Evaluating the factors influencing the tropospheric ozone distribution over the North Atlantic during summer 2010 with GEOS-Chem and its adjoint

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1. Introduction

Boreal forest fires are a significant source of atmospheric trace gases and aerosols and consequently play an increasingly important role in the chemical state of the atmosphere and global climate. Accurately estimating their impact relies on understanding the connection between measurements over the source region and downwind, involving uncertain atmospheric chemistry. In particular, there are serious gaps in our understanding of NO_y and organic chemistry within biomass burning plumes.

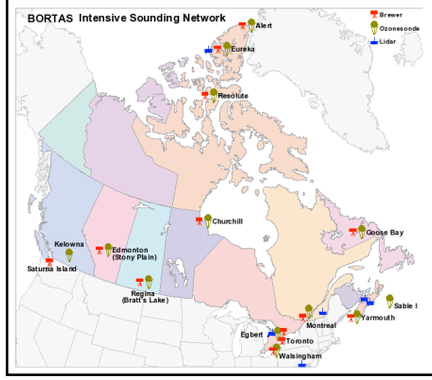
BORTAS is a multi-national project, led by the University of Edinburgh, with the overall goal of quantifying the connections between the composition and the distribution of pollutant outflow from boreal forest fires, ozone production and loss within that outflow, and the resulting perturbation to oxidant chemistry in the troposphere. To achieve this, the main objectives of BORTAS are to:

1. Measure biomass burning outflow from boreal North America using satellite, aircraft and surface instruments.
2. Describe observed chemistry within plumes with models of atmospheric chemistry and transport.
3. Quantify the impact on tropospheric oxidant chemistry by combining measurements and models.

This poster presents the first results from phase A of the BORTAS project, held in the summer of 2010, to evaluate the tropospheric ozone distribution over North America and the North Atlantic and its sensitivity to boreal biomass burning relative to other emission sources.

2. BORTAS-A measurement campaign

Phase A of the BORTAS project was a ground-based measurement campaign throughout July and August 2010, centred around the Dalhousie University Raman Lidar which provided continuous, under clear sky conditions, measurements of tropospheric aerosol profiles throughout the campaign period.



Support measurements for the Dalhousie Lidar included FTIR, Sun photometer, wind profiler, surface ozone, nephelometer, and AMS measurements.

Sampling of the outflow from biomass burning in central Canada as it was transported eastward to the Maritime provinces and the North Atlantic was provided by an intensive sounding network of Sun photometer, lidar, and ozonesonde measurements.

Daily ozonesonde launches were made from 12 July to 4 August 2010 at Bratt's Lake, Egbert, Goose Bay, Montreal, Sable Island, Walsingham, and Yarmouth.

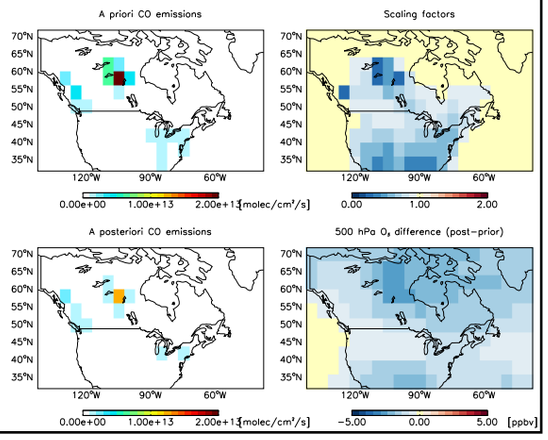
3. Model analysis

BORTAS model analysis is performed with GEOS-Chem v8-02-04 with biomass burning emission estimates provided by the Fire Locating and Monitoring of Burning Emissions (FLAMBE) inventory (http://www.nrlmry.navy.mil/flambe/).

