

Identifying fire plumes in the Arctic with tropospheric FTIR measurements and transport models

C. Viatte^{1*}, K. Strong¹, J. Hannigan², E. Nussbaumer², L. K. Emmons², S. Conway¹, C. Paton-Walsh³, J. Hartley¹, J. Benmergui^{4,**}, and J. Lin^{4,5}

¹Department of Physics, University of Toronto, Toronto, ON, Canada

²National Center for Atmospheric Research, Boulder, CO, USA

³Department of Chemistry, University of Wollongong, Wollongong, New South Wales, Australia

⁴Department of Earth and Environmental Sciences, University of Waterloo, Waterloo, ON, Canada

⁵Department of Atmospheric Sciences, University of Utah, Salt Lake City, UT, USA

*now at: Division of Geological and Planetary Sciences, California Institute of Technology, Pasadena, CA, USA

**now at: School of Engineering and Applied Sciences, Harvard University, Cambridge, MA, USA

Fires release trace gases in the atmosphere, such as carbon monoxide (CO), hydrogen cyanide (HCN), and Non-Methane HydroCarbons (NMHCs), including ethane (C₂H₆), acetylene (C₂H₂), methanol (CH₃OH), formic acid (HCOOH), and formaldehyde (H₂CO). These species can be transported to the Arctic (Shindell et al., 2008) and affect tropospheric chemistry (Tilmes et al., 2011), oxidizing power (Olson et al., 2012), and radiative transfer (Wang et al., 2011) of this sensitive polar region, which has been warming rapidly over the past century (Lesins et al., 2010). The frequency and intensity of biomass burning are strongly linked to climate change, and constitute a large source of the variability in Arctic tropospheric composition. However, our knowledge concerning transport, emissions from fires and sources of Arctic pollution

remains incomplete. Our recent study investigates pollution from biomass burning events that occurred in extratropical forests and were transported to the high Arctic with two sets of FTIR measurements, located in Eureka (Nunavut, Canada, 80°05'N, -86°42'W) and Thule (Greenland, 76°53'N, -68°74'W) (Viatte et al., 2015).

Biomass burning tracer species: CO, HCN, and C₂H₆ total columns measured at Eureka and Thule from 2008 to 2012 are shown on the left and right panels, respectively, of Figure 1 (next page). They exhibit strong seasonal cycles, reflecting the importance of chemistry and transport processes in their Arctic budget. In addition to these cycles, simultaneous enhancements of the CO, HCN, and C₂H₆ total columns can be seen in their day-to-day variabilities, such as in April and July 2008 (red circles, Figure 1), and in August 2010 (green squares, Figure 1).

Fire events are identified in the FTIR timeseries by selecting all days that have simultaneous enhancements of these three primary tracers. Using this methodology, ten biomass burning events have been identified as reaching Eureka and eight at Thule, between 2008 to 2012. In order to match the biomass burning candidate events identified in the timeseries with actual plumes, it is necessary to find the source fires and show that the plumes generated there are capable of travelling to the Arctic stations where they were observed. Figure 2 shows an example of the source attribution and the travel duration of a plume that reached Eureka on the 10th of July, 2008. As a priori information, STILT footprints (Stochastic Time-Inverted Lagrangian Transport model) are generated to show the source region influencing an atmospheric measurement at Eureka, which for that day is located in Eastern Russia (light blue region inside the red box, Figure 2a). Then the FIRMS map (Fire Information for Resource Management System, which provides MODIS hot spot data) is used to verify

