Electronic Supplementary Material (ESI) for Environmental Science: Advances.

Table A1 : List of stations used for PSCF calculations.

Station	NAPS ID	Latitude	Longitude	Pollutants
Auclair	052801	47.72	-68.71	$PM_{2.5}$, O_3
Saguenay - UQAC	050504	48.42	-71.05	SO_2
Rés. Faun. Ashuapmushuan	053201	48.81	-72.74	$PM_{2.5}$
Québec - Vieux-Limoilou	050308	46.82	-71.22	$PM_{2.5}$, CO, SO_2, NO_X
Lac-Édouard	053901	47.63	-72.29	O_3
La Patrie	054901	45.37	-71.25	$PM_{2.5}$, O_3
Montréal - Rivière Des Prairies	050129	45.65	-73.57	$PM_{2.5}$, CO, SO_2, NO_X, EC
Saint-Anicet	054401	45.12	-74.29	$PM_{2.5}$, CO, SO_2, O_3, NO_X, EC
Gatineau - Hull	050204	45.44	-75.72	$PM_{2.5}$, CO, SO_2, NO_X
Parc de la Gatineau - La-Pêche	052401	45.62	-76.02	O_3
Rouyn-Noranda - Parc Tremblay	050604	48.23	-78.98	$PM_{2.5}$, SO_2
Senneterre	055101	48.43	-77.20	$PM_{2.5}$, O_3
Lemieux	055201	46.30	-72.06	$PM_{2.5}$, CO, O_3
L'Assomption	054501	45.81	-73.43	CO
Varennes	052601	45.72	-73.38	NO_X

Table A2 : List of RSQAQ stations used to evaluate GEOS-Chem.

Station	NAPS ID	Latitude	Longitude	Pollutants
Auclair	052801	47.72	-68.71	$PM_{2.5}$, O_3
Saguenay - UQAC	050504	48.42	-71.05	$PM_{2.5}$, O_3
Rés. Faun. Ashuapmushuan - Pemonca	053201	48.81	-72.74	$PM_{2.5}$, O_3
Québec - Vieux-Limoilou	050308	46.82	-71.22	$PM_{2.5}$, O_3, NO_X, SO_2, CO
Québec - École Les Primevères	050311	46.77	-71.37	$PM_{2.5}$, O_3, NO_X
Québec - Collège St-Charles-Garnier	050310	46.79	-71.25	$PM_{2.5}$, O_3, NO_X
Saint-François-de-l'Île-d'Orléans	053501	47.02	-70.86	O_3
Deschambault	053301	46.68	-71.97	$PM_{2.5}$, O_3
Notre-Dame-Du-Rosaire	053601	46.85	-70.45	$PM_{2.5}$, O_3
Saint-Hilaire-de-Dorset	053701	45.82	-70.86	$PM_{2.5}$, O_3
Trois-Rivières - Cap-de-la-Madeleine	055401	46.36	-72.51	SO_2
Bécancour	054703	46.35	-72.43	$PM_{2.5}$, SO_2
Saint-Zéphirin-de-Courval	051501	46.04	-72.66	$PM_{2.5}$, O_3
Tingwick	053801	45.91	-71.95	$PM_{2.5}$, O_3
Charette	052001	46.44	-72.89	$PM_{2.5}$, O_3
Lac-Édouard	053901	47.63	-72.29	$PM_{2.5}$, O_3
Sherbrooke - Parc Cambron	050404	45.41	-71.87	$PM_{2.5}$, O_3
La Patrie	054901	45.37	-71.25	$PM_{2.5}$, O_3
Montréal - Saint-Jean-Baptiste	050103	45.64	-73.50	$PM_{2.5}$, O_3, NO_X, SO_2
Montréal - Rivière Des Prairies	050129	45.65	-73.57	$PM_{2.5}$, O_3, NO_X, CO, EC
Montréal - Aéroport de Montréal	050128	45.47	-73.74	$PM_{2.5}$, O_3, NO_X, CO
Montréal - Saint-Joseph	050134	45.54	-73.57	$PM_{2.5}$, O_3, NO_X, SO_2
Montréal - Sainte-Anne-De-Bellevue	050126	45.43	-73.93	$PM_{2.5}$, O_3, NO_X, SO_2
Laval – Chomedey	050113	45.55	-73.75	$PM_{2.5}$, O_3, NO_X, CO
Saint-Faustin–Lac-Carré	052301	46.04	-74.48	$PM_{2.5}$, O_3
Longueuil	050119	45.52	-73.49	$PM_{2.5}$, O_3, NO_X
Saint-Jean-sur-Richelieu - L'Acadie	055301	45.29	-75.35	$PM_{2.5}$, O_3, NO_X
Saint-Simon	052201	45.72	-72.84	$PM_{2.5}$, O_3
Saint-Anicet	054401	45.12	-74.29	$PM_{2.5}$, O_3, SO_2, CO, EC
Gatineau - Hull	050204	45.44	-75.72	$PM_{2.5}$, O_3, NO_X, SO_2, CO
Mont-St-Michel	055001	46.77	-75.43	$PM_{2.5}$, O_3
Parc de la Gatineau - La-Pêche	052401	45.62	-76.02	$PM_{2.5}$, O_3
Senneterre	055101	48.43	-77.20	$PM_{2.5}$, O_3

Table A3 : List of RSQAQ stations excluded from GEOS-Chem evaluation.

Station	NAPS ID	Latitude	Longitude
Saguenay - Parc Powell	050902	48.43	-71.19
Shawinigan - Saint-Marc	051201	46.55	-72.74
Montréal - Échangeur Décarie	050109	45.50	-73.66
Saint-Joseph-de-Sorel - École Martel	051801	46.05	-73.13
Rouyn-Noranda - Parc Tremblay	050604	48.23	-78.98
Témiscaming	052701	46.72	-79.10
Rouyn-Noranda - Mgr Rhéaume Est	08044*	48.24	-79.02

* This station is not part of the NAPS network, and therefore does not have a NAPS ID number. We list the RSQAQ ID number for this station instead.

Table A4: Percentage of trajectories missing for NO_x.

Station	NAPS ID	Annual	Summer (JJA)	Winter (DJF)
L'Assomption	054501	16.52 %	22.46 %	43.91 %
Saint-Anicet	054401	16.52 %	22.46 %	43.91 %
Varennes	052601	16.52 %	22.46 %	43.91 %
Québec - Vieux-Limoilou	050308	16.52 %	22.46 %	43.91 %
Gatineau - Hull	050204	16.52 %	22.46 %	43.91 %
Montréal - Rivière Des Prairies	050129	16.52 %	22.46 %	43.91 %

Table A5: Percentage of trajectories missing for SO_2.

Station	NAPS ID	Annual	Summer (JJA)	Winter (DJF)
Saint-Anicet	054401	17.04 %	22.96 %	45.08 %
Gatineau - Hull	050204	16.36 %	22.02 %	43.91 %
Montréal - Rivière Des Prairies	050129	16.52 %	22.46 %	43.91 %
Québec - Vieux-Limoilou	050308	16.19 %	22.46 %	43.51 %
Rouyn-Noranda - Parc Tremblay	050604	16.42 %	22.46 %	43.07 %
Saguenay - UQAC	050504	16.52 %	22.46 %	43.91 %

Table A6: Percentage of trajectories missing for O_3.

