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Aviation-Climate Change Research Initiative  
(ACCRI)

**Subject specific white paper (SSWP) on  
UT/LS chemistry and transport**

**SSWP # II**

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## Executive Summary

The global commercial aircraft fleet currently numbers about 10,000 and flies several billion kilometres per year while burning more than 100 MT of fuel per year at high temperatures producing mostly water and CO<sub>2</sub>. However, NO<sub>x</sub> (= NO+NO<sub>2</sub>), other minor gaseous species, organic aerosols from unburnt fuel and soot and ions are also injected at cruise altitudes located in upper troposphere and lower stratosphere (UT/LS), a region particularly sensitive to atmospheric climate change.

The demand for air transportation in the US is projected to grow three fold by 2025 while similar growth is projected for the aviation industry world wide. Future climate impacts are expected to increase based on this projected aviation growth and resulting changing atmospheric conditions. These impacts relate to the impact of tripling aviation system capacity and the resulting global impact of these additional engine emissions which are estimated to be approximately twice as large as at the turn of the last century. However, if current economic projections obtain for this period, boundary layer (BL) NO<sub>x</sub> emissions may also double and hence their contribution to the UT region. In addition to global climate impacts there is also potential for even greater regional or local effects. The growth of emissions of both BL and aircraft NO<sub>x</sub> will likely lead to an increased production of ozone in the UTLS. This increase in UT/LS ozone will cause a significant increase in the radiative forcing, which in turn will contribute to global warming.

Jet traffic spends 60% of the time in the upper troposphere (UT) where current information is insufficient to make an accurate prediction of the climate impacts of increased jet traffic due to tropospheric ozone generated from aircraft NO<sub>x</sub> emissions. A review of the HO<sub>x</sub>/NO<sub>x</sub> chemistry concludes that the chemistry is fairly well known in the lower stratosphere. However, the upper troposphere HO<sub>x</sub>/NO<sub>x</sub> chemistry is uncertain as revealed by measurements of NO and OH concentrations which conflict with current model simulations from various aircraft campaigns. The aerosols from aviation emissions can also interact with the background constituents and alter the NO<sub>x</sub> and ClO<sub>x</sub> chemistry with resulting changes in regional ozone in the UT/LS.

In order to evaluate the impact of NO<sub>x</sub> from aircraft on UT ozone, the other sources of NO<sub>x</sub> in this region, such as lightning and transport from the boundary layer must be better quantified than at present. Aircraft measurements reveal “high” levels of NO<sub>x</sub> in the summer UT over North America, generated by lightning. Satellite measurements have recently been analyzed to give a more precise estimate of the global lightning source to be about 5±3 MT per year which makes it an important NO<sub>x</sub> in-situ source for the UT. Upward transport of NO<sub>x</sub> from the boundary layer is also significant. The fraction of the deep convection source of NO<sub>x</sub> from the surface sources which reaches the 10 to 13 km level is

1 estimated to be around 10-50 %. While the uncertainty in this estimate may  
2 simply reflect the difference in the meteorology in the measurement regions, this  
3 needs to be better characterized. The parameterization for convective transport  
4 in 3D models needs to be improved. The effects of the vertical transport by the  
5 Asian monsoon and the Madden-Julian oscillation should be modelled or  
6 parameterized by models as they impact transport into the upper troposphere  
7 and stratosphere via the tropical transition layer.

8  
9 In the aviation corridors, NO<sub>x</sub> is elevated above the background by aircraft  
10 emissions; the aircraft contribution may be dominant under certain meteorological  
11 conditions. The elevated NO<sub>x</sub> and ozone may persist for some time and be  
12 transported to other regions. The radiative forcing due to ozone may be much  
13 higher in some areas than on a global basis. Hence characterization of the  
14 aviation perturbations within the flight corridors needs to be improved and should  
15 be the focus of aircraft and satellite studies. This calls for additional extensive  
16 aircraft campaigns focused on the flight corridors in order to quantify the regional  
17 climate effects of aviation.

18  
19 There are several recent satellites which provide new information on the NO<sub>x</sub>  
20 and nitric acid at flight levels. The data from MIPAS, ACE and  
21 AURA/MLS/HIRDLS is being applied to the NO<sub>x</sub>/HO<sub>x</sub> chemistry of the upper  
22 troposphere. Despite the excellent scientific progress now being made, future  
23 satellite instruments with enhanced capabilities are required. These enhanced  
24 capabilities should include improved vertical resolution to study the UT/LS region.  
25 Denser sampling and higher horizontal resolution are required to address the  
26 corridors issue.

27  
28 There is a real concern that there will be a gap in satellite instruments suitable for  
29 UT/LS investigations in the next 5 years. Deploying ozonesondes as satellite  
30 observation gap fillers would seem to be the minimum requirement. The  
31 SHADOZ/IONS ozonesondes have proven to be highly useful for investigating  
32 ozone in the upper troposphere since they have excellent vertical resolution.

33  
34 The real verification of the climate impact of increased upper troposphere ozone  
35 is the detection of changes in the ozone radiative forcing (RF) at the surface and  
36 at the top of the atmosphere. There are difficulties in measuring changes in the  
37 IPCC radiative forcing metric because of the way in which it is defined at the top  
38 of the tropopause. There are large uncertainties in the calculations of the  
39 radiative forcing metric due to a lack of knowledge of cloud effects. There need to  
40 be verifications of the radiative forcing metric by comparison against real  
41 measurements of observed surface radiative forcing and with satellite radiative  
42 trapping at the top of the atmosphere. This will need to be accomplished by  
43 concurrent simulations of surface forcing and top of the atmosphere radiative  
44 trapping with the same models used to calculate the RF metric.

45  
46 There have been important advances in models since 1999. Data assimilation

1 has proven very valuable in providing a “value-added” component to satellite  
2 data. There are new satellite instruments taking global measurements which  
3 could be used to compare with the model outputs. Multiscale models are needed  
4 to investigate the corridors aspect of the aircraft emissions and the transition to  
5 regional scale climate impacts. Parameterizations used for deep convection need  
6 to be both used consistently (with the basic dynamical model) and verified  
7 according to the scale of model.

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1

## 2 **1. Introduction**

3

4 Currently, world wide, the commercial aircraft fleet numbers about 10,000 and  
5 flies several billion kilometres per year while burning more than 100 MT of fuel  
6 per year at high temperatures producing mostly water and CO<sub>2</sub>. However, NO<sub>x</sub> (=   
7 NO+NO<sub>2</sub>), other minor gaseous species, organic aerosols from unburnt fuel and  
8 soot and ions are also injected at cruise altitudes located in upper troposphere  
9 and lower stratosphere (UT/LS) region. This region is particularly sensitive to  
10 atmospheric climate change: in the tropical tropopause layer (TTL) net heating is  
11 particularly weak and the dynamics is impacted by non-local effects such as  
12 wave breaking in the stratosphere (Holton et al., 1995; Leblanc et al, 2003).  
13 Ozone in this region is particularly important as a greenhouse gas (GHG). At mid-  
14 and high-latitudes stratospheric/tropospheric exchange (STE) occurs and  
15 delivery of ozone and other species to the troposphere is important. The state of  
16 the tropical cold point tropopause is affected by the composition and the  
17 associated thermal balance. This regulates the entry of water vapour into the  
18 middle atmosphere and hence the temperature and polar ozone chemistry in the  
19 stratosphere, which has an impact on the dynamics and thereby the UT/LS  
20 region.

21

22 Most aircraft NO<sub>x</sub> emissions are released directly into the chemically complex  
23 and radiatively sensitive UT/LS between 8-13 km. At the time of the IPCC  
24 (Penner et al., 1999) assessment, there was concern that heterogeneous  
25 chemistry following immediate conversion of sulfur to aerosols from the aircraft  
26 engines could affect the impact on ozone from the NO<sub>x</sub> emissions. Recent  
27 measurements suggest that this immediate conversion is sensitive to background  
28 conditions.

29

30 The effect of aircraft emissions on atmospheric ozone concentration depends on  
31 the altitude at which the emissions are injected. The importance of the NO  
32 catalytic production of ozone from the NO<sub>x</sub> emissions through the oxidation of  
33 methane and hydrocarbons become less effective with altitude while the catalytic  
34 ozone loss cycles become more efficient. Any uncertainties in how well we  
35 understand the atmospheric chemical and physical processes in the UT/LS affect  
36 our ability to understand the magnitude of the aviation effects on ozone and  
37 methane.

38

39 As noted above, the impact of aviation in the UT/LS was subject of an IPCC  
40 report (Penner et al., 1999) and most recently was the subject of workshop in  
41 Boston (Wuebbles, 2006). When aircraft fly in this region the NO<sub>x</sub> emitted reacts  
42 with HO<sub>x</sub> (=OH + HO<sub>2</sub>), CO and sunlight and leads to the generation of ozone.  
43 The water emitted and the ozone generated also impact the HO<sub>x</sub> which can  
44 impact methane, another GHG. Thus, from these simple examples, we see that  
45 there is an intimate and complex link between aircraft emissions and the potential

1 for climate impact. And as the size of the fleet is expected to increase in the first  
2 part of this century it is important to consider the future impacts.

3  
4 The demand for air travel in the US is projected to grow by about three fold by  
5 2025 (see below). World wide aviation growth is also expected. Estimates of  
6 annual fuel use by 2020 annual for commercial air traffic are ~ 350 MT or 2.6  
7 times the estimated fuel use by the global 1999 commercial fleet. This translates  
8 into global NO<sub>x</sub> emissions of ~ 1.5 MT-N from commercial air traffic or about 2.8  
9 times the estimated 1999 NO<sub>x</sub> emissions levels. At the same time total revenue  
10 passenger kilometers are projected to increase from 3,170 billion in 1999 to  
11 8,390 billion in 2020, or by a factor of 2.65 (Sutkus et al., 2003) These estimated  
12 increased emissions are expected to lead to global increase in ozone production  
13 with the potential for larger regional effects. The increase in UT/LS ozone will  
14 cause a significant increase in the radiative forcing, which in turn will contribute to  
15 global warming.

16  
17 In this report, in addition to dealing with the topic of ozone generation and the  
18 associated direct radiative forcing and indirect forcing via its impact on methane,  
19 we were also asked to attempt an assessment of the uncertainty of dynamical  
20 influences on the impacts of aircraft emissions. We have approached this aspect  
21 of our directive by integrating it with the discussion on chemistry as it is difficult to  
22 separate the chemical impacts from transport influences.

23  
24 One of the problems that arise with respect to the impact of the commercial fleet  
25 of aircraft is that we would like to be able to characterize the natural or  
26 unperturbed atmosphere so that we have a baseline for comparison.  
27 Unfortunately, we do not have that luxury since the current fleet has been flying  
28 in the UT/LS before the region has been well characterized. Thus the  
29 assessment of current impact of aircraft must be addressed via modelling studies  
30 combined with the aircraft measurement programs and models require careful  
31 evaluation.

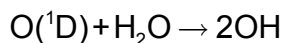
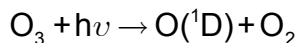
32  
33 The outline of the remainder of the report is as follows. In section two we  
34 summarize the background science focusing on the chemical and transport  
35 issues, which includes gas phase and aerosol chemistry, outlining the current  
36 state of the science including modelling, and we attempt to locate the impact of  
37 aviation within the climate change context. In section three we extract the  
38 problem areas, the areas that we see developing and areas that need substantial  
39 improvement. And we look to the other areas of the study, particularly the  
40 relationship with condensation trails and condensation cirrus. Section four sets  
41 priorities on what can be done on the short term, while section five sets  
42 recommendations and section six provides a summary.

43

## 2. Current state of chemical and dynamical issues in the UT/LS

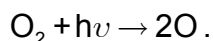
### 2.1 Current state of atmospheric chemistry in the UT/LS

In the UT/LS the chemistry is driven by the presence of ozone and water vapour: the ozone is photolysed to produce O(<sup>1</sup>D) which, with water vapour, produces HO<sub>x</sub> radicals viz.,



In the lower stratosphere the HO<sub>x</sub> radicals interact with NO<sub>y</sub><sup>1</sup>, Cl<sub>y</sub><sup>2</sup> and Br<sub>y</sub><sup>3</sup> and impact the ozone budget in this region. Any NO generated rapidly gets converted to a suite of NO<sub>y</sub> species in the stratosphere, including NO, NO<sub>2</sub>, NO<sub>3</sub>, N<sub>2</sub>O<sub>5</sub>, HNO<sub>3</sub>, HNO<sub>4</sub>, ClONO<sub>2</sub>, BrONO<sub>2</sub> (and in the mesosphere N can be included).

The major source of ozone in the stratosphere is via the photolysis of O<sub>2</sub>



As can be seen in [Figure 1](#) the net ozone source is below about 10 mb and is principally in the tropics while at high latitudes net chemical loss occurs (e.g. Cunnold et al., 1980). From this source structure the stratosphere supplies about 500 MTs of ozone annually to the troposphere, principally at high latitudes, and this represents an important component of the *net* photochemical budget of tropospheric ozone. For the most part our understanding of atmospheric chemistry outside of polar regions in the lower stratosphere appears to be reasonably well understood from satellite, balloon and aircraft measurements (e.g. Zellner, 1999).

One of the problem areas in the stratosphere is the level of Br<sub>y</sub>. Analysis of BrO stratospheric measurements suggest that the levels of Br<sub>y</sub> in the stratosphere are about 24 pptv which is somewhat larger than that supplied by halons and CH<sub>3</sub>Br (e.g., Salawitch et al., 2005; WMO, 2007) The discrepancy between the observations and the sources is probably due to the supply of halogenated very short lived species (WMO, 2006) which are not well mixed in the troposphere. Since Br<sub>y</sub> is important particularly for ozone loss in polar regions, knowledge of future emissions of these short lived species is important.