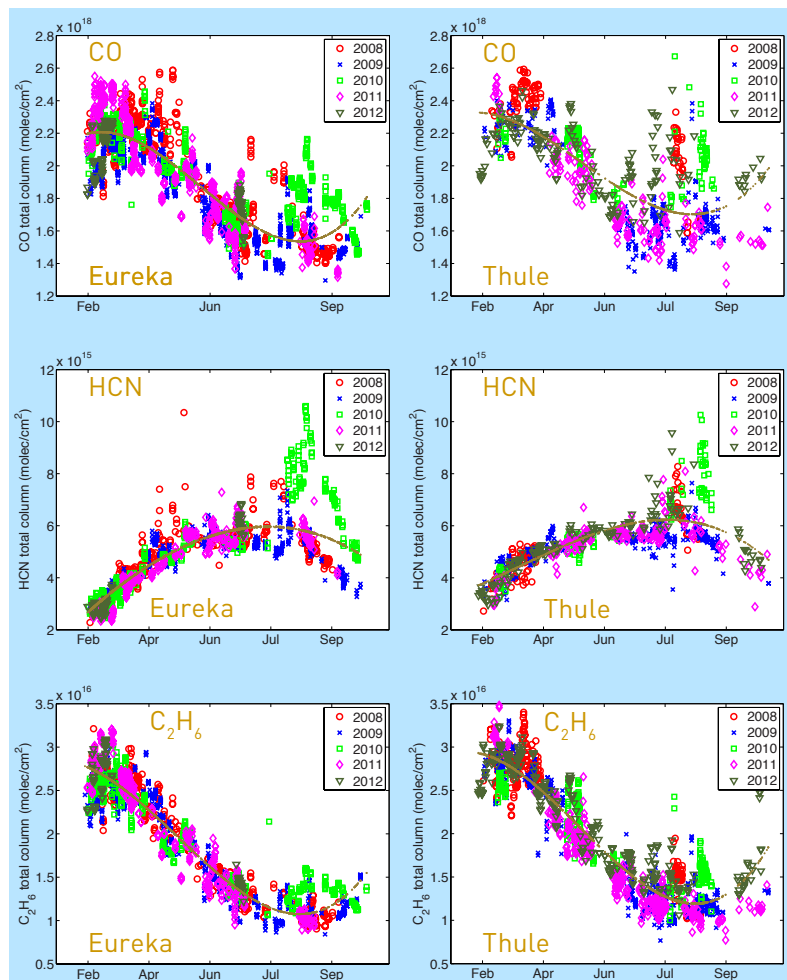


Figure 1: Annual cycles of multiyear CO, HCN, and C₂H₆ total columns measured at Eureka (left panels) and Thule (right panels) from 2008 to 2012. The brown lines represent the polynomial fits to the data.

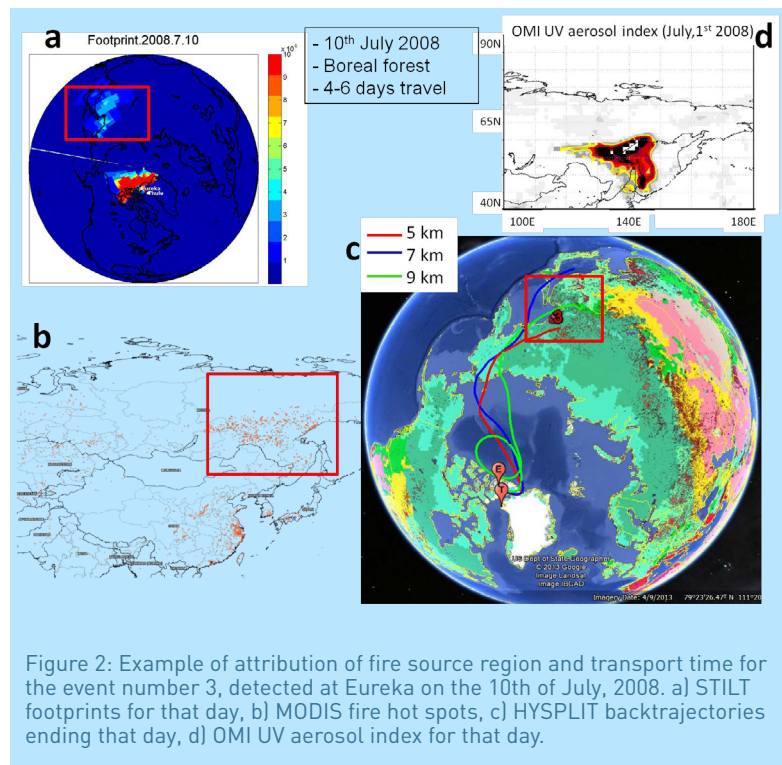


Figure 2: Example of attribution of fire source region and transport time for the event number 3, detected at Eureka on the 10th of July, 2008. a) STILT footprints for that day, b) MODIS fire hot spots, c) HYSPLIT backtrajectories ending that day, d) OMI (Ozone Monitoring Instrument) aerosol index map is used to confirm the presence

that a significant fire event occurs in that specific region, within a 10-day period (red dots in Figure 2b). To assess the travel duration of that plume from the fire region to Eureka, an ensemble of HYSPLIT back-trajectories (Hybrid Single Particle Lagrangian Integrated Trajectory Model) is generated, for several travel times, end times of the calculated trajectories, and air-parcel altitudes. In Figure 2c, air masses ending at Eureka at 5, 7, and 9 km (red, blue, and green lines, respectively) on July 10th, come from the fire region (red box). Then finally, the OMI (Ozone Monitoring Instrument) aerosol index map is used to confirm the presence

of a significant fire event in that region, as shown in Figure 2d (colored area within the red box). Consistent results from these multiple datasets provides confidence in the attribution of trace gas enhancements to specific fire events.