Station	NAPS ID	Annual	Summer (JJA)	Winter (DJF)
Parc de la Gatineau - La-Pêche	052401	16.24 %	23.66 %	43.70 %
Senneterre	055101	16.59 %	22.43 %	43.91 %
Lac-Édouard	053901	15.89 %	23.22 %	40.63 %
Lemieux	055201	18.31 %	22.46 %	48.76 %
La Patrie	054901	16.37 %	22.46 %	43.07 %
Rés. Faun. Ashuapmushuan	053201	17.36 %	22.71 %	45.21 %
Saint-Anicet	054401	16.52 %	22.46 %	43.91 %
Auclair	052801	16.58 %	22.46 %	44.57 %

Table A7: Percentage of trajectories missing for $PM_{2.5}$.

Station	NAPS ID	Annual	Summer (JJA)	Winter (DJF)
Gatineau - Hull	050204	16.52 %	22.46 %	43.91 %
Montréal - Rivière Des Prairies	050129	16.52 %	22.46 %	43.91 %
Rouyn-Noranda - Parc Tremblay	050604	16.52 %	22.46 %	43.91 %
Québec - Vieux-Limoilou	050308	16.19 %	22.46 %	43.51 %
Auclair	052801	16.52 %	22.46 %	43.91 %
Rés. Faun. Ashuapmushuan	053201	16.52 %	22.46 %	43.91 %
La Patrie	054901	16.71 %	22.47 %	44.32 %
Lemieux	055201	16.37 %	22.46 %	43.07 %
Senneterre	055101	16.59 %	22.43 %	43.91 %
Saint-Anicet	054401	16.52 %	22.46 %	43.91 %

Table A8: Percentage of trajectories missing for CO.

Station	NAPS ID	Annual	Summer (JJA)	Winter (DJF)
Montréal - Rivière Des Prairies	050129	16.52 %	22.46 %	43.91 %
Gatineau - Hull	050204	16.52 %	22.46 %	43.91 %
Québec - Vieux-Limoilou	050308	16.19 %	22.46 %	43.51 %
Saint-Anicet	054401	16.52 %	22.46 %	43.91 %
L'Assomption	054501	16.58 %	22.46 %	44.57 %
Lemieux	055201	16.37 %	22.46 %	43.07 %

Table A9: Percentage of trajectories missing for EC.

Station	NAPS ID	Annual	Summer (JJA)	Winter (DJF)
Montréal - Rivière Des Prairies	050129	16.52 %	22.46 %	43.91 %
Saint-Anicet	054401	16.52 %	22.46 %	43.91 %

Table A10: Evaluation of the GEOS-Chem base case simulation against observations from the RSQAQ stations given in Table A2 for winter (December, January and February), spring (March, April, and May), summer (June, July and August), and fall (September, October, and November). The units of ME and MB are μg m^{-3} for $PM_{2.5}$ and EC, ppm for CO, and ppb for all other species.

Pollutant	Season	ME	NME [%]	MB	NMB [%]
$PM_{2.5}$ [μg m^{-3}]	winter	2.70	34	-0.871	-11
	spring	1.74	29	-0.177	-3
	summer	1.73	23	-0.421	-6
	fall	1.96	33	0.554	9
CO [ppm]	winter	0.058	24	-0.045	19
	spring	0.049	24	-0.041	-21
	summer	0.043	21	-0.029	-14
	fall	0.055	26	-0.054	-26
NO_X [ppb]	winter	7.10	52	-6.93	-50
	spring	4.45	53	-4.41	-52
	summer	2.95	48	-2.91	-47
	fall	4.67	46	-4.55	-45
SO_2 [ppb]	winter	0.468	46	0.052	5
	spring	0.435	51	-0.162	-19
	summer	0.438	82	0.223	42
	fall	0.535	75	0.306	43
O_3 [ppb]	winter	4.49	17	4.12	15
	spring	5.54	16	5.34	16
	summer	8.24	33	8.20	33
	fall	11.00	54	11.00	54
EC [μg m^{-3}]	winter	0.160	30	-0.079	-15
	spring	0.103	29	-0.047	-13
	summer	0.160	35	-0.154	-34
	fall	0.125	29	-0.054	-13

Table A11: Relative and absolute (ppb) differences between the sensitivity simulations and the base-case simulation O_3 concentrations, spatially averaged over a region that encompasses the locations of the RSQAQ stations and the large population centres (45° N to 49° N, 78.75° W to 68.75° W).

Season	noQC	noCA	noUS
winter	-2.2 %	-0.4 %	4.3 %
	-0.733	-0.138	1.441
spring	1.1 %	2.6 %	11.0 %
	0.42	1.046	4.379
summer	4.5 %	5.2 %	16.6 %
	1.368	1.601	5.099
fall	0.4 %	2.5 %	12.8 %
	0.131	0.819	4.236

Table A12: Percentage of the area of Quebec exceeding the 2020 Canadian Ambient Air Quality Standards annual $PM_{2.5}$ concentration threshold of 8.8 µg m^{-3} in each simulation.

Year	Base Case	noQC	noRoC	noUS
2012	0.7 %	0.2 %	0.3 %	0.0 %
2013	11.7 %	10.9 %	11.2 %	11.1 %
2014	0.2 %	0.0 %	0.0 %	0.0 %
2015	0.2 %	0.0 %	0.0 %	0.0 %
2016	0.2 %	0.0 %	0.0 %	0.0 %

Table A13: Percentage of the area of Quebec exceeding the World Health Organization (WHO) air quality guidelines annual $PM_{2.5}$ concentration threshold of 5.0 µg m^{-3} in each simulation.

Year	Base Case	noQC	noRoC	noUS
2012	4.3 %	0.3 %	3.5 %	2.0 %
2013	31.1 %	24.3 %	29.2 %	26.4 %
2014	3.7 %	0.0 %	2.8 %	1.3 %
2015	3.8 %	0.0 %	3.2 %	1.5 %
2016	2.7 %	0.0 %	1.5 %	1.2 %

Table A14: Percentage of the population of Quebec exceeding the 2020 Canadian Ambient Air Quality Standards annual $PM_{2.5}$ concentration threshold of 8.8 µg m^{-3} in each simulation.

Year	Base Case	noQC	noRoC	noUS
2012	49.5 %	0.0 %	39.0 %	0.0 %
2013	49.5 %	0.0 %	39.0 %	0.0 %
2014	39.0 %	0.0 %	0.0 %	0.0 %
2015	39.0 %	0.0 %	0.0 %	0.0 %
2016	39.0 %	0.0 %	0.0 %	0.0 %

Table A15: Percentage of the population of Quebec exceeding the World Health Organization (WHO) air quality guidelines annual $PM_{2.5}$ concentration threshold of 5.0 µg m^{-3} in each simulation.

