Potential impacts of aircraft emissions on the air quality near the ground (importance of heterogeneous chemistry and nitrate radicals)

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Introduction

- Rapid increase of the demand for aviation transport
- Recent studies [e.g. *Barrett et al.*, 2010] suggest that current non-LTO (non-landing and takeoff) aviation emissions (climb/descent + cruise altitude emissions) impact local air quality increasing premature deaths.
- Impacts of aircraft emissions on the distribution of gases and aerosols near the ground

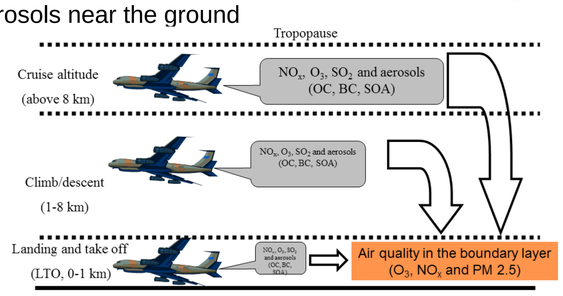

Table 1. The total annual emissions from aircraft used in this study. Unit of the emissions is Tg (teragram)/year.

	NO_x (as NO)	CO	SO_2	black carbon (BC)	organic carbon (1/3 of BC)
total emissions	1.347	1.692	0.164	0.007	0.002
LTO emissions	0.126 (9.5 %)	0.624 (37.3 %)	0.0167 (10.3 %)	0.00134 (19.9 %)	0.000446
climb/descent emissions	0.489 (36.9 %)	0.732 (43.8 %)	0.0518 (32.0%)	0.00296 (44.1 %)	0.000985
cruise altitude emissions	0.712 (53.7 %)	0.315 (18.8 %)	0.0931 (57.6 %)	0.00242 (36%)	0.000805

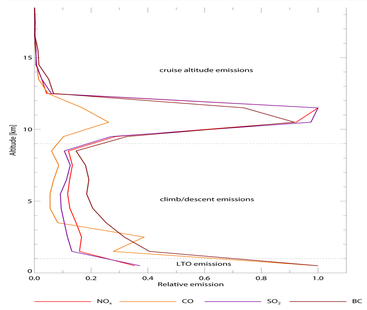

Figure 1. A vertical profile of the total annual emissions of NO_x (red), CO (orange), SO_2 (purple) and black carbon (brown) from aircraft emission database representing 1999.

- Factors determining impacts of non-LTO emissions in the boundary layer
  - Background concentration of aerosols
  - Key chemical reactions during wintertime
- Sensitivity of aerosol formation to background NH_3 concentrations

Model and Data

- CAM (Community Atmosphere Model)-chem in offline mode
- 26 vertical levels covering up to 3.5 hPa, with the horizontal resolution of approximately a 2.5° (longitude) × 2.0° (latitude)
- Meteorological fields from online CAM-chem run representing 2002
- Aviation emission data from the Boeing Company for year of 1999

Case	LTO emissions (0-1 km)	Climb/descent emissions (1-8 km)	Cruise altitude emissions (above 8 km)
control	No	No	No
aircraft	Yes	Yes	Yes
aircraft_non_LTO	No	Yes	Yes
cruise	No	No	Yes
air_2x_NH_3 (double NH_3 flux)	Yes	Yes	Yes
no_air_2x_NH_3 (double NH_3 flux)	No	No	No

Aviation emissions and air quality in the boundary layer

- Overall, wintertime (January) perturbation due to aviation emissions is larger than summer (July).
- Non-LTO emissions, especially emissions from cruise altitudes cause the largest perturbation.
- Perturbations in O_3, NO_y and PM 2.5 are statistically significant at 95% confidence level in the winter.
- **However,** considering uncertainties in the emission index (a factor to convert fuel burn rate to emission of each species) and amount of NH_3, **impacts of aviation emissions on air quality in the boundary layer (e.g. several ppbv of O_3 and less than 1% increase of PM 2.5) are negligible. Statistical significance is meaningless here.**
- Concentrations of hydrophilic aerosols and free ammonia could amplify aviation impacts from above the boundary layer.
- Non-LTO emissions decrease NO_y near the ground and limit O_3 increase.

