake and photochemistry. We find that transport and energy sector emissions have a more significant impact on surface ozone compared to fire emissions during our study period, with the relative importance of fire influence increasing south of 60°N. .

Siberian forests act as an important surface sink for ozone, especially during June, July and August when ozone surface fluxes are largest, and account for 37% of all ozone deposition in this period (Fig. 10 & 11). With future northward migration of the treeline at high latitudes, understanding how this can act as a sink for ozone in the future is important, as this could go towards helping alleviate the high latitude tropospheric ozone burden.

Code and data availability. The code and data used in this study is available from the authors upon reasonable request.

5 *Supplement.*

Author Contributions. Conceptualization of research by TT and SRA, with support from DVS. TT
10 performed model simulations with support from LC. TT performed model evaluation and data analysis with support from SRA and RJP. CK provided WRFotron modelling scripts. TT wrote the manuscript. All authors provided comments on manuscript.

15 *Competing Interests.* The authors declare that they have no conflict of interests.

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