<sup>1</sup> NO<sub>y</sub> = NO + NO<sub>2</sub> + NO<sub>3</sub> + 2N<sub>2</sub>O<sub>5</sub> + HNO<sub>2</sub> + HNO<sub>3</sub> + ClONO<sub>2</sub> + BrONO<sub>2</sub>

<sup>2</sup> Cl<sub>y</sub> = Cl + ClO + HOCl + ClONO<sub>2</sub> + HCl + 2Cl<sub>2</sub>O<sub>2</sub> + BrCl + 2Cl<sub>2</sub>

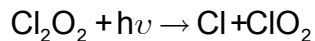
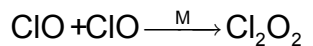
<sup>3</sup> Br<sub>y</sub> = Br + BrO + HOBr + BrONO<sub>2</sub> + HBr + BrCl + 2Br<sub>2</sub>



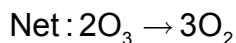
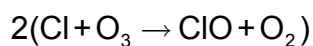
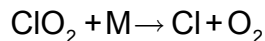
## Chemical and dynamical processes: revised final

1 Another uncertainty at this point in polar regions is that about 2/3 of the loss of  
2 ozone is thought to be via the formation of the Cl<sub>2</sub>O<sub>2</sub> dimer via the reaction  
3 sequence

4



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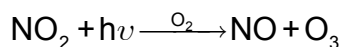
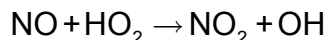
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7 However, recent laboratory measurements of Cl<sub>2</sub>O<sub>2</sub> cross sections applied to the  
8 observations of the important species in the above reaction sequence, Cl<sub>2</sub>O<sub>2</sub>,  
9 ClO, and O<sub>3</sub> suggest that the reaction sequence is not rapid enough to account  
10 for the observed ozone loss (von Hobe et al., 2007; Pope et al., 2007).

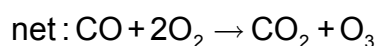
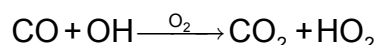
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12 In the troposphere O<sub>3</sub>, O(<sup>1</sup>D) and water play a similar role as in the stratosphere,  
13 acting as a source of HO<sub>x</sub> radicals. In particular, the OH generated can attack  
14 many species, both organic and inorganic in the troposphere, acting as a  
15 “detergent”. The main chemical source of ozone in the troposphere is via the  
16 reactions

17



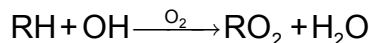
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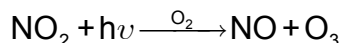
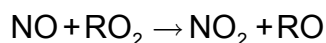
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20 where the first reaction breaks the O<sub>2</sub> bond. Organic peroxy radicals can also  
21 generate O<sub>3</sub> via a similar suite of reactions

22



23



24

25 where RH is a gaseous organic species and further reaction of the RO can  
26 generate HO<sub>x</sub> and catalyze the formation of ozone. Thus it is clear that, under  
27 suitable conditions, NO can catalyze the generation of O<sub>3</sub>. In addition, NO also  
28 converts HO<sub>2</sub> to OH.

28

29 Photochemical production of ozone in the troposphere is estimated to be about  
30 5,000 MT/year while photochemical loss is about 4,500 MT/year leading to a net  
31 photochemical source of about 500 MT/year, i.e. of the same order to the source  
32 from the stratosphere (e.g. Stevenson et al., 2006). An important loss process for

1 ozone is deposition to the surface over land, sea and to a less extent over ice,  
2 which amounts to a global loss of about 1,000 MT/year. In general an increase of  
3 ozone in the troposphere will lead to an increase of OH.

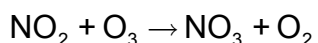
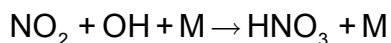
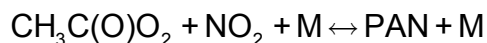
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5 One of the species generated by the VOC-NO<sub>x</sub> chemistry is PAN (CH<sub>3</sub>CO<sub>2</sub>NO<sub>2</sub>)  
6 which can be produced by the breakdown of isoprene and acetone for example.  
7 Measurements indicate that PAN is an important carrier of NO<sub>x</sub> in the UT region  
8 as it is stable at low temperatures and is relatively insoluble and slow to  
9 photolyse (e.g. Singh et al., 2006; 2007).

10  
11 One aspect of UT chemistry that needs to be more carefully studied is the  
12 sometimes-used assumption of photochemical steady state (PCSS) for analysis  
13 of measurements. For example, a recent study by Bertram et al. (2007) suggests  
14 that the measurement of the ratio of NO<sub>x</sub> and HNO<sub>3</sub> in the UT region can reveal  
15 lightning sources of NO<sub>x</sub> that occur with large scale convection. But an important  
16 corollary to their work is that quite often NO<sub>x</sub> and HNO<sub>3</sub> are not in PCSS due to  
17 the interaction of convection and chemistry, each with similar time scales. (See  
18 also Prather and Jacob, 1997; Lawrence and Jacob, 1998). But since convection  
19 is, in some sense, stochastic then simple photochemical calculations will often be  
20 misleading and different process metrics need to be developed such as PDFs  
21 which carry the statistical information.

22  
23 As noted above OH can attack organic species and one of the most important  
24 organics is methane, which is a greenhouse gas. Thus if tropospheric ozone  
25 were to increase, as a result of increased NO<sub>x</sub>, (to which aviation would  
26 contribute several percent) it is expected that OH would increase leading to a  
27 decrease in CH<sub>4</sub>. Thus, as noted in Penner et al. (1999), there will be  
28 compensating effects in terms of radiative forcing, positive from ozone increases  
29 and negative from methane decreases. However, due to their differing lifetimes,  
30 the ozone effect will be more regional (cf. [Figure 2](#)) whereas the methane impact  
31 will be global in extent (see also work by Stevenson et al., 2004).

32  
33 Thus sources of NO<sub>x</sub> and hydrocarbons (HCs) are important to the ozone budget  
34 in the troposphere and it is important to have reliable estimates of their  
35 emissions.

36  
37 In the cold, especially winter, free troposphere, NO<sub>x</sub> is converted and  
38 sequestered as PAN, HNO<sub>3</sub>, HNO<sub>4</sub>, and N<sub>2</sub>O<sub>5</sub>:



40

1 Here the  $\text{CH}_3\text{C}(\text{O})\text{O}_2$  radical is most likely produced by photochemical  
2 degradation of acetaldehyde and acetone. There are substantial contributions to  
3 the  $\text{HO}_x$  radical source in the upper troposphere from acetone and other  
4 oxygenated organic compounds but with some uncertainty as regards  
5 quantitative understanding (see Section 2.2.2).  $\text{N}_2\text{O}_5$  is also transformed to  $\text{HNO}_3$   
6 heterogeneously on “hygroscopic” aerosols ubiquitous in the troposphere:



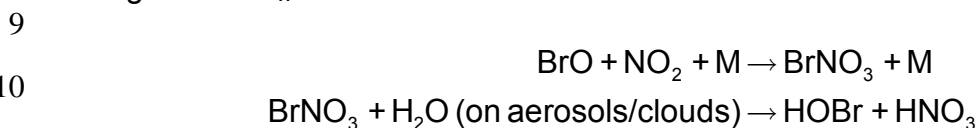
10 According to 3-D model simulations by Dentener and Crutzen [1993], this  
11 heterogeneous reaction provides a dominant pathway for the conversion of  $\text{NO}_x$   
12 to  $\text{HNO}_3$  in the winter troposphere and leads to significant decrease in the  
13 concentrations of ozone and OH radical (20% and 25%, respectively, on average  
14 in the northern hemisphere) particularly from winter to spring because  $\text{HNO}_3$  is  
15 much more stable than  $\text{N}_2\text{O}_5$  under sunlight.

17 Comparisons between observed and modeled  $\text{NO}_x$  concentrations in the mid- to  
18 upper-troposphere from tropics to high latitudes confirmed the role of  
19 heterogeneous  $\text{N}_2\text{O}_5$  hydrolysis, but also indicated that its reaction probability ( $\gamma$ )  
20 should be generally smaller (by a factor of 2 or more) than assumed in Dentener  
21 and Crutzen’s pioneering work ( $\gamma = 0.1$ ) [Schultz et al., 2000; Tie et al., 2003].  
22 More stringent evidence has been obtained in the summertime lower troposphere  
23 over the United States by airborne in-situ measurements of  $\text{N}_2\text{O}_5$  and  $\text{NO}_3$  along  
24 with detailed aerosol measurements [Brown et al., 2006], in which the analysis of  
25 photochemical steady-state  $\text{N}_2\text{O}_5$  concentrations inferred significant decrease in  
26  $\gamma(\text{N}_2\text{O}_5)$  on aerosols by more than an order of magnitude in air masses likely  
27 enriched in organic coating, nitrate content, or efflorescence of aerosols in  
28 agreement with available experimental data [Kane et al., 2001; Folkers et al.,  
29 2003; Hallquist et al., 2003; Thornton et al., 2003]. The same methodology,  
30 however, may not work well to estimate the role of the heterogeneous  $\text{N}_2\text{O}_5$   
31 hydrolysis in the UTLS because (as noted above) of long timescales needed to  
32 establish the photochemical steady state at cold temperatures [Brown et al.,  
33 2003]. Evans et al. [2005] compiled a new parameterization for  $\gamma(\text{N}_2\text{O}_5)$  as a  
34 function of aerosol composition, relative humidity and temperature for the GEOS-  
35 CHEM global chemical transport model and obtained the global mean  $\gamma(\text{N}_2\text{O}_5) =$   
36 0.02. This resulted in increases in  $\text{NO}_x$ ,  $\text{O}_3$  and OH concentrations by 7%, 4%,  
37 and 8%, respectively, relative to those simulated by assuming constant  $\gamma(\text{N}_2\text{O}_5) =$   
38 0.1. The largest changes were found in descending branches of the Hadley  
39 circulation where relative humidity is very low. Also, the new parameterization  
40 was shown to better simulate climatological values of  $\text{O}_3$  and OH in the global  
41 troposphere for their model.

43 The understanding of the source and role of organic aerosols, which are very  
44 often found to be mixed internally with sulfate aerosols even in the UTLS, is still  
45 quite uncertain and requires further theoretical and experimental work for their  
46 source identification and chemical characterization and for their impacts on  $\text{N}_2\text{O}_5$

1 hydrolysis [Iraci and Tolbert, 1997; Zhao et al., 2005; Heald et al., 2005; Murphy  
2 et al., 2007].

3  
4 A few ppt of inorganic bromine background are likely to exist in the free  
5 troposphere (mainly via photodecomposition of bromo-carbons) as well as in the  
6 marine boundary layer (mainly via volatilization from sea-salt aerosols) and thus  
7 BrNO<sub>3</sub> hydrolysis may add to the conversion of NO<sub>x</sub> to HNO<sub>3</sub> contributing to the  
8 budgets of NO<sub>x</sub> and ozone:



11  
12 and also leads to the conversion of water vapour to HO<sub>x</sub> since both HOBr and  
13 HNO<sub>3</sub> photolyze producing OH (Lary et al., 1996).

14  
15 Two model studies have been performed regarding this issue in the free  
16 troposphere; von Glasow et al. [2004] found no more than a marginal impact from  
17 the heterogeneous BrNO<sub>3</sub> hydrolysis on aerosols, whereas Yang et al. [2005]  
18 showed that the surface of cloud droplets may activate this process. Since  
19 significant uncertainties exist with regard to the tropospheric source of inorganic  
20 bromine and the role of clouds for the heterogeneous reactions as well as for the  
21 scavenging of reactive gases, more work is needed for better estimates.

22  
23 Aerosol emission from aircraft can be impacted by the role of organic material in  
24 the plume. However the impact appears greatest for low sulphur fuels (Yu et al.,  
25 1999).

26  
27 One of the possibilities suggested for the difference between model and  
28 measured NO<sub>x</sub>/HNO<sub>3</sub> ratio was the uptake of HNO<sub>3</sub> by cirrus which also lead to  
29 denitrification if the cirrus particles are large and rapidly sedimented (e.g.  
30 Lawrence and Crutzen, 1998).

31  
32 Measurements in the SOLVE (SAGE III Ozone Loss and Validation Experiment)  
33 and BIBLE (Biomass Burning and Lightning Experiment) campaigns and reported  
34 by Kondo et al. (2003) indicated that the HNO<sub>3</sub> uptake on cirrus clouds is very  
35 strongly temperature dependent and uptake would only be important in the UT  
36 with temperature less than ~ 215K. Ziereis et al. (2004) during the INCA  
37 campaign also found a strong temperature dependence of uptake but that, on  
38 average, only ~ 1% of NO<sub>y</sub> was found as particulate NO<sub>y</sub>. Some of the early  
39 laboratory studies suggested that ice could sequester enough HNO<sub>3</sub> to affect the  
40 NO<sub>x</sub>/HNO<sub>3</sub> photochemical ratio and this was also addressed by laboratory and  
41 model studies (cf. Hudson et al., 2002; Tabazedeh et al., 1999).

42  
43 Gamblin et al. (2006a,b) have also addressed the issue of HNO<sub>3</sub> and NO<sub>y</sub> on  
44 cirrus using measurements from the SOLVE-I field campaign in the UT and LS  
45 over Scandinavia. They find that often NO<sub>y</sub> uptake on cirrus is important but, in

1 the troposphere, that HNO<sub>3</sub> is not necessarily the main species deposited to  
 2 cirrus ice and suggest that perhaps N<sub>2</sub>O<sub>5</sub>, HNO<sub>4</sub> or PAN may preferentially  
 3 deposit to ice; the amount of uptake also appears sensitive to the length of time  
 4 the air parcel may have been exposed to sunlight. They also consider that the  
 5 apparent variability of the results in the UT may indicate that the region may not  
 6 be in photochemical or physiochemical equilibrium in the mixed media situation.  
 7 They also suggest (Gamblin et al., 2007) a possible time marker or clock for  
 8 cloud parcel lifetime in the UT.