To improve our knowledge concerning the dynamical and chemical processes associated with Arctic pollution from fires, the two sets of FTIR measurements were compared to the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4, Emmons et al., 2010). The 2008 timeseries of daily mean CO, HCN, C₂H₆, C₂H₂, CH₃OH, and H₂CO total columns measured by the FTIRs at Eureka and Thule (Figure 3, blue and green dots, respectively), and calculated by MOZART-4 at these two sites (Figure 3, black and red dashed lines, respectively) are used to compare their seasonal cycles. This year was chosen because the April and July biomass burning events have been studied during the ARCTAS (Arctic Research of the Composition of the Troposphere from Aircrafts and Satellites) campaign.

In winter, CO, HCN and C₂H₂ total columns estimated by MOZART-4 agree very well with the FTIR measurements, leading to high confidence in the transport mechanisms in the model, since it is the major process controlling the Arctic budget of these long-lived gases. However, for C₂H₆ which is also a long-lived tracer, the underestimation of its concentrations by MOZART-4 in winter confirms an underestimation in anthropogenic emissions in the model. Also we note that in spring and summer, the overestimation of modeled concentrations suggest that loss processes for HCN are missing, confirming that its sinks are not well quantified (Zeng et al., 2012). The CH₃OH seasonal cycle estimated by MOZART-4 exhibits the best agreement with the observational datasets at Eureka. Focusing on the July 2008 biomass burning event, the CH₃OH enhanced concentrations are well captured by the model, suggesting that its fire emissions are correct. For CO

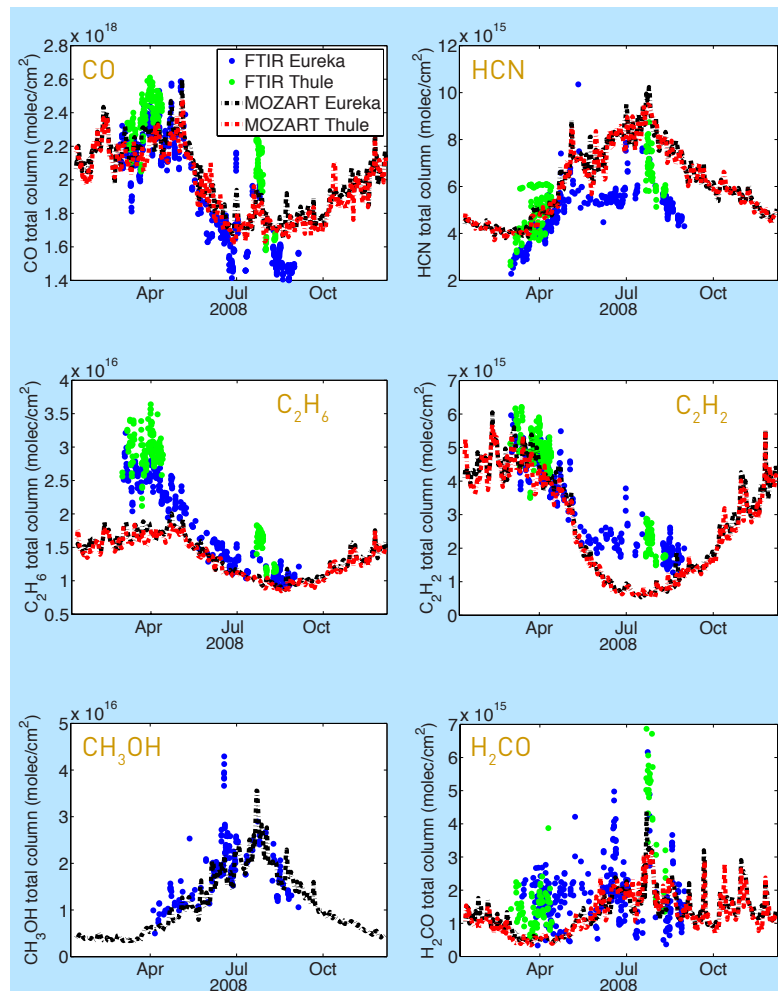


Figure 3: Timeseries of CO, HCN, C₂H₆, C₂H₂, CH₃OH, and H₂CO total columns measured by the FTIRs at Eureka (blue) and Thule (green) and calculated by MOZART-4 at Eureka (black) and Thule (red) for 2008.

and H₂CO, enhancements estimated by the model are too low compared to the measurements. This might indicate that their fire emissions are too low in the model. In contrast, the modelled and measured HCN enhancements are in good agreement. For C₂H₆ and C₂H₂, the modelled enhancements are extremely low compared to the measurements, indicating missing sources.

