A perspective on the requirements for

Emission Inventories of Short-Lived Climate Forcers needed to assess human role and to project SLCFs



The step from human activities/emissions to radiative forcing:

Emission ► Production/Loss ► Abundance/Burden ► RF

- All steps are diurnally, seasonally, geographically variant
- Many steps depend on local chemical-physical environment
- Many steps depend on co-emitted species
- Many steps are not linearly scalable
- No simple, fungible rate-of-exchange (like CO₂-eq for WMGHG)

O. Wild et al., 2001, Indirect long-term global cooling from NOx emissions, GRL 28:1719-1722

This seminal paper by Oliver Wild demonstrated that:

- 1. location of emissions (latitude, altitude) and co-species emitted (CO, VOCs) changed the SLCF RF, and
- 2. emissions excite chemical modes, producing indirect effects across most SLCFs



O. Wild, et al., 2004, Chemical transport model ozone simulations for spring 2001 over the western Pacific: Regional ozone production and its global impacts, JGR 109(D15): D15S02.

This paper followed the O_3 caused by SLCF emissions for several E. Asian metropolitan regions, following the steps from the first slide and demonstrating how they can differ by day and type of pollution.



Altitude and amount of the O_3 increase depends depends on the meteorology of the day, and even the decay (lifetime) of O_3 .



Figure 5. Additional mass of O_3 from 1 day's emissions of precursors from Shanghai on (top) 12 March and (bottom) 16 March. Note that the altitude regions are additive; together with the stratospheric impacts (which are small) they give the global perturbation. See color version of this figure in the HTML.

O. Wild, et al., 2004, Chemical transport model ozone simulations for spring 2001 over the western Pacific: Regional ozone production and its global impacts, JGR 109(D15): D15S02.

This paper followed the O_3 caused by SLCF emissions for several E. Asian metropolitan regions, following the steps from the first slide and demonstrating how they can differ by day and type of pollution. #2



Table 2. Ozone Production From 1 Day's Emissions of Precursors in March 2001

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	Gross Production			Mean Burden	
Region	Region,	Globe,	Fraction	Region,	Globe,
	10 ⁶ kg	10 ⁶ kg	in region	10 ⁶ kg	10 ⁶ kg
Beijing	2.4 ± 1.3	16.9 ± 2.6	0.14	0.03 ± 0.15	$\begin{array}{c} 5.67 \pm 1.39 \\ 7.77 \pm 1.43 \\ 5.41 \pm 0.80 \\ 6.43 \pm 1.66 \end{array}$
Tokyo	3.6 ± 1.5	18.7 ± 2.3	0.19	0.29 ± 0.33	
Shanghai	8.8 ± 1.7	21.5 ± 1.8	0.41	1.86 ± 0.96	
Hong Kong	9.9 ± 2.2	24.8 ± 3.8	0.40	2.14 ± 0.74	

M.J. Prather et al,(2017) Global Atmospheric Chemistry – Which Air Matters, ACP, 17(14), 9081-9102.

RESOLUTION: As we move to higher resolution chemistry models we see incredible granularity in SLCF-critical chemistry, and this is also seen with the in situ observations. Below shows the fine-grain structure in column CH_4 loss by OH. We will need SLCF emissions inventory on the model grid.



Figure 1. (a) Column tropospheric loss frequency (1 yr^{-1}) for CH₄ :

Lu Hu et al., (2018). Global simulation of tropospheric chemistry at 12.5 km resolution: performance and evaluation of the GEOS-Chem chemical module (v10-1) within the NASA GEOS Earth System Model (GEOS-5 ESM), GMDD, gmd-2018-111.

RESOLUTION: As we move to higher resolution chemistry models we see incredible granularity in SLCF-critical chemistry, and this is shown with mid-tropospheric O_3 below



Figure 2. **500 hPa** ozone distribution on August 1, 2013 at 0Z simulated by GEOS-5 with the GEOS-Chem chemical module at cubed-sphere C720 (~12.5×12.5km) resolution

C.D. Holmes et al. (2014) The climate impact of ship NOx emissions: an improved estimate accounting for plume chemistry, ACP 14:6801–6812, acp-14-6801-2014.

Intermediate modeling of SLCF emissions:

Even with <1° resolution, there will be a need to model the complex, non-linear processes that occur within fresh plumes.

Work by Chris Holmes shows the importance of plume models vs. instant dilution over the grid cell.

It also shows failings of previous plume models that calculated O_3 production and NOx loss, but **NOT** CH₄ loss.

LESSON: We will always develop more⁻¹⁴ accurate models for the SLCF inventories. If you build it they will come!



Figure 5. Steady-state RF (mW m⁻²) from O_3 and CH₄ caused by ship NO_x emissions. Values are scaled to emissions of $1 \text{ Tg}(N) \text{ yr}^{-1}$. Dashed lines link estimates that are made with 5%

M.J. Prather (1994) Lifetimes and eigenstates in atmospheric chemistry, GRL, 21, 801-804. M.J. Prather, J. Hsu (2010) Coupling of N_2O and CH_4 by global atmospheric chemistry, Science, 330:952-954

Chemical feedbacks affect residence times and perturb other gases:

CO makes CH₄

A perturbation to CO alone, δ [CO]=1, causes a significant perturbation to CH₄ that is "unmasked" in a few months:

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\delta[CO](t) \approx + 0.005 e<sup>-t/13.6</sup> + 0.995 e<sup>-t/0.285</sup>
\delta[CH_{4}](t) \approx +0.15 \text{ e}^{-t/13.6} - 0.15 \text{ e}^{-t/0.285}
                                                                #1 (13.6 y)
                                                                               #2 (0.285 y)
                                                                                              #3 (0.56 s)
                                                      δ[CH4]
                                eigenvactors aco
                                                       δ[OH]
                                                                                         1
                                                                              0
                                                                                             Ω
                                                                0
                                                                           1
                                                                    change in abundance (ppb)
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M.J. Prather (1994) Lifetimes and eigenstates in atmospheric chemistry, GRL, 21, 801-804. *M.J. Prather, J. Hsu* (2010) Coupling of N_2O and CH_4 by global atmospheric chemistry, Science, 330:952-954

Chemical feedbacks affect residence times and perturb other gases:

N₂O destroys CH₄

Due to Interactions of N₂O with stratospheric O₃ chemistry, a relatively large <u>reduction in CH₄ (-3.6 ppb) is tied to an</u> <u>increase in N₂O (+10 ppb)</u> that decays with a 108-yr time scale.

Thus N_2O 's climate impact through radiative forcing is reduced*:

(i) -8.4% because the decay of a pulse is faster than the e-fold of the steady-state lifetime and

(ii) a further -4.5% to account for the reduction in CH_4 .

Some thoughts on SLCF inventories:

Emission ► Production/Loss ► Abundance/Burden ► RF

- Be consistent on co-emitted species
- Maintain full traceability, so that previous work can be adapted to new knowledge and so that the modeling/assessment community can scale by sectors/etc.
- Use as high resolution as possible, sometimes avoiding fixed regulargrid results that cannot be disaggregated.
- A full inventory of all species that the modeling community would 'like' is simply not practical, so keep track of key marker species by source-type of emission, allowing emission factors to be used by the models.