

**Fugitive emissions from the Bakken shale illustrate role
of shale production in global ethane shift**

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Introduction

Supporting text S1 includes further details on the GEOS-Chem model setup used in simulations. Supporting Figure S1 highlights more data from the mass balance analysis. Supporting Tables S1 and S2 provide more information on the mass balance analysis and model simulations.

Text S1.

GEOS-Chem Model description

The GEOS-Chem chemical transport model (version 10_01; <http://www.geos-chem.org>) is a global-to-regional 3-D model of tropospheric ozone- HO_x - NO_x - BrO_x -CO-VOC-aerosol chemistry that has frequently been applied to both global and regional air quality studies [Fiore *et al.*, 2014; Zhang *et al.*, 2014]. In this study, we use GEOS-Chem to estimate the ability of the observed Bakken fugitive flux of C₂+ alkanes to affect hemispheric-scale photochemistry in the global configuration, as well as archive boundary conditions for a regional North American simulation to assess the potential for fugitive Bakken emissions to perturb summertime regional surface photochemistry.

We use the model driven by the GEOS-5 FP meteorological reanalysis provided by the NASA Global Modeling and Assimilation Office of the Goddard Space Flight Center. Meteorological fields are provided at the native model horizontal resolution of 5/16° longitude by 1/4° latitude, with 72 vertical layers up to 0.01 hPa (approximately 38 in the troposphere), with 1-hour and 3-hour temporal resolution for 2- and 3-dimensional fields. Here we degrade total vertical resolution to 47 levels, maintaining native vertical resolution in the troposphere. For global simulations, we degrade the horizontal resolution to 2.5° longitude by 2° latitude and integrate transport over 15-min intervals. For regional North American simulations, we maintain the native horizontal resolution, prescribe boundary conditions that update each hour, and integrate transport over 10-min intervals. The chemical solver integrates over 60-min time steps for chemistry and emissions in all simulations.

We use the standard GEOS-Chem tropospheric chemical mechanism with recent significant updates to its isoprene oxidation mechanism [Mao *et al.*, 2013]. Alkane emissions and chemistry are isolated for ethane, propane, and lumped C₄+ species; methane concentrations are prescribed. Tropospheric photolysis rates are calculated online within the model using the Fast-J code [Wild *et al.*, 2000] with overhead ozone constrained by satellite observations. Aerosol chemistry includes inorganic, carbonaceous, mineral dust, and sea salt particles that interact with gas-phase species by influencing photolysis rates, through heterogeneous reactions, and via partitioning of semi-volatile compounds between gas and condensed phases.

Extended Data Table 2 summarizes the emissions used in the model simulations. Emission inventories are processed using the Harvard-NASA Emissions COmponent (HEMCO), which automatically converts an inventory to the spatiotemporal resolution of a given simulation [Keller *et al.*, 2014]. Base anthropogenic surface emissions are from the Emissions Database for Global Atmospheric Research (EDGAR) version 4.2 [EDGAR 2011] for inorganic compounds and the REanalysis of the TROpospheric chemical composition (RETRO) inventory for organic compounds [Schultz *et al.*, 2007]. We overwrite these base emissions in the United States with the most recent Environmental Protection Agency National Emission Inventory (NEI2011); input at 0.1° by 0.1° horizontal and 1-h temporal resolution, and integrated to each horizontal resolution. We also overwrite the base emissions with regional inventories for Canada (CAC), Mexico (BRAVO), Europe (EMEP), and South and East Asia (MIX v1.1). Anthropogenic emission inventories have diurnal and seasonal variability, some with

additional weekly cycles, and are scaled each year relative to their reference periods on the basis of economic data to allow for trends [van Donkelaar *et al.*, 2008].