9 **2.2 Emissions**

10  
 11 **2.2.1 NO<sub>x</sub> emissions:** Emissions of NO<sub>x</sub> into the troposphere come from a  
 12 variety of sources: anthropogenic emissions, biomass burning, boreal forest fires,  
 13 biogenic emissions, lightning emissions, cosmic rays and transport from the  
 14 stratosphere. In the stratosphere the main source of NO<sub>x</sub> is via the oxidation of  
 15 N<sub>2</sub>O with O(<sup>1</sup>D) with contributions from cosmic rays, auroral precipitation and  
 16 sporadic solar proton events (e.g. Jackman et al., 1985; 2005).  
 17

18 The stratospheric source of NO to the troposphere can be estimated from the  
 19 loss of N<sub>2</sub>O in the stratosphere which is ~ 12 MT-N/year (IPCC,2001, Table 4.4).  
 20 About 10% of the N<sub>2</sub>O loss occurs with reaction with O(<sup>1</sup>D) and 6% of the total  
 21 yields NO (e.g. Olsen et al., 2001). There are also “small” contributions from  
 22 cosmic ray precipitation ~ 0.08 MT-N/year and also auroral precipitation  
 23 (Jackman et al., 1980). Most of the NO<sub>y</sub> exits to the troposphere in the form of  
 24 HNO<sub>3</sub> with a global flux of about 0.7 MT-N/year.  
 25

26 Burning of fossil fuel represents an important source of NO<sub>x</sub> in the atmosphere  
 27 with a total of about 33 MT/year and of which aviation currently contributes about  
 28 2% (see Table 1). However, most of the anthropogenic emissions are into the  
 29 boundary layer where the lifetime is of order of a few days whereas aviation  
 30 emissions are mostly injected into the UT/LS region where the lifetime is longer,  
 31 ~ months.  
 32

33 Table 1 Global NO<sub>x</sub> sources (MT-N/year) for 2000 (IPCC, 2001) and 2030  
 34 Dentener et al. (2005)\*  
 35

Global NO <sub>x</sub> sources	2000	Above 7 km	*2030
Fossil Fuel	33.0		40-50
Aircraft	0.7	0.6	1.5†
Biomass Burning	7.1	??	7.1
Soils	3.0**		3.0
Lightning	6*	3.6	6
Stratosphere	0.7***	0.7	0.7
Total	50.5		

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\* Martin et al, 2007 (The figure in brackets is the emission above 7 km.)

\*\* Jaeglé et al., 2005.

\*\*\* Olsen et al., 2001 (and text)

† Sutkis et al. (2003)

Biomass burning arises mostly in the tropics due to human activities (e.g. Crutzen and Andreae, 1990) such as land clearance etc and amounts to ~ 7.1T MT-N/year of which Boreal forest fires contribute about 3-8%. There is a substantial year to year variation. Boreal sources in particular may be expected to increase with changing climate (Stocks et al., 1998; Gillett et al., 2004; Flannigan et al., 2005). Emission estimates for the 1997-2006 period are available online (Randerson et al., 2007). The approach used to calculate these emissions is described in van der Werf et al. (2006), Giglio et al. (2006) and Randerson et al. (2005). The NO<sub>x</sub> emissions follow the dry seasons in the Northern and Southern hemispheres and have a year to year variability reflecting the movement of the ITCZ and the influence of El Niño (van der Werf et al., 2004, 2006). One of the uncertainties is the effective injection height of biomass burning emissions.

The amount of NO<sub>x</sub> generated by lightning is relatively uncertain and values between 2 and 12 MT-N/year are common in the literature (e.g. IPCC, 2001). Recent estimates appear to have reduced the uncertainty to about 6±2 MT-NO<sub>x</sub>/year (Martin et al, 2007) much of which is generated in the upper atmosphere. This is similar to estimate in the comprehensive review of Schumann and Huntrieser (2007) of 5±3 MT-N/year.

However, we note that the analysis was a multistep process depending on chemical and dynamical modelling and observations from several different satellites and the associated error bars may be optimistic. Also reasonable estimates indicate that more than 60% of the NO<sub>x</sub> is created above 7 km at continental mid-latitudes (e.g. Pickering et al., 1998). This source of about 3.6 MT-N represents the largest in-situ source of the NO<sub>x</sub> in the upper troposphere. However, the vertical distribution of the emission source needs evaluation and towards that end the observations from the OSIRIS instrument on the Odin satellite should be useful (Sioris et al., 2007).

Cosmic ray production of NO is modulated by the Sun's magnetic field and so maximizes during solar minimum. Production maximizes ~ 12 km in polar regions due to deflection of the ions by the Earth's magnetic field. The source is ~ 0.08 MT-N/year (e.g. Jackman et al., 1980) mostly concentrated at higher latitudes.

Another "source" of NO<sub>x</sub> in the upper troposphere is that due to convection (see below). As noted above a large fraction of the NO<sub>x</sub> emissions occur at or near the surface. Thus if there is large scale convection the NO<sub>x</sub> can be lofted to the upper troposphere. Current estimates are very model sensitive and not always

1 available from model output. Of the ~ 45 MT-N/year emitted in or near surface if  
 2 10% was lofted to the UT this would significantly impact the NO<sub>x</sub> budget in this  
 3 region and this is within the range of estimates, certainly for North America as  
 4 noted by Singh et al. (2007). However, during the lofting process the soluble  
 5 HNO<sub>3</sub> would be preferentially removed in the convective tower by rainout leaving  
 6 behind the NO<sub>x</sub>.

7  
 8 **2.2.2 CO and VOC emissions:** As was seen above CO plays a major role in  
 9 the generation of ozone and thus knowledge of its distribution and how it might  
 10 change in the future is important. Future emissions we shall address below.  
 11 Current emissions, shown in Table 2 are taken from the IPCC (2001) report. The  
 12 sources are similar as for NO<sub>x</sub>, viz. anthropogenic, biomass burning, ocean,  
 13 oxidation of methane and VOCs etc. One point to note, however, is that CO  
 14 emissions from aircraft play a much more muted role than those of NO<sub>x</sub>.

15  
 16 The situation regarding current CO emissions seems rather uncertain, but it may  
 17 also relate to model uncertainties. For example, modeling studies undertaken for  
 18 the IPCC (2007) (e.g. Shindell et al., 2006) ostensibly with the similar CO  
 19 emissions give very different CO fields. This is a question that needs to be  
 20 addressed.

21  
 22 Table 2. Sources of CO (MT/year) (IPCC, 2001)  
 23

Oxidation of methane	800
Oxidation of isoprene	270
Oxidation of industrial NMHC	110
Oxidation of biomass NMHC	30
Oxidation of acetone	20
Sub-total in-situ oxidation	1230
Vegetation	150
Oceans	50
Biomass burning	700
Aircraft (Sutkis et al., 2003)	0.7
Fossil fuel	650
Sub-total direct emissions	1550
Total Sources	2780

24  
 25 Volatile organic compounds (VOC) include a wide variety of non-methane  
 26 hydrocarbons (NMHC) and oxygenated NMHC. There are three main sources:  
 27 (a) anthropogenic emissions (b) biomass burning, and (c) vegetation [IPCC,  
 28 2001] with vegetation supplying two-thirds of the global source, emitted primarily  
 29 in the tropics. VOC emissions from fossil fuel usage (approximately 25% of total  
 30 emissions) and biomass burning (about 5% of total emissions) have distributions  
 31 similar to NO<sub>x</sub>.

Table 3 VOC emissions \*MT-C/year (IPCC, 2001)

	VOCs	Isoprene	terpene	acetone
Fossil Fuel	161			
Biomass Burning	33			
Vegetation	377	220	127	30
Total	571			

We note that the major source of emissions are natural and often are temperature sensitive and thus represent a potential changing source in a climate change scenario.

## 2.3 Measurements

**2.3.1 Aircraft measurements:** The chemical composition of the UT/LS region has been explored by many missions using aircraft (compare Fehsenfeld et al., 2006 for a review of past work). One of the more recent experiment suites has been by the INTEX (Intercontinental Chemical Transport Experiment) mission. Singh et al. (2007), for example, found that there were occasions when the measurements of  $\text{NO}_x$  and  $\text{HO}_x$  and complementary species were not consistently modelled in the upper troposphere while in the lower stratosphere there was consistency between measurements and modelling. The disagreement suggests either errors in the measurements or perhaps more likely important species have not been definitively identified (such as  $\text{Br}_x$ ,  $\text{Cl}_y$ , aerosols) or error in rate constants which seems less likely.

There also has been important European research studying emissions from aircraft and their impact on atmospheric composition and climate in the UT/LS region. These studies included measurements of  $\text{O}_3$ ,  $\text{NO}_x$ , OH,  $\text{CH}_4$  and water vapour (e.g. Sausen et al, 2005; Gauss et al., 2004).

**2.3.2 Satellite experiments:** While aircraft campaigns reveal details of atmospheric processes satellite measurement programs with their associated validation campaigns provide global perspective. Since IPCC (1999) many satellites have been launched that have revealed the global nature of air quality in the troposphere and the status of the stratospheric ozone (e.g. Odin, Terra, Aura, SCISAT-I, ENVISAT). Most recently Calipso and CloudSat have added to the A-Train capability. MetOp, launched in 2006, has as its primary objective the gathering of meteorological data (~1 km vertical resolution of T and water vapour to about 10 km and a horizontal resolution of about  $10 \times 10 \text{ km}^2$ ) for improving weather forecasting. However, it also has an important air quality (AQ) capability via instruments such as GOME2, IASI and AVHRR ([http://www.esa.int/esaEO/SEM9NO2VQUD\\_index\\_0\\_m.html](http://www.esa.int/esaEO/SEM9NO2VQUD_index_0_m.html)) which can provide



1 column or partial column information on many chemical species such as NO<sub>2</sub>,  
2 CO, ozone, HNO<sub>3</sub> and aerosols of tropospheric and stratospheric interest. Global  
3 CO partial columns with ~ 20x20 km horizontal resolution have been measured  
4 by MOPITT (References for a suite of tropospheric instrument details and  
5 retrievals are given in IGAC, 2007). TES has reported partial vertical columns for  
6 CO and ozone (both tropospheric and stratospheric) while MLS/AURA operating  
7 in limb mode measures CO, as does AIRS. In spite of problems with tape on the  
8 front aperture the HIRDLS team have managed to obtain very interesting data on  
9 O<sub>3</sub>, H<sub>2</sub>O, CH<sub>4</sub>, N<sub>2</sub>O, HNO<sub>3</sub>, CFC-11, CFC-12, temperature, cloud top pressure,  
10 and four aerosol extinctions on a standard pressure grid with ~ 1 km vertical  
11 resolution for temperature (see  
12 <http://daac.gsfc.nasa.gov/Aura/HIRDLS/index.shtml>, and Massie et al., 2007).  
13 Many nadir viewing surface remote sensing instruments provide estimates of  
14 aerosol optical depth (AOD), e.g. MODIS (IGAC, 2007) and aerosol properties  
15 (Hu et al., 2007). Limb viewing instruments such as OSIRIS (Murtagh et al.,  
16 2002), ACE-FTS (Bernath et al., 2005), MAESTRO (McElroy et al., 2007), the  
17 SAGE suite (e.g. Wang et al., 2006; <http://www-sage3.larc.nasa.gov/>)  
18 measurements provide measurements of aerosols and cirrus clouds and sub-  
19 visible cirrus in the UT/LS region.

20  
21 Furthermore this information can be ingested into models via data assimilation  
22 and the information transported to other locations.

23  
24 Although the prime mission of many of the satellite programs mentioned above  
25 was not to address UT/LS issues important to aviation, careful improvements in  
26 retrieval method and validation programs have lead to the generation of  
27 quantitative measurements for the UT/LS region. For example, we note that the  
28 ACE-FTS instrument on SCISAT-I simultaneously measures 20-30 species down  
29 to 5 km including many species related to AQ and biomass burning and forest  
30 fires (Bernath et al., 2005). Likewise the MLS and TES instruments, both on  
31 EOS-Aura, are being used to address AQ issues, long range transport and  
32 impacts of large scale convection (Jiang et al., 2007). However the vertical  
33 resolution for the above instruments is at least 3-4 km (e.g. ACE). Nevertheless,  
34 instruments such as SAGE, OSIRIS, MAESTRO and HIRDLS have resolution ~  
35 1 km suitable for UT/LS studies.

36  
37 **2.3.3 Aircraft measurements investigating flight corridors:** The  
38 POLINAT (Pollution From Aircraft Emissions in the North Atlantic Flight Corridor  
39 (POLINAT) field campaign, using 3 aircraft and lasting from 1994 to 1998 was  
40 designed to assess the impact of commercial aircraft on the atmosphere in the  
41 vicinity of the eastern North Atlantic flight corridor by means of measurements  
42 (and modelling support) of the distribution of NO, NO<sub>x</sub>, NO<sub>y</sub> and other species  
43 (e.g. Schumann et al., 2000; Schlager et al., 1999; Singh et al., 1999; Ziereis et  
44 al., 2000). The measurements indicated strong latitudinal gradients in NO, NO<sub>x</sub>,  
45 and NO<sub>y</sub>. The research aircraft sampled plumes of various ages and saw clear  
46 signatures of air having been lofted from the lower atmosphere and also

1 signatures of lightning  $\text{NO}_x$ . Complementary modelling studies indicated that  
2 about 50% of the  $\text{NO}_x$  in the UT/LS was from the boundary layer. This should be  
3 compared with the later work on INTEX-A over north America where the results  
4 suggested that most of the  $\text{NO}_x$  was from lightning (10% from the boundary layer)  
5 (Singh et al., 2006; 2007). This difference may reflect the different meteorological  
6 situations over the summer hot convectively unstable central USA as compared  
7 to wave cyclone systems impacting Western Europe and eastern Atlantic or  
8 simply the uncertainty in the analysis. Cooper et al. (2006) found, after allowing  
9 for recent stratospheric intrusions using FLEXPART (Stohl et al., 1998; 2005),  
10 that ~ 75% of the upper tropospheric “ozone excess” over North America is  
11 generated by lightning  $\text{NO}_x$  emissions.  
12

13 The MOZAIC project, with instruments flown on commercial flights, (e.g., Cooper  
14 et al., 2006) flies in the flight corridors and represents a unique dataset for  
15 corridor studies. The MOZAIC package carries sensors to measure ozone,  $\text{NO}_y$   
16 compounds, water vapour, temperature and relative humidity: it has been flown  
17 on over 26,600 flights from 1994 to 2006 (e.g., Marenco et al., 1998; Nedelec et  
18 al., 2003; Volz-Thomas, 2005).  
19

## 20 **2.4 Modelling studies**

21  
22 Modelling studies are important for studying impacts of changing conditions  
23 either natural or anthropogenic. Current (and past) data are used for diagnosis  
24 and evaluation of models (e.g. IPCC2001, 2007) so that they may be used with  
25 confidence but with the limitations transparent.  
26

27 Our understanding of the UT/LS region of the atmosphere has been advanced by  
28 chemical and dynamical modelling using different but related models spanning  
29 many scales from microphysics of cloud formation to global scale transports.  
30 Global scale models include chemical transport models (CTMs), on-line weather  
31 forecast models and climate models. Mesoscale and cloud resolving models are  
32 also being applied in an attempt to improve our understanding of processes and  
33 in particular the exchange of water between the troposphere and stratosphere.  
34

