Development of furan oxidation mechanism from OH and NO$_3$ oxidation within biomass-burning regimes via chamber experiments

AER:
Benjamin Brown-Steiner
Matthew Alvarado

Georgia Tech:
Nga Lee (Sally) Ng;
Taekyu Joo
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Teakyu Joo
Furans

- Smell your coffee.
  - Furan exposure!
  - Darker roasts = more furans
    “roasted coffee”: furan-2-ylmethanethil

- Furans emitted directly (biomass burning) or form from degradation of VOCs
  - Rapidly oxidize to form furan derivatives (lifetime ~ hours)
  - Collectively, these aren’t good to breathe
  - Are known to produce SOA, which aren’t good to breathe

- Some measurements and laboratory studies exist, but much is still uncertain, unknown, and unconstrained
  - Branching ratios, SOA formation potential, etc.

- Most mechanisms include furans as lumped form (e.g. MCM)

Schoenauer and Schieberle (2018)
Furans – Quick Review

Hatch et al. (2015)

Colmenar et al. (2012)

Furan
2-methylfuran
2-furanaldehyde
3-methylfuran
3-furanaldehyde
2,5-dimethylfuran
5-methyl-2-furanaldehyde
Table 3. Calculated SOA mass (as g kg⁻¹ fuel burned) produced from the measured precursors in each chemical class after 6 h of oxidation (at [OH] = 2 × 10⁶ molecules cm⁻³) using representative rate constants. The values in parentheses reflect the estimated SOA mass assuming 100% reaction of all compounds in each class.

<table>
<thead>
<tr>
<th>Category</th>
<th>Black spruce</th>
<th>Pond. pine</th>
<th>Cutgrass</th>
<th>Wiregrass</th>
<th>Rice straw</th>
<th>Ind. peat</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aromatic HCs⁹</td>
<td>0.18 (0.63)</td>
<td>0.21 (0.77)</td>
<td>0.08 (0.22)</td>
<td>0.01 (0.04)</td>
<td>0.05 (0.18)</td>
<td>0.31 (1.19)</td>
</tr>
<tr>
<td>Phenols</td>
<td>0.03 (0.04)</td>
<td>0.11 (0.16)</td>
<td>0.03 (0.04)</td>
<td>0.02 (0.02)</td>
<td>0.06 (0.09)</td>
<td>0.11 (0.16)</td>
</tr>
<tr>
<td>Aliphatic HCs⁹</td>
<td>0.05 (0.05)</td>
<td>0.09 (0.11)</td>
<td>0.001 (0.001)</td>
<td>0.00 (0.00)</td>
<td>0.01 (0.01)</td>
<td>0.47 (0.61)</td>
</tr>
<tr>
<td>Oxy. aliphatics</td>
<td>0.02 (0.06)</td>
<td>0.04 (0.11)</td>
<td>0.003 (0.009)</td>
<td>0.01 (0.02)</td>
<td>0.02 (0.05)</td>
<td>0.03 (0.08)</td>
</tr>
<tr>
<td>Terpenes</td>
<td>0.47 (0.51)</td>
<td>0.42 (0.43)</td>
<td>0.001 (0.001)</td>
<td>0.001 (0.001)</td>
<td>0.01 (0.01)</td>
<td>0.009 (0.009)</td>
</tr>
<tr>
<td>Furans</td>
<td>0.07 (0.08)</td>
<td>0.13 (0.14)</td>
<td>0.01 (0.01)</td>
<td>0.04 (0.04)</td>
<td>0.06 (0.06)</td>
<td>0.10 (0.11)</td>
</tr>
<tr>
<td>Total potential SOA</td>
<td>0.81 (1.37)</td>
<td>0.99 (1.70)</td>
<td>0.12 (0.29)</td>
<td>0.08 (0.12)</td>
<td>0.21 (0.40)</td>
<td>1.01 (2.15)</td>
</tr>
<tr>
<td>Typical POA EF⁹</td>
<td>9.92</td>
<td>28.16</td>
<td>4.16</td>
<td>5.6</td>
<td>9.92</td>
<td>9.92</td>
</tr>
<tr>
<td>OA enhancement ratio</td>
<td>1.08 (1.14)</td>
<td>1.04 (1.06)</td>
<td>1.03 (1.07)</td>
<td>1.01 (1.02)</td>
<td>1.02 (1.04)</td>
<td>1.10 (1.22)</td>
</tr>
</tbody>
</table>

10% - 15% of Black Spruce and Ponderosa Pine
Up to 30% - 50% of total SOA in other fuels
### Existing Studies Examining Furan Oxidation