Model analysis of the BORTAS-A period is supported by the GEOS-Chem adjoint to:

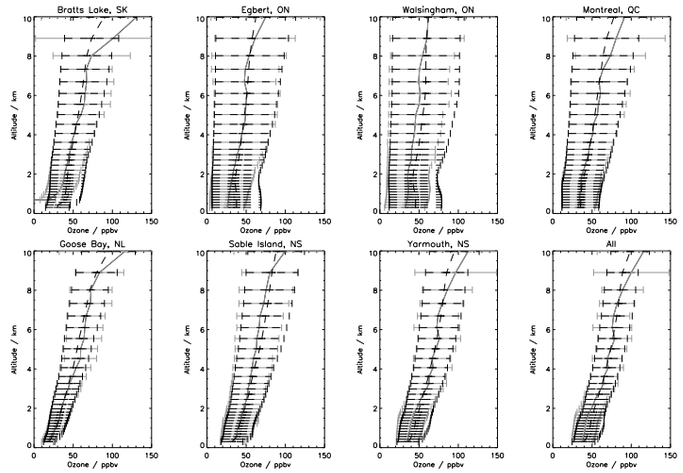
1. calculate scaling factors to reduce uncertainty associated with model inputs, e.g. FLAMBE, and
2. calculate sensitivity of modelled ozone distribution to precursor emissions (see section 5).

CO inversion with total columns retrieved from MOPITT (v4) show FLAMBE emissions to be a factor of ~2 too high in boreal regions for July 2010. The a posteriori FLAMBE emissions reduce ozone in the free troposphere by ~1-2 ppbv.

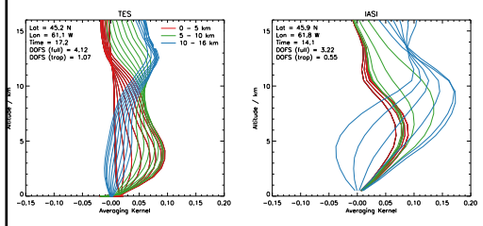


4. Summer 2010 tropospheric ozone distribution

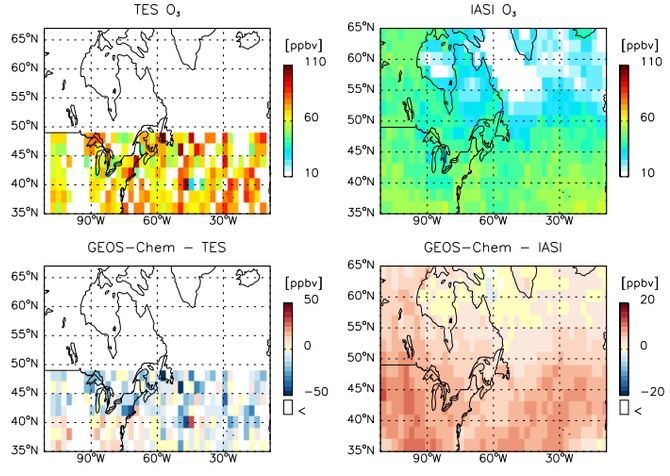
The modelled tropospheric ozone distribution is evaluated over North America with daily ozonesonde profiles measured during BORTAS-A, and extended across the North Atlantic using ozone profiles retrieved from the Tropospheric Emission Spectrometer (TES) and Infrared Atmospheric Sounding Instrument (IASI) satellite instruments.



The mean model ozone profiles, co-located in space and time to the launch of each ozonesonde, are shown as solid lines in the figure above and are generally in reasonable agreement with the ozonesonde profiles averaged over the GEOS-Chem vertical levels (dashed lines). Error bars show the 1-sigma standard deviation.



The averaging kernels for the TES and IASI ozone profile retrievals show slightly different sensitivities to the vertical distribution of ozone in the troposphere over the North Atlantic related to different spectral resolutions (0.1 cm^{-1} for TES, 0.5 cm^{-1} for IASI).

