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Article

Ethanol and Methanol in South Korea and China: Evidence for Large Anthropogenic Emissions Missing from Current Inventories

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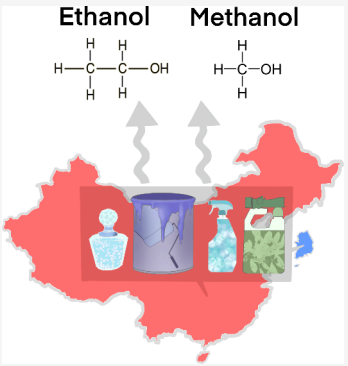
ABSTRACT: Observations during the KORUS-AQ, MAPS-Seoul, and APHH-Beijing field campaigns of 2015−2017 reveal high concentrations of ethanol and methanol in urban air over South Korea and China, with median concentrations of 2−4 ppb for ethanol and 12−18 ppb for methanol. Simulations with the GEOS-Chem model show that these values cannot be captured by current emission inventories. They could originate from volatile chemical products (VCPs). Fitting observed ethanol concentrations with GEOS-Chem would imply per capita VCP emissions 2.4 times higher in South Korea and 1.5 times higher in China than in the U.S. The strong ethanol−methanol correlation suggests a major methanol component in VCP emissions, unlike in the U.S. where methanol use is largely banned. Including these emissions in GEOS-Chem increases the level of surface ozone over South Korea and China by 1−3 ppb. KORUS-AQ aircraft profiles also indicate a high free tropospheric methanol background of 3.2 ppb, which appears to be of terrestrial biospheric origin but cannot be reproduced by GEOS-Chem.



KEYWORDS: *Volatile Chemical Products, Volatile Organic Compounds, Emissions, Environmental Modeling, East Asia Air Quality*

I. INTRODUCTION

Understanding the sources of anthropogenic nonmethane volatile organic compounds (NMVOCs) is important for air quality management, especially in East Asia where they are important sources of fine particulate matter[1] and where ozone production is often NMVOC-limited.[2] Recent surface and aircraft campaigns have shown high concentrations of ethanol and methanol including the Megacity Air Pollution Study (MAPS)- Seoul in May−June 2015,[3] the Korea-United States Air Quality (KORUS-AQ) study over Korea in May−June 2016,[4] and the Air Pollution and Human Health (APHH)-Beijing study in November-December 2016 and May−June 2017.[5] Here we interpret these observations of ethanol and methanol with the GEOS-Chem chemical transport model to reveal large unaccounted anthropogenic sources for both.

The dominant global sources of methanol and ethanol are from the terrestrial biosphere.[6−8] Ocean emissions,[9,10] biomass burning,[11] and secondary atmospheric production[12,13] are also important. Anthropogenic sources include solvent use, industrial processes, and fuel use.[14] Oxidation by the hydroxyl radical (OH) is the main sink with corresponding atmospheric lifetimes of 1−3 days for ethanol and 5−12 days for methanol.[15,16] Both species can serve as precursors for ozone,[4,17] while ethanol is also a precursor of acetaldehyde and from there peroxyacetyl nitrate (PAN), which contributes to the transport of ozone pollution.[18]

VCPs such as adhesives, coatings, printing inks, pesticides, cleaning agents, and personal care products are an increasingly important source of NMVOCs in many urban environments as emissions from combustion have decreased.[19−21] VCP emissions are generally missing or under-accounted in emission inventories, particularly from the residential sector.[19] Ethanol is the most abundant VCP emitted.[22,23] It is a common solvent used in personal care, cleaning, and other household products. Ethanol emissions have shifted from fuel use to VCPs in U.S. cities, with 70% of ethanol in New York City coming from VCPs.[23] Methanol is also present in VCPs as a component in many solvent mixtures, although regulations due to its toxicity minimize its use in the U.S.[24]

Several studies in East Asia have pointed out the importance of VCP emissions, including ethanol. One such NMVOC inventory[25] for the Pearl River Delta region includes a high contribution from VCP emissions. An alternative NMVOC emission inventory[26] for China based on product sales reports found a much larger contribution from residential sources compared to the widely used Multi-resolution Emission Inventory for China (MEIC). MEIC includes VCPs but focuses on industrial sources, with residential sources contributing less than 5% of total VCP emissions.[27] Ethanol was the top emitted species in 2017 from industrial and