<table>
<thead>
<tr>
<th>year</th>
<th>citation</th>
<th>furan</th>
<th>oxidant</th>
<th>objective</th>
</tr>
</thead>
<tbody>
<tr>
<td>1995</td>
<td>Bierback et al.</td>
<td>F, 2MF, FFs</td>
<td>OH</td>
<td>mechanism</td>
</tr>
<tr>
<td>2011</td>
<td>Tapia et al.</td>
<td>3MF</td>
<td>NO₃, OH, Cl</td>
<td>mechanisms</td>
</tr>
<tr>
<td>2012</td>
<td>Colmenar et al.</td>
<td>FFs</td>
<td>NO₃</td>
<td>mechanism</td>
</tr>
<tr>
<td>2013</td>
<td>Aschmann et al.</td>
<td>F, MF, DMF</td>
<td>OH, NO</td>
<td>mechanism</td>
</tr>
<tr>
<td>2013</td>
<td>Strollo and Ziemann</td>
<td>3MF</td>
<td>OH, NO</td>
<td>SOA formation</td>
</tr>
<tr>
<td>2013</td>
<td>Liljegren and Stevnes</td>
<td>3MF</td>
<td>OH</td>
<td>low-pressure</td>
</tr>
<tr>
<td>2013</td>
<td>Aschmann et al.</td>
<td>F, MFs, DMFs</td>
<td>OH</td>
<td>mechanism</td>
</tr>
<tr>
<td>2014</td>
<td>Alwe et al.</td>
<td>DHFs</td>
<td>O₃, OH, Cl</td>
<td>mechanism</td>
</tr>
<tr>
<td>2015</td>
<td>Hatch et al.</td>
<td>F</td>
<td></td>
<td>biomass burning</td>
</tr>
<tr>
<td>2016</td>
<td>Müller et al.</td>
<td>F</td>
<td></td>
<td>biomass burning</td>
</tr>
<tr>
<td>2017</td>
<td>Hatch et al.</td>
<td>F</td>
<td></td>
<td>biomass burning</td>
</tr>
<tr>
<td>2017</td>
<td>Zhao and Wang</td>
<td>FFs</td>
<td>OH</td>
<td>mechanism</td>
</tr>
<tr>
<td>2017</td>
<td>Yuan et al.</td>
<td>F, MFs, DMFs</td>
<td>OH</td>
<td>mechanism</td>
</tr>
<tr>
<td>2018</td>
<td>Li et al.</td>
<td>F, MFs, DMFs</td>
<td>O₃</td>
<td>mechanism</td>
</tr>
</tbody>
</table>
Furan chemistry is complicated

Bierbach et al. (1995)

Zhao and Wang (2017)
Furan chemistry is complicated

Yuan et al. (2017)
Furan chemistry is complicated

So there is a clear need to organize, simplify, and constrain furan chemistry if we are going to be able to simulate it.
# Furan and Furan Derivative Oxidation

*I. Colmenar et al. / Atmospheric Environment 54 (2012) 177–184*

## Table 2
Comparison of the literature rate coefficients for the reactions of NO$_3$, OH and Cl atoms with various furan derivatives at 300 K.

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<tr>
<th>Compounds</th>
<th>Structure</th>
<th>$k \times 10^{12}$/cm$^3$ molecule$^{-1}$ s$^{-1}$</th>
<th>NO$_3$</th>
<th>OH</th>
<th>Cl</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Furan</td>
<td><img src="image" alt="Furan Structure" /></td>
<td>1.4 ± 0.2$^a$</td>
<td>1.3 ± 0.2$^b$</td>
<td>0.99 ± 0.06$^c$</td>
<td>40.1 ± 3$^g$</td>
</tr>
<tr>
<td>2-Methylfuran</td>
<td><img src="image" alt="2-Methylfuran Structure" /></td>
<td>25.7 ± 1.7$^c$</td>
<td>61.9 ± 3$^h$</td>
<td>73.1 ± 3.5$^m$</td>
<td>410 ± 30$^k$</td>
</tr>
<tr>
<td>2-Furanaldehyde</td>
<td><img src="image" alt="2-Furanaldehyde Structure" /></td>
<td>1.20 ± 0.28$^d$</td>
<td>35.1 ± 1$^i$</td>
<td></td>
<td>261 ± 27$^l$</td>
</tr>
<tr>
<td>3-Methylfuran</td>
<td><img src="image" alt="3-Methylfuran Structure" /></td>
<td>28.6 ± 0.6$^c$</td>
<td>93.5 ± 18$^j$</td>
<td>113 ± 22$^e$</td>
<td>87.3 ± 1.8$^m$</td>
</tr>
</tbody>
</table>
Chamber Studies