Year	Base Case	noQC	noRoC	noUS
2012	88.2 %	0.0 %	86.6 %	68.4 %
2013	91.9 %	0.2 %	91.0 %	72.9 %
2014	87.7 %	0.0 %	84.8 %	60.0 %
2015	87.9 %	0.0 %	86.6 %	61.6 %
2016	85.1 %	0.0 %	61.6 %	58.5 %

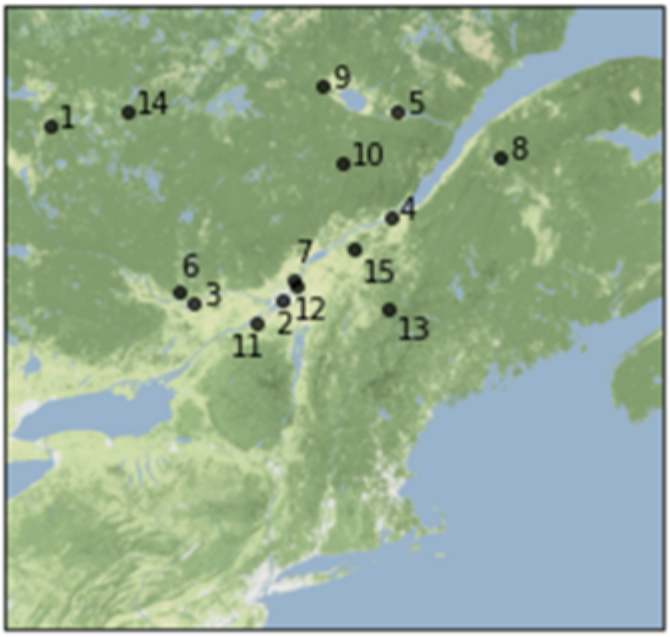

Figure A1: Locations of stations used for the PSCF analysis: 1. Rouyn-Noranda, 2. Montréal, 3. Gâtineau - Hull, 4. Québec, 5. Saguenay, 6. La Peche, 7. Varennes, 8. Auclair, 9. Rés. Faun. Ashuapmushuan, 10. Lac-Edouard, 11. Saint-Anicet, 12. L'Assomption, 13. La Patrie, 14. Senneterre, 15. Lemieux

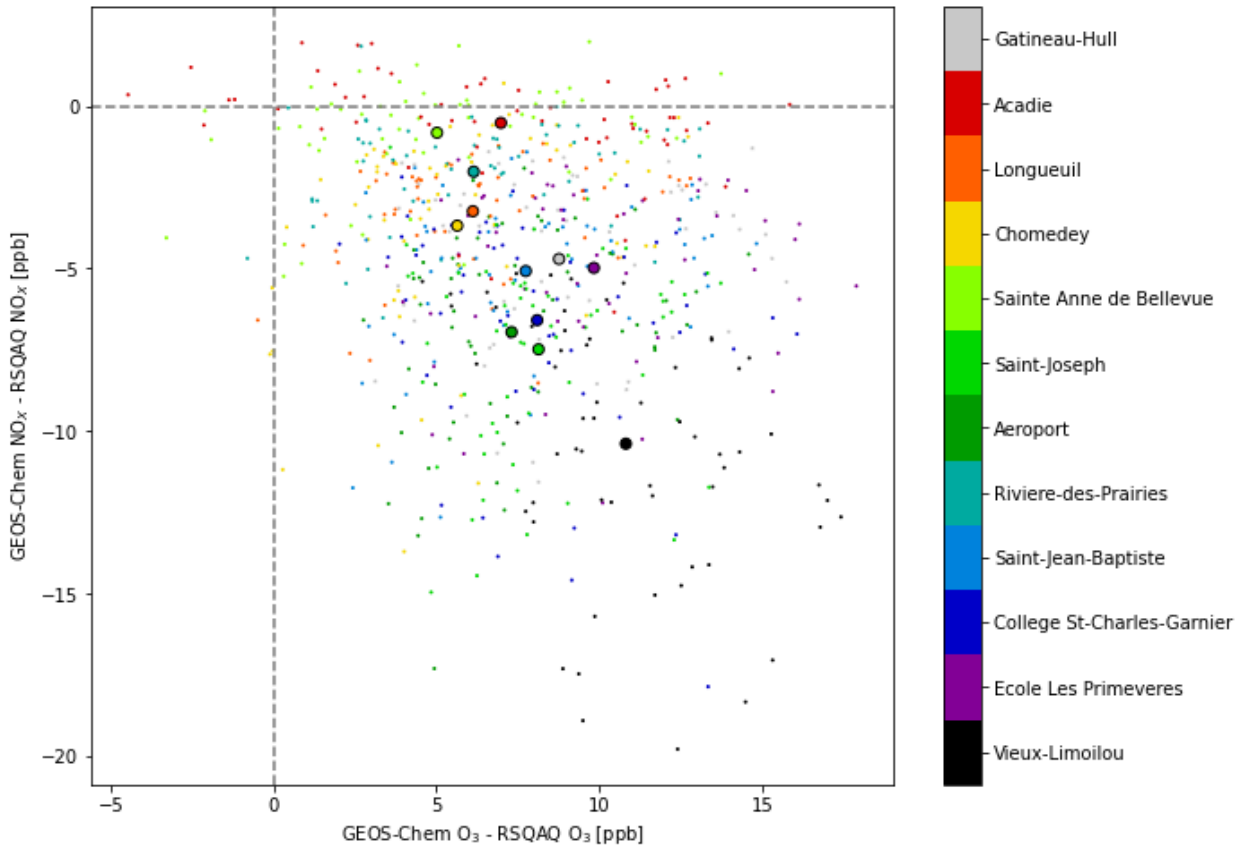

Figure A2: Scatterplot of model bias in NO_X concentrations vs. model bias in O_3 concentrations. Temporal averages at each station are displayed as large dots.