35 **2.4.1 Plume to global scale modelling:** The injection of aircraft emissions  
36 into the atmosphere occurs at the engine scale but we seek the impacts on a  
37 global scale. Current global models are not able to resolve at this scale and the  
38 process of accounting for the injection of aircraft emissions has to be  
39 parameterized and this is usually done by using models that can resolve the  
40 scales from the engine to the plume scale (loosely defined). Mesoscale modelling  
41 is then required to assess the processes of dilution from the plume scale to the  
42 grid scale that might be typical of mesoscale processes. The impact of the  
43 various assumptions of the larger scale chemistry needs to be carefully  
44 assessed.  
45

1 The emitted jet engine exhaust plume is initially hot; it rapidly becomes trapped in  
2 the turbulent twin vortexes which meld in the wake and then dissipates into the  
3 background atmosphere on a longer time scale. The effect of wake dynamics on  
4 the dispersal of  $\text{NO}_x$  and  $\text{HO}_x$  was investigated by Lewellen and Lewellen (2001).  
5 It is important to consider the chemistry occurring in the aircraft plume and wake  
6 before it has been expanded to the model grid scale. Initial attempts to combine  
7 near field, far field, and global models in series (Danilin et al., 1997) were the first  
8 global impact studies to be based directly on detailed microphysics and chemical  
9 kinetics occurring in the aircraft plume and wake. From the perspective of ozone  
10 production in the UT/LS, removal of active species can occur by irreversible  
11 deposition of  $\text{HO}_x$  and  $\text{NO}_x$  source species or by conversion of more reactive  
12 nitrogen- and hydrogen-containing species into less reactive ones. Conversion of  
13  $\text{N}_2\text{O}_5$  into  $\text{HNO}_3$  on sulfate or ice particles is the best established example of the  
14 latter mechanism. Soot, sulfuric acid ("sulfate"), and water-ice particles are the  
15 main condensed-phase species found in the exhaust of jet aircraft. The extent to  
16 which aircraft aerosols offset the effects of aircraft  $\text{NO}_x$  emissions on  
17 atmospheric ozone depends on a variety of chemical and dynamical factors. The  
18 reactions are likely to be enhanced in the presence of increased atmospheric  
19 particulates in the plume, in contrails and in aircraft-induced cirrus clouds, which  
20 correspond to PSC type II ice from a chemical point of view. The most important  
21 heterogeneous reaction is the heterogeneous hydrolysis of  $\text{N}_2\text{O}_5$ . An important  
22 model simulation of effect of wake dynamics chemistry on  $\text{NO}_x$  and  $\text{HO}_x$   
23 concentrations showed that the effective  $\text{NO}_x$  emissions were reduced by about  
24 40% and the ozone perturbation by 30% (Valks and Velders, 1999).  
25

26 The interaction of plume and global modelling was also investigated by Kraabøl  
27 et al. (2002) who studied the impact of aircraft emissions in an early version of  
28 the Oslo CTM including a plume model (Kraabøl et al., 2000a; b). The plume  
29 model used was a multi-cylinder model to allow for mixing and with  
30 comprehensive tropospheric chemistry. They found that the oxidation of  $\text{NO}_x$  in  
31 the plume reduces the efficiency of aircraft  $\text{NO}_x$  emissions for ozone generation  
32 by converting  $\text{NO}_x$  to  $\text{HNO}_3$  (Kraabøl et al., JGR 2002) and must be taken into  
33 account.  
34

35 Meilinger et al. (2005) have developed a two-box plume model with complex gas  
36 phase and heterogeneous chemistry with a microphysical scheme. The two  
37 boxes, representing the plume and the background atmosphere, are allowed to  
38 mix using a turbulence time constant. They find that the development is quite  
39 sensitive to the conditions of the background atmosphere. They also find, similar  
40 to global scale models that above 210K hydrolysis of  $\text{BrNO}_3$  and  $\text{N}_2\text{O}_5$ , enhanced  
41 by virtue of the increased aircraft induced aerosol sulphate area, is important in  
42 the ozone budget. Below 210 K growth of background particles and suppressed  
43 chlorine activation is important.  
44

45 Kärcher et al. (2000) have presented an analytic parameterization for the  
46 development of nano-sized particles in the near field plume based on the amount

1 of chemi-ions emitted, the sulphuric acid (and also the S-conversion factor) and  
2 condensable organic species which can be couple with far-plume models such  
3 as that of Danilin et al. (1997) and 3-D models to estimate the growth of aerosols  
4 in the plume and impacts in the atmosphere.  
5  
6

7 **2.4.2 Global, regional, multiscale models:** Three-dimensional models used  
8 for studying atmospheric chemistry come in different flavours. Perhaps the least  
9 complex is the chemical transport model (CTM). In this type of model the main  
10 transport is by resolved winds supplied by another source such as weather  
11 forecast or climate model. The transport scheme adopted is critical as it must be  
12 mass conserving, stable, and not too computer intensive. Methods such as the  
13 Prather scheme (Prather, 1986) and the van Leer scheme (van Leer, 1977) seem  
14 to be among the most robust.  
15

16 Many models for the IPCC(1999) report (Penner et al, 1999) were tropospheric in  
17 domain but the nature of the aircraft problem suggests that the dynamical and  
18 chemical interaction between the troposphere and stratosphere should be  
19 accurately captured. For example, in the Northern hemisphere winter more than  
20 half of aircraft emissions are directly into the lowermost stratosphere (e.g. Köhler  
21 et al., 1997). For models which do encompass the troposphere and stratosphere  
22 the resolved circulation utilized is likewise critical as it must simulate the Brewer-  
23 Dobson circulation with fidelity and stratospheric/tropospheric exchange  
24 accurately (as for example in ozone transport to the troposphere) which is  
25 important with the transport of species such as water vapour to the stratosphere.  
26 It must also represent tropical and polar quasi-horizontal transport barriers. Other  
27 features that appear to be important will be the representation of the Asian  
28 summer monsoon in transporting material to the UT/LS region (e.g. Park et al.,  
29 2007). In addition, the Madden-Julian oscillation (MJO) which is a region of  
30 intense convection in the tropics (e.g. Miura et al., 2007) is linked to the delivery  
31 of chemical species, including water to the lower stratosphere via the TTL (e.g.  
32 Wong and Dessler, 2007). Baldwin et al. (2007) also point to the influences of the  
33 stratosphere on tropospheric dynamics and also the need for more  
34 comprehensive (in terms of including the stratosphere) models.  
35

36 We note that part of the ability to well represent these and other dynamical  
37 features will rest with the resolution adopted for the global CTM model as well as  
38 the metrological fields adopted. Currently the resolution for a state-of-the-art  
39 global CTM with comprehensive chemistry with both “stratospheric” and  
40 “tropospheric” chemistry is in the region of about  $1^{\circ} \times 1^{\circ}$  (and that is continuously  
41 improving with increasing computer power. Currently global weather forecast  
42 models are running with resolution  $\sim 25$  km (e.g. ECWMF) and  $\sim 33$  km (e.g.  
43 Canadian Meteorological Centre, Belair et al., 2007)  
44

45 Other transport processes are included such as the impact of the planetary  
46 boundary layer which is an important two-way filter between the surface and the

1 free troposphere. Large scale convection is parameterized by one of a number of  
2 schemes which can also be linked to lightning generation (e.g., Tost et al., 2007).  
3 Emissions are included but there is no standard emission suite, although several  
4 comprehensive data bases are available (e.g. GEIA, EMEFS). Comprehensive  
5 chemical schemes are now standard although what is often missing are  
6 interactive aerosol schemes and washout and rainout schemes can be quite  
7 primitive. Many species, such as ozone, are deposited on the surface with rates  
8 that depend on wind speed and surface roughness and surface type.

9  
10 Regional models (or limited area models, LAMs) have a role to play in studying  
11 the dissipation of plumes of emissions perhaps injected from convection towers  
12 or aircraft. Such models enable processes to be studied at higher resolution with,  
13 one expects, higher fidelity. However, a problem of how to validate (or evaluate)  
14 these models is an issue as there is not enough data available. Nevertheless,  
15 many air quality CTM regional models have operated in the past with very limited  
16 vertical domains and physical processes are not suitable for aircraft studies.

17 **Figures 3a and 3b** show NO and O<sub>3</sub> fields at mesoscale (15x15 km<sup>2</sup>) resolution  
18 using a global multiscale model, GEM-AQ (Kaminski et al., 2007) and give some  
19 idea of the heterogeneity of the region.

20  
21 One of the modelling tools that should be important in studying the different  
22 scales inherent in aircraft studies are multiscale models. These operate in (at  
23 least) two ways. One method is to run the model over the same time domain with  
24 higher and higher spatial (and temporal) resolutions and shrinking the interior grid  
25 such as has been done for air quality studies, for example, by MC2-AQ (Kaminski  
26 et al., 2002; Plummer et al., 2001; Yang et al., 2003) and also MM5 (e.g. Grell et  
27 al., 2000). A different approach is to have the interior high spatial resolution  
28 domain fixed within a lower resolution exterior domain which can be global or  
29 regional (e.g. Kaminski et al., 2007; O'Neill et al., 2007; Grell et al., 2005).

30  
31 McKenna et al. (2002) have developed a new Lagrangian model for the  
32 stratosphere, CLaMS (Chemical Lagrangian Model of the Stratosphere) where  
33 the quasi-horizontal flow is driven by meteorological analysis winds with cross-  
34 isentropic flow (vertical) driven by heating rates from a radiation calculation. The  
35 model has recently been extended to the surface using a hybrid coordinate  
36 allowing a transition from potential temperature to pressure coordinates  
37 (Konopka et al., 2004) and mixing is driven by wind shear.

38  
39 Another very useful analysis tool is the Lagrangian trajectory models such as  
40 FLEXPART (Stohl et al., 1998; 2005).

41  
42 **2.4.3 Weather forecast and Climate models:** CTMs are powerful tools for  
43 analysis and limited forecasting. However, for certain types of problems, such as  
44 the long terms effects of aviation, they are limited because they lack the  
45 capability to incorporate feedbacks. For tropospheric chemistry problems the  
46 main feed backs are via changing GHG fields such as methane and ozone.

1 Additionally, aerosols can interact with liquid and frozen water to alter cloud fields  
2 and thus impact solar and IR heating. In the stratosphere, changing water, CO<sub>2</sub>  
3 and ozone fields impact the heating. Modification of the heating function then  
4 alters the dynamical fields and transport characteristics.

5  
6 Of course, weather forecast and climate models (with and without interactive  
7 oceans) allow for some (and in some cases all) of the above feedbacks. And  
8 gaseous and aerosol fields have been added in many cases (Eyring et al.,  
9 2007b). Many models only treat the troposphere in detail (e.g., Stevenson et al.,  
10 2006; Kaminski et al., 2007) while others only have detailed chemistry with  
11 feedbacks for stratospheric species (e.g. see Eyring et al., 2006 for a suite of 13  
12 models). But even more comprehensive on-line models with both detailed  
13 tropospheric and stratospheric chemistry with feedbacks on the meteorology are  
14 being developed (see the list in Annex I) and will be necessary for future  
15 forecasts (cf. Baldwin et al., 2007).

16  
17 We note that for problems that deal with the UT/LS region that tropospheric  
18 models are not adequate. Furthermore, in order to develop an adequate Brewer-  
19 Dobson circulation the lid of the climate model must be above ~ 80 km  
20 (0.01hPa); the development of a quasi-biennial oscillation (QBO) signal requires  
21 higher vertical resolution in the stratosphere than most models have at this  
22 juncture. But that will soon change.

23  
24 These models are complex and, as noted above, it is important to have  
25 confidence in the similarities while understanding the differences, the latter of  
26 which permits an estimation of our confidence in model structures. In the recent  
27 past, there have been comparison experiments of models for IPCC reports but  
28 these have not focused on the UT/LS regions (e.g, Shindell et al, 2006;  
29 Stevenson et al., 2006). In addition, the application of important metrics to  
30 evaluate the models (such as STE of ~ 500 MT-O<sub>3</sub>/year) has only been applied in  
31 a limited fashion. This is partly because models have been in the throes of  
32 development; comparisons have been used more to deal with the issue of  
33 uncertainty in modelling rather than adopting a more systematic approach (e.g  
34 Wild, 2007). Along these lines we note that recently Eyring et al. (2006; 2007a)  
35 have published papers comparing the forecasts of chemistry climate models  
36 applied to the problem of stratospheric ozone depletion and recovery. Their  
37 study has underscored that there still are serious issues with models at the  
38 fundamental level of transport and emissions.

39  
40 There are other metrics or diagnostics that can be used as a measure of how  
41 well a model is accurately representing the atmospheric situation modelled.  
42 Some of these are discussed by Pan et al. (2007) for example. CO varies rapidly  
43 across the tropopause with larger values in the troposphere. Similarly, ozone  
44 varies rapidly across the troposphere but with more in the stratosphere. Thus  
45 CO/O<sub>3</sub> correlations represent a very useful diagnostic to test the  
46 representativeness of model transport. Similarly, in the UT/LS regions, NO<sub>y</sub>/N<sub>2</sub>O

1 correlations can be revealing of model transport. And as noted above  $\text{NO}_x/\text{HNO}_3$   
2 ratios in the UT should be very variable, ranging from  $> 1$  to  $< 1$  depending on  
3 how recent convection and lightning have occurred. Forming a suitable PDF for  
4 measurement and model ratio might be a useful diagnostic.

## 5 **2.5 Future climate impacts**

6  
7 **2.5.1 Climate change:** In the context of climate change it is to be expected that  
8 a future fleet will operate in a different atmosphere both in terms of meteorology  
9 (transport) and chemistry. In addition, these changes will also impact the  
10 chemistry of the UT/LS region. So that it is important to have a better  
11 understanding of changes in relative humidity in the UT/LS region (of course this  
12 also impacts contrail formation and the development of cirrus clouds.). This  
13 problem is dealt with in a different white paper. However, there are also obvious  
14 impacts on the chemistry via  $\text{HO}_x$  abundances and the production and loss of  
15 ozone and also its impact on the radiation budget in the TTL region. However,  
16 there are also chemistry-related impacts on humidity and that relates to the  
17 supply of freezing nuclei to the UT/LS region either from the lower troposphere  
18 via delivery of aerosols or from the stratosphere and mesosphere via sulphate or  
19 metallic ions generated in the mesosphere. These are issues that must be  
20 resolved in an integrated fashion rather than thought of simply as chemical or  
21 transport issues.  
22

23 Future climate is likely to be different: this means that it can be anticipated that  
24 winds (transport times), cloud processes, water amounts,  $\text{NO}_x$  generation by  
25 lightning will be different. And we have noted that the relative contributions of  
26 aircraft  $\text{NO}_x$  and lightning  $\text{NO}_x$  (with a small contribution from stratospheric  $\text{NO}_y$ )  
27 are important. Nevertheless, aircraft impacts cannot be divorced from possible  
28 impacts of future surface emissions. Most climate studies using standard  $\text{CO}_2$   
29 increase scenarios suggest that the main impact in the future will be due to  
30 increased anthropogenic emissions (e.g. Stevenson et al., 2006) with climate  
31 impacts on biogenic emissions being of secondary import. However, with a more  
32 rapid rise in  $\text{CO}_2$  and increased temperatures biogenic emissions grow in a non-  
33 linear fashion to become more important. In the future, anthropogenic, biogenic  
34 and biomass burning emissions, are expected to grow as population expands  
35 and the economies of India and China grow. And we note that current  $\text{CO}_2$   
36 emissions are growing faster than envisaged by any of the IPCC scenarios.

