

PERSPECTIVES: ATMOSPHERIC CHEMISTRY

AIRCRAFT IMPACTS
Unraveling Contrail Chemistry [JUI]

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As common as contrails are in the sky today, it is easy to imagine that the (about) 10,000 jet aircraft operating every day alter the composition of the atmosphere substantially at aircraft cruising [JU2] altitudes. But do they? And if so, by how much? Two recent campaigns in the North Atlantic region are now providing much-needed observational constraints on the chemical composition of the atmosphere at the relevant altitudes. Such studies are particularly timely as airline industry forecasts are predicting a doubling in the number of operational aircraft over the next 20 years (1).

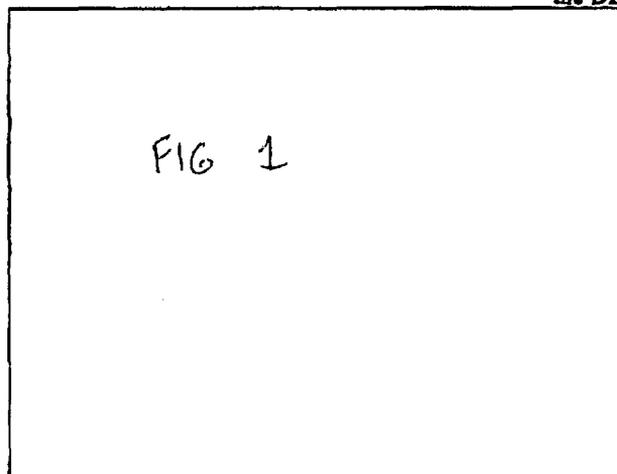
Jet aircraft emit several chemical species that affect atmospheric chemistry and climate, including carbon dioxide (CO_2), water (H_2O), nitrogen oxides (NO_x), sulfur oxides (SO_x), and soot. Over the last decade, NO_x emissions have attracted the most attention because of their role in forming ozone, a potent greenhouse gas throughout the atmosphere and a common air pollutant in the lower troposphere. Considerable research has been focused on low NO_x combustor technologies, and the International Civil Aviation Organization (ICAO) has twice reduced the recommended ceiling on aircraft NO_x emissions during landing and takeoff.

The NO_x emissions from present-day aircraft into the atmosphere are now relatively well quantified at about 0.5 teragrams of N per year. But their impact on ambient atmospheric chemistry [JU4] is obscured by other NO_x sources such as lightning and surface emissions (see the figure). Surface emissions, in particular, are substantially larger than aircraft emissions. For example, NO_x emissions from automobiles and biomass burning amount to about 20 and 10 teragrams of N per year, respectively. However, most of these emissions are thought to be removed from the atmosphere by uptake on cloud drops followed by precipitation, before they can diffuse or be lifted to aircraft cruise [JU5] altitudes. If this view holds [JU6], then the upper troposphere-lower stratosphere (UT-LS) region

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(at heights of 9 to 13 km), where most aircraft fly, represents a relatively pristine environment that is affected only episodically during weather events that rapidly lift surface air or bring down stratospheric air. Even the small fraction of surface emissions transported to high altitude is similar in magnitude to [JU7] the aircraft source—although quantifying it [JU8] is a major challenge for atmospheric scientists. To complicate matters, the same weather events responsible for surface NO_x transport may also be associated with lightning that can produce and inject NO_x directly at the upper altitudes. But detailed understanding of NO_x generation and transport in lightning events is lacking.

The various source contributions to up-



per tropospheric NO_x concentrations must be quantified to reach an understanding of aircraft impacts on ozone. Atmospheric photochemical models incorporating the best available parameterizations of NO_x sources and atmospheric transport have hitherto provided the only quantitative guidance (2-4). These models have predicted that current aircraft operations perturb NO_x concentrations at cruise altitudes in the northern midlatitudes by about 20%, but possibly by up to 100%. The ozone response to this perturbation is moderated by the amount of hydrogen oxide (hydroxyl radical and its peroxy precursors) available for ozone-forming chemical reactions. Predicted ozone perturbation ranges between 2 and 14%. The wide range of predictions reflects the substantial uncertainties associated with model process parameterizations and source strength es-

timates.