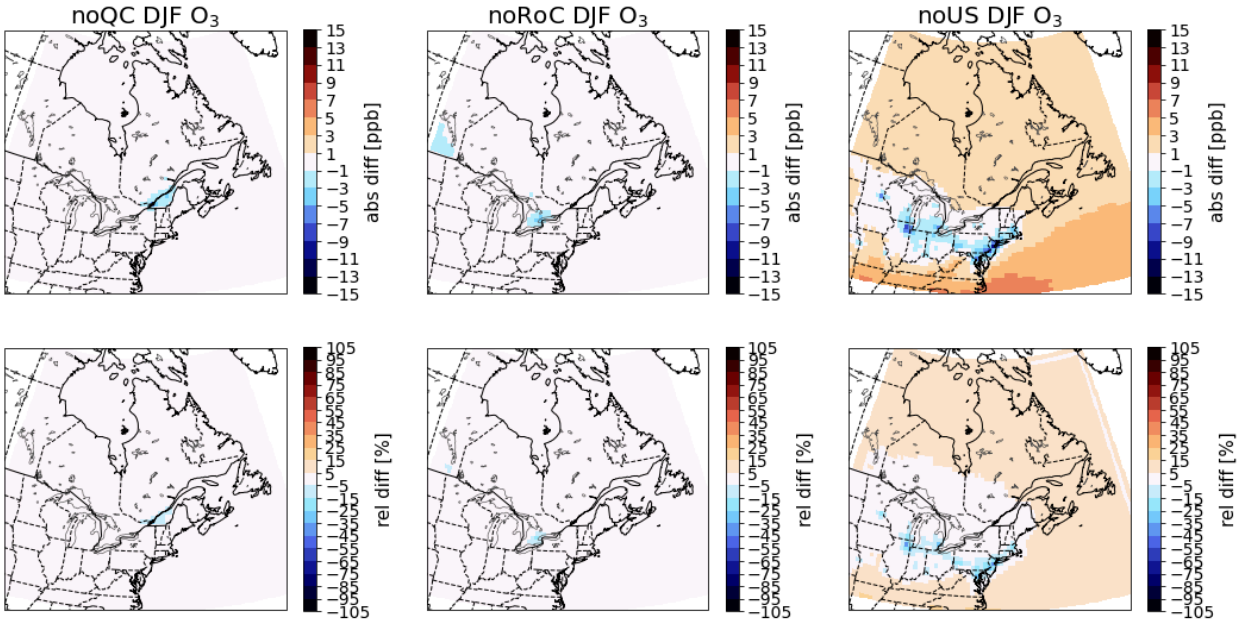

Figure A3 : Differences in winter surface O_3 concentrations between the sensitivity simulations and the base case simulation. Absolute differences are shown in the top row and relative differences in the bottom row. The three columns show the results of the noQC (left), noRoC (center) and noUS (right) sensitivity simulations.

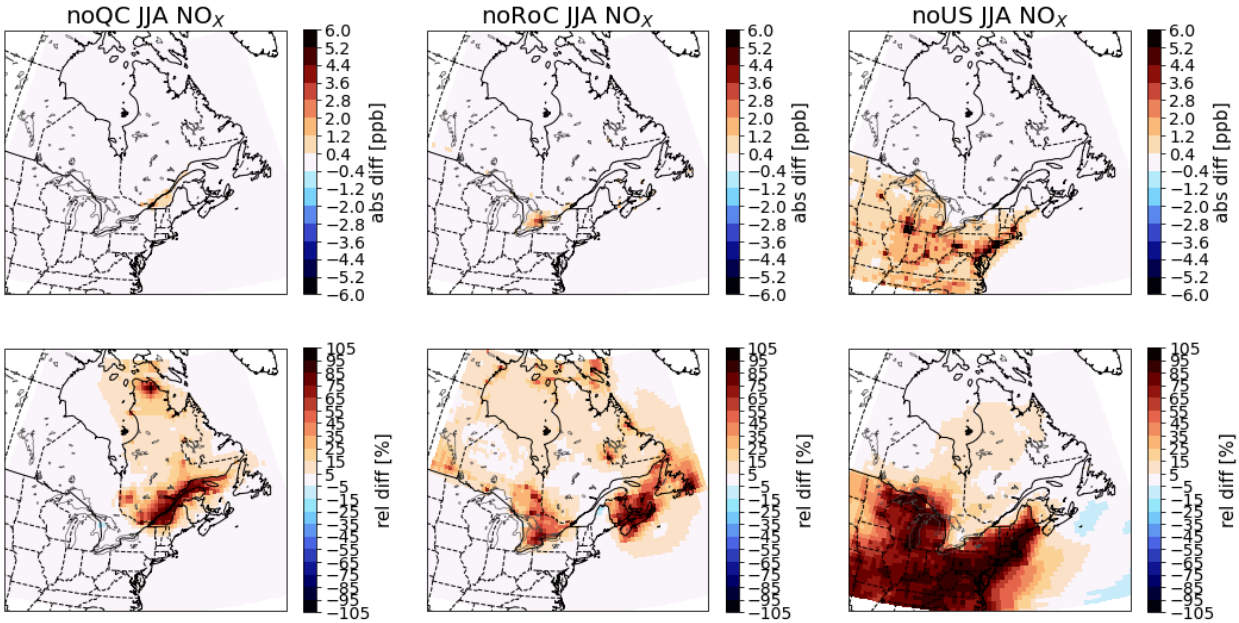

Figure A4: Differences in summer surface NO_x concentrations between the sensitivity simulations and the base case simulation. Absolute differences are shown in the top row and relative differences in the bottom row. The three columns show the results of the noQC (left), noRoC (center) and noUS (right) sensitivity simulations.

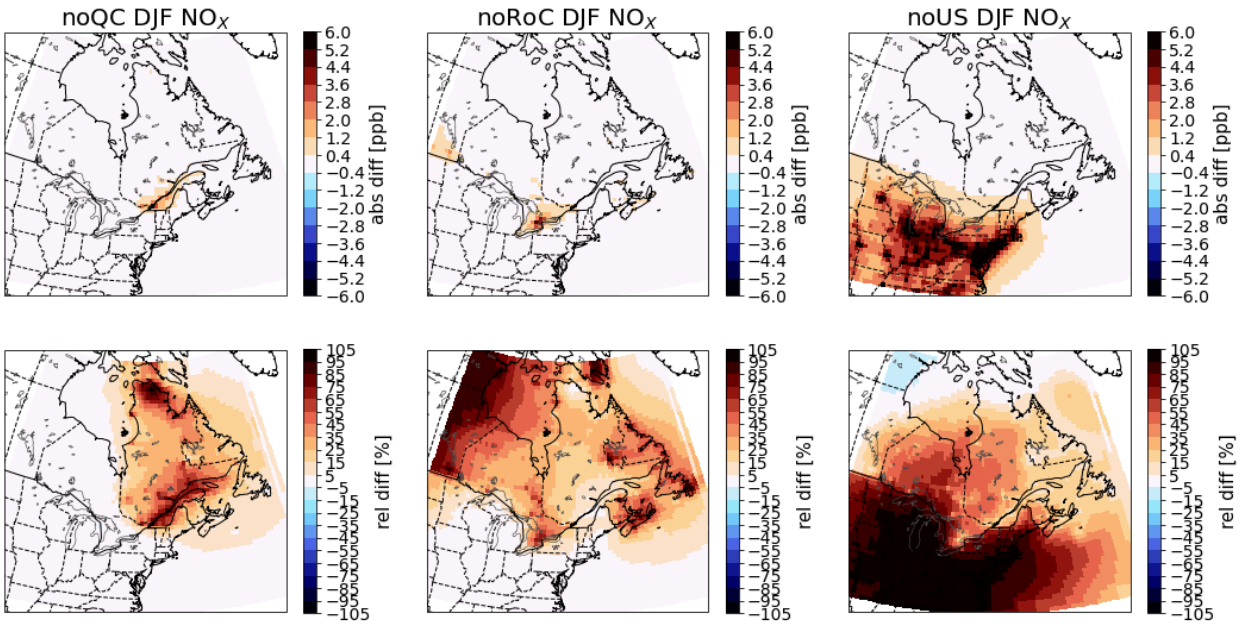

Figure A5 : Differences in winter surface NO_x concentrations between the sensitivity simulations and the base case simulation. Absolute differences are shown in the top row and relative differences in the bottom row. The three columns show the results of the noQC (left), noRoC (center) and noUS (right) sensitivity simulations.

