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Sensitivity analysis of ambient NO2 concentration to primary emission sources in Alberta, Canada using WRF/CMAQ modeling

Erfan Hajiparvaneh [a], Hossein Alizadeh [b], Charles Robert Koch [a], Vahid Hosseini* [b]

a Mechanical Engineering Department, University of Alberta, T6G 2R3, Alberta, Canada
b School of Sustainable Energy Engineering, Simon Fraser university, V3T 0N1, British Columbia, Canada

1. INTRODUCTION

The adverse impacts of air pollution on socioeconomic factors have prompted the revision of air quality standards worldwide (McCarthy and Lattanzio 2014, Wang, Yin et al. 2019, Anjum, Ali et al. 2021, Strak, Weinmayr et al. 2021). In Canada, these concerns have led to the recent revision of Canadian air quality standards (CAAQS) for Nitrogen dioxide (NO2), with new stringent standards set to take effect in 2025 (Canada 2021). The revised CAAQS for NO2 aims to further improve human health and encourage continuous efforts to mitigate air pollution. Accordingly, the threshold for NO2 concentration was set at an annual average of 17 ppb in 2020. However, this threshold will be reduced by 30% to 12 ppb in 2025.

In 2019, Alberta province, home to the fourth-largest oil reserves in the world, emerged as the highest emitter of nitrogen oxides (NOx), which includes NO2 and NO, in Canada, with emissions totalling 638,099 tonnes/year (Environment 2021). The upstream oil and gas (UOG) sector, encompassing oil and gas extraction, production, and heavy transportation activities, accounted for 49% of the total NOx emissions in Alberta (Nopmongcol 2018). Transportation, especially in urban areas, was another significant contributor, responsible for 15% of NOx emissions (Nopmongcol 2018). Understanding the sensitivity of ambient NO2 concentrations to these major NOx emitters, particularly the UOG sector, is crucial for policymakers, researchers, and stakeholders concerned with reducing air pollution and promoting public health. This understanding can inform the development of effective strategies to comply with the new CAAQS standards.

Increased UOG development and production activities have raised public concerns about the potential health impacts (Vlavianos 2006). However, there is still a lack of comprehensive understanding regarding the contribution and influences of upstream oil and gas production on ambient air pollution (Johnston, Lim et al. 2019, Gonzalez, Francis et al. 2022). Air Quality Model (AQM) is deployed to evaluate the contribution of the major NO2 sources, namely UOG and transportation, and understand the sensitivity of ambient NO2 concentrations to these. Previous studies (Cho, McEachern et al. 2012, Cho, Morris et al. 2012, Vijayaraghavan, Cho et al. 2016, Bari and Kindzierski 2018, Zhang, Moran et al. 2018) have primarily focused on the northern part of Alberta and the impacts of the Athabasca oil sands field. In 2019, the effects of oil sand regions on ambient ozone concentrations were analyzed, considering current and future emission scenarios (Vijayaraghavan, Cho et al. 2016). The most recent modelling report by the Alberta government developed a WRF-CMAQ model for the entire province and validated it for PM2.5 concentrations (Nopmongcol 2018). However, none of the mentioned modelling studies exclusively analyzed NO2 concentrations, and the sensitivity of ambient NO2 concentrations to different emission change scenarios remains unknown.

The WRF-CMAQ model is deployed and evaluated in this study to capture and analyze NO2 concentrations. The unique topography of Alberta enables us to determine the sensitivity of ambient NO2 concentrations to emissions from UOG and transportation. Unlike previous studies focusing solely on the Athabasca oil sands, this model covers the entire province and includes emissions from all UOG activities, transportation, and other industrial or non-industrial sources. By assessing the contributions of primary NO2 sources in Alberta, this study analyzed potential emission reduction scenarios to meet the new CAAQS NO2 standards. This AQM allows for analyzing the impact and effectiveness of emission control strategies in reducing NO2 levels.