These results indicate that long-term and continuous measurements of Arctic tropospheric composition are important for quantifying emissions from fire plumes transported from lower latitudes and improving the prediction of trace gas concentrations and variability in chemical transport model simulations. This would help in assessing the atmospheric impact of biomass burning pollution on the Arctic climate system. Further, that mid-latitude land use and pollution sources are changing we can expect increased pollution and biomass burning events. The warming Arctic is rapidly changing independently. We should expect transport pathways from mid-latitudes to the Arctic to carry more pollutants northward. Our ability to observe and model these events will be critical to our evaluation of the overall anthropogenic effects on the Arctic regions.

References

- Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), *Geosci. Model Dev.*, 3, 43–67, doi:10.5194/gmd-3-43-2010, 2010.
- Lesins, G., Duck T. J., and Drummond, J. R.: Climate trends at Eureka in the Canadian high Arctic, *Atmosphere Ocean*, 48, 59–80, doi:10.3137/AO1103.2010, 2010.
- Olson, J. R., Crawford, J. H., Brune, W., Mao, J., Ren, X., Fried, A., Anderson, B., Apel, E., Beaver, M., Blake, D., Chen, G., Crouse, J., Dibb, J., Diskin, G., Hall, S. R., Huey, L. G., Knapp, D., Richter, D., Riemer, D., St. Clair, J., Ullmann, K., Walega, J., Weibring, P., Weinheimer, A., Wennberg, P. O., and Wisthaler, A.: An analysis of fast photochemistry over high northern latitudes during spring and summer using in-situ observations from ARCTAS and TOPSE, *Atmos. Chem. Phys.*, 12, 6799–6825, doi:10.5194/acp-12-6799-2012, 2012.
- Shindell, D. T., Chin, M., Dentener, F., Doherty, R. M., Faluvegi, G., Fiore, A. M., Hess, P., Koch, D. M., MacKenzie, I. A., Sanderson, M. G., Schultz, M. G., Schulz, M., Stevenson, D. S., Teich, H., Textor, C., Wild, O., Bergmann, D. J., Bey, I., Bian, H., Cuvelier, C., Duncan, B. N., Folberth, G., Horowitz, L. W., Jonson, J., Kaminski, J. W., Marmer, E., Park, R., Pringle, K. J., Schroeder, S., Szopa, S., Takemura, T., Zeng, G., Keating, T. J., and Zuber, A.: A multi-model assessment of pollution transport to the Arctic, *Atmos. Chem. Phys.*, 8, 5353–5372, doi:10.5194/acp-8-5353-2008, 2008.
- Tilmes, S., Emmons, L. K., Law, K. S., Ancellet, G., Schlager, H., Paris, J.-D., Fuelberg, H. E., Streets, D. G., Wiedinmyer, C., Diskin, G. S., Kondo, Y., Holloway, J., Schwarz, J. P., Spackman, J. R., Campos, T., Ndiec, P., and Panchenko, M. V.: Source contributions to Northern Hemisphere CO and black carbon during spring and summer 2008 from POLARCAT and START08/preHIPPO observations and MOZART-4, *Atmos. Chem. Phys. Discuss.*, 11, 5935–5983, doi:10.5194/acpd-11-5935-2011, 2011.
- Viatte, C., Strong, K., Hannigan, J., Nussbaumer, E., Emmons, L. K., Conway, S., Paton-Walsh, C., Hartley, J., Benmergui, J., and Lin, J.: Identifying fire plumes in the Arctic with tropospheric FTIR measurements and transport models, *Atmos. Chem. Phys.*, 15, 2227–2246, doi:10.5194/acp-15-2227-2015, 2015.
- Wang, Q., Jacob, D. J., Fisher, J. A., Mao, J., Leibensperger, E. M., Carouge, C. C., Le Sager, P., Kondo, Y., Jimenez, J. L., Cubison, M. J., and Doherty, S. J.: Sources of carbonaceous aerosols and deposited black carbon in the Arctic in winter-spring: implications for radiative forcing, *Atmos. Chem. Phys.*, 11, 12453–12473, doi:10.5194/acp-11-12453-2011, 2011.
- Zeng, G., Wood, S. W., Morgenstern, O., Jones, N. B., Robinson, J., and Smale, D.: Trends and variations in CO, C₂H₆, and HCN in the Southern Hemisphere point to the declining anthropogenic emissions of CO and C₂H₆, *Atmos. Chem. Phys.*, 12, 7543–7555, doi:10.5194/acp-12-7543-2012, 2012.