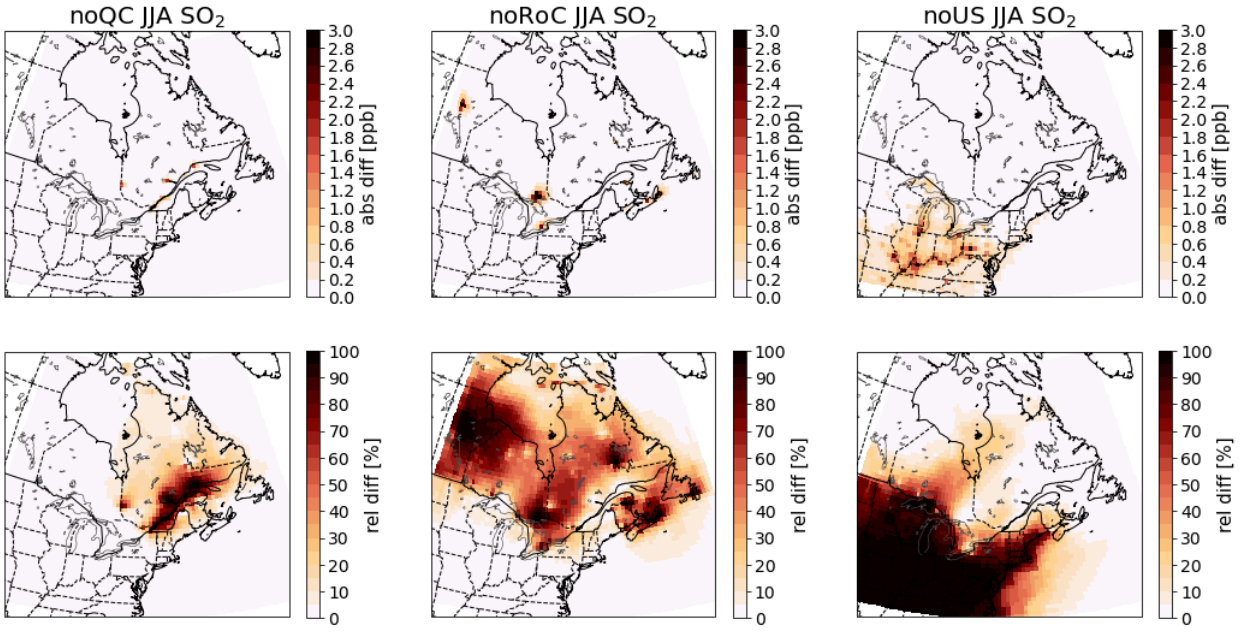

Figure A6 : Differences in summer surface SO_2 concentrations between the sensitivity simulations and the base case simulation. Absolute differences are shown in the top row and relative differences in the bottom row. The three columns show the results of the noQC (left), noRoC (center) and noUS (right) sensitivity simulations.

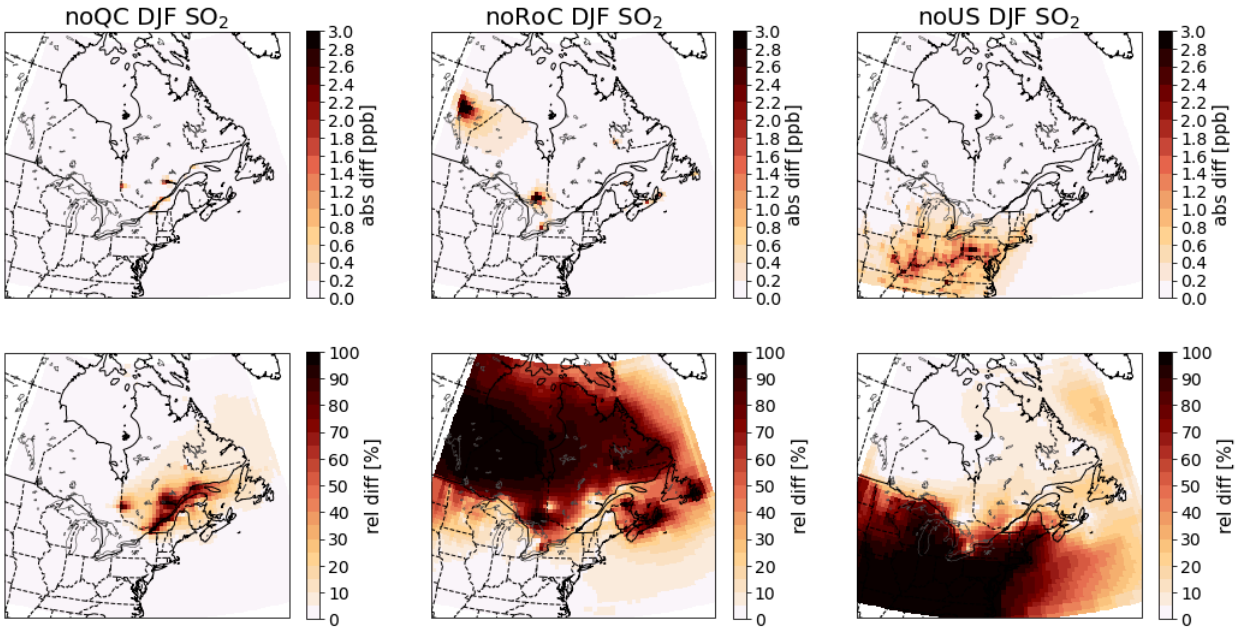

Figure A7 : Differences in winter surface SO_2 concentrations between the sensitivity simulations and the base case simulation. Absolute differences are shown in the top row and relative differences in the bottom row. The three columns show the results of the noQC (left), noRoC (center) and noUS (right) sensitivity simulations.