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Table 1. Ethanol and Methanol Emissions (Tg a^{-1}) in South Korea and China[a]

country	South Korea			China		
emission scenario	baseline	+U.S.-based VCPs	adjusted	baseline	+U.S.-based VCPs	adjusted
			Ethanol			
total	0.029	0.12	0.24	1.3	4.5	6.0
anthropogenic	<0.01	0.089	0.22	0.14	3.3	4.9
terrestrial biogenic	0.019			1.1		
open fires	<0.01			<0.01		
			Methanol			
total	0.13	0.14	0.62	4.9	5.1	14.1
anthropogenic	<0.01	0.01	0.49	0.05	0.25	9.3
terrestrial biogenic	0.12			4.8		
open fires	<0.01			0.11		

[a]Baseline anthropogenic emissions for 2016 are from the KORUS-AQ v5 inventory for South Korea and from the MEIC inventory for China. Baseline + U.S.-based VCP anthropogenic emissions add 1.9 kg of ethanol and 0.12 kg of methanol per capita per year based on U.S. data (McDonald et al., 2018). Adjusted anthropogenic emissions fit the MAPS-Seoul, KORUS-AQ, and APHH-Beijing observations. Terrestrial biogenic emissions from MEGANv2 and open fire emissions from GFED4 are the same in all scenarios.

domestic solvent use in the product sales report based inventory.[26] Methanol was also included, but it had low emissions. An additional NMVOC emission inventory for China[28] based on product usage also found large ethanol emissions from cleaning products and printing inks, but they did not report methanol emissions. Modeling of KORUS-AQ observations[29] shows that the KORUSv5 inventory for South Korea greatly underestimated NMVOC emissions from VCPs, liquefied petroleum gas (LPG), and natural gas use.

Another source of methanol in China could be vehicle fuel. Methanol is used as a gasoline fuel oxygenate in China, and total methanol fuel use increased over 25 fold between 2000 and 2016.[30] MEIC includes methanol blended fuels in their residential heating and power usage emissions but not in their vehicle emissions.[27]

Here we use the GEOS-Chem model to show that current emission inventories cannot account for the high ethanol and methanol concentrations observed in the KORUS-AQ, MAPS-Seoul, and APHH-Beijing campaigns. From the observed correlations between these two species and with other species, we infer that the missing emissions are anthropogenic and from common sources, suggest that unaccounted VCPs could be important contributors, and revise emission estimates. We also examine the origin of the elevated background methanol in the free troposphere observed in KORUS-AQ.

II. DATA AND METHODS

1. Observations. The KORUS-AQ campaign included 20 daytime flights carried out from May 2 to June 10, 2016 over the Korean Peninsula and Yellow Sea.[31] Most flights included a low-altitude route with a missed approach over the Seoul Air Base, sampling near-surface air over Seoul. Ethanol and methanol concentrations were measured by proton transfer reaction time-of-flight mass spectrometry (PTR-ToF-MS), but the instrument was not calibrated to measure ethanol, resulting in at least ±50% uncertainty in the measurements.[32] Methanol data was of higher quality with an average uncertainty of 4.5% due to availability of a standard. Additional NMVOCs were measured using whole air samples analyzed by gas chromatography-mass spectroscopy (GC-MS).[4]

The MAPS-Seoul campaign included ethanol and methanol GC-MS measurements from whole air samples collected at a surface site on the grounds of the Korean Institute of Science and Technology (KIST) in Seoul (37.604 N, 127.045 E) from May 23 to June 10, 2015.[33] Samples were collected twice per day in the morning and afternoon, yielding 23 total measurements. Measurement uncertainty was 20−30%.

The APHH-Beijing campaign included measurements from a meteorological mast located at the Institute of Atmospheric Physics (IAP) in Beijing (39.976 N, 116.378 E) from November 12 to December 10, 2016 and May 15 to June 24, 2017.[34] Gas chromatography flame ionization detection (GC-FID) ethanol and methanol measurements were collected at 5 m altitude, and PTR-ToF-MS eddy covariance flux measurements were collected at 102 m altitude.[35] The measurements were calibrated using standards. Measurement precision was 58% for ethanol and 40% for methanol.