2. PROBLEM DEFINITION, MODEL & METHODOLOGY

2.1 CAAQS Status in Alberta

**Corresponding author:* Vahid Hosseini, School of Sustainable Energy Engineering, Simon Fraser university, V3T 0N1, British Columbia, Canada; e-mail: vahid_hosseini@sfu.ca

The Canadian Ambient Air Quality Standards (CAAQS) were developed to assess the realization of actions to improve air quality across Canada (CCME). These standards have been defined for various chemical substances, including nitrogen dioxide (NO2) (Canada 2021). CAAQS divides the concentration level of intended pollutants at a certain location into four categories: Red, Orange, Yellow, and Green. The Red category aims to reduce pollutant levels below the CAAQS through advanced air management actions. The Orange category aims to improve air quality and prevent exceedance of the CAAQS. The Yellow category aims to improve air quality through early and ongoing actions. The Green category requires air quality stakeholders to maintain clean air levels.

An analysis was conducted to assess the status of monitoring stations based on the new CAAQS by considering the availability of observation data. If the emission levels from 2019 remain constant, the analysis shows that 13% of CAAQS stations will be categorized as red, 30% as orange, 55% as yellow, and one station as green. It is important to note that all the red stations will be located in large urban areas, indicating that nearly 55% of the province's total population is exposed to high levels of NO2 concentration. The exceedance of the CAAQS for NO2 is a pressing issue that requires targeted actions and the implementation of new policies to prevent further exceedance.

2.2 Emission Inventory Summary

An accurate AQM requires a comprehensive consideration of all emission sources in an emission inventory. The quality and accuracy of these inventory files directly impact the performance of the modelling system (Zhu, Mac Kinnon et al. 2019). In the Alberta province, multiple anthropogenic emission inventory databases are available (Nopmongcol 2018). For this study, the most up-to-date anthropogenic emission inventory, which combines available inventories, is utilized. Compiled by the Ramboll Company and NOVUS Environmental, this emission inventory is the most current and reliable data source for air quality modelling in Alberta. It has also been employed in the latest Alberta photochemical modelling report (Nopmongcol 2018).

Notably, the total NOx emission in Alberta exceeds 600,000 tons per year (CCME). When examining the contribution of different NOx emission sources, it becomes evident that Upstream Oil and Gas (UOG) emissions are nearly three times higher than any other NOx source, accounting for approximately 48% of the total NOx emissions in Alberta (Nopmongcol 2018). The second and third highest contributors to NOx emissions are non-industrial point and non-point sources, and on-road mobile sources, respectively (Nopmongcol 2018). These sectors contribute approximately 20% and 15% of the NOx emissions, respectively.

2.3 Modeling Domain

The focus of this study is to be consistent with the government of Alberta's air quality modeling requirements (Alberta 2021). The computational domain consists of three nested domains for the WRF model and one finer domain for the CMAQ model. The largest WRF domain covers northern America and captures large-scale low-pressure atmospheric conditions. The mid-scale atmospheric phenomena are considered in a smaller domain. The finest domain includes the geographical features of Alberta at a high resolution. The computational domain is a one-way nested domain, and the results from the coarse domain are used as initial and boundary conditions for the finer domain.