37 In future climate change scenarios, cloudiness is projected to decrease at  
38 latitudes below about 50 degrees and to increase at latitudes above 50 degrees  
39 (IPCC; 2007.p767). These changes will affect the radiative forcing from aviation  
40 ozone on a regional basis.

41  
42

43 **2.5.2 Future aircraft emissions:** The emissions from aircraft will depend on  
44 the composition of the fuel. For example the sulphur fraction can impact the

1 chemistry in the plume and also the generation of aerosols which are deposited  
2 in the UT/LS and may impact chemistry and cirrus formation. In the future this will  
3 depend on the development of new engines and constraints imposed by noise  
4 control for take off and landing and possibly of aircraft flying a few kilometres  
5 higher (not SSTs). For example, over the next 20 years the replacement of old  
6 aircraft with new aircraft such as the Airbus 380 and the Boeing 787 may impact  
7 future emission scenarios. Estimates of annual fuel use by 2020 annual for  
8 commercial air traffic are ~ 350 MT or 2.6 times the estimated fuel use by the  
9 global 1999 commercial fleet. This translates into global NO<sub>x</sub> emissions of ~ 1.5  
10 MT-N from commercial air traffic or about 2.8 times the estimated 1999 NO<sub>x</sub>  
11 emissions levels. At the same time total revenue passenger kilometers are  
12 projected to increase from 3,170 billion in 1999 to 8,390 billion in 2020, or by a  
13 factor of 2.65 (Sutkus et al., 2003) These estimated increased emissions are  
14 expected to lead to global increase in ozone production with the potential for  
15 larger regional effects.

16  
17 Perhaps of more importance is the advent of new air traffic control mechanisms  
18 to cut “wasted” air time and reduce fuel consumption and perhaps also the  
19 development of new routes both intra- and inter-continental. Of particular  
20 importance of the development of new routes may be the impact of increased  
21 flights over the Arctic regions, a possible impact which needs to be explored (e.g.  
22 Gauss et al., 2006).

23  
24 There is also the possibility of the emission character of a future fleet to be  
25 considered. The European Commission within the fifth Framework Programme  
26 on competitive and sustainable growth supported the CRYOPLANE project to  
27 investigate aircraft fueled with liquid-hydrogen (LH<sub>2</sub>). Since there would be no  
28 hydrocarbons (HCs) or sulphur in the fuel these type of aircraft would not emit  
29 CO<sub>2</sub>, CO, soot or SO<sub>2</sub>. They would emit NO<sub>x</sub> and water. While the NO<sub>x</sub> emissions  
30 are likely to decrease the water emissions would increase ~ 2.6. Gauss et al.  
31 (2003) investigated the impact of cryoplanes for the year 2015 using the Oslo  
32 CTM. They find that the replacement aircraft would increase water vapour near  
33 the tropopause by about 250 ppbv. Although we should note that with the current  
34 lifetime of a fleet of aircraft being ~ 30 years LH<sub>2</sub> planes will not be a major  
35 component of the commercial fleet for quite a few years to come.

36  
37 Recent work by Søvde et al. (2007) has looked at the impact of a “mixed” fleet  
38 using an emission scenario for 2050. The “mixed” refers to a combination of  
39 subsonic and supersonic aircraft. They found impacts on the ozone due to  
40 aerosol emissions. Their analysis is limited from a climate perspective as they  
41 used meteorology for the year 2000 for both current and future scenarios.

42  
43 Connectivity: Transport issues identified here are also important for water vapour  
44 and related meteors, such as cirrus, while cirrus may play a role controlling NO<sub>x</sub>  
45 distribution in the UT/LS. The aerosol, whether sulphate, organic, soot may play  
46 an important role in the chemistry of the engine vortex and plume and generation  
47 of ozone. However, perhaps more important is their putative role as FN/IN for



1 cirrus. Again, transport is an issue.

## 2 **3. Focus on uncertainties**

3

4 In this section we address the issues of uncertainties in the basic science issues  
5 concerning impacts of aircraft but with a focus more on chemistry and transport  
6 issues but include some brief comments on radiative forcing.

7

### 8 **3.1 Chemistry and emissions**

9 For chemistry the issues of major uncertainty include an accurate knowledge of  
10 the tropospheric ozone budget, HO<sub>x</sub>, NO<sub>x</sub> chemistry in the UTLS which may be  
11 impacted by issues of convection lofting precursor species, possibly aerosols and  
12 affecting loss by washout. A related cloud issue is the role of HNO<sub>3</sub> take-up on  
13 cirrus clouds. In addition, the role of halogen chemistry is lurking on the sidelines.  
14 Certainly lower stratospheric Br<sub>y</sub> distribution (and sources) need to be resolved  
15 as does the Cl<sub>2</sub>O<sub>2</sub> dimer problem in springtime polar regions.

16

17 The issue of the uncertainty of the background NO<sub>x</sub> emissions from lightning in  
18 the UT region and convection from the boundary layer to the UT appear to be the  
19 major issues and related corridor issues.

20

21 3.1.1 Chemistry: From section 2 it is clear that over the past decade  
22 measurements in the UT have revealed problems with our understanding of the  
23 NO<sub>x</sub>, HO<sub>x</sub> budget. Recent work as part of the INTEX-A campaign has been  
24 presented by Singh et al. (2006) (see also Ren et al., 2006) who, for example,  
25 note that OH measurements are substantially lower than model values for the  
26 INTEX and other campaigns (see [Figure 4](#)). This uncertainties may be related to  
27 issues of steady state (used for analysis) versus a more dynamic non-steady  
28 state chemical regime driven by removal of HNO<sub>3</sub> within clouds and NO<sub>x</sub>  
29 replacement by lightning and STE. Or it may be due to the lofting of precursors  
30 which may have not be measured or have escaped detection.

31

32 Issues regarding the importance of halogen chemistry are much less well  
33 defined, but could be playing a role in the HO<sub>x</sub>/NO<sub>x</sub> UT chemistry issue. For  
34 example hydrolysis of BrNO<sub>3</sub> does lead to the creation of more HO<sub>x</sub>. But at this  
35 point is more speculation on the authors' part.

36

37 Freezing nuclei (FN) or ice nuclei (IN) act as sources for the sublimation of water  
38 in the UT. The main sources are likely to be the lower troposphere although there  
39 is an influx of stratospheric sulphate aerosols via STE and also the possibility of a  
40 cosmic ray ionization contribution to aerosol and thus ice formation. Thus it  
41 becomes important to understand and quantify the fraction of aerosols generated  
42 that become IN and this requires more study. The issue of lower tropospheric  
43 sources of aerosols and their size distribution is likewise important although  
44 processing in clouds tends to produce aerosols in the 0.1-1.0 micron range. Thus

1 quantification of direct sources of aerosols is quite uncertain although the  
2 situation is improving. The issue of the generation of secondary aerosols is  
3 somewhat more uncertain, e.g. SO<sub>2</sub> to sulphate oxidation and, as noted above  
4 the issue of the generation of secondary organic aerosols in the UT is still an  
5 issue for research but the empirical evidence suggests that their abundance is  
6 related to tropical and Boreal fires.

7  
8 The status of our knowledge of the ozone budget in the UT/LS still is more  
9 uncertain that one would like especially for the estimates of potentially small  
10 impacts of aircraft. From the above discussion we consider that the percentage  
11 contribution of air traffic emissions versus natural and other anthropogenic  
12 sources is quite uncertain in the UT.

13  
14 Although we have not dealt with it, the uncertainty in the physics and  
15 microphysics of water vapour in the UT/LS is important for chemistry and ozone  
16 generation, radiative forcing and generation and sustenance of cirrus clouds.

17  
18 And related to this is the issue of the impact of HNO<sub>3</sub> on freezing of water vapour  
19 into cirrus crystals is still uncertain.

20  
21 3.1.2 Emissions: In terms of the global NO<sub>x</sub> source the contribution from aircraft  
22 is only a few percent. However, as noted above, as viewed from the situation of  
23 the UT/LS the contribution of aircraft, ~ 0.7 MT-N/year is rather larger (~ 7%)  
24 measured against the context of ~ 4 MT-N/year from lightning in the UT, ≥ 4 MT-  
25 N/year from large scale convection (using a 10% delivery fraction) and resolved  
26 transport and 0.8 MT-N/year from STE (N<sub>2</sub>O and cosmic rays). However, the  
27 delivery of the aircraft NO<sub>x</sub> is to a relatively narrow corridor which tends to  
28 enhance its impact so that locally the impact could be much larger (e.g.  
29 Schumann et al., 2000). The uncertainty in the STE of NO<sub>y</sub> in terms of absolute  
30 amount seems rather small as it is largely constrained by the stratospheric  
31 source. However, the lightning source is still rather uncertain, although there  
32 appears to some convergence on the absolute amounts. Interestingly, the lower  
33 bounds that have appeared (~ 2 MT-N/year) are such that it increases the  
34 potential impact of aircraft. This is certainly an area that requires further study,  
35 both in terms in absolute amounts and also the vertical distribution of the source.  
36 If, as seems likely, delivery of NO<sub>x</sub> by lightning often occurs in association with  
37 convective activity then the contributions from transport from the surface,  
38 concomitant with washout of soluble components such as HNO<sub>3</sub>, and in-situ  
39 lightning source tends to make the unambiguous evaluation of separate lightning  
40 and convection sources difficult. It also means that the knowledge (or lack) of  
41 lower tropospheric NO<sub>x</sub>/HNO<sub>3</sub> ratio takes on a greater importance. This ratio is  
42 impacted by heterogeneous chemistry and knowledge of the aerosol distribution.  
43 Furthermore the status of the amount and distribution of anthropogenic, biomass  
44 burning and biogenic tropospheric sources becomes important.

45

1 As noted in section 2, estimates of delivery of NO<sub>x</sub> from the boundary layer are  
2 uncertain and range from 10% to 50%, although this may simply reflect the  
3 different dynamical conditions over summertime north America and Europe: this  
4 may also change in a future climate state. As noted above the fraction of aircraft  
5 NO<sub>x</sub> to other sources for the year 2000 was about  $0.7/(4+5+0.8) \sim 7\%$ . As noted  
6 in the introduction, for the time frame 2020/2030 aircraft NO<sub>x</sub> emissions are  
7 expected to increase to  $\sim 1.5$  MT-N (Sutkiss et al, 2003). Anthropogenic emissions  
8 are expected to rise by a factor of 1.5 to 2 depending on the scenario followed  
9 (e.g. Dentener et al., 2005) which using 10% for the contribution to the UT give a  
10 source of  $\sim 7.5$  to 10 MT-N/year. Even though one might anticipate more  
11 convective activity and thus more lightning in a future climate state based on  
12 energy considerations we will take the lightning contribution as fixed. In which  
13 case the ratio of aircraft to other sources in the UT is about  $1.5/(4+0.8+7.5)$  or  
14 about 12% or less. So that the relative impact of aircraft in 2025 could be higher  
15 than at present. Moreover, if economic conditions are not conducive to such  
16 growth it is likely to impact both the air transportation sector as well as other  
17 sources of anthropogenic NO<sub>x</sub>.

### 19 **3.1.3 Comprehensive Tropospheric/stratospheric Chemistry models:**

20 In the IPCC1999 report most of the 3D CTMs used in the study had either  
21 comprehensive tropospheric or stratospheric chemistry with background  
22 tropospheric chemistry but not both. There were few climate models with  
23 comprehensive tropospheric or stratospheric chemistry. Now both CTMs and  
24 climate models are incorporating comprehensive tropospheric and stratospheric  
25 chemistry (see annex I). Future studies of aircraft models will need to include  
26 comprehensive UT/LS chemistry.

### 27 **3.2 Measurements - species and winds – and analysis**

28  
29 It is still important to have aircraft campaigns since they can provide detailed and  
30 comprehensive in situ measurements. However, any analysis and  
31 parameterizations derived from these need to be verified for a variety of  
32 conditions. Thus the use of satellite measurements to evaluate the information on  
33 a global scale seems important. However, what is required are instruments to  
34 measure species and temperature with high vertical resolution, and in the context  
35 of contemporary experiments (see above) this means vertical resolution  $\sim 1$  km  
36 for UT/LS measurements. These types of instruments are under study using  
37 millimetre and mid-IR in Europe and one of the experiments on ESA's list of  
38 Explorer possibilities is PREMIER (PRocess Exploration through Measurements  
39 of Infrared and millimetre-wave Emitted Radiation)  
40 ([http://www.esa.int/esaCP/SEMhQH9ATME\\_Protecting\\_0.html](http://www.esa.int/esaCP/SEMhQH9ATME_Protecting_0.html)) which could  
41 produce 3D imaging of water vapour, ozone, and other species as well as  
42 temperature, in the UT/LS and with the required vertical resolution. Interestingly,  
43 one of the goals of PREMIER is to operate synergistically with METOP (see  
44 above) together with use of models and data assimilation systems to transfer the  
45 information to the lower troposphere.

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In addition, to attack the relative humidity problem in the UT, there needs to be improved instrumentation to measure air ambient temperature accurately (and precisely) to  $\sim 0.2^\circ\text{K}$ .