2. GEOS-Chem Simulation. We use GEOS-Chem v13.3.0 (https://zenodo.org/record/5703095#.YwIkMBDMJAc) in a nested-grid simulation of the KORUS-AQ period at a horizontal resolution of 0.5° x 0.625° over East Asia (15−59.5° N, 70−150° E).[36,37] GEOS-Chem has been used previously to simulate oxidant chemistry during KORUS-AQ.[38−42] Transport is driven by Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2) meteorological data assimilated by the NASA Global Modeling and Assimilation Office (GMAO). We generate boundary conditions updated every 3 h from a global simulation at a horizontal resolution of 4° x 5° from November 1, 2015, to June 24, 2016, with 6 months used for model spin-up. We run the nested simulations from April 1 to June 24, 2016, with the first month used for spin-up. An overestimate of OH in GEOS-Chem over Korea relative to KORUS-AQ observations was previously found that was corrected by increasing carbon monoxide (CO) boundary conditions by 50%.[43] We apply the same correction here.

Table 1 lists the ethanol and methanol emissions in the model for South Korea and China. Our baseline simulation uses MEIC anthropogenic emissions for China,[44] 2015 KORUSv5 anthropogenic emissions for South Korea and other Asian countries,[45] and CEDS anthropogenic emissions for the rest of the world.[46] Plant growth and decay emissions are from MEGANv2.[8] Open fire emissions are from GFED4.[47] Anthropogenic emissions of ethanol and methanol in the MEIC inventory are 0.081 and 0.024 kg per capita per year, respectively. They are negligibly small in the KORUSv5 inventory.

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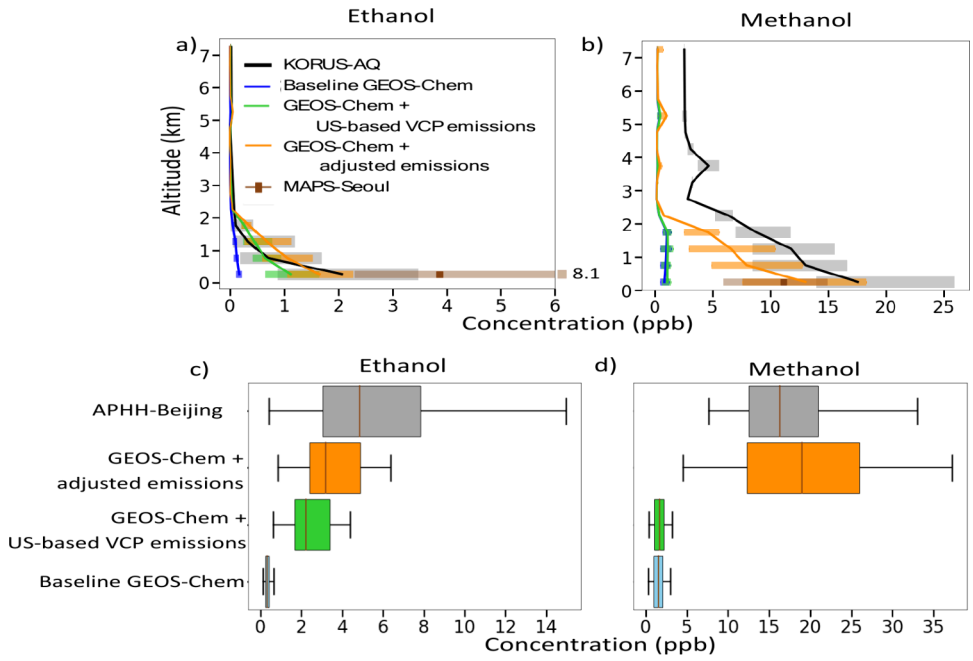

Figure 1. Ethanol and methanol concentrations over Seoul and Beijing. Observations are compared to GEOS-Chem simulations with baseline ethanol and methanol emissions from the standard model, added U.S.-based VCP emissions, and adjusted emissions to match the observations (Table 1). Panels a and b show median concentrations and interquartile ranges for vertical profiles over Seoul from the KORUS-AQ aircraft observations in May−June 2016 (126.7−127.3 E, 37.3−37.8 N) with the GEOS-Chem simulation sampled along the flight tracks. Also shown are surface observations from MAPS-Seoul (37.604 N, 127.045 E). The methanol vertical profiles for the baseline and U.S.-based VCP emissions overlap almost completely, as U.S. methanol VCP emissions are small. Panels c and d show concentration boxplots (medians, interquartile ranges, and full ranges) for APHH-Beijing tower observations (116.378 E, 39.976 N) in May−June 2017 compared to the GEOS-Chem simulations for that grid cell.

Ethanol and methanol emissions in this baseline simulation are dominated by biospheric sources (plant growth and decay). Anthropogenic emissions are small for ethanol and insignificant for methanol. The main sink for both ethanol and methanol in the GEOS-Chem simulation is oxidation by OH. GEOS-Chem also includes deposition sinks for both, but these are negligibly small because of limited solubility in water.