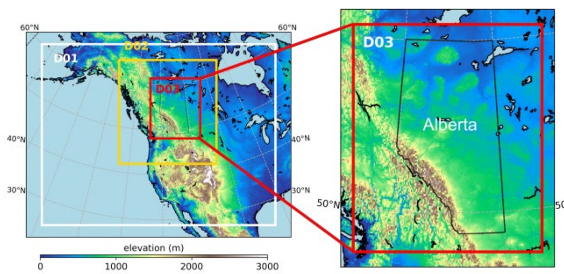


Fig. 1 Nested Computational Domain

2.4 WRF-CMAQ Model

The AQM used in this study consists of the WRF v4.2.2 model (Skamarock, Klemp et al. 2019) and the CMAQ v5.3.3 model (Appel, Bash et al. 2021). The WRF-Advance Research WRF (ARW) model was used in this analysis to resolve meteorological fields such as temperature and wind speed (Skamarock, Klemp et al. 2019). The model configuration used in this study includes the Thompson scheme for microphysics parameterization (Thompson, Rasmussen et al. 2004), for long and short wave radiation the rapid radiative transfer model for global climate models (RRTMG) scheme (Iacono, Delamere et al. 2008), the revised MM5 scheme for surface layer (Jiménez, Dudhia et al. 2012), unified Noah land surface model (Mukul Tewari, Tewari et al. 2004) and Yonsei University scheme for boundary layer (Hong, Noh et al. 2006). The US National Center for

Atmospheric Research (NCAR) final operational global analysis dataset was used to generate boundary and initial conditions.

The CMAQ model is a state-of-the-art Eulerian photochemical grid-based model that analyzes and simulates the transport and chemical interactions of multiple primary and secondary pollutants (Appel, Bash et al. 2021). In this study, the piece-wise parabolic method (PPM) is utilized for resolving horizontal advection (Colella and Woodward 1984) and parameterizes horizontal diffusion fluxes using eddy diffusion theory. The carbon-bond 06 model is used for analyzing gas-phase chemistry (Appel, Bash et al. 2021). Boundary and initial conditions for the CMAQ domain are generated using the northern hemisphere monthly modeling results of CMAQ available online through the CMAS Center database (Agency). The coupling between the CMAQ and WRF models is offline one-way coupling.

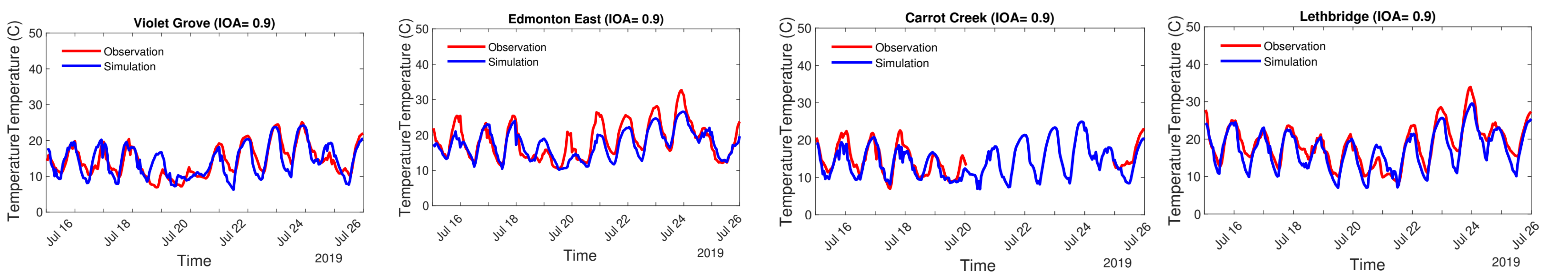
2.5 Sensitivity Analysis and Scenarios

In this study the Brute Force Sensitivity Analysis (BFSA) method was implemented to determine the contribution of major sources to ambient NO2 concentration and evaluate potential strategies for NO2 concentration reduction. BFSA is a forward sensitivity analysis method that calculates the impacts of input changes through time and space. It is particularly useful when the effects of a limited number of inputs on all outputs need to be assessed (Chen, Chang et al. 2019, Thunis, Clappier et al. 2019). Zero-out scenarios and perturbation cases were considered to evaluate the impacts of different scenarios. In the zero-out scenarios, the total emissions from a specific source were eliminated, while in the perturbation cases, 1%, 5%, 25%, 50%, and 75% of the emissions from either UOG or transportation sources were eliminated.