Of course from a dynamical point of view, which is critical, improved wind measurements are important and ADM-aeolus ([http://www.esa.int/esaLP/SEM3Y0LKKSE\\_LPAdmaeolus\\_0.html](http://www.esa.int/esaLP/SEM3Y0LKKSE_LPAdmaeolus_0.html)) will provide improved wind measurements in the near future using Doppler wind lidar measurements with a late 2008 launch. In addition, the Canadian Space Agency is still planning to launch SWIFT (Stratospheric Wind Interferometer For Transport) (<http://www.space.gc.ca/asc/eng/sciences/swift.asp>) which uses ozone lines in the mid-IR and a Michelson interferometer to measure winds in the lower stratosphere.

For the past number of years it has been a goal to use chemical measurements to improve wind estimates. Recent work between ESA, Environment Canada and BIRA (Belgisch Instituut voor Ruimte-Aëronomie, Institut d'Aéronomie de Belgique: BIRA-IASB, Belgium) (e.g. de Grandpré et al., 2007) has used 4DVar and assimilation of MIPAS ozone data to improve wind estimates in the lower stratosphere using the Canadian weather forecast model, GEM (Global Environmental Multiscale) (Belair et al., 2007), and the BIRA chemical module. This is encouraging for future meteorological data. However, it is not clear in the coming years that there will be enough satellites available to provide the necessary chemical data. This is a serious problem (that also will impact the monitoring of the stratosphere and ozone recovery over the next decades).

From the above discussion it is clear that the lower atmosphere has a major impact on the UT/LS and the UT in particular on relatively short time scales. One the major impacts is the transport of species important in the ozone budget such as CO and NO<sub>x</sub>. However, bottom up budgeting of emissions, while useful, is still quite uncertain (uncertainty depends on species). Top down assessment of tropospheric emissions (e.g. Martin et al., 2007; Jaegle et al., 2005) using satellite data, while it has limitations, is proving very useful and continued work should improve knowledge of surface emissions. Thus satellites dedicated to air quality studies such as TRAC (TRopospheric composition and Air Quality) ([http://www.esa.int/esaCP/SEMHQH9ATME\\_Protecting\\_0.html](http://www.esa.int/esaCP/SEMHQH9ATME_Protecting_0.html)) or that can be combined with other information is being done using GOME, SCHIACHY etc (Martin et al, 2007) data should be continued (see comment on PREMIER and METOP mission above).

With the rapidly changing climate over polar regions and the Arctic in particular, and with the potential for increased air traffic over the Arctic, it would seem prudent to have been observing capability for meteorology (climate) and chemical species over this region. Thus there has been recent activity to study the use of Molniya orbits (e.g. Riishojgaard, 2005). It is clear that imaging nadir viewing FTS

1 mid-IR instruments, multi-channel near UV, Visible, NIR imagers and  
2 SCIAMACHY-type instruments can provide detailed capability to monitor this  
3 region with quasi-geostationary viewing. Several satellites with 12 hour orbits can  
4 provide continuous coverage of polar regions and extensive coverage down to  
5 mid-latitudes.  
6

### 7 **3.3 Modeling capability**

8  
9 Since the IPCC 1999 report models have improved. However, actual model  
10 performance on NO<sub>x</sub> in the UT has not improved over the last decade (e.g.,  
11 Singh et al, 2007). The availability of multiscale models which can be used to  
12 study plume dispersion problems has improved. Increasing computer power and  
13 storage has enabled much improved resolution for the combination of chemistry  
14 and meteorology. Gas phase and aerosol chemistry running together is now  
15 almost standard.  
16

17 It is important to recall that the aviation problem is not run in isolation so that any  
18 improvement in the basic atmospheric model will lead to a better assessment of  
19 aircraft impacts. Thus we expect improved spatial resolution to continue.  
20 Currently weather forecast models are being used globally ~ 25-35 km horizontal  
21 spatial resolution (e.g. Belair et al., 2007; Jung and Leutbecher, 2007). It is likely  
22 that in the next year or two it will be possible to run global chemical weather  
23 models with similar horizontal resolution. Of course one of the problems will be  
24 the evaluation of models at this resolution which is why such as satellite with the  
25 features of PREMIER (see above) would be invaluable. In addition to improved  
26 horizontal resolution it will be important to have concomitant improved vertical  
27 resolution in order to better represent transport from the troposphere to the  
28 stratosphere and also vertical wave propagation.  
29

30 There will be associated problems to be solved with the vast amounts of data  
31 generated. Some will be associated with storage, access and transfer, problems  
32 of analysis – simple comparison is limited and limiting, and, as discussed above,  
33 there will need to be much improved protocols (including correlations) for  
34 comparing chemical weather and chemistry climate models with each other and  
35 with data. Statistical analysis such as use of PDFs may become more common.  
36

37 The distinction between weather forecast and climate models is fast eroding as  
38 weather forecast models are run out for longer times, have improved heating  
39 codes, include ozone chemistry, have better surface schemes. However, in the  
40 short term chemistry climate models will be useful for longer simulations and thus  
41 have lower spatial resolution. Some of the issues surrounding the magnitude of a  
42 climate signal can be temporarily addressed by the using chemical weather  
43 models in a time-slice mode run with appropriate (future) SSTs from ocean-  
44 atmosphere models.  
45

1 Climate chemistry models are in a state of flux at the moment. As noted above  
2 SPARC comparison studies CCMval (Chemistry-Climate Model Validation  
3 Activity for SPARC) (<http://www.pa.op.dlr.de/CCMVal/>) (Eyring et al., 2006;  
4 2007a) have revealed a number of problems. One such problem is the status of  
5 the total chlorine,  $Cl_y$ , distribution of the models in the stratosphere. The model  
6 distributions are significantly different and the problems certainly involve  
7 transport, dynamical, age-of-air issues that need to be resolved. In addition, the  
8 input of the CFCs and related species may also be contributing to the  
9 discrepancies. The problem may be that most models use a few CFCs to  
10 represent the total input of chlorine, all with different lifetimes. Thus each model  
11 may have a distinctly different input function with an associated different time  
12 constant for flushing from the stratosphere aside from any issues regarding age  
13 of air. In addition Waugh et al. (2007) emphasize the importance of resolved  
14 transport in the lower stratosphere and the interaction between that and  
15 resolution and the representation of transport barriers.

16  
17 There is a related issue with the total amount of bromine in the stratosphere.  
18 There is an important role for halogenated very short lived species and their  
19 contribution needs to be (a) resolved by measurements and (b) included in all  
20 models.

21  
22 **3.3.1 Resolved Transport Issues:** One of the issues mentioned above is that  
23 of an apparent discrepancy in  $Cl_y$  distributions between models. Some of this  
24 may be attributed to the internal dynamical properties of the model (winds etc)  
25 which translate to different age-of-air between the models (e.g. Waugh et al.,  
26 2007). However, some of the discrepancy is likely due to differences in transport  
27 schemes (e.g. Wild et al., 2007). Some schemes such as semi-Lagrangian are  
28 efficient but sometimes do not have adequate conservation properties, while  
29 spectral methods, which generally have good conservation properties when  
30 strong gradients are not an issue, have problems when strong horizontal  
31 gradients create Gibbs fringes and then various types of hole-filling techniques  
32 are applied (and rarely discussed) in an attempt to maintain mass conservation  
33 which can have various disturbing pathologies. And of course, there is the  
34 associated issue of the positive part of the Gibbs fringe – how to constrain it?  
35 This is not simply a stratospheric issue. Strong species gradients arise in the  
36 troposphere (e.g. rainout of soluble species is very “spotty”, in the stratosphere  
37 uptake of various species on PSCs can be very irregular, and in the mesosphere  
38 and stratosphere the formation of a strong vortex and associated descent of low  
39 mixing ratio air can create very strong horizontal gradients. Some of these issues  
40 can be ameliorated by going to higher resolution but it seems more reasonable  
41 that the community should start using more robust (if more computationally  
42 expensive) and reliable transport schemes such as the Prather scheme (Prather,  
43 1986) or the van Leer scheme (van Leer, 1977). While some variety of transport  
44 schemes is useful, even necessary as it allows for uncertainties and variation  
45 between models some more rigorous comparisons seem necessary.

46

1 A related issue is transport by sub-grid scale motions such as large scale  
2 convection. In CTMs these schemes are often/sometimes applied in an  
3 inconsistent manner as the parametrizations as the resolved winds utilized to  
4 drive the model will often have used a different parameterization and most likely  
5 at a different resolution as that used in the CTM. It would be useful to quantify the  
6 impact of such uncertainties.  
7

### 8 ***3.4 Radiative Forcing comments***

9  
10 The increased emissions from future air traffic will alter radiative forcing due to  
11 generation of increased ozone, increased carbon dioxide and associated  
12 decreases in methane concentrations. The major change will be due to the  
13 increased ozone in the upper troposphere, particularly in aviation corridor regions  
14 which, to some extent, is counteracted by a globally reduced methane  
15 abundance due to increased OH. And of course the relevant time constants for  
16 ozone and methane are quite different. This issue was identified at the time of the  
17 IPCC (1999) report. A recent study by Stevenson et al (2004) applied a  
18 chemistry-climate model to study the radiative forcings generated by aircraft NO<sub>x</sub>  
19 emissions through changes in ozone and methane. They injected pulses of  
20 aircraft NO<sub>x</sub> the model for a various months and investigated the impacts on  
21 ozone and methane radiative forcings. Given that the spatial distributions of the  
22 RF from aviation ozone and the RF from methane will be considerably spatially  
23 different the question remains how much compensation will occur.  
24

25 The mechanism is the increase in radiative forcing at the earth's surface which  
26 causes surface and atmospheric heating. The real verification of the climate  
27 impact of increased upper troposphere ozone is the detection of changes in the  
28 ozone radiative forcing at the surface and at the top of the atmosphere.  
29 Radiative Forcing (RF) calculated at the top of the troposphere at the tropopause  
30 is used as a metric for measuring the potential for climate change. And it is often  
31 given as a simple global number (NRC, 2005). However, RF is far from uniform  
32 as is clear from the regional changes in ozone which translate into regional  
33 changes in RF (cf. Figure 2), as opposed to the situation for methane where,  
34 because of its ~ 10 year lifetime, the changes in methane, and so the RF, is  
35 much more spatially uniform. Clouds can cause large changes in RF and  
36 represent a major uncertainty on the model calculation of RF.  
37

38 There are difficulties in detecting changes in the IPCC radiative forcing metric  
39 because of the way in which it is defined at the top of the troposphere at the  
40 tropopause. There are large uncertainties in the calculations of the radiative  
41 forcing metric due to a lack of knowledge of cloud effects. There need to be  
42 verifications of the radiative forcing metric by comparison against real  
43 measurements of observed surface radiative forcing and with satellite radiative  
44 trapping at the top of the atmosphere. This will need to be accomplished by  
45 concurrent simulations of surface forcing and top of the atmosphere radiative

1 trapping with the same climate models used to calculate the RF metric (Puckrin  
2 et al, 2004).  
3

4 In addition, we note that RF can be readily *measured* at the surface where it is  
5 used as more of a climate observable than a metric. For example, the current  
6 network of pygeometers measures the long wave component of surface radiative  
7 forcing. Philipona et al. (2005) have detected the increase in total surface  
8 radiative forcing from the increase in all of the greenhouse gases. The surface  
9 forcing from tropospheric ozone itself has been measured by Evans et al. (1999)  
10 using spectral measurements of long wave radiation. The effects of clouds on  
11 surface radiative forcing need to be investigated by extensive ground  
12 measurements of surface radiative forcing by the individual greenhouse gases  
13 and particularly by tropospheric ozone. Satellite measurements of nadir outgoing  
14 long wave radiation can also be used to investigate the effects of clouds on the  
15 top of the atmosphere radiative forcing (Harries et al, 2001).

16  
17 Measurements can supply RF on a regional basis. The regional nature was aptly  
18 demonstrated by the change in surface temperature range (and RF) from Sept  
19 11-14 with no aircraft flying. For three days after September 11, the Federal  
20 Aviation Administration grounded commercial aircraft in the U.S.; there was an  
21 anomalous increase in the average diurnal temperature range of 1.2°C for the  
22 period Sept. 11-14, 2001, a change not matched in the last 30 years (Travis et al.  
23 (2002). This temporary “climate change” was due to decreased surface radiative  
24 forcing from both contrails and tropospheric ozone.  
25

### 26 **3.5 Future Climate and related feedbacks**

27  
28 For assessments of future impacts of aviation, i.e. which might occur in a future  
29 climate, chemistry-climate models need to be used in order to permit feedbacks;  
30 CTMs will not permit feedbacks and hence the need for metrics such as radiative  
31 forcing as a proxy for change. This will apply for our subject area(s) of interest,  
32 both chemistry and transport. However, if the changes are small, several long  
33 runs will be required, with slightly different boundary conditions (such as SSTs) in  
34 order to extract statistically significant findings. This is necessary since emissions  
35 will be changing, the ozone layer will be changing, STE may alter the influx of  
36 stratospheric ozone to the troposphere, lightning production of NO<sub>x</sub> may change  
37 in a future climate and the strength of the Brewer Dobson circulation may alter,  
38 affecting water vapour amounts in the UT/LS region and this will modify the  
39 formation and impacts of contrails, contrail-cirrus and the background cirrus  
40 clouds.  
41

### 42 **3.6 Interconnectivity with other SSWP theme areas**

43



1 The same type of measurements and regional modelling for water vapour and  
2 NO<sub>x</sub> are required for the contrail cirrus radiative forcing theme area. Modelling of  
3 plume dispersion into the ambient atmosphere is also a common problem with  
4 the contrail cirrus theme. Another common link is the upward convection  
5 transport of smoke from biomass fires to provide CCN Cloud Condensation  
6 Nuclei) for the formation of cirrus in the UT.  
7

## 8 **4. Priorities**

9 The following list summarizes the major uncertainties from section 3. In the list  
10 below, HP, MP and LP stand for highest priority, medium priority, and lower  
11 priority, respectively.