We show in Section III that this baseline simulation greatly underestimates the ethanol and methanol concentrations observed in KORUS-AQ, MAPS-Seoul, and APHH-Beijing due to missing anthropogenic sources. It was previously found more generally that there are large underestimates of VOC emissions, including from VCPs and fugitive gas emissions, in a GEOS-Chem simulation of the KORUS-AQ observations.[29] We conducted a sensitivity simulation adding global VCP emissions based on a speciated per capita U.S. inventory[19] with a corresponding diurnal profile.[23] These include ethanol and methanol as well as ethane, propane, toluene, xylene, C4+ alkanes, C3+ alkenes, limonene, formaldehyde, acetone, and methylethylketone. Ethanol emissions are 1.9 kg per capita per year, and methanol emissions are 0.12 kg per capita per year. An independent U.S. VCP inventory found similar emission factors of 1.7 kg per capita per year for ethanol and 0.18 kg per capita per year for methanol.[21] We applied the VCP per capita emissions to a 2015 global gridded population map.[48] This caused ethanol anthropogenic emissions in South Korea and China to increase to 0.089 and 3.3 Tg a^{-1}, respectively (Table 1), with VCPs now dominant over biospheric emissions. However, as shown below, this simulation still underestimates the observed ethanol concentrations. In addition, it does not significantly correct for modeled methanol even though methanol and ethanol are strongly correlated in the observations.

We conducted an additional simulation in which we adjusted the ethanol and methanol per capita emissions in South Korea and China to match the surface and aircraft observations over Seoul and Beijing. This hypothesizes per capita VCP emissions higher than in the U.S. or other missing emissions such as from cooking. Observations in the U.S. show large cooking emissions of both ethanol and methanol,[49−51] but these emissions are not included in the MEIC and KORUSv5 inventories. The adjusted simulation represents our best estimate of emissions, as given in Table 1. In this adjusted simulation, the ethanol emission factors per capita in South Korea and China are respectively 2.4 and 1.5 times higher than in the U.S. The methanol emission factors are 10 and 5.5 kg per capita per year in South Korea and China, respectively.

III. RESULTS AND DISCUSSION

Figure 1 shows model comparisons to measurements of ethanol and methanol concentrations over Seoul and Beijing during KORUS-AQ, MAPS-Seoul, and APHH-Beijing. The measurements feature very high concentrations near the surface, with median values for ethanol of 2 ppb (KORUS-AQ below 0.5 km altitude), 4 ppb (MAPS-Seoul), and 5 ppb (APPH-Beijing) and for methanol of 18 ppb (KORUS-AQ), 12 ppb (MAPS-Seoul), and 16 ppb (APHH-Beijing). For comparison, measurements in urban areas of U.S. and Europe show typical concentrations of 1−9 ppb for ethanol and 3−11 ppb for methanol.[52−55] Ethanol blended fuel, which contributes to urban ethanol concentrations in the U.S., was not used in South Korea at the time of the measurements.[40] The

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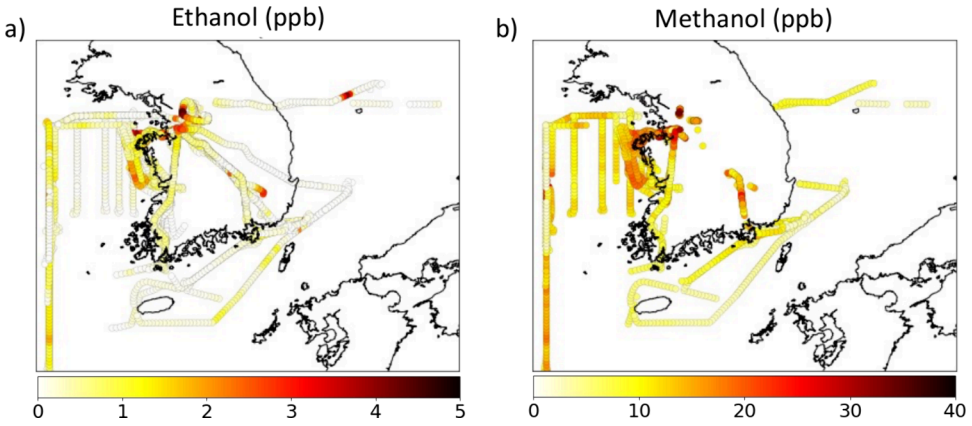

Figure 2. Ethanol and methanol concentrations below 0.5 km altitude during KORUS-AQ. Observations along the flight tracks are shown for the ensemble of May−June 2016 flights.