Further model development is hindered by the lack of observational data. Satellite-based instruments have encountered substantial difficulties in probing the chemical composition of the upper troposphere and lower stratosphere, leaving sporadic aircraft- and balloon-based measurements as the primary data source. In addition to providing a basis for model improvement, observational data may enable an empirically based, model-independent assessment of aircraft impacts. This was one of the motivations for two recent aircraft-based field campaigns investigating cruise-level aviation impacts on NO_x and ozone photochemistry in and around the North Atlantic flight corridor between September and November 1997: the Subsonic Assessment Ozone and NO_x Experiment (SONEX) and the Pollution from Aircraft Emissions in the North Atlantic Flight Corridor (POLINAT) experiment (5-7). Investigations were carried out onboard the DLR Falcon and NASA DC-8 aircraft,

which carried 10 and 14 chemical and radiation measuring instruments, respectively. The measurement efforts were complemented by advanced flight planning and data analysis modeling tools.

Empirical NO_x source deconvolution [JU9] is possible only under favorable atmospheric conditions. The atmospheric NO_x impact of an individual source must have a geographically distinct pattern, preferably peaked close to its origin. Such patterns can develop if atmospheric mixing is slow relative to NO_x removal. For an aircraft source,

maximum concentrations over the United States and Europe and in the North Atlantic flight corridor would be a likely part of the pattern. In addition, source separation is facilitated if the source emissions, atmospheric mixing processes, and chemical loss processes are steady, continuous processes that combine to produce smooth concentration patterns.

Neither of these conditions are expected to be met for NO_x in the UT-LS. Nevertheless [JU10], the SONEX and POLINAT findings have revealed more fully the extent of NO_x variability as a result of atmospheric dynamics and source fluctuations and the resultant difficulties with an empirical analysis. The picture that emerges from these studies is of a region with frequent, but irregular, injections of NO_x from the surface, from lightning, and from stratosphere-troposphere exchange.

1 Ambient NO_x perturbations as a result of
2 aircraft emissions are readily observed a
3 few kilometers behind individual aircraft,
4 but the signatures are quickly lost at
5 greater spatial scales in a mostly incoher-
6 ent NO_x landscape (8). Observed NO_x
7 concentrations are unevenly distributed
8 over a wide range, making it difficult to
9 define average UT-LS background con-
10 centrations.

11 A degree of coherence is imposed on
12 the NO_x observations through correlations
13 with other tracers of emission sources and
14 by air mass trajectory analysis. In par-
15 ticular, measurements of CO , O_3 , and to-
16 tal particle density have been used to
17 identify surface and stratospheric influ-
18 ences on sampled air masses. Combined
19 with analysis of wind speed and direction
20 over a 3- to 5-day period before an obser-
21 vation, these diagnostics provide insights
22 into the latitudinal and altitudinal history
23 of the air parcel. On the basis of these
24 analyses, the derived magnitude of the
25 aircraft contribution to the regional scale
26 NO_x and ozone environment (9, 10) is
27 consistent with the low-end range of
28 model estimates. At this impact level, the
29 global warming potential of aircraft-
30 induced ozone change is predicted to be
31 comparable to that of aircraft CO_2 emis-
32 sions (2[JUL]).

33 Even with the SONEX and POLINAT
34 efforts, the UT-LS remains substantially
35 undersampled. This scarcity of data is a
36 considerable impediment to an improved
37 assessment of aircraft impacts. Future sat-
38 ellite-based measurements of UT-LS
39 chemical composition, such as those
40 planned from the NASA Earth Observing
41 System (EOS) platform, promise to im-
42 prove this situation substantially. In the
43 meantime, the SONEX and POLINAT
44 findings argue strongly for the need to
45 improve model treatment of tropospheric
46 transport and chemistry through carefully
47 designed modeling and data studies (2,
48 11, 12).

49 Although our understanding of aircraft
50 effects on ozone remains incomplete, fu-
51 ture studies of aircraft impacts are likely
52 to focus increasingly on atmospheric
53 cloudiness. The Intergovernmental Panel
54 on Climate Change concludes that air-
55 craft-induced cloudiness, either through
56 direct contrail formation or through indi-
57 rect effects on cirrus cloud formation, may
58 increase global warming to a greater ex-
59 tent than aircraft CO_2 and NO_x emissions
60 (2). SONEX and POLINAT measure-
61 ments of condensation nuclei densities
62 have provided initial constraints on the
63 aircraft contribution to cloud precursors in
64 the traffic corridors (10, 13). However, the
65 current level of scientific understanding of

cloud-forming processes is considerably
poorer than that of ozone chemical pro-
cesses.

References and Notes

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Sources of atmospheric NO_x . Biomass burn-
ing, automobile and industrial emissions, light-
ning, and aircraft emissions all contribute to the
upper troposphere-lower stratosphere NO_x
budget, but quantifying these sources remains
difficult. Values are given in units of teragrams
of N per year.