12

### 13 **Chemistry**

- 14 1. Our understanding of NO<sub>x</sub>/HO<sub>x</sub> chemistry in the UT is uncertain;  
15 measurements are not well reproduced by model simulations and may  
16 also be influenced by items (4) and (5) below and possibly (2) and (3).  
17 (HP)
- 18 2. There is a need to improve our understanding of the impact of background  
19 aerosols and those from aviation emissions on the background  
20 constituents. They can alter the NO<sub>x</sub> and ClO<sub>x</sub> chemistry with resulting  
21 changes in regional ozone in the UT/LS. (HP)
- 22 3. Improved estimates of the potential role of halogen chemistry are required.  
23 (MP)
- 24 4. Conversion or uptake of HNO<sub>3</sub> on cirrus clouds needs to be better  
25 understood. (HP)

26

### 27 **Emissions**

- 28 5. The relative contribution of aviation NO<sub>x</sub> to NO<sub>x</sub> from lightning and NO<sub>x</sub>  
29 lofted by convection from boundary layer pollution sources in the flight  
30 corridors is uncertain and needs to be better defined. (HP)
- 31 6. Chemistry processes in the UT are strongly influenced by convective  
32 processes for which species ratios can be used as measures of model  
33 performance. (HP)

34

### 35 **Models, multiscale, global and climate**

- 36 7. More plume to regional scale models for plume processing of NO<sub>x</sub>  
37 capability are required as there are very few groups at present capable of  
38 making detailed calculations. (MP)
- 39 8. Model representations of vertical transport processes from surface to  
40 tropopause need improvement (see above). (HP)
- 41 9. Ideally, models to address the climate problem should be climate models  
42 with comprehensive tropospheric and stratospheric chemistry in order to  
43 better incorporate chemical and dynamical feedbacks. If CTMs are used  
44 then they also should have comprehensive tropospheric and stratospheric  
45 chemistry. (HP)

- 1 10. Ideally, models should be able to address/simulate dynamical issues such  
2 as the Asian monsoon and the Madden-Julian oscillation in order to  
3 properly characterize upward tropical transport (MP).  
4 11. Predictions of future climate conditions, composition and emissions are  
5 needed but should be addressed with (8) above. (HP)

6  
7 **Measurements**

- 8 12. Measurements of vertical profiles in UT with high vertical and horizontal  
9 resolution are required and there is a role for O<sub>3</sub>, NO<sub>2</sub> and CO sondes.  
10 (HP)  
11 13. There needs to be support for continued use of current satellites and  
12 analysis of concomitant data for now and new satellite instruments in the  
13 future. (HP)

14  
15 **Radiative Forcing Issues**

- 16 14. Verification of the radiative forcing metric for ozone and methane is  
17 needed and cloud effects need to be quantified. (MP)

18  
19 The understanding of aviation impacts on the climate system clearly requires a  
20 deep understanding of the *natural* atmosphere. Thus much of the above list  
21 itemizes a lack of understanding of the natural atmosphere. The following  
22 outlines ongoing work and future plans that will assist in improving our  
23 understanding of the atmosphere of the atmosphere in the UT/LS. Of course the  
24 UT/LS does not exist in isolation. Thus improved understanding of the  
25 troposphere and stratosphere in general are important.

26  
27 **Ongoing measurement programs**

28  
29 Clearly it is important to continue monitoring in concert with modelling and  
30 analysis. This provides continuity with current measurements and allows the time  
31 series to be extended into the future (e.g. AERONET (Aerosol Robotic Network)  
32 (<http://aeronet.gsfc.nasa.gov/>). Maintenance of ozonesonde capability and in  
33 particularly programs such as SHADOZ (Southern Hemisphere Additional  
34 Ozonesondes) (Thompson et al., 2003a, b; <http://croc.gsfc.nasa.gov/shadoz/>) in  
35 concert with modelling have allowed important new understanding of tropical  
36 ozone behaviour and IONS (INTEX ozone networks study) (Thompson et al.,  
37 2007a, b; <http://croc.gsfc.nasa.gov/intexb/ions06.html>) for ozone transport and  
38 the frequency of STE over North America.

39  
40 In addition, there should be continuing support for analysis of current satellite  
41 experiments/instruments that have the capacity to probe the UT/LS, viz., ACE,  
42 MAESTRO, MIPAS, OSIRIS, SMR, MLS, OMI, SCHIAMACHY, TES, AIRS, IASI,  
43 HIRDLS and also instruments such as MODIS, MISR, AVHRR etc that can  
44 provide aerosol column optical depth will be important to maintain continuity into  
45 the future of basic tropospheric science. But we note that in 3-5 years there will  
46 be limited satellite availability that will be useful for UT/LS studies which is

1 particular to aviation impacts.

### 3 **Future work**

4  
5 We discuss campaigns below. A critical part of the infrastructure for campaigns  
6 are aircraft, particularly aircraft that can probe the UT/LS region, such as the ER2  
7 and Geophysica which can probe the lowermost stratosphere while other  
8 instrumented aircraft can probe the upper troposphere. It is important that these  
9 aircraft be supported. Currently the Geophysica is in need of upgrades which are  
10 ~ \$1M. In addition, instrumented commercial aircraft such as the MOZAIC  
11 (Measurement of OZone by Airbus In-service aircraft) fleet also have an  
12 important role to play (<http://www.cnrm.meteo.fr/dbfastex/datasets/moz.html>). It  
13 would be important if other governments/aviation companies could be persuaded  
14 to participate.

15  
16 The major chemistry and related issues summarized above can only be  
17 addressed with new research. For example to better characterize lightning NO<sub>x</sub>  
18 sources, and convection etc will require dedicated aircraft campaigns with  
19 possibly new instrumentation to attack problems of heterogeneous chemistry on  
20 ice and aerosols and possibly measurements of “missing” species. In addition, it  
21 will be necessary to support laboratory chemistry, particularly heterogeneous  
22 chemistry.

23  
24 With respect to the development of new instrumentation an interesting possibility  
25 is the development of a NO<sub>2</sub> sonde (e.g., Pisano et al., 1996; Sitnikov et al.,  
26 2005). This would complement the ozonesonde and would be a major asset in  
27 researching the production of ozone by aircraft since it would provide high  
28 vertical resolution profiles for NO<sub>2</sub>. Some infusion of support could lead to  
29 instruments suitable for release in concert with ozonesondes. This could be done  
30 on relatively a short time scale.

### 32 **Campaigns**

33  
34 We consider it important continue to continue to conduct aircraft experiments and  
35 these should be coordinated where possible with satellite UT/LS measurements  
36 of gases (e.g. ACE, MIPAS, AURA (MLS, TES, HIRDLS), METOP) and aerosols.  
37 Clearly aircraft will continue to play a leading role in investigations over the next  
38 three years. The aircraft include but are not limited to the ER2, DC-8,  
39 Geophysica, WB57 and HIAPER. UAVs are a developing technology which  
40 should be exploited since they can remain aloft for several days. These will be  
41 important in addressing both summer and winter conditions and ideally there  
42 should a major campaign conducted in most continents. These would investigate  
43 the issues of (a) lofting PBL NO<sub>x</sub> sources and (b) lightning NO<sub>x</sub> (c) better  
44 characterization of corridor issues.

45  
46 The impact of improving knowledge in these areas will be high as it is basic

1 knowledge of the background atmosphere. And models simulations of same will  
2 be necessary for the simulation of aircraft impacts.

### 3 4 **Modelling**

5  
6 Climate impact forecasts are dependent on models. Thus it is important to  
7 maintain and improve modelling capabilities. This would include improved  
8 horizontal and vertical resolution, improved physical processes, better emission  
9 data bases, more accurate resolved and sub-gridscale transport processes. The  
10 development of multi-scale models is needed to investigate the corridors aspect  
11 of the aircraft emissions and the transition to regional scale climate impacts.  
12 Parameterizations used for deep convection need to be both used consistently  
13 (with the basic dynamical model) and verified according to the scale of model.

14  
15 It would be useful for plume-to-corridor scale models to run comparison  
16 simulations of the conversion of NO<sub>x</sub> into nitric acid in the near field plumes to  
17 better characterize the in-plume conversion of NO into HNO<sub>3</sub> by heterogeneous  
18 chemistry in the plume and better resolve the issue of EEIs (effective emission  
19 indices) for use in global models.

20  
21 Also it will be important to develop more challenging protocols for comparing  
22 models and devise improved diagnostic schemes or metrics for comparing with  
23 measurements. This latter is particularly important as it provides useful diagnostic  
24 tools for assessing model capability. An essential component of this is improved  
25 computer resources.

26  
27 Better characterization and parameterization of convection will remain an issue.  
28 One means to attack the problem is by the use of cloud resolving models (CRMs;  
29 see for example, Xie et al., 2006; Xu et al., 2006; Lopez et al., 2006) to provide a  
30 first order basis for improved parameterizations. Nevertheless, any  
31 parameterization will need to be verified against measurements.

32  
33 Better models will lead to improved forecasts in terms of reliability and given  
34 current constraints the goals are eminently achievable.

35  
36 Elements required to address the various problems outlined above

- 37
- 38 1) Support for 3 modelling groups (a single model is useful but using different  
39 models is a more basic way of addressing uncertainty issues, as long as  
40 certain basic metrics (to be identified) are complied with.
  - 41 2) Each group may require 3 PY over three years to address some of the  
42 issues.
    - 43 a. Campaign modelling
    - 44 b. Climate effects forecasting
    - 45 c. Access to substantial computer power.
- 46

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## Radiative Forcing Issues

An important goal is to reduce the errors associated with radiative forcing (RF) issues. There have only been very few and limited validation comparisons of RF with real measurements. In particular, research is necessary for the evaluation of cloud attenuation effects which represent a substantial uncertainty on model calculation of the RFM. Validation of the actual RF observable is needed to reduce GCM climate simulation errors of surface RF and surface temperatures.

In terms of impact, improving knowledge and accuracy of RF estimates would improve the uncertainties of estimates of relative contribution of aviation to global warming which are estimated using the RFM. The data could, perhaps, also lead to improvements in GCM simulations of climate changes for regions and also reduce the current error associated with the impacts of a tripled future aircraft fleet contribution to the total RFM.

The achievability is high since satellite measured databases of radiative trapping and ground surface measures of surface RF already exist. The work would consist mainly of data analysis of existing Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) databases (<http://www.arm.gov/>) . It would be prudent to expand the current AERI (Atmosphere Emitted Radiance Interferometer) stations into a future network for surface radiative forcing.

The costs are estimated to be :

- a) analysis of satellite databases 5 FTEs,
- b) analysis of existing ARM AERI database 3 FTE,
- c) and network expansion \$300K per station.

Timelines: analysis of AERI database 2 years, analysis of existing satellite databases: 3 years and network development 10 years.

## 5. Recommendations

In this section we address the “recommendations for the optimum use of current tools for modeling and data analysis” and interpret this to mean what can be done with a limited expenditure of funds (and assuming with insufficient funds for major new measurement programs initiatives). Also for the scientific domain of this SSWP we address chemistry-climate impacts of aviation. Priority one would be the impact on ozone and associated RF effects and second priority would be the effects of aerosols in the plume/contrail. In terms of uncertainty, we have noted above that the UT is a region of considerable uncertainty both in terms of background emission sources and chemistry, both gas phase and heterogeneous.

1 The issues raised in Sections 2 and 3 have led us to a number of  
2 recommendations for 'practical' application of the currently available information  
3 involving available measurements and current modeling capabilities. This can,  
4 perhaps, be separated into three aspects. The analysis of observations, currently  
5 collected, continuing analysis of observations taken as part of an on-going  
6 program such as a satellite experiment and the analysis using currently available  
7 models.

8  
9 As noted in sections 2 and 3, recent plume and CTM modeling studies have  
10 emphasized the impacts of background aerosols (natural and non-aviation  
11 anthropogenic) and those generated by aviation combustion on the UT/LS ozone  
12 budget: plume aerosols can modify UT/LS chemistry by activating Cly to destroy  
13 ozone, while ozone oxidation reactions followed by heterogeneous reactions can  
14 result in a reduction of ozone within the plume. But these conditions are quite  
15 sensitive to background conditions. In addition, once the plume aerosols become  
16 part of the background the enhanced aerosol field can have similar impacts.  
17 These aspects could be addressed by a modelling study where the goals would  
18 be to come up with a "best" estimate of UT/LS background aerosols for current  
19 and future (~2050) conditions and also appropriate future meteorological  
20 conditions. Several models could repeat the earlier work of Meilenger et al.  
21 (2005) and Søvde et al. (2007) for these future conditions to better characterize  
22 impacts.

23  
24 One issue that could also be addressed without new data would be to confirm the  
25 results of recent modelling studies on the impacts of a putative fleet of supersonic  
26 aircraft (SSA). Current estimates suggest that a fleet of SSAs would result in an  
27 increase in column ozone. But as this is impacted by aircraft-generated aerosols  
28 it would be useful to reexamine this with different fuel type.

29  
30 Another recommendation is to continue extensive analysis of the current suite of  
31 satellite UT/LS measurements of gases (ACE, MIPAS, AURA/MLS,  
32 AURA/HIRDLS) in order *to better characterize the UT/LS region* so that we can  
33 be more confident of the putative impacts of aviation. But we also note that nadir  
34 viewing instruments (e.g. MOPITT, AURA/TES, MODIS etc) also yield important  
35 information on emissions and convection and also aerosol distributions.

36  
37 We also recommend the analysis of current data sets be accompanied by  
38 modelling analysis using 3D models and that at least a few of these should  
39 include evaluated "comprehensive" tropospheric and stratospheric chemistry in  
40 order to provide a more solid baseline. This could include both CTMs and GCMs.  
41 But there should also be an assessment of the robustness of the using  
42 appropriate diagnostics such discussed above such as STE ozone fluxes,  
43 tropospheric <OH> densities and correlations. These models could be used to  
44 assess the impact of aviation in climate mode or time-slice mode using SSTs and  
45 related surface properties from climate models. The rationale for using time-slice  
46 mode is that the aircraft climate/dynamical impacts are likely to be small and

1 looking for climate signatures could require running many ensembles, although if  
2 sufficient models were run this might be taken as equivalent to ensembles. This  
3 would certainly supplement the assessment of climate impacts using a RF type  
4 metric.

5  
6 An important recommendation concerns the radiative forcing changes due to the  
7 increased UT ozone from aircraft traffic. The uncertainty that clouds introduce to  
8 model simulations of this parameter was noted in Section 2. A request could be  
9 made to DOE ARM to process the large existing AERI database for surface  
10 radiative forcing from ozone and methane. With such a database, investigations  
11 of cloud effects on radiative forcing in corridor regions and in several climate  
12 regimes would be facilitated.

13  
14 Of the issues that could be addressed is the re-evaluation of data from previous  
15 campaigns in the light of more recent understanding. And this brings up the issue  
16 of data accessibility. Is the older aircraft data accessible and could it be made  
17 available with such tools as GIOVANNI, developed by NASA for several satellite  
18 instruments? GIOVANNI is fast to use and easy to use; it permits scientists to  
19 access and work with the data easily online without importing large volumes of  
20 data.