Changes in NO_y

[defined as N + NO +NO_2 + NO_3 + HNO_3 + HO_2NO_2+ 2×N_2O_5 + $CH_3CO_3NO_2$ (PAN) + $CH_3COCH_2ONO_2$ (organic nitrate) + $CH_2CCH_3CO_3NO_2$ (MPAN, methacryloyl peroxynitrate) + $CH_2CHCCH_3OOCH_2ONO_2$ (ISOPNO3, peroxy radical from NO_3 + isoprene) + $CH_2CCH_3CHONO_2CH_2OH$ (lumped isoprene nitrates)]

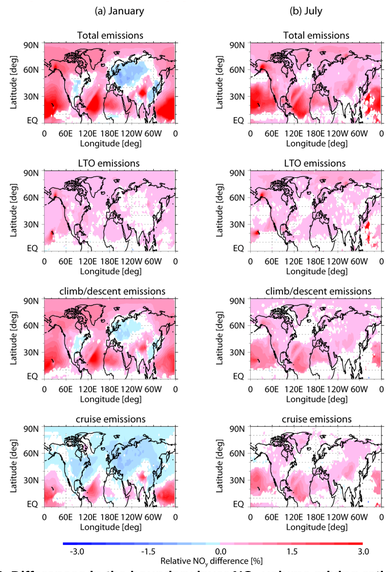

Figure 2. Differences in the boundary layer NO_y volume mixing ratio relative to the background NO_y between the baseline control and the simulation with aircraft emissions (air – control) in (a) January (left column) and (b) July (right column). From top to bottom, [top] (aircraft – control), (aircraft – aircraft_non_LTO), (aircraft_non_LTO – cruise) and [bottom] (cruise – control).

- O_3 is increased by aviation emissions (not shown), but NO_y (mostly NO_x) is **decreased** up to 2% by non-LTO emissions in winter.

Q. What causes the seasonal differences in NO_y and O_3 between January and July?

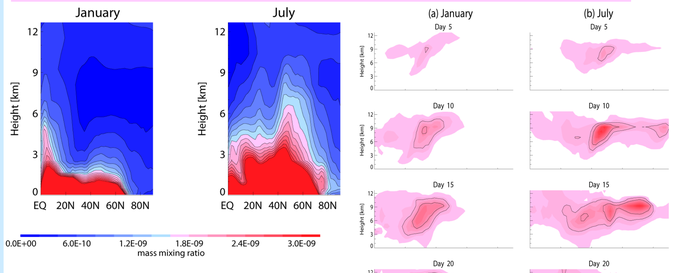

Figure 3. Latitude-altitude distribution of monthly averaged mass mixing ratios of PM 2.5. PM 2.5 was averaged over longitude 0E and 90E in January (left) and July (right).

- Higher background concentration of aerosols in summer
- Heterogeneous reactions on aerosols (sulfate, NH_4NO_3, hydrophilic OC and SOA)

$N_2O_5 \rightarrow 2\ HNO_3$

$NO_3 \rightarrow HNO_3$

$NO_2 \rightarrow 0.5 * (OH + NO + HNO_3)$

- In summary, these are NO_x to NO_y reactions.
- These heterogeneous reactions can explain the smaller NO_y perturbation in July.

Figure 4. Propagation of O_3 perturbation resulted from suddenly imposed cruise emissions for 30 days on control_CAM outputs at the beginning of (a) January and (b) July. Only the perturbations larger than 0.1 ppbv are shaded. Solid lines indicate where the perturbations are 0.5 and 1 ppbv.

Q. Why does the surface NO_y decrease due to aviation emissions from the upper troposphere in the winter?

- $NO_2 + OH + M \rightarrow 2\ HNO_3 + M$, OH is not largely affected by aviation emissions.

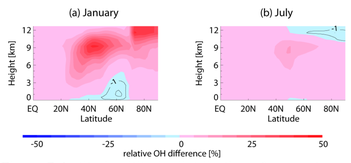

Figure 5. Latitude-altitude distribution of percentage difference to the background NOx concentration, (aircraft_non_LTO – control)/(aircraft_non_LTO) x 100 %, in OH between the control and the simulation with non-LTO aircraft emissions averaged over longitude 0E and 90E in January (left) and July (right).

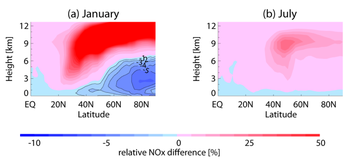

Figure 6. Latitude-altitude distribution of percentage differences in NO_x between the control and the simulation with non-LTO aircraft emissions averaged over longitude 0E and 90E in January (left) and July (right).

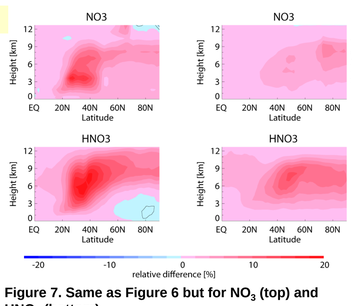

Figure 7. Same as Figure 6 but for NO_3 (top) and HNO_3 (bottom).

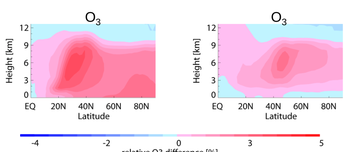

Figure 8. Same as Figure 7 but for O_3.

