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**Supplemental Material**

An investigation of petrochemical emissions during KORUS-AQ: Ozone Production, Reactive Nitrogen Evolution, and Aerosol Production.

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| **Table S1.** Aircraft Measurements aboard the NASA DC-8 research aircraft during the KORUS-AQ used in this study | Reference | Slusher et al. (2004) | Day et al. (2002) | Weinheimer et al. (1994) | Huey et al. (2004) | Müller et al. (2014) |
| Calibration Method | In-flight calibration by periodic addition of isotopically labeled PAN standard generated from a photolytic source described by Warneck and Zerbach (1992). | Kenagy et al. (2021) | In-flight calibration by periodic addition of NO standard (Scott-Marrin, 1.98 ppmv). In-flight determination of the photolytic conversion efficiency of NO2 to NO using NO2 formed from reaction between the standard NO and high concentration of O3. Periodic intercomparison of O3 instrument onboard the DC-8 with primary standard (Thermo Scientific Model 49i). | In-flight calibration by periodic addition of isotopically labeled SO2 standard. Laboratory calibration of the relative sensitivity of HCl to SO2 using the method described in Kim et al. (2007). | In-flight calibration by addition of VOCs standards dynamically diluted to 5 ppbv. The instrument sensitivity of acetone used as proxy for that of acrolein. The sensitivity of benzaldehyde estimated from ion-molecule reaction kinetics. |
| Uncertainty | 20-30% | 10% | 10-30% | 30-40% | 9-11% |
| Method | I- Chemical Ionization Mass Spectrometry (CIMS) | Thermal Dissociation Laser-Induced Fluorescence (TD-LIF) | Chemiluminescence | SF6- CIMS | Proton Transfer Reaction Time-of-Flight Mass Spectrometry (PTR-ToF-MS) |
| Measurement | PANs | PNs, ANs | NO, NO2, NOy, O3 | SO2 and HCl | VOCs, OVOCs |

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| **Table S1 (continued).** Aircraft Measurements aboard the NASA DC-8 research aircraft during the KORUS-AQ used in this study | Reference | Blake et al. (2003), Simpson et al. (2020) | Sachse et al. (1987,1991) | Vay et al. (1999) | Richter et al. (2015),  Fried et al. (2020)  fF | Crounse et al. (2006),  Teng et al. (2015) | Bahreini et al. (2009),  Nault et al. (2018) | Faloona et al. (2004) | Shetter and Muller, (1999) |
| Calibration Method | Simpson et al. (2020) | In-flight single point calibration periodically using gas standard by NOAA Earth System Research Laboratory and traceable to WMO scales (CO: x2014A; CH4: x2004A). | In-flight single point calibration periodically using gas standard by NOAA Earth System Research Laboratory and traceable to WMO scales (CO2: x2007). | Fried et al. (2020) | Laboratory characterization of HNO3 absolute sensitivities using HNO3 permeation tube. Laboratory calibration of HNs using the method described in Lee et al. (2014) and Teng et al. (2015). Laboratory sensitivities were proxied to flight sensitivities using in-flight standard addition for H2O2 and ethylene glycol. | Nault et al. ( 2018) | Brune et al. (2022) | The absolute spectral sensitivity determined with 1000 W NIST-traceable tungsten-halogen sources. |
| Uncertainty | 5% | 2% | 0.25 ppm | 6% | 30-50%  + 3-50 pptv | 35% (NO3-), 38% (Organic aerosol) | 35% | 12% (j-NO2), 20% (j-O3) |
| Method | Whole Air Sampling and Gas Chromatography (WAS and GC) | Mid-IR Wavelength Modulation Spectroscopy | Non-dispersive IR spectrometer | Compact Atmospheric Multi-Species Spectrometer (CAMS) | CF3O- CIMS | High-Resolution Time of Flight Aerosol Mass Spectrometer | Airborne Tropospheric Hydrogen Oxides Sensor Spectroscopy | Charged-coupled device Actinic Flux Spectroradiometers (CAFS) |
| Measurement | NMHCs | CO and CH4 | CO2 | HCHO | HNO3, hydroxy-nitrates (HNs) | Nitrate, Organic- Aerosol | OH, HO2 | Photolysis frequencies |