Biomass burning emissions of NO_x, CO and volatile organic compounds (VOCs) from open fires are prescribed monthly from the Global Fire Emissions Database (GFED) version 4 [Giglio *et al.*, 2013]. Biogenic VOC emissions from terrestrial plants follow the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1, respond to meteorological conditions, and are constrained by satellite observations of leaf area indices, although monoterpenes are emitted directly as CO following fixed yield assumptions [Barkley *et al.*, 2011]. Biogenic marine VOC emissions are included. Production of NO_x by lightning is released locally into deep convective plumes [Murray *et al.*, 2012] and soil microbial production of NO_x responds to meteorology and fertilization [Hudman *et al.*, 2012]. Tropospheric methane concentrations are fixed to annual mean observations across four equal-area latitudinal bands.

We prescribe the estimated observed Bakken alkane fluxes by applying uniform scaling factors for each species to the local anthropogenic inventory (EPA NEI 2011) over a rectilinear region bounded by 46.7°N and 48.8°N and 104.08°W and 101.5°W. The emissions used are defined in Extended Data Table 2. We do not alter the local alkane emissions from biomass burning or anthropogenic stack point sources not associated with oil and gas production.

Following recommendations from ongoing GEOS-Chem analyses of the NASA SEAC⁴RS aircraft field campaign of summer 2013, we reduce the EPA NEI2011 NO_x emissions by 60% throughout the United States, and the source of NO_x from soils by 50% during late summer months in the Great Plains.

For our base simulation, we perform a global simulation for January 1, 2013 through December 31, 2014, and discard the first year as initialization. We then run the model in its nested North American configuration for the sub-period July 1, 2013 through August 31, 2014. We re-grid the coarse-model instantaneous conditions at July 1, 2013 00:00 UTC for the fine-model initial conditions, and periodically update boundary conditions every 60 min from the global model. We repeat this process for one additional sensitivity simulation in which we zero the Bakken source of C₂+ alkanes from fugitive emissions. We use the difference of the base simulation with this sensitivity simulation to estimate the impact of the Bakken on global and regional photochemistry from fugitive C₂+ alkanes.