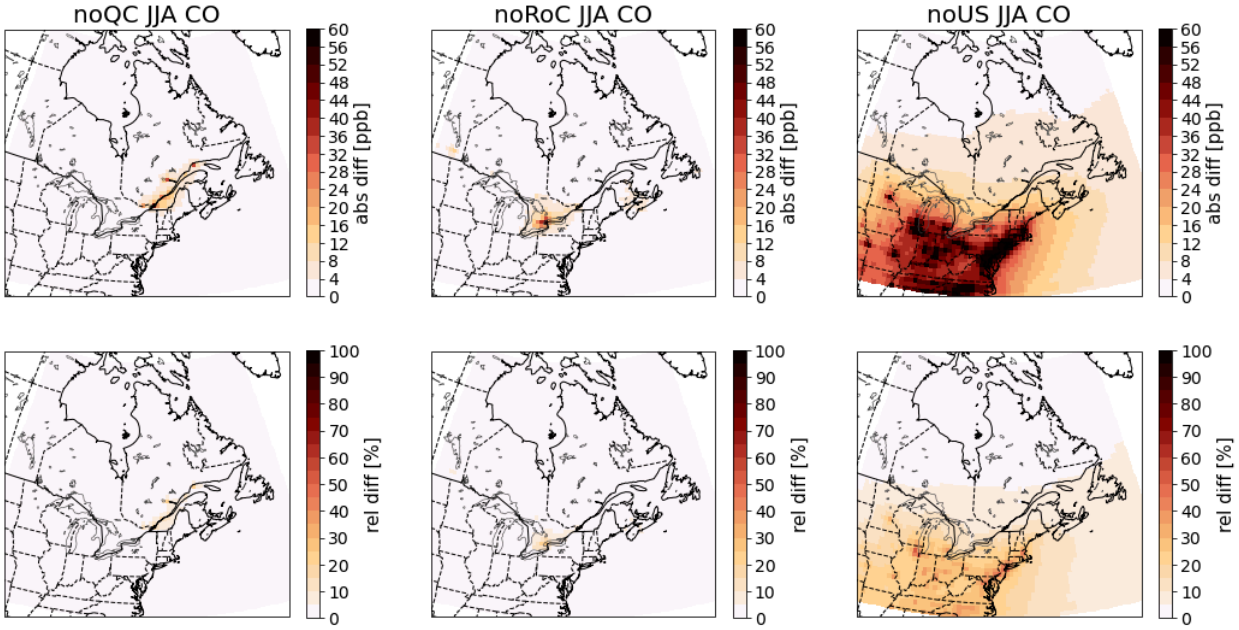

Figure A8: Differences in summer surface CO concentrations between the sensitivity simulations and the base case simulation. Absolute differences are shown in the top row and relative differences in the bottom row. The three columns show the results of the noQC (left), noRoC (center) and noUS (right) sensitivity simulations.

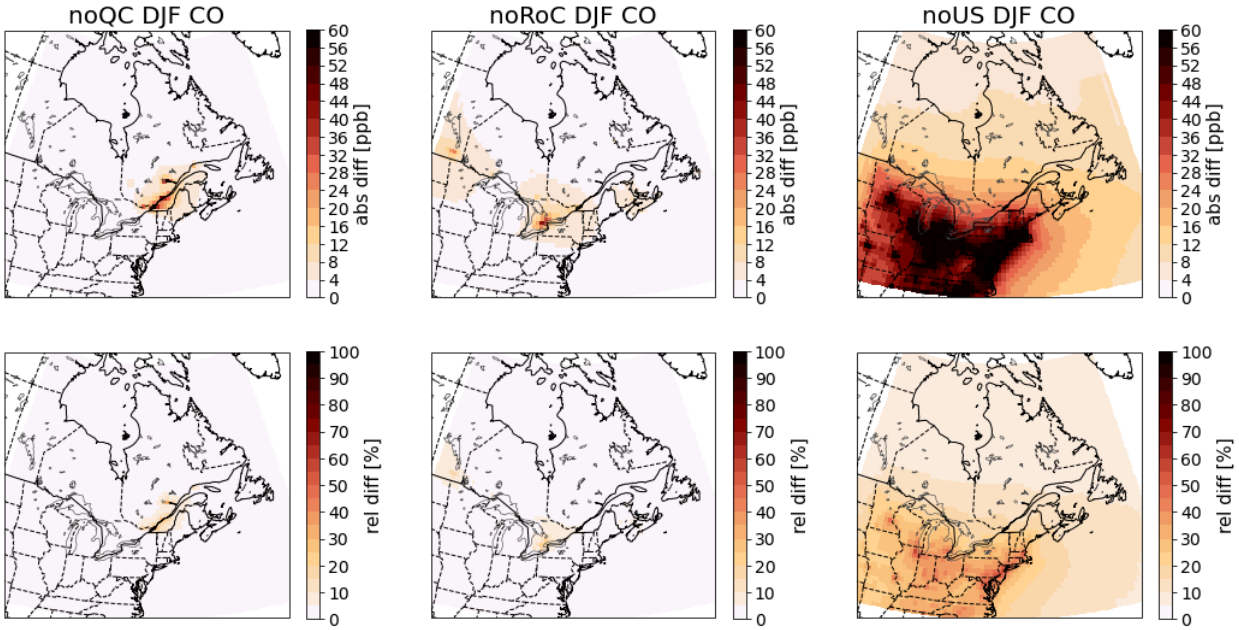

Figure A9: Differences in winter surface CO concentrations between the sensitivity simulations and the base case simulation. Absolute differences are shown in the top row and relative differences in the bottom row. The three columns show the results of the noQC (left), noRoC (center) and noUS (right) sensitivity simulations.

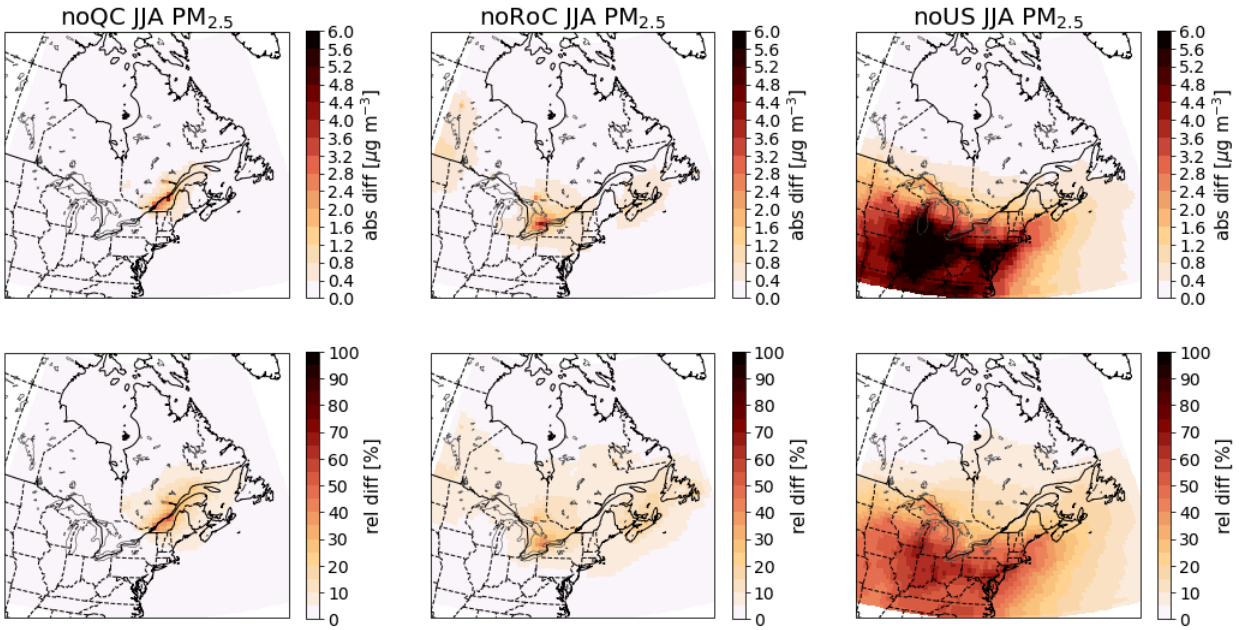

Figure A10: Differences in summer surface $PM_{2.5}$ concentrations between the sensitivity simulations and the base case simulation. Absolute differences are shown in the top row and relative differences in the bottom row. The three columns show the results of the noQC (left), noRoC (center) and noUS (right) sensitivity simulations.