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| **Table S2.** Details of petrochemical plume transects used in this study during RF11 and RF18 | | | |
| Date (RF) | Plume Number | Local Time | Locations |
| 22 May (RF11) | 1 | 9:47–9:49 | Yellow Sea |
| 22 May (RF11) | 2 | 11:50–11:55 | Yellow Sea |
| 22 May (RF11) | 3 | 12:46–12:48 | Yellow Sea |
| 5 June (RF18) | Near-source | 10:49–10:58 | DPCC |
| 5 June (RF18) | Near-source | 14:27–14:33 | DPCC |
| 5 June (RF18) | 4 | 11:37–11:40 | Yellow Sea |

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| **Table S3.** The SOA yields of VOC precursors accounting for losses of semi-volatile gases to chamber walls adapted from Ma et al. (2017). | | | | | |
| Class | Compound | Stoichiometric SOA yield (c\* in μg m-3) | | | |
| 1 | 10 | 100 | 1000 |
| Alk5 | n-heptane, n-octane, n-nonane,  n-decane, cyclohexane | 0.157 | 0.000 | 0.000 | 0.000 |
| Ole1 | propene, 1-butene, i-butene,  cis-butene, trans-butene | 0.014 | 0.000 | 0.098 | 0.088 |
| Ole2 | styrene, 1,3-butadiene | 0.052 | 0.000 | 0.183 | 0.157 |
| Aro1 | benzene, toluene, ethylbenzene, i-propylbenzene, n-propylbenzene | 0.276 | 0.002 | 0.431 | 0.202 |
| Aro2 | m+p-xylene, o-xylene, ethyltoluenes, trimethylbenzenes | 0.310 | 0.000 | 0.420 | 0.209 |
| Isop | isoprene | 0.034 | 0.000 | 0.005 | 0.000 |
| Terp | alpha-pinene, beta-pinene | 0.210 | 0.000 | 0.348 | 0.297 |

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| **Table S4.** Model constraints used in 0-D box model simulations. | | | | | | | | | | |
| Meteorology |  | Model Inputa |  | Compound |  | Model Initial Constraint (ppbv) |  | Background  (ppbv) |  | Near  Source (ppbv)b |
| Pressure (mbar) |  | 995 |  | **Inorganics** |  |  |  |  |  |  |
| Temperature (K) |  | 295 |  | O3 |  | 70 |  | 96 |  | 70 |
| RH (%) |  | 27.4 |  | NO |  | 10 |  | 0.61 |  | 10 |
| j-NO2 (s-1) |  | 9×10-3 |  | NO2 |  | 20 |  | 2.45 |  | 20 |
| j-O3 (s-1) |  | 3.2×10-5 |  | PAN |  | 0.49 |  | 0.49 |  | 1.4 |
|  |  |  |  | PPN |  | 0.04 |  | 0.04 |  | 0.09 |
|  |  |  |  | **Alkanes** |  |  |  |  |  |  |
|  |  |  |  | Ethane |  | 4.24 |  | - |  | 5.2 |
|  |  |  |  | Propane |  | 3.16 |  | - |  | 5.7 |
|  |  |  |  | n-Butane |  | 4.73 |  | - |  | 4.1 |
|  |  |  |  | i-Butane |  | 6.85 |  | - |  | 4 |
|  |  |  |  | n-Pentane |  | 2.3 |  | - |  | 1.8 |
|  |  |  |  | i-Pentane |  | 3.74 |  | - |  | 2.7 |
|  |  |  |  | n-Hexane |  | 6.67 |  | - |  | 6.8 |
|  |  |  |  | **Alkenes** |  |  |  |  |  |  |
|  |  |  |  | Ethene |  | 22.3 |  | - |  | 21.4 |
|  |  |  |  | Propene |  | 6.76 |  | - |  | 5.9 |
|  |  |  |  | Butene |  | 1.73 |  | - |  | 0.8 |
|  |  |  |  | trans-2-Butene |  | 0.14 |  | - |  | 0.2 |
|  |  |  |  | cis-2-Butene |  | 0.25 |  | - |  | 0.3 |
|  |  |  |  | iso-Butene |  | 1.15 |  | - |  | - |
|  |  |  |  | 1,3-Butadiene |  | 5.4 |  | - |  | 2.5 |
|  |  |  |  | Isoprene |  | 0.39 |  | - |  | 0.2 |
|  |  |  |  | **Aromatics** |  |  |  |  |  |  |
|  |  |  |  | Benzene |  | 2.82 |  | - |  | 3.9 |
|  |  |  |  | Toluene |  | 0.92 |  | - |  | 2.6 |
|  |  |  |  | Ethylbenzene |  | 0.34 |  | - |  | 0.6 |
|  |  |  |  | o-Xylene |  | 0.15 |  | - |  | 0.2 |
|  |  |  |  | m,p-Xylene |  | 1.08 |  | - |  | 0.7 |
|  |  |  |  | Styrene |  | 1.62 |  | - |  | 0.6 |
|  |  |  |  | **Oxygenates** |  |  |  |  |  |  |
|  |  |  |  | HCHO |  | 1.9 |  | 1.9 |  | 8.5 |
|  |  |  |  | **Others** |  |  |  |  |  |  |
|  |  |  |  | C2H2 |  | 1.55 |  | - |  | 1.4 |
|  |  |  |  | CO |  | 340 |  | 245 |  | 340 |
|  |  |  |  | CO2 |  | 4.47x105 |  | 4.15 x105 |  | 4.47 x105 |
|  |  |  |  | CH4 |  | 2028 |  | 1933 |  | 2179 |
| aMedian of measurements made within P1-P3 of RF11. bTransect average of near-source plume transects during RF18. | | | | | | | | | | |