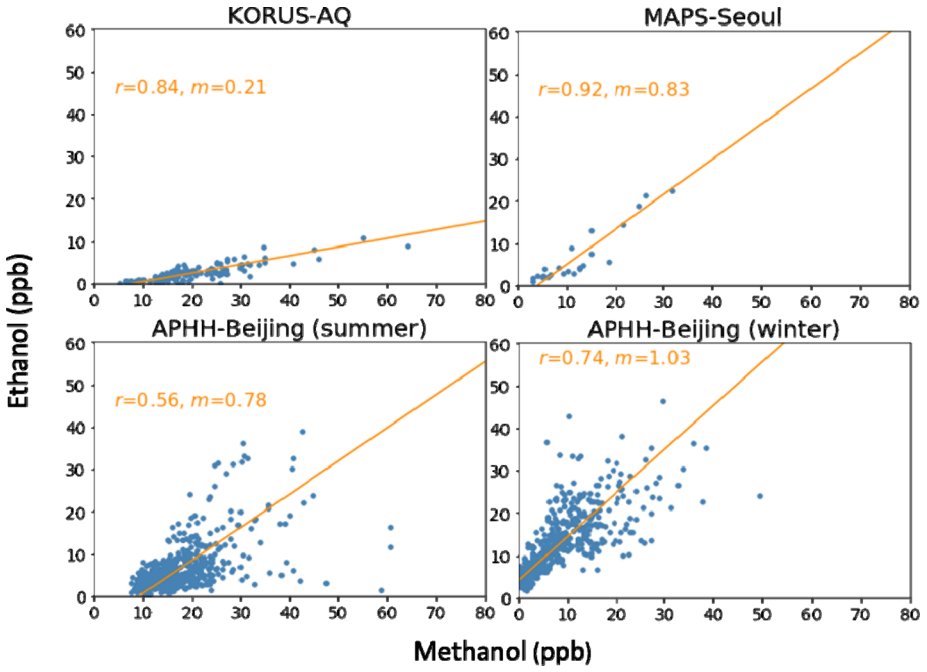

Figure 3. Relationship between observed methanol and ethanol concentrations over Seoul and Beijing. KORUS-AQ measurements over Seoul are for the 126.7−127.3 E, 37.3−37.8 N, 0−0.5 km altitude domain. Points represent 1 min averaged measurements for KORUS-AQ, twice daily measurements for MAPS-Seoul, and 10 min measurements for APHH-Beijing. Pearson correlation coefficients (r) and reduced-major-axis (RMA) regression lines (slopes m) are also shown.

KORUS-AQ vertical profiles show decreasing concentrations with altitude in the planetary boundary layer (PBL), consistent with the short model lifetimes against oxidation by OH in the region (0.44 days for ethanol and 1.4 days for methanol). Lifetimes are shorter than the global averages reported in the introduction due to high OH concentrations.[43] Methanol retains a high background of 3.2 ppb in the free troposphere above a 3 km altitude.

The baseline simulation underestimates the ethanol and methanol concentrations over Seoul by an order of magnitude. This cannot be simply attributed to the 50 km resolution of the model simulation because the lifetimes of ethanol and methanol are relatively long and the aircraft observations average over relatively long fetches. Primary NMVOCs originating from combustion showed much lower biases (30−50%) in the model relative to KORUS-AQ observations.[29] Using the CMAQ model results in a similar model underestimate in methanol during the KORUS-AQ period, pointing to a missing source in the emissions inventory.[56] Our simulation with adjusted emissions reproduces the observed variability of concentrations, further indicating that the 50 km resolution does not cause excessive smoothing. That simulation still cannot reproduce the observed free tropospheric background of methanol, which contributes to the PBL enhancement. We discuss the origin of this free tropospheric background later in the paper.