• This project:
  – Identify organic compounds produced from OH and NO$_3$ oxidation of furan compounds and their SOA production potential
  – Under different:
    • VOC:NO$_x$ ratios
    • RH
    • T conditions (chamber)
  – Develop a mechanism for biomass burning plumes (ASP)
  – Quantify O$_3$ and PM$_{2.5}$ (SOA) impacts
Georgia Tech Environmental Chamber Facility

Gas-phase measurements
- Gas Chromatograph Flame Ionization Detector (GC-FID): VOC measurements
- High Resolution Time of Flight Chemical Ionization Mass Spectrometer (HR-ToF-CIMS): oxidized gas-phase compounds, radicals
- UV absorption O₃ analyzer
- Chemiluminescence NO/NO₂/NOₓ analyzer
- CAPS NO₂ monitor

Particle-phase measurements
- High Resolution Time of Flight Aerosol Mass Spectrometer (HR-ToF-AMS): Aerosol composition (organics, sulfate, nitrate, ammonium, chloride), mass loading, aerosol size distribution
- Filter Inlet for Gases and AERosols High Resolution Time-of-Flight Chemical Ionization Mass Spectrometer (FIGAERO-HR-ToF-CIMS): molecular level particle-phase composition
- Scanning Mobility Particle Sizer (SMPS): aerosol size distribution, volume distribution
- Condensation Particle Counter (CPC): aerosol number concentration
- Offline filter characterization
Secondary Organic Aerosol Formation and the Oxidation Mechanism of Methylfurran by Nitrate Radical Oxidation

Taekyu Joo, Jean C. Rivera-Roisín, Fobang Liu, Masayuki Takahashi, Matthew J. Alvarado, Nga Lee (Early) Ng

Introduction
- Methylfuran (MF) is abundant during biomass burning.
- One of the minor species from biomass burning (Bruns et al., 2010; Stockwell et al., 2011; Heaton et al., 2017; Takahashi et al., 2018)
- Methylfuran accounts for 23-32% of all furan derivatives, and 1% of total measured VOCs from biomass burning (Budisulistiorini et al., 2014; Gilman et al., 2018)
- One of the most reactive species in biomass burning flames (Hummelzее et al., 2018)

SOA formation yield & growth behavior
- SSA yields (Zimmer et al., 2006)
  - NO, NO2, HNO3, NO3
  - Mass efficiency: 70% for SSA yield in higher NO2 concentrations.
  - NO3 reacts more efficiently in higher NO2 concentrations.
  - NO3 reacts more efficiently in higher NO2 concentrations.

Particle-phase composition
- NO3 (0.1 M) reaction with MF.
- NO3 radicals can react with MF to form SOA.
- NO3 radicals can react with MF to form SOA.

High-resolution aerosol mass spectra
- MF (100 ng/L)
  - Mass spectra of the secondary aerosols (SSA) formation

Gas-phase composition & proposed oxidation pathway
- MF + NO3 experiments
  - Products observed: nitrate esters, formic acid, oxalic acid.
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Summary & Conclusion
- MF + NO3 aerosol yield is lower compared to BrC (BrC = bisnorcamphor or mononorcamphor in NOx + SOA experiments).
- Detection of a large range of bisnorcamphor species in the gas phase and particulate phase.
- Oxidation products all methylfuran metabolites and NOx radical products for SSA formation. Oxidation products in the gas phase would have functional groups that can undergo partitioning.

Acknowledgement
- This work was supported by NSF (1839727) and NIH (1R43GM128518).
### ASP (Aerosol Simulation Program)

#### Table 2. Lumped Chemical Mechanism for Furan Used in This Work

<table>
<thead>
<tr>
<th>Reaction</th>
<th>$A$</th>
<th>$n$</th>
<th>$E_a (K)$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>FURAN + OH → HO₂ + RP20</td>
<td>6.2e-11</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>RP20 + OH → 0.53 RO₂59 + 0.47 RO₂60 + RO₂T</td>
<td>5.6e-11</td>
<td>0</td>
<td>0</td>
<td>2</td>
</tr>
<tr>
<td>RP20 → UR25</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>3</td>
</tr>
<tr>
<td>RO₂59 + NO → NO₂ + HO₂ + UR24</td>
<td>2.08e-12</td>
<td>0</td>
<td>180.0</td>
<td>4</td>
</tr>
<tr>
<td>RO₂59 + HO₂ → OH + HO₂ + UR24</td>
<td>3.14e-13</td>
<td>0</td>
<td>800.2</td>
<td>4</td>
</tr>
<tr>
<td>RO₂59 + RO₂⁻ → HO₂ + UR24 + RO₂T</td>
<td>1.0e-15</td>
<td>0</td>
<td>0</td>
<td>4</td>
</tr>
<tr>
<td>RO₂60 + NO → NO₂ + HO₂ + 2.0 MGLY</td>
<td>2.08e-12</td>
<td>0</td>
<td>180.0</td>
<td>4</td>
</tr>
<tr>
<td>RO₂60 + HO₂ → OH + HO₂ + 2.0 MGLY</td>
<td>3.14e-13</td>
<td>0</td>
<td>800.2</td>
<td>4</td>
</tr>
<tr>
<td>RO₂60 + RO₂⁻ → HO₂ + 2.0 MGLY + RO₂T</td>
<td>1.0e-15</td>
<td>0</td>
<td>0</td>
<td>4</td>
</tr>
</tbody>
</table>