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| **Table S5.** Summary of PANs mixing ratios measured by TD-CIMS instrument at altitudes <500 m. | | | | | | | | | | | |
|  | Mixing ratioa  (All) | | |  | Mixing ratiob  (DPCC+Yellow Sea) | | |  | Mixing ratioc  (Seoul) | | |
| Avg (1) | Med | Max |  | Avg (1) | Med | Max |  | Avg () | Med | Max |
| PAN | 8.4102 (6.1) | 6.6102 | 6.0103 |  | 1.4103 (0.7) | 1.3103 | 6.0103 |  | 9.5102 (7.0) | 8.0102 | 4.5103 |
| PPN | 62 (49) | 48 | 6.4102 |  | 1.1102 (0.62) | 96 | 6.4102 |  | 64 (50) | 54 | 3.4102 |
| APAN | 8.7 (20) | 4.5 | 8.5102 |  | 18 (38) | 11 | 8.5102 |  | 9 (7.8) | 7.2 | 61 |
| (a-c) All mixing ratios in pptv. The statistics are produced using the data points filtered by boundaries in Figure S3. | | | | | | | | | | | |

Diagram

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**Figure S1.** Time series of related species and relative abundance of alkenes to ethyne. (a) KORUS-AQ Research Flight (RF) 11 on 22 May 2016 and (b) RF18 on 5 June 2016.

**Estimation of OH exposures**

OH exposure, defined as the product of OH concentration and time averaged over the photochemical lifetime of an air mass, can represent a qualitative measure of photochemical processing in air masses (McKeen and Liu, 1993; Parrish et al., 2007). We estimate OH exposure based on the WAS-averaged (i.e., averaged over the WAS sampling interval) concentration ratios of hydrocarbon pairs (SE1) (de Gouw et al., 2005) and alkene to hydroxy nitrate in parent and daughter relationship (SE2) (Bertman et al., 1995). Emission ratios of ethyne and alkene (ERethyne/alkene) in SE1 are derived from the average instantaneous DPCC emissions flux of ethyne to that of alkenes in Fried et al. (2020). Hydroxy nitrate branching ratios of alkenes ( in SE2 are from Teng et al. (2015).

(SE1)

(SE2)

Figure S2 illustrates the OH exposures estimated using equation SE1 and SE2 for P1-P3 along with the observed NOx to NOy ratios. In addition, the OH exposure of 2 x 1010 molecules cm-3 s based on the transect average OH concentration from the ATHOS measurements and plume transport time of ~2 hours at P1 was within the range of the OH exposures based on the concentration ratios.

Chart, scatter chart

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Figure S2. Estimates of the WAS-averaged OH exposures for P1, P2 and P3 (left axis). The transect averaged NOx to NOy ratios with vertical error bar indicating the standard deviation (right axis). The bottom and top axes illustrate distance from the DPCC and plume age based on 4 ms-1 wind speed.

Graphical user interface, chart

Description automatically generated with medium confidenceFigure S3. (a) Flight path of the DC-8 color-coded by the boundaries of Seoul (blue) and DPCC/Yellow Sea (green), which are used to filter data and produce statistics in Table S5. (b) The DC-8 data of PAN (red line), PPN (blue line), APAN (green line) and HCHO (cyan line) are averaged in 1-degree longitude bins and normalized to their peak value. The yellow marker indicates the location of the Seoul Metropolitan Area.