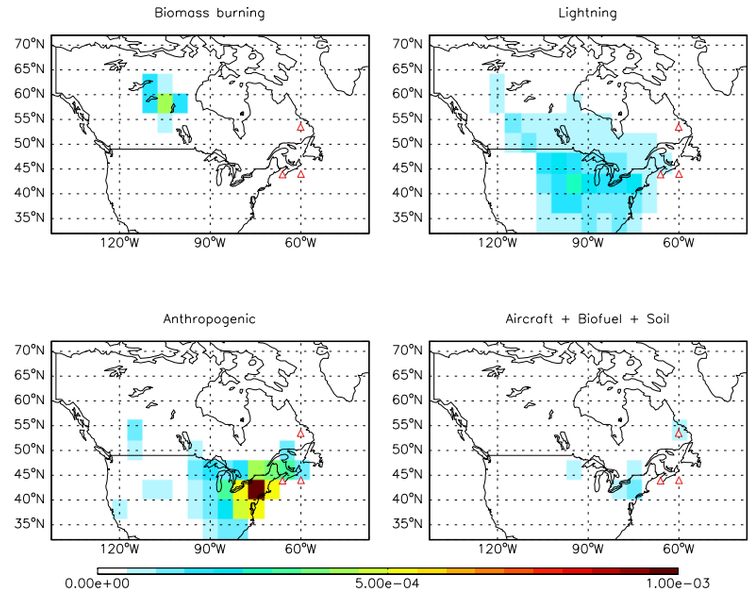


The modelled ozone distribution is sampled at the satellite observation locations and smoothed with the averaging kernels for comparison to the to TES and IASI. The mean difference (layer averages between 4 and 6 km) between the model and TES is -6.6 ppbv, consistent with the reported high bias of up to 15%, and with large variability reflecting the relatively coarse spatial sampling. The mean difference relative to IASI is +4.4 ppbv, consistent with a known low bias of ~15-20% compared to independent data.

5. North Atlantic ozone sensitivity to precursor emissions

The sensitivity of the modelled summertime tropospheric ozone distribution over the North Atlantic to precursor emissions is evaluated with the full chemistry adjoint of GEOS-Chem. The cost function is defined as the free tropospheric ozone profile over three ozonesonde launch sites in Maritime Canada: Goose Bay, Sable Island, and Yarmouth - shown as red triangles in the figure below.

The figure below shows the normalised sensitivity (gradient of the cost function with respect to each emission source) of the modelled ozone to NO_x emissions estimates across North America for July 2010. The largest sensitivity is to anthropogenic NO_x emissions along the east coast of the USA with biomass burning in central Canada showing the next highest sensitivity. Of the other NO_x emissions, lightning is the dominant source with peak sensitivity across central USA to the west of the Great Lakes.



6. Summary and outlook for BORTAS-B

The preliminary analysis presented here shows that the tropospheric ozone distribution modelled by GEOS-Chem for the summer of 2010 is in reasonable agreement with independent observations over North America and the North Atlantic. Detailed calculations with the GEOS-Chem adjoint show the free tropospheric ozone distribution to be largely sensitive to emissions from boreal biomass burning, anthropogenic, and lightning sources.

Phase B of the BORTAS project is a measurement campaign with the UK Facility for Airborne Atmospheric Measurements BAe146 research aircraft, to be based out of Halifax, Nova Scotia, Canada between 12 July and 3 August 2011. The BORTAS-B campaign will allow us to build extensively on the analysis presented here providing:

- airborne measurements of a comprehensive suite of species associated with biomass burning in the outflow to the North Atlantic throughout the free troposphere, including an LIF instrument providing detailed measurements of NO_y speciation;
- additional ground-based instrumentation at Dalhousie University, including an ozone profiling Lidar;
- detailed chemical modelling along aircraft, and biomass burning plume, trajectories with a comprehensive chemical mechanism derived from the Master Chemical Mechanism (MCM).
- comprehensive analysis of tropospheric composition across the North Atlantic with satellite observations of key biomass burning species (e.g. HCN, PAN) from TES, IASI, and ACE-FTS.

Acknowledgements

BORTAS is funded by the Natural Environment Research Council under grant NE/F017391/1. DKH supported through NASA New Investigator Program. FLAMBE emissions provided by Edward Hyer at the Naval Research Laboratory. Bob Yantosca, Jingqiu Mao and Jenny Fisher at Harvard provided the FLAMBE implementation in GEOS-Chem. MOPITT CO and TES O_3 data were obtained from the NASA Langley Research Center Atmospheric Science Data Center.