*References are as follows: 1, Bierbach et al. [1995]; 2, Bierbach et al. [1994]; 3, assumed equal to ALD2 in CACM [Griffin et al., 2002, 2005]; 4, stoichiometry adapted from University of Leeds Master Chemical Mechanism (http://mcm.leeds.ac.uk/) with reaction rates adapted from CACM and Jenkin et al. [1997].

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Alvarado and Prinn (2009)
OH Oxidation of 2- and 3-methylfuran

Aschmann et al. (2013)
OH Oxidation of 2- and 3-methylfuran

Strollo and Ziemann 2013
OH Oxidation of 2- and 3-methylfuran

Strollo and Ziemann 2013
OH Oxidation

Aschmann et al. (2013)
OH Oxidation

Aschmann et al. (2013)
25 potential products
25 14 potential products

hydroxyfuranones
2MF + OH → 2MF_D_OH
2MF_D_OH + O2 → 2MF_D_OH_OO
2MF_D_OH_OO + NO → 2MF_D_OH_O + NO2
2MF_D_OH_O → 2MF_D_P_ALDEST
2MF_D_P_ALDEST + O2 → ALDEST_G + HO2
2MF_D_P_ALDEST + O2 → ALDEST_H + HO2
2MF + OH → 2MF_C_OH
2MF_C_O_H + O2 → 2MF_C_OH_OO
2MF_C_OH_OO + NO → 2MF_C_OH_O + NO2
2MF_C_OH_O → HDXYFN_K + CH3
2MF_C_OH_O → HDXYFN_L + HO2
2MF_C_OH → A1_2MF
A1_2MF + O2 → A2_2MF
A2_2MF → DIALD_A + HO2
2MF_C_OH_O + NO → B_INTER_2MF + NO2
B_INTER_2MF → ALDACID_C + CH3
B_INTER_2MF → ALDACID_D + HO2
ALDACID_C ↔ HDXYFN_K
ALDACID_D ↔ HDXYFN_L

3MF + OH → 3MF_D_OH
3MF_D_OH + O2 → 3MF_D_OH_OO
3MF_D_OH_OO + NO → 3MF_D_OH_O + NO2
3MF_D_OH_O → 3MF_D_P_ALDEST
3MF_D_P_ALDEST + O2 → ALDEST_J + HO2
3MF_D_P_ALDEST + O2 → ALDEST_I + HO2
3MF + OH → 3MF_C_OH
3MF_C_O_H + O2 → 3MF_C_OH_OO
3MF_C_OH_OO + NO → 3MF_C_OH_O + NO2
3MF_C_OH_O → HDXYFN_M + HO2
3MF_C_OH_O → HDXYFN_N + HO2
3MF_C_OH → A1_3MF
A1_3MF + O2 → A2_3MF
A2_3MF → DIALD_B + HO2
3MF_C_OH_O + NO → B_INTER_3MF + NO2
B_INTER_3MF → ALDACID_E + HO2
B_INTER_3MF → ALDACID_F + HO2
ALDACID_C ↔ HDXYFN_K
ALDACID_D ↔ HDXYFN_L
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<td>1.4 ± 0.2$^a$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.3 ± 0.2$^b$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.99 ± 0.06$^c$</td>
</tr>
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</tr>
<tr>
<td></td>
<td></td>
<td>12.6 ± 1.9$^e$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>13 ± 5$^f$</td>
</tr>
</tbody>
</table>
Chamber Simulations

ICs: MF2, MF3, HONO
Lights on @ 75 minutes
25% each branching ratio
50% B Branch, 16.7% to each of the others
Gas-Phase

25% each branching ratio

100% B Branch

50% B Branch, 16.7% to each of the others

OH

Ozone

30 ppb

42 ppb

35 ppb
Next Steps

• Chamber studies with OH oxidation
• Add to ASP:
  – NO$_3$ oxidation
  – NO $\rightarrow$ NO$_2$ chemistry (organic nitrates)
  – Furfural
  – Oligomerization parameterizations
• Synthesis and mechanism constraints
• 1$^{st}$ year of a 3-year project…so stay tuned!
Secondary Organic Aerosol Formation and the Oxidation Mechanism of Methylfuran by Nitrate Radical Oxidation

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Questions?