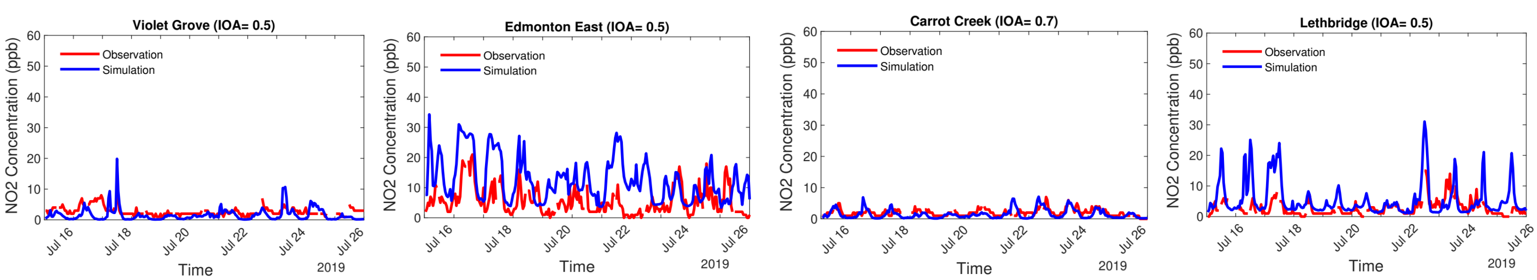
3. MODEL VALIDATION & PERFORMANCE

The correlation between modeled and observed temperature can be analyzed using the time series in Figure 2a. The temperature's mean bias (MB) of -1.14 K indicates an underestimation of the modeling results. However, the model's mean error (ME) value of 2.18 and the index of agreement (IOA) value of 0.91 demonstrate that the model meets the Bowden and Emery criteria (Emery, Liu et al. 2017). The biases in meteorological fields can be attributed to factors such as grid resolution, land-use model resolution, proximity to open water bodies, and elevation sensitivity of wind speed sensors.

Regarding the performance of the photochemical model, the time series of NO2 concentration is shown in Figure 2b. The Normalized Mean Square Error (NMSE), the Fractional Bias (FB) and the Factor of 2 (FAC2) for the averaged NO2 concentration of all stations calculated by the model are 2.23, 0.23, and 0.41, respectively, which satisfy the benchmark criteria introduced by Hannah (Hanna and Chang 2012). The results of the model reveals that most monitoring stations during the modeling period did



(a) Temperature



(b) NO_2

Fig. 2 Ground level temperatures model vs. experimental. Hourly time series of observed (red line) and modeled (blue); (a) Temperature (°C), (b) NO2 (ppb) at selected monitoring stations.

not experience NO2 concentrations greater than 20 ppb, with a total average of less than 10 ppb. The model accurately captures this trend, with only minor over-prediction observed during the middle of the day and rush hour.

4. RESULTS & DISCUSSION

4.1 Source Contribution (Zero-out)

Zero-out sensitivity scenarios provide a comprehensive understanding of UOG and transportation contribution to the total NO2 concentration. Fig 3, shows the average NO2 concentration reduction in percentage due to the source elimination of either mobile transportation Fig 3a or UOG Fig 3b. The impact of emission reduction in the modelling period is more local so the maximum concentration reduction is in the vicinity (4km distance) of the sources. One of the reasons for such behaviour is the short life cycle of NO2 in warm temperatures and atmospheric instability in warm seasons. That made the impacts of the sources more local. As it is evident from Fig 3, the transportation contribution is more pronounced in the vicinity of populated cities and along the highway that connects these cities. Comparing the impacts of UOG and transportation, the UOG considerably affects NO2 concentration across the whole province. However, its emissions affect rural and background areas and alongside the Rocky Mountains from west to west south of Alberta.