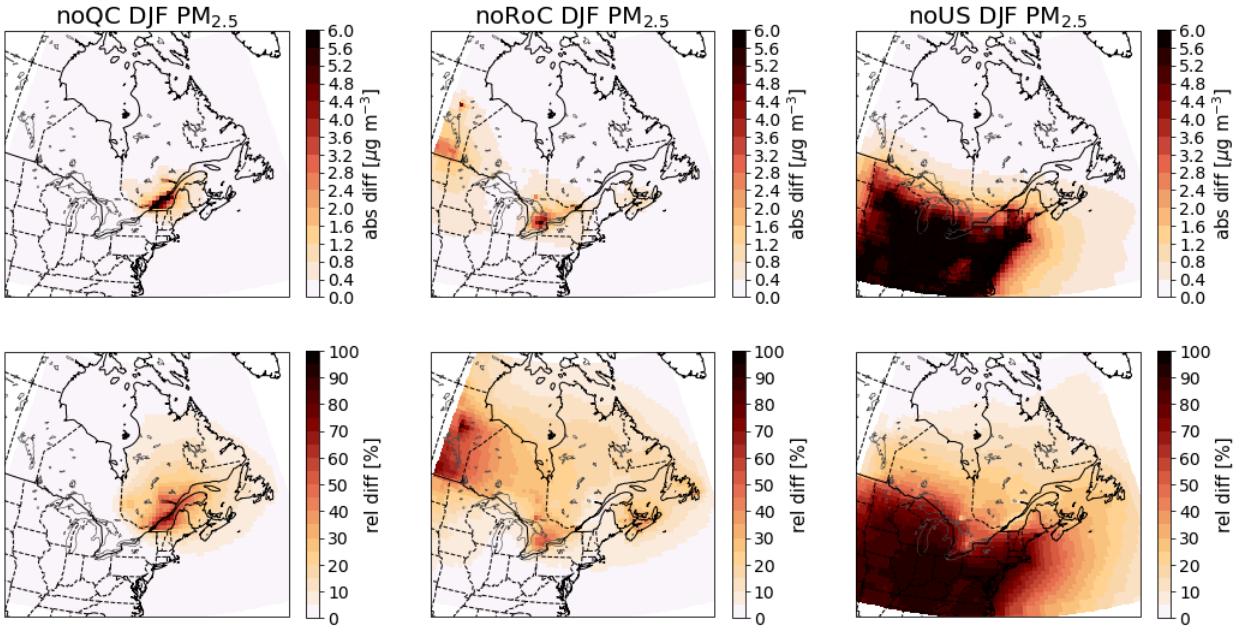

Figure A11: Differences in winter surface $PM_{2.5}$ concentrations between the sensitivity simulations and the base case simulation. Absolute differences are shown in the top row and relative differences in the bottom row. The three columns show the results of the noQC (left), noRoC (center) and noUS (right) sensitivity simulations.

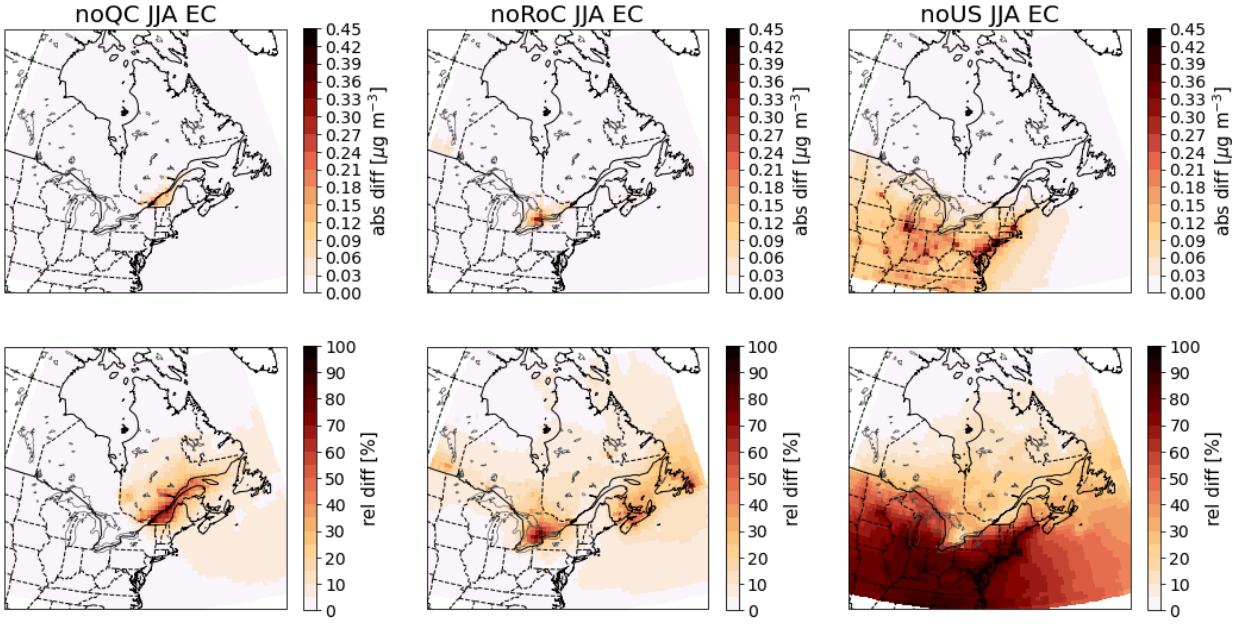

Figure A12: Differences in summer surface EC concentrations between the sensitivity simulations and the base case simulation. Absolute differences are shown in the top row and relative differences in the bottom row. The three columns show the results of the noQC (left), noRoC (center) and noUS (right) sensitivity simulations.