Figure 2 shows the spatial distributions of ethanol and methanol concentrations observed by KORUS-AQ above and around the Korean peninsula below 0.5 km altitude. The concentrations are highest for both regions over the Seoul Metropolitan Area, indicating a missing urban source. Figure S1 shows strong correlations with CO over Seoul for both ethanol (r = 0.84 in MAPS-Seoul) and methanol (r = 0.80). The ethanol/CO enhancement ratio is 53 ppt/ppb for MAPS-

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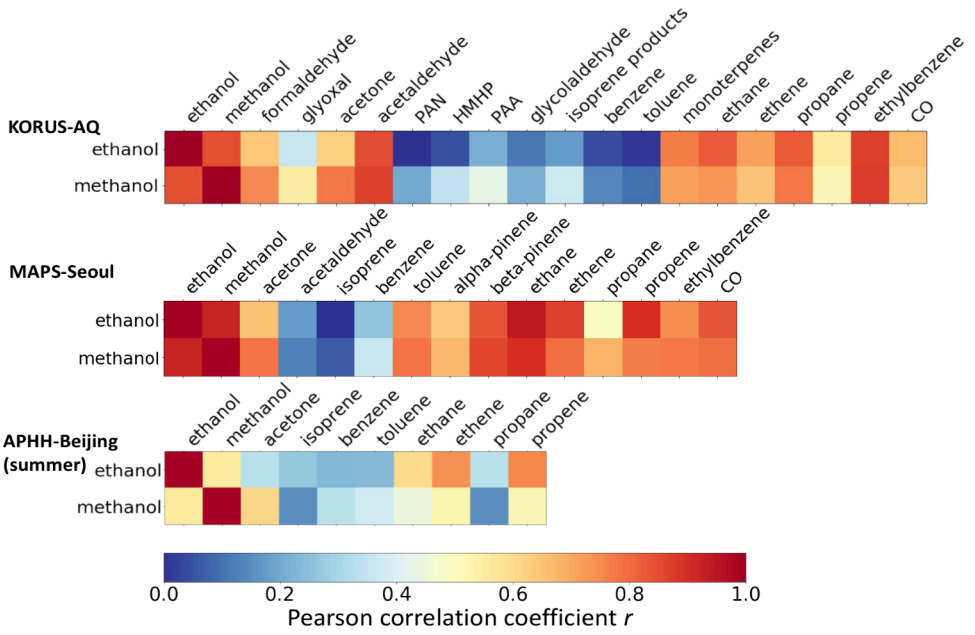

Figure 4. Correlations of ethanol, methanol, and other NMVOC concentrations over Seoul and Beijing. Pearson correlation coefficients are shown for individual observations as defined in Figure 2. KORUS-AQ measurements over Seoul are for the 126.7−127.3 E, 37.3−37.8 N, 0−0.5 km altitude domain. Only summer measurements are included for APHH-Beijing. Winter measurements show high correlations among all species that may reflect the longer lifetimes. Figure S1 shows scatterplots for correlations between selected species.