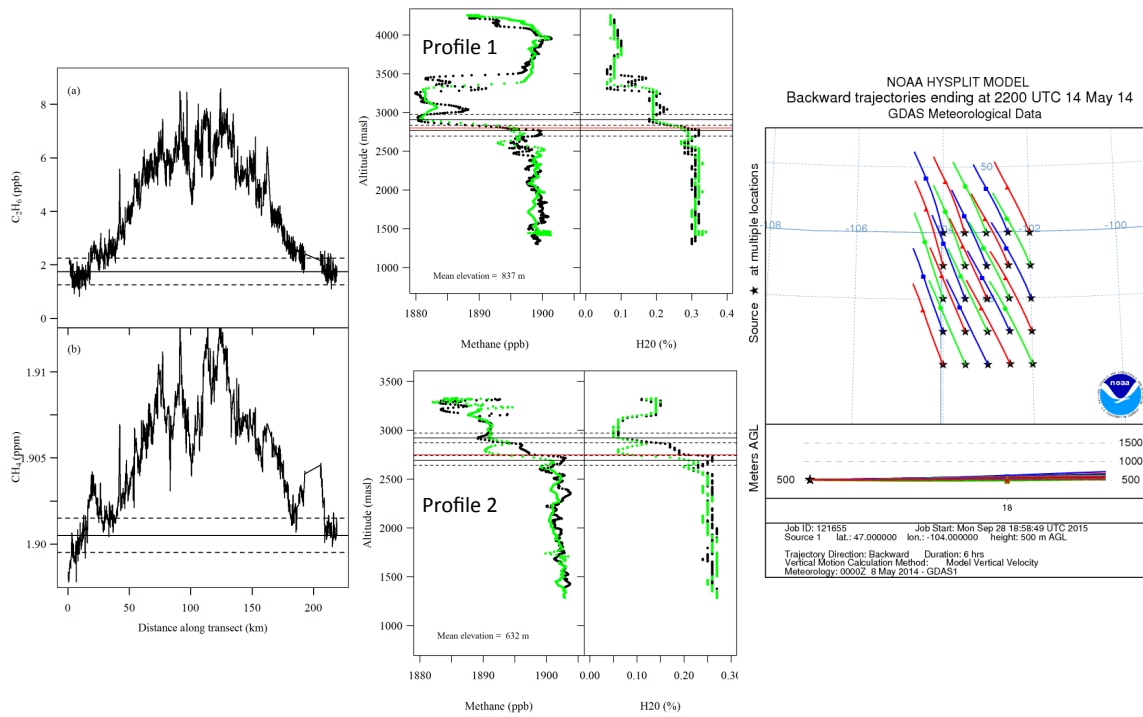


Figure S1. Downwind cross-sections, vertical profiles, and wind back trajectories for transect 2 on May 14, 2014. Illustrates the downwind enhancements (with background value and uncertainty illustrated), vertical profiles with Z_{pbl} and Z_e illustrated with uncertainty, and wind vectors from the Hysplit model [Draxler *et al.*, 2015] showing steady unidirectional winds.

Date/Transect	V (m/s) $\pm\sigma$	Wind (deg.) $\pm\sigma$	Transect Elevation (masl)	Z _{pbl} (magl)	Z _e (magl)	Z ₁ (magl) $\pm\sigma$	X _{C₂H₆} $\pm\sigma$ (ppb)	Flux _{C₂H₆} $\pm\sigma$ (Tg/yr)
20140513/1	13.4 \pm 1.8	313 \pm 9	1034	1840	2908	2107 \pm 1073	1.93 \pm 0.58	0.28 \pm 0.14
20140514/1	7.80 \pm 1.73	337 \pm 12	1052	1930	2070	1965 \pm 172	2.32 \pm 0.58	0.19 \pm 0.06
20140514/2	8.23 \pm 1.57	339 \pm 11	1328	1995	2180	2041 \pm 210	2.48 \pm 0.58	0.26 \pm 0.08
20140521/1	3.16 \pm 1.38	198 \pm 40	1175	1980	2285	2056 \pm 321	10.5 \pm 1.04	0.26 \pm 0.13
20140521/2	2.79 \pm 1.28	205 \pm 41	1704	2240	2270	2098 \pm 104	8.52 \pm 1.04	0.21 \pm 0.12
20140522/1	7.05 \pm 1.16	174 \pm 11	1138	1750	2290	1859 \pm 549	3.38 \pm 1.04	0.24 \pm 0.07
20140522/2	7.37 \pm 1.23	181 \pm 9	1709	1675	2610	1909 \pm 940	3.60 \pm 1.04	0.20 \pm 0.07

Table S1. Summary information for mass balance calculations.

	NO	CO	C ₂ H ₆	C ₃ H ₈	C ₄₊	Other NMVOCs
Global (Jan-Dec 2014)						
<i>Anthropogenic</i>	2100	22000	180	150	510	330
<i>Open Fires</i>	330	13000	80	20	14	410
<i>Biogenic</i>	520	1500				6900
<i>Lightning</i>	460					
Total	3400	36000	260	170	520	7700
North America (9.625°N-60.125°N; 130.156°W-60.156°W; Jul-Aug 2014)						
<i>Anthropogenic</i>	210	2900	49	16	150	43
<i>Open Fires</i>	45	1600	9.0	2.5	2.4	53
<i>Biogenic</i>	140	290				1300
<i>Lightning</i>	240					
Total	640	4852	58	18	153	1380
Bakken Region (46.7°N-48.8°N; 104.08°W-101.5°W; Jul-Aug 2014)						
<i>Anthropogenic</i>	0.71	2.4	10	7.3	5.3	0.19
Oil & Gas Production			10	7.3	5.2	
Other	0.71	2.4	0.02	0.010	0.090	0.19
<i>Open Fires</i>	<0.005	0.020	<0.005	<0.005	<0.005	<0.005
<i>Biogenic</i>	0.52	0.050				0.49
<i>Lightning</i>	0.73					
Total	2.7	4.9	20	15	11	0.87

Table S2. Emission rates of reactive nitrogen and carbon species used in the model simulations. All units in Gmol (species) yr⁻¹. Methane levels are prescribed in the model and is thus not included in the emissions model.

Movie S1. Change in surface ozone from fugitive Bakken shale alkane emissions.