- Low actinic flux at high latitudes in the winter: longer lifetime of NO_3 radicals
- O_3 perturbation is positive in the boundary layer

- Important nighttime chemistry

(titration of O_3 & NO_x to NO_y conversion)

$NO_2 + O_3 \rightarrow NO_3$

$NO_3 + NO_2 \rightarrow N_2O_5$

$N_2O_5 + H_2O \rightarrow 2\ HNO_3 \rightarrow$ wet deposition

- Net reaction

$2NO_2 + O_3 + H_2O\ (s) \rightarrow 2\ HNO_3$

(a more efficient sink for NO_x than O_3)

Summary of mechanisms reducing NO_y

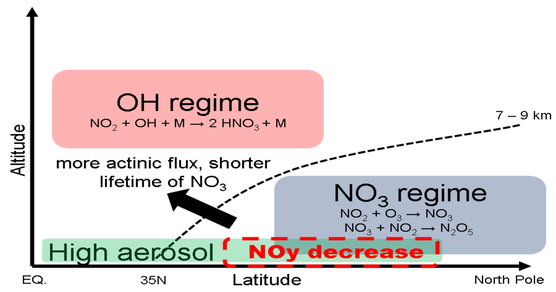

Figure 9. A diagram describing the boundary between OH regime and NO_3 regime and the intersection of the NO3 regime and the high aerosol concentration near the surface.

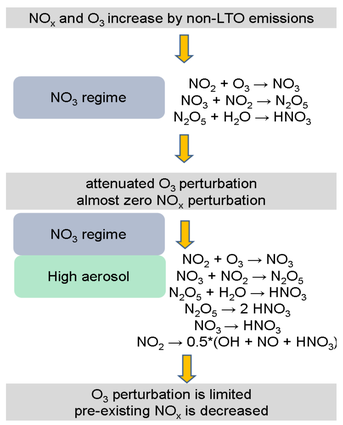

Figure 10. A schematic diagram explaining the changes in perturbation made by non-LTO emissions while propagating and why the perturbations decrease the NOx in the boundary layer during the wintertime.

Changes in aerosols

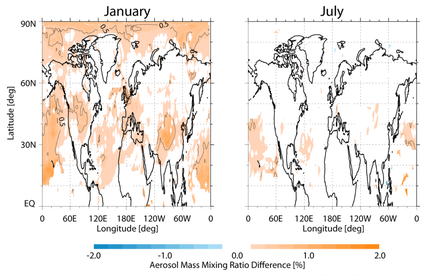

Figure 11. Percentage differences in the boundary layer PM 2.5 between the control and the simulations with aircraft emissions in January. [Top] (aircraft – control) [bottom left] (aircraft_non_LTO – control) and [bottom right] (aircraft – non_LTO).

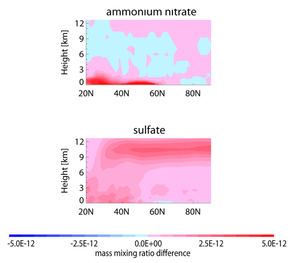

Figure 12. Latitude-altitude distribution of differences in [top] ammonium nitrate and [bottom] sulfate between the control and non_LTO simulation. The differences were averaged over longitude 0E and 90E in January.

- PM 2.5 in Midwest and East Coast of the US, Europe and East Asia show statistically significant influence of aviation emissions. (about 0.1 ppb ~ 0.1 μg/m^3).
- Even the relatively large PM 2.5 perturbation in January changes at most 1% of the background PM 2.5
- The increase of PM 2.5 in the lower troposphere in January is mostly due to NH_4NO_3 due to increased HNO_3.

Sensitivity of aerosol formation to the ground NH3 flux

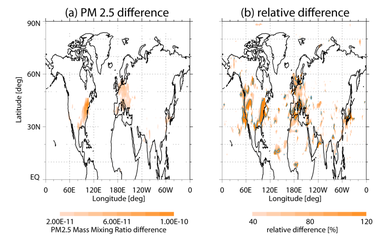

Figure 13. (a) Differences in the boundary layer PM 2.5 due to the doubled NH_3 flux (air_2X_ NH_3 - no_air_2x_NH_3 – aircraft + control) in January. (b) The relative PM 2.5 perturbation (air_2X_ NH_3 - no_air_2x_NH_3)/(aircraft - control) * 100 [%].

- With higher NH_3, the sensitivity of non-LTO emissions effects on PM 2.5 is strongest in the East Coast of the US and Europe.
- In these regions with heavy air traffic, doubled ground NH_3 fluxes increase more than 100% PM 2.5.
- Remaining uncertainties of NH_3 and background aerosols could amplify or diminish impacts of non-LTO emissions on air quality.
- However, even the doubled PM 2.5 increase with doubled NH_3 fluxes on PM 2.5 is not critical.

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