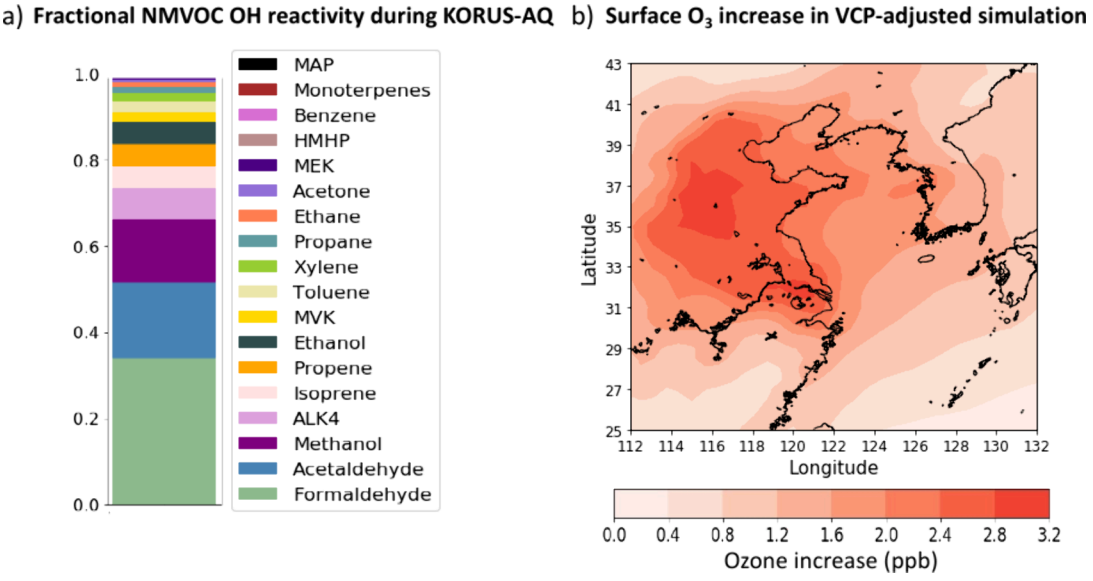

Figure 5. Effect of anthropogenic ethanol and methanol emissions on total NMVOC reactivity and surface ozone in GEOS-Chem. Panel a shows the fractional contributions of individual NMVOCs to total NMVOC reactivity in the adjusted simulation over the KORUS-AQ domain. Model reactivity was sampled and summed for the whole campaign along the NASA DC-8 flight tracks. The reactivity of a NMVOC species is defined as the product of its concentration by its local rate constant for reaction with OH. Panel b shows the GEOS-Chem increase in 24-h average surface ozone in the adjusted simulation relative to the baseline simulation for the May−June 2016 KORUS-AQ period.

Seoul, consistent with 46 ppt/ppb observed in Los Angeles (deGouw et al., 2018) and 54 ppt/ppb observed in New York City in the summer[23] (Coggon et al., 2021). The methanol/CO enhancement ratio for MAPS-Seoul is 64 pptv/ppbv, whereas 21 ppt/ppb was observed in Los Angeles with low correlation (r = 0.39) suggesting a biogenic source for the methanol.[57] A bottom-up VCP emission inventory for the U.S.[21] including both anthropogenic and biogenic sources finds ethanol/CO and methanol/CO emission ratios in Los Angeles of 35 and 16 ppt/ppb, respectively.

Figure 3 shows the correlations between the observed ethanol and methanol concentrations over Seoul and Beijing. The correlations are strong, indicating a commonality of sources. The correlation between ethanol and methanol persists in the winter Beijing measurements when there is no biogenic influence, implying that the missing sources must be anthropogenic. The ethanol/methanol enhancement ratios (slopes of the regression lines) are similar for MAPS-Seoul and APHH-Beijing, suggesting similar sources in both cities. The enhancement ratio is lower in KORUS-AQ, due to lower

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ethanol levels, which might reflect not only the sampling of more aged air in which some of the ethanol has been oxidized (Figure 1) but also possibly a low bias in the aircraft measurements. The summer Beijing measurements show a subpopulation with high methanol and low ethanol that might be due to vehicular emissions, as discussed below.

Figure 4 shows the correlation coefficients of ethanol and methanol with other NMVOC concentrations for different measurement campaigns. Scatterplots for strongly correlated species are shown in Figure S1. Ethanol and methanol are most strongly correlated to each other, further supporting the commonality of sources. They have low correlations with isoprene and its oxidation products, arguing against a biogenic source. The correlations with monoterpenes, in opposition to isoprene, may be explained by the use of monoterpenes in fragranced products.[19,49] The KORUS-AQ data show strong correlation with ethylbenzene, which originates from paint solvents.[4] The strong correlation with acetaldehyde may result from chemical production from ethanol.[29] The negative correlations with benzene argue against direct emission from vehicles being a major source of either ethanol or methanol.

The simulated ethanol concentrations in GEOS-Chem are substantially improved when we implement U.S.-based per capita VCP emission factors in the model, and they match the observations when we increase these emission factors by 50% in the adjusted scenario (Figure 1). However, the U.S.-based emission factors include very little methanol due to regulations against the use of methanol in commercial products. Apparently, these regulations have not been as strong in South Korea and China, although China introduced new standards in 2020 that limited the amount of methanol in certain products.[58]

There were strong correlations between methanol and NO_x vertical fluxes at the APHH-Beijing site that implied a vehicle source for methanol,[5] consistent with the use of methanol as fuel in China.[59] But the strong correlations between ethanol and methanol in the APHH-Beijing data and the similarity of ethanol/methanol enhancement ratios with Seoul (MAPS-Seoul) where methanol is not used in fuel argue more for a dominant VCP source for methanol in Beijing. The APHH-summer data in Figure 2 show a subpopulation with high methanol and low ethanol that might reflect vehicle emissions. More scatter in the summer compared to that in the winter may also reflect the influence of biogenic emissions and the shorter lifetimes.

Increasing ethanol and methanol emissions in the adjusted simulation to match the observations in South Korea and China has limited impacts on other aspects of the GEOS-Chem oxidant simulation, previously described for KORUS-AQ.[43] Total NMVOC reactivity with OH in KORUS-AQ increased by 14% relative to the baseline simulation. Figure 5 shows that formaldehyde and acetaldehyde are the principal contributors to NMVOC reactivity for the KORUS-AQ conditions, with methanol in the third position. Acetaldehyde and formaldehyde are produced from ethanol and methanol, but these contributions are minor. Surface ozone concentrations increase by 1−3 ppb in the adjusted simulation compared with the baseline, as shown in Figure 5.