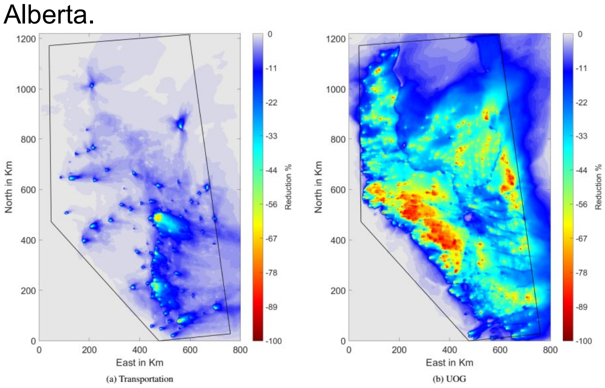


Fig. 3 NO2 concentration reduction due to zeroing out of Mobile sector and UOG emission

Emission sources affect each station differently. For example, at Violet Grove and Carrot Creek, UOG sources are dominant. While at Edmonton East and Lethbridge, the mobile source is the dominant source. Considering the stations' classification, Violet Grove and Carrot Creek are RB stations, while Edmonton East and Lethbridge are PE stations. Other stations like Fort Fort Saskatchewan, Grand Prairie, Edson, Calgary Central, and Airdrie, which show the same pattern as Edmonton East and Lethbridge station, are all PE stations. The NO2 level in these PE stations is considerably higher than in the RB stations. Overall, RB stations are more sensitive to UOG effects compared with mobile and other sources. Meanwhile, PE stations are more sensitive to mobile sources. The Fort McMurray Athabasca Valley station, which is PE station, is within 30 km distance from one of the largest oil sands in the world, but still, the transportation effects in this station are more pronounced than UOG effects. The maximum NO2 concentration in stations where transportation dominates other sources is between 20-40 ppb while the maximum of NO2 level in stations where transportation is not dominant is between 7-15 ppb.

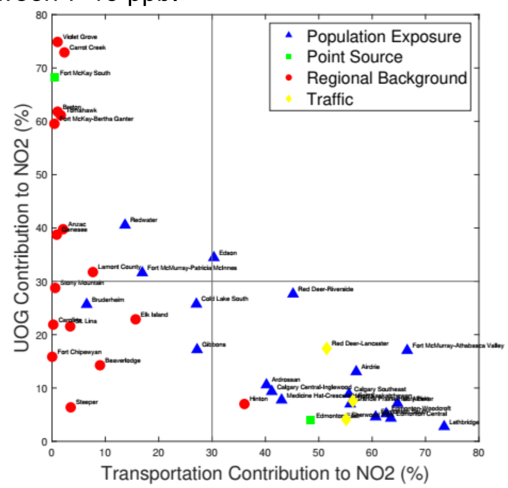


Fig. 4 Comparison of Mobile Transportation and UOG sources contributions on the NO2 concentration for four categories of stations

The scatter plot in Fig. 4 is provided to further assess the sensitivity of NO2 levels to transportation from vehicles and emissions from unconventional oil and gas (UOG) sources. In this figure, the contribution of major NO2 sources to the overall NO2 concentration is compared across 4 categories of monitoring stations. Evidently, in 73% of the PE stations, transportation accounts for more than 30% of the total NO2 concentration, while the impact of UOG sources is less than 30%. Conversely, in most of the RB stations, UOG sources are the dominant contributors to NO2. For instance, in Violet Grove and Carrot Creek, approximately 75% of the total NO2 concentration originates from UOG sources. As for the two PS stations, one is located near Fort Mckay oil industries and is more influenced by UOG sources (68%), while the other is in Edmonton and is affected by mobile sources (48%). Fig. 5 illustrates how the effects of UOG on NO2 concentration are primarily observed in Regional Background stations and rural areas, while the NO2 concentration in the majority of urban areas and Population Exposure stations is influenced by mobile sources.