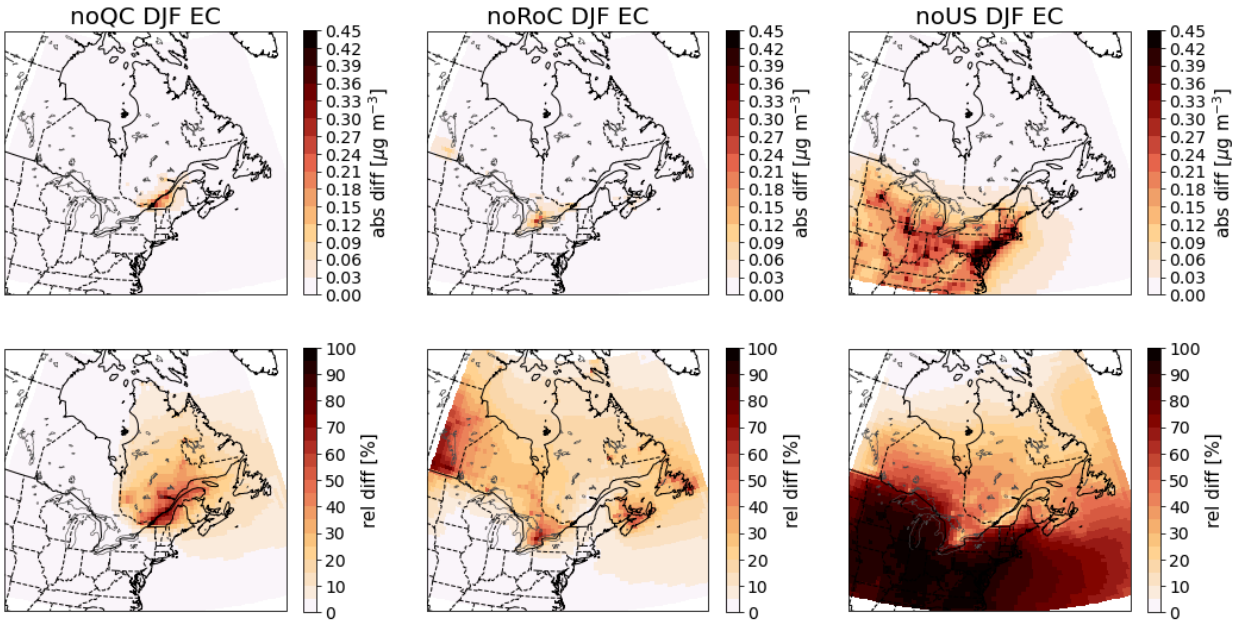

Figure A13: Differences in winter surface EC concentrations between the sensitivity simulations and the base case simulation. Absolute differences are shown in the top row and relative differences in the bottom row. The three columns show the results of the noQC (left), noRoC (center) and noUS (right) sensitivity simulations.

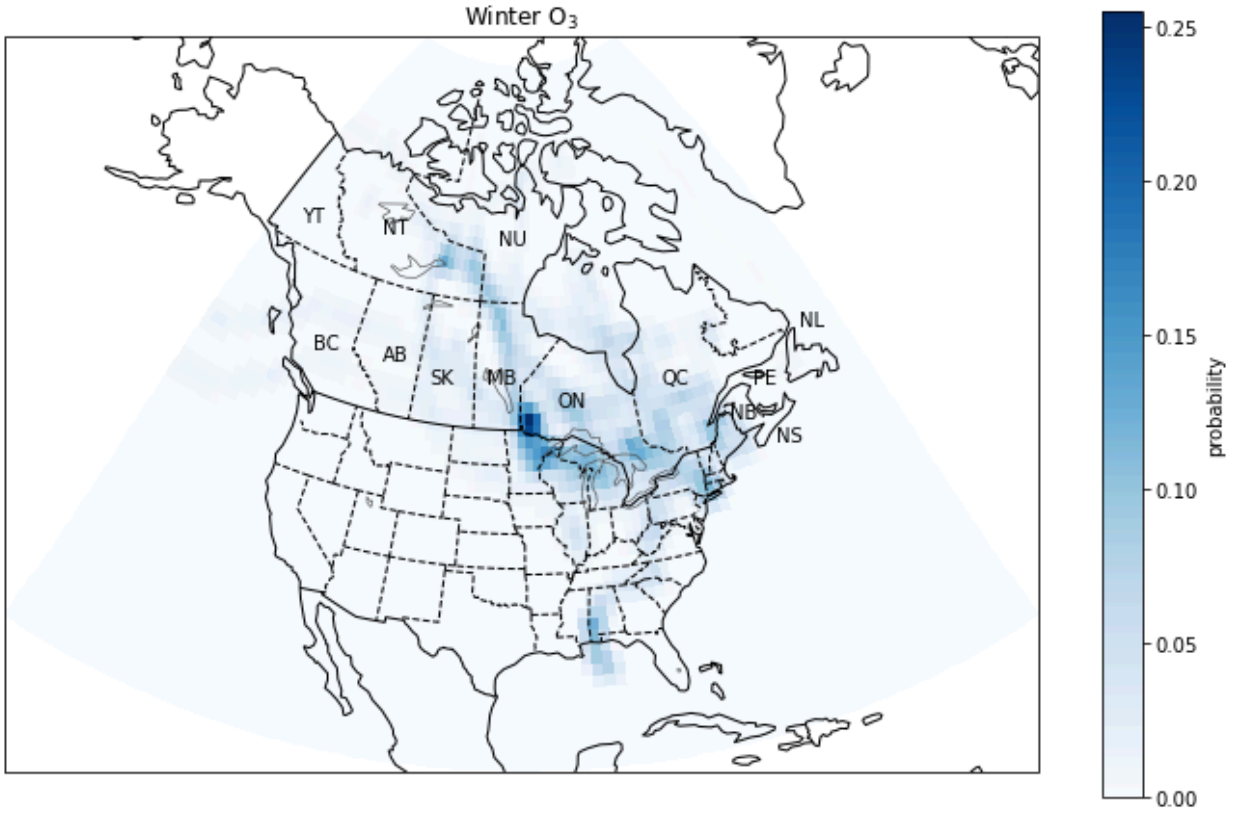

Figure A14: Winter probability source contribution function map calculated using the O_3 measurements. Canadian provinces and territories are labeled by postal abbreviation. NL: Newfoundland and Labrador, PE: Prince Edward Island, NS: Nova Scotia, NB: New Brunswick, QC: Quebec, ON: Ontario, MB: Manitoba, SK: Saskatchewan, AB: Alberta, BC: British Columbia, YT: Yukon, NT: Northwest Territories, NU: Nunavut.

In winter, the PSCF map for O_3 shows a medium probability that is widespread over a large territory, including Quebec, Ontario, Manitoba and the northeastern United States. Interestingly, there is a high probability near the Ontario-Manitoba border. According to the National Pollutant Release Inventory (NPRI),[1] a major source of VOCs in this region is the Gimli plant of Diageo Canada Inc., a distillery. This facility is the largest point source of VOCs in Manitoba, but according to the NPRI other point sources also contribute to VOC emissions in this region, including wood product manufacturers and pulp and paper mills. According to the GEOS-Chem modeling, it is possible that ozone production is limited by the relatively low concentrations of VOCs in winter for certain regions in southern Quebec. Thus, transboundary transport of anthropogenic VOCs from outside the province could increase ozone in winter.

References:

1 Government of Canada, National Pollutant Release Inventory Dashboard, https://www.canada.ca/en/environment-climate-change/services/national-pollutant-release-inventory/tools-resources-data/all-year-dashboard.html, (accessed June 27, 2022).