Although implementing VCP emissions improves the model representation of near surface ethanol and methanol measurements, it does not address the elevated background concentration of methanol in the free troposphere. During KORUS-AQ, measured methanol above 3 km altitude averaged 3.2 ppb, with little dependence on altitude and little overall variability (Figure 1). As shown in Figure 1, this elevated background is not present for ethanol and does not respond significantly in the model to increases in emissions from South Korea or China. A 7 ppb methanol background is apparent in the APHH-Beijing data for summer but not for winter (Figure 2). Free tropospheric methanol during KORUS-AQ is most strongly correlated with acetone (Figure 6), and acetone is understood to be of dominant biogenic

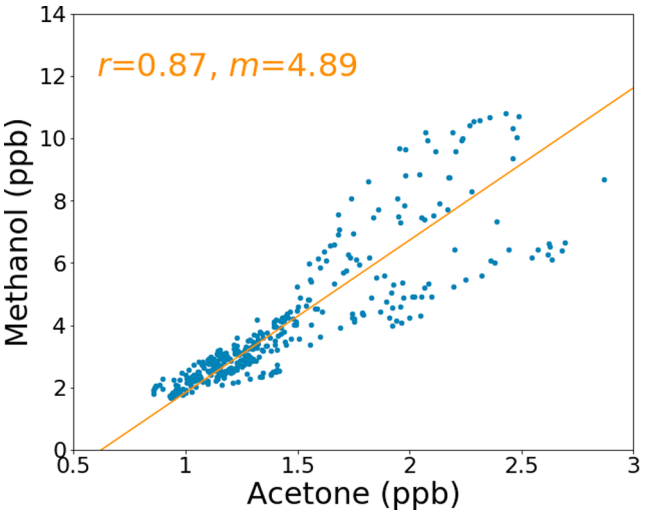

Figure 6. Correlation of methanol and acetone in the free troposphere over South Korea. Individual points represent 1 min averaged observations above 3 km altitude during the KORUS-AQ aircraft campaign (May−June 2016). Pearson correlation coefficient and the reduced-major-axis (RMA) regression line and slope (m) are also shown.

origin.[60] This suggests a missing biogenic source of methanol in the global GEOS-Chem simulation that serves as a boundary condition for our regional East Asia simulation. An older global GEOS-Chem simulation of methanol found that East Asia in summer had the highest free tropospheric (500 hPa) concentrations in the world, exceeding 5 ppb, a consequence of much higher biogenic sources than in the present model.[61] A model inversion of IASI methanol satellite data over the Eurasian continent found that large increases in biogenic methanol emissions over the Middle East and central Asia are needed.[62]

In summary, analysis of aircraft and ground-based observations of ethanol and methanol over Seoul and Beijing reveals a large anthropogenic source missing from the current emission inventories. Attributing this source to volatile chemical products (VCPs) would imply a population-based VCP emission factor for ethanol that is 2.4 times higher in South Korea and 1.5 times higher in China than in the U.S. Strong correlation with ethanol suggests that methanol has major VCP sources in South Korea and China, in contrast to the U.S. where its use in VCPs is largely banned. Including these emissions of ethanol and methanol in an atmospheric chemistry model simulation for East Asia results in ethanol and methanol becoming the seventh and third most important contributors to NMVOC reactivity and increases model surface ozone by 1−3 ppb. Methanol also shows an elevated free tropospheric background over South Korea that appears to

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be of terrestrial biospheric origin but is not captured by the model.

■ ASSOCIATED CONTENT

Data Availability Statement

KORUS-AQ data was obtained from https://www-air.larc.nasa.gov/cgi-bin/ArcView/korusaq. APHH-Beijing data was obtained from https://catalogue.ceda.ac.uk/uuid/77e44e0d5df14e89ab35c0af1f7cb726. The anthropogenic emission inventory for China is from www.meicmodel.org. The KORUSv5 emission inventory is developed by Konkuk University, available at http://aisl.konkuk.ac.kr/#/emission_data/korus-aq_emissions. MERRA-2 reanalysis data are from https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/data_access/.

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestair.4c00210.

Relationships between observed methanol, ethanol, and selected species concentrations during KORUS-AQ over Seoul (PDF)

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Notes

The authors declare no competing financial interest.

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