Anthropogenic Sources of Methanol and Ethanol in East Asia

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Introduction

High concentrations of methanol and ethanol have been measured in China and South Korea, greatly exceeding the concentrations found in U.S and Europe. Both species are underestimated by up to order of magnitude in atmospheric models, suggesting missing emission sources in standard inventories. Aircraft and surface observations point to Volatile Chemical Product (VCP) emissions serving an important role in the underestimate of these two species in East Asia. Emissions from blended fuel use in China also provide an additional source of methanol. We implement population-density scaled VCP emissions in GEOS-Chem to correct model underestimates.

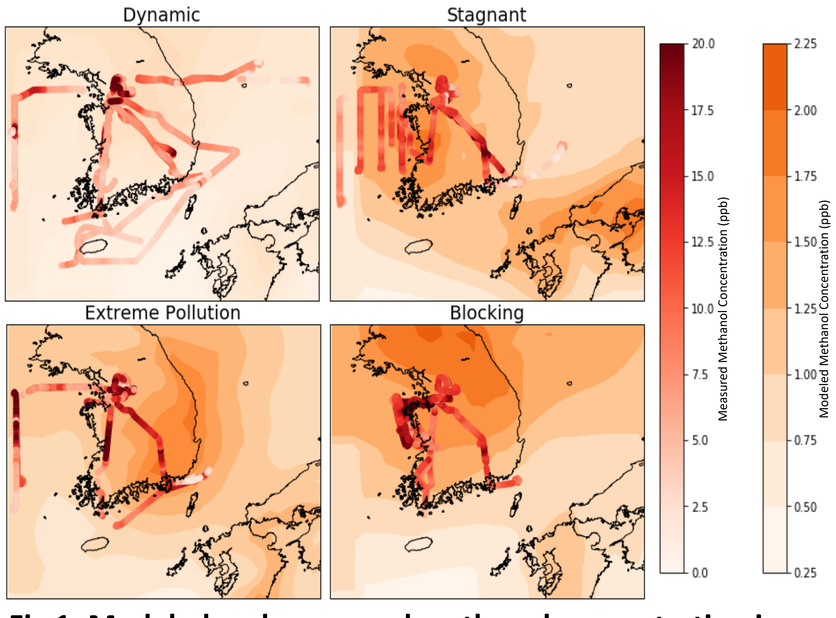

Fig 1: Modeled and measured methanol concentration in South Korea during KORUS-AQ GEOS-Chem methanol concentrations averaged over the PBL (lower 2 km) are in orange. Measured methanol is overlayed in red. Panels are split by meteorological period, where the dynamic and extreme pollution periods were characterized by favorable winds for pollution transport from China whereas stagnant and blocking periods were more influenced by local emissions. Methanol measurements during KORUS-AQ were led by Armin Wisthaler.

Analysis of Observations

Fig. 1 compares modeled and measured methanol during the KORUS-AQ campaign (May-June 2016). The spatial distribution of the model bias is strongest around major cities of Seoul and Daegu as well as over the Yellow Sea, suggesting missing methanol from anthropogenic sources in both Korea and China. Correlation with monoterpenes in Seoul indicate VCPs may be a missing source of methanol. This is supported by a strong correlation with ethanol during MAPS-Seoul (Fig. 2), which recent U.S studies show comes from personal care products. VCP, and especially personal care product emissions, are largely not included in the KORUSv5 and MEIC emission inventories used in this work.
KORUS-AQ correlations over the yellow sea suggest a vehicle source of methanol in China. Surface measurements in Beijing also show strong correlations between methanol and vehicle emission tracers, as well as a rush hour peak in its diurnal profile (Acton et al. 2020). Methanol blended fuel is widely used in China but not included in emission inventories, suggesting blended fuels are an important missing source of methanol.

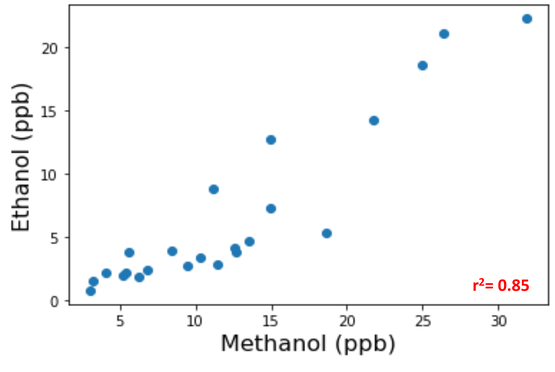

Fig 2: Methanol and Ethanol Concentrations during MAPS-Seoul. Whole air samples were collected on site at the Korean Institute of Science and Technology in May 2015 and analyzed using gas chromatography by Isobel Simpson.

Implementation of Volatile Chemical Product Emissions

Fig. 3 shows preliminary results of incorporating VCP emissions based on US emission factors as compared to KORUS-AQ measurements. Methanol and ethanol near-surface concentrations increase by 25% and 450% respectively. Acetaldehyde is shown as a high-yield product of ethanol, although primary emissions were also included in our VCP emissions inventory. Free troposphere ethanol does not increase after including VCP emissions, likely due to its short lifetime (~3 days). Underestimates of measured concentrations of all species remain large, indicating that U.S emission factors do not apply to East Asia.
Future work will involve scaling these emissions to match measurements in Seoul to obtain VCP emission factors applicable to East Asia. A source of methanol from blended fuel use will also be added to anthropogenic emissions in China by scaling up MEIC mobile emissions to match measurements in Beijing. This is expected to improve free troposphere bias.

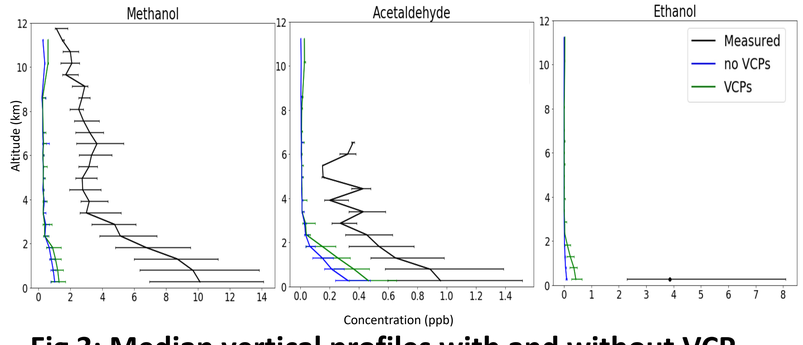

Fig 3: Median vertical profiles with and without VCP emissions as compared to measurements. Error bars show first and third quartiles. Modeled profiles use GEOS-Chem v13.1.0. The baseline no VCP simulation uses 4x5 resolution while the VCP simulation uses 0.5x0.625 resolution. KORUS-AQ measured vertical profiles are shown for methanol and acetaldehyde. Ethanol was not measured during KORUS-AQ so surface ethanol during MAPS-Seoul is shown instead.