4.1 NO2 composition in major cities

The impact of transportation emissions on NO2 concentration in populated areas of Alberta has been investigated through sensitivity analysis and zero-out scenarios. The results reveal that all the stations exceeding the Canadian Ambient Air Quality Standards (CAAQS) are located in populated areas. To determine the composition of NO2 concentration in stations located in two major cities, Edmonton and Calgary, the NO2 reduction due to the zeroing out of each source was averaged over the simulation period. The sensitivity simulations demonstrate that the contribution of unconventional oil and gas (UOG) sources to NO2 emission at these stations is relatively low, accounting for approximately one-tenth and one-fifth of the impact of mobile sources in Edmonton and Calgary, respectively. In Edmonton, eliminating mobile sources could lead to a significant reduction of 60% in NO2 concentration during the modelling period, while the elimination of UOG sources would only result in a 6% decrease.

Similarly, in Calgary, the sensitivity of UOG sources is slightly higher, but their contribution remains below 10% when zeroed out. Therefore, the impact of mobile sources on NO2 concentration in both cities is substantial, with transportation emissions accounting for approximately 53% of the total. These findings highlight the importance of considering transportation emissions when proposing NO2 abatement policies to meet the new 2025 CAAQS standards in Alberta.

4.2 2025 CAAQS achievement

The new stringent Canadian Ambient Air Quality Standards (CAAQS) have set a lower threshold for NO2 concentration. The annual average threshold of 17 ppb in 2020 has been reduced by 30% to 12 ppb for the year 2025. To analyze the potential NO2 mitigation plan in urban areas, the concentration of NO2 in the year 2019, which represents a pre-COVID-19 situation, was considered. Urban areas are particularly sensitive to transportation-related emissions, with mobile sources contributing approximately 53% to NO2 concentration. Therefore, the potential abatement strategies, taking into account different transportation scenarios, were investigated. By considering the 2019 annual average of observed NO2 and the linear response of NO2 concentrations to emission changes, the required emission reductions to achieve the new standards were calculated for two major cities in Alberta. The results of this analysis are summarized in Table 4.

According to the findings presented in Table 1, reducing the NO2 concentration by an average of 7.8% in Edmonton and 16.2% in Calgary is crucial to meet the 2025 standards. To achieve this target, the modelling results indicate that the transportation sector needs to reduce its emissions by an average of 16% in Edmonton and 33.6% in Calgary. Overall, a 23% reduction in mobile source emissions is necessary for both cities to transition from the red to yellow management level, in accordance with the Canadian Ambient Air Quality Standards (CAAQS) for the year 2025.

Table 1 Reduction of mobile source emission required for realizing the CAAQS

	Annual Averaged (PPb)	*Required Conc. Change (%)*	*Required Emission Change (%)*
Edmonton Central	14.9	19.5	40.6
Edmonton East	12.1	0.8	1.2
Edmonton Woodcroft	12.4	3.2	6.2
Calgary Inglewood	15.5	22.6	47.2
Calgary Southeast	13.3	9.8	20.1

5. SUMMARY AND CONCLUSION

The study investigates the exceedance of the new Canadian Ambient Air Quality Standards (CAAQS) for NO2 in Alberta. An air quality atmospheric model is used to address the NO2 exceedance from the 2025 CAAQS in Alberta. The contribution of major NO2 sources, including unconventional oil and gas (UOG) and transportation, to ambient NO2 concentration is examined through sensitivity analysis.

The WRF-CMAQ model is validated against observation data from 40 monitoring stations across Alberta. The validation shows small biases in temperature and wind speed, attributed to factors such as grid resolution and land-use model resolution. The photochemical model simulation results for NO2 satisfy benchmark criteria with minor over-predictions during certain periods.

A zero-out case study reveals that UOG contributes less than 10% to ambient NO2 concentration in urban areas, while transportation is responsible for 53% of ambient NO2 concentration in urban areas. A sensitivity analysis confirms a linear correlation between emission changes and NO2 concentration, with urban areas being more sensitive to transportation emissions and rural areas more sensitive to the UOG source.

The study emphasizes the importance of reducing transportation emissions to meet the new CAAQS standards in populated cities and urban areas. Based on the base emission of 2019, the study calculates that transportation emissions in Edmonton and Calgary need to be reduced by at least 23% by 2025 to achieve the 12 ppb threshold.

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