Diagram

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Figure S4. Enhancements of PAN, APAN and butadiene-hydroxy nitrate (HN) over the South Sea. The DC-8 flight tracks are color-coded by the mixing ratios of APAN. The inset shows time series of PAN (red line), APAN (green line with circle marker) and butadiene-HN (solid cyan lines) along with latitudes (dashed line).

Chart, pie chart

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Figure S5. Fractional contributions of VOC class to P(O3) for the plume transect P1, P3 and P4.

Chart, histogram

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Figure S6. Histograms of instantaneous ozone production (P(O3)) and ozone production efficiency (OPE) for all measurements when the WAS collected samples at altitude <500 m during RF11 and RF18. The calculated P(O3) and OPE are based on the 1s-merged data and averaged to the collection interval of the WAS. P(O3) and OPE for the DPCC and Yellow Sea (blue bars) include all measurements over the DPCC and Yellow Sea (i.e., both petrochemical and non-petrochemical samples).

Chart, bar chart

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Figure S7. (a) VOC-OH reactivity and (b) effective SOA yield of the speciated VOCs. The inset in panel b shows OA mass concentrations measured by AMS within petrochemical plume transects.

Chart

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Figure S8. Correlation plots of (a) HCHO, (b) APAN, (c) butadiene-HN, (d) PAN, (e) dihydroxytoluene, and (f) benzaldehyde versus OA, where r2 values are included in the upper right corner of each panel. Gray markers and solid lines in (a-f) correspond to the measurements within P1 and linear regression fit to the data, respectively.

Chart, scatter chart

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Figure S9. Comparison of SOA mass yield as a function of organic mass concentrations (Mo) from Ma et al. (2017) and Wood et al. (2010) to the empirical SOA yield from Ge et al. (2017) and Sato et al. (2011) for (a) propene and (b) 1,3-butadiene, respectively.

Chart, histogram

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Figure S10. The model simulated results of (a) 1,3-butadiene and (b) acrolein over 7-hour model integration time. (a) The solid blue line, cyan dashed line, and black dashed line correspond to the base case, slower dilution case and faster dilution case for the modelled 1,3-butadiene, respectively. The magenta dashed line shows model sensitivity test with an additional first-order loss given to OH to reproduce the observed OH level (2.7×106 molecules cm-3) at P1. Note that the better agreement between the model and the observations in the sensitivity test (magenta dashed line) indicates the impact of higher modelled OH in the base case on the fast decay of the modelled 1,3-butadiene. (b) The solid green line, lighter green dashed line, and darker green dashed line correspond to the base case, slower dilution case and faster dilution case for the modelled acrolein, respectively. The open markers in both (a) and (b) represent the averaged mixing ratios observed within the DPCC plume transects (P1-P3). Notably, the model agreed better with acrolein with 1,3-butadiene even with potential positive interference in acrolein signal detected by PTR-MS.

Graphical user interface

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Figure S11. Zero-out model simulations by which the initial constraints of speciated and class of VOCs are set to zero. Model output of (a) PAN and (b) PPN from the base simulation (blue) and zero-out simulations without the initial constraints of alkanes (red line), alkenes (green) and orange (aromatics). Model output of (c) APAN, (d) PBzN, and (e) PHAN from base case (blue solid line) without their hydrocarbon precursors (dashed lines).

Chart

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Figure S12. (a) Mixing ratios of acetaldehyde, acrolein and APAN observed over the Yellow Sea during RF11. Note that the mixing ratios of acrolein are upper limits due to potential interference on acrolein signal at 57 m/z detected by PTR-MS. (b) Mixing ratios of glycolaldehyde and benzaldehyde observed over the Yellow Sea during RF11. Gray shaded areas in panel (a) and (b) indicate the DPCC plume transects. (c) The simulated glycolaldehyde over 7-hour model integration time. The open circles represent the peak values of glycolaldehyde mixing ratios within the DPCC plumes transects (P1-P3). (d) The simulated benzaldehyde over 7-hour model integration time. The open circles represent the averaged mixing ratios of benzaldehyde within the DPCC plume transects (P1-P3). The solid blue lines, cyan dashed lines, black dashed lines, and vertical error bars in panel (c) and (d) correspond to the base case, slower dilution case, faster dilution case, and measurement uncertainties, respectively.

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