## 21 **6. Summary**

22  
23 The current state of knowledge on the formation of ozone and related chemistry  
24 and transport in the upper troposphere at air traffic flight levels has been  
25 surveyed. About 40% of the time aircraft fly in the lower stratosphere where the  
26 chemistry is well known and there is confidence in the projections of models. In  
27 the upper troposphere where jet traffic spends 60% of the time, the chemistry is  
28 insufficiently defined to make an accurate prediction of the climate impacts of  
29 increased jet traffic. In the upper troposphere HO<sub>x</sub>/NO<sub>x</sub> chemistry is uncertain as  
30 revealed by various aircraft campaigns. Measurements indicate that PAN is an  
31 important component of NO<sub>y</sub> in the mid to upper troposphere and is likely  
32 involved in long range transport of NO<sub>x</sub>. Also aircraft measurements reveal “high”  
33 levels of NO<sub>y</sub> in the summer UT over NA, similar to those in the LT, but  
34 generated by lightning rather than anthropogenic emissions. The sources of NO<sub>x</sub>  
35 in the UT are not all well quantified, despite recent progress on quantifying the  
36 lightning source. The fraction of the deep convection source of NO<sub>x</sub> from the  
37 surface sources which reaches the 10 to 13 km level is estimated to be around  
38 10 % for North American summer meteorological conditions but possibly 50% for  
39 European summer meteorological conditions. There is a need for new aircraft  
40 campaigns focused on the quantification of the NO<sub>x</sub> sources in the flight  
41 corridors.

42  
43 Heterogeneous chemistry on aerosols/contrails from aircraft emissions may alter  
44 ozone loss chemistry in the regional background atmosphere and modify ozone  
45 production from NO<sub>x</sub> emissions. Plume to corridor studies would be useful to

1 better characterize the conversion of NO<sub>x</sub> into HNO<sub>3</sub> in the near field plumes.  
2 These effects need to be characterized/confirmed for different fuel types.

3  
4 The evidence of the climate impact of increased upper tropospheric ozone due to  
5 air traffic is the detection of changes in the ozone radiative forcing at the surface  
6 and at the top of the atmosphere. The radiative forcing due to ozone may be  
7 higher in some regional areas than on a global basis. There are uncertainties in  
8 the calculations of the radiative forcing metric mainly due to a lack of knowledge  
9 of cloud effects. There have been few verifications of the RF metric with real  
10 measurements of observed surface radiative forcing or with satellite radiative  
11 trapping at the top of the atmosphere.

12  
13 The characterization of aviation impacts in details within the corridor is limiting  
14 progress and should be the focus of aircraft and satellite studies. However, it is  
15 rendered difficult by the relatively small effect on synoptic scales. There are  
16 several recent satellites which provide new information on the NO<sub>x</sub> and nitric acid  
17 at flight levels and this information could perhaps be applied to the problem.

18  
19 In concert with aircraft campaigns, satellites experiments with improved vertical  
20 resolution are needed to study the UT/LS region.

21  
22 There is a concern that there will be a gap in satellite instruments suitable for  
23 UT/LS investigations. The SHADOZ/IONS ozonesondes have proven to be highly  
24 useful for investigating ozone in the upper troposphere and could be used to  
25 partially fill such a gap. There have been too few aircraft campaigns focused on  
26 the flight corridors and which are coordinated with satellite overpass  
27 measurements.

28  
29 There have been large advances in models since 1999. Data assimilation has  
30 proven very valuable in providing “value added” information from satellite data.  
31 Multiscale models are needed to investigate the corridors aspect of the aircraft  
32 emissions and the transition to regional scale climate impacts. Parameterizations  
33 used for deep convection need to be both used consistently (with the basic  
34 dynamical model) and verified according to the scale of model. Finally it is  
35 recommended that climate assessments of the impacts of a future fleet should  
36 use chemistry-climate models with comprehensive tropospheric and stratospheric  
37 chemistry at as high a resolution as feasible.

38  
39 We caution that although current measurements can yield improved results by  
40 the application of more sophisticated models, it is unlikely that accurate  
41 simulations of aircraft emissions impacts on UT/LS ozone and resulting radiative  
42 forcing of climate will be possible without information from new satellite and  
43 aircraft missions and expanded sounding systems such as ozonesondes and  
44 lidars.

45



## 7. References

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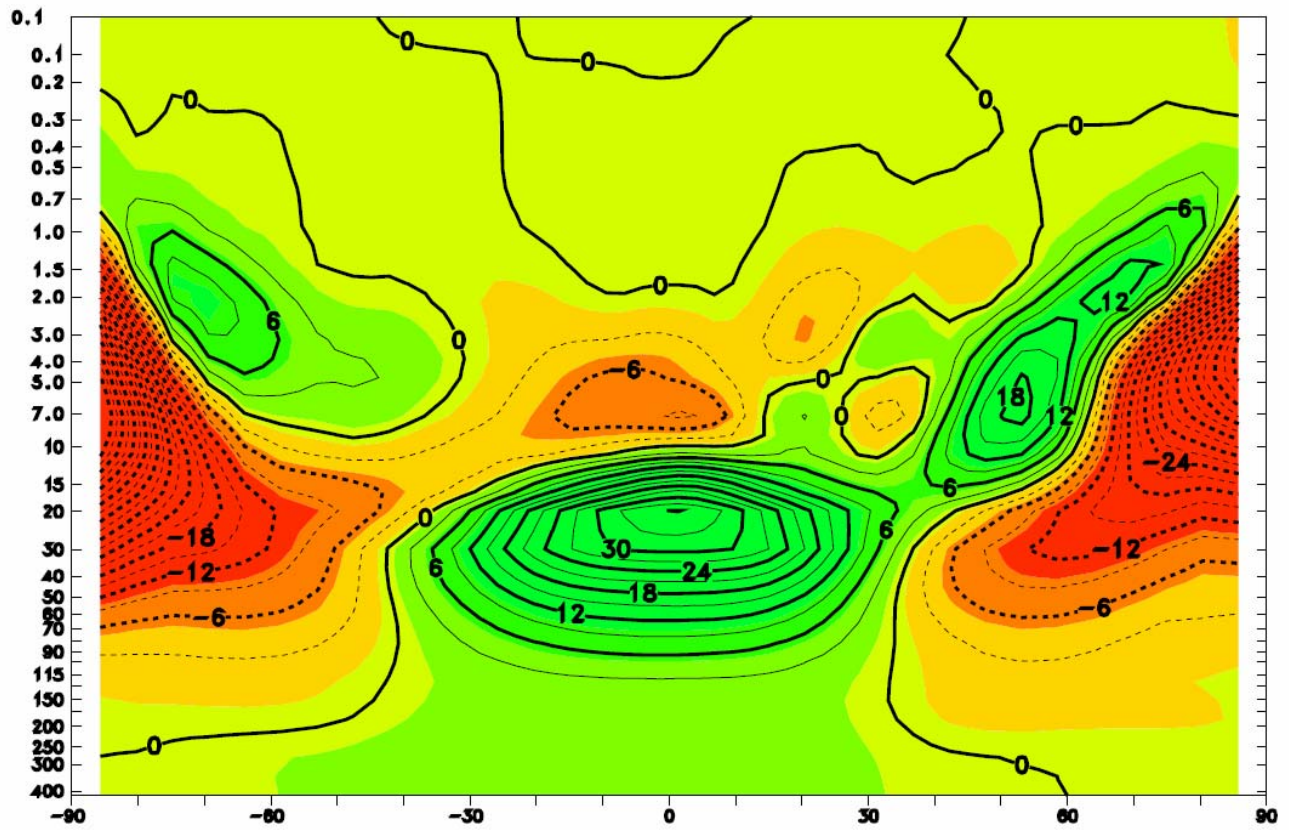
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1 **Figures**

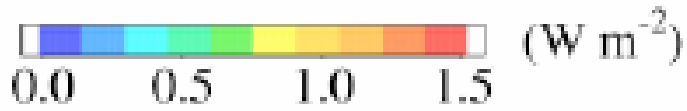
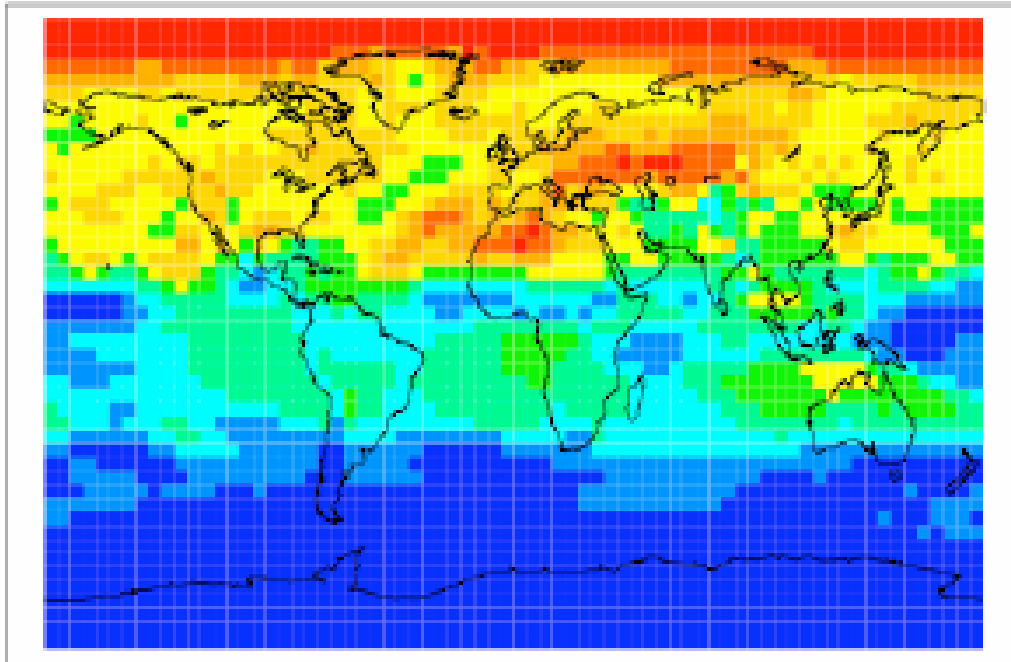
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3 Figure 1. Net ozone production for equinox from CMAM model ( $10^9$  ozone  
4 molecules  $\text{cm}^{-3} \cdot \text{day}^{-1}$ ) (courtesy of Stephen Beagley, 2007). [Will be cut off at  
5 10 mb]



- 1 Figure 2 The instantaneous radiative forcing from tropospheric ozone since pre-
- 2 industrial times of 0.49 W/m<sup>2</sup> (Mickley et al., 2004)
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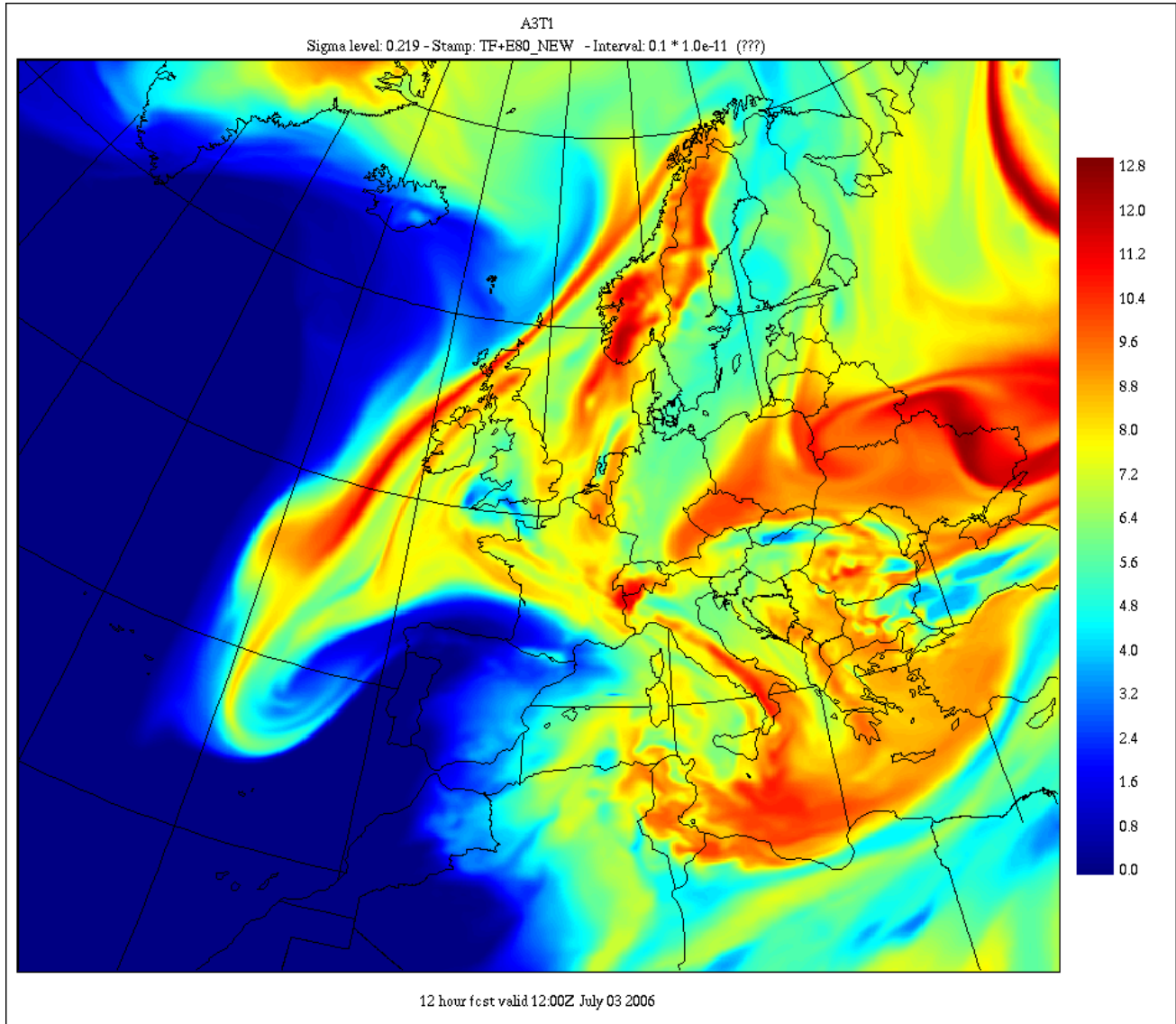
d. JJA O<sub>3</sub> forcing



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Chemical and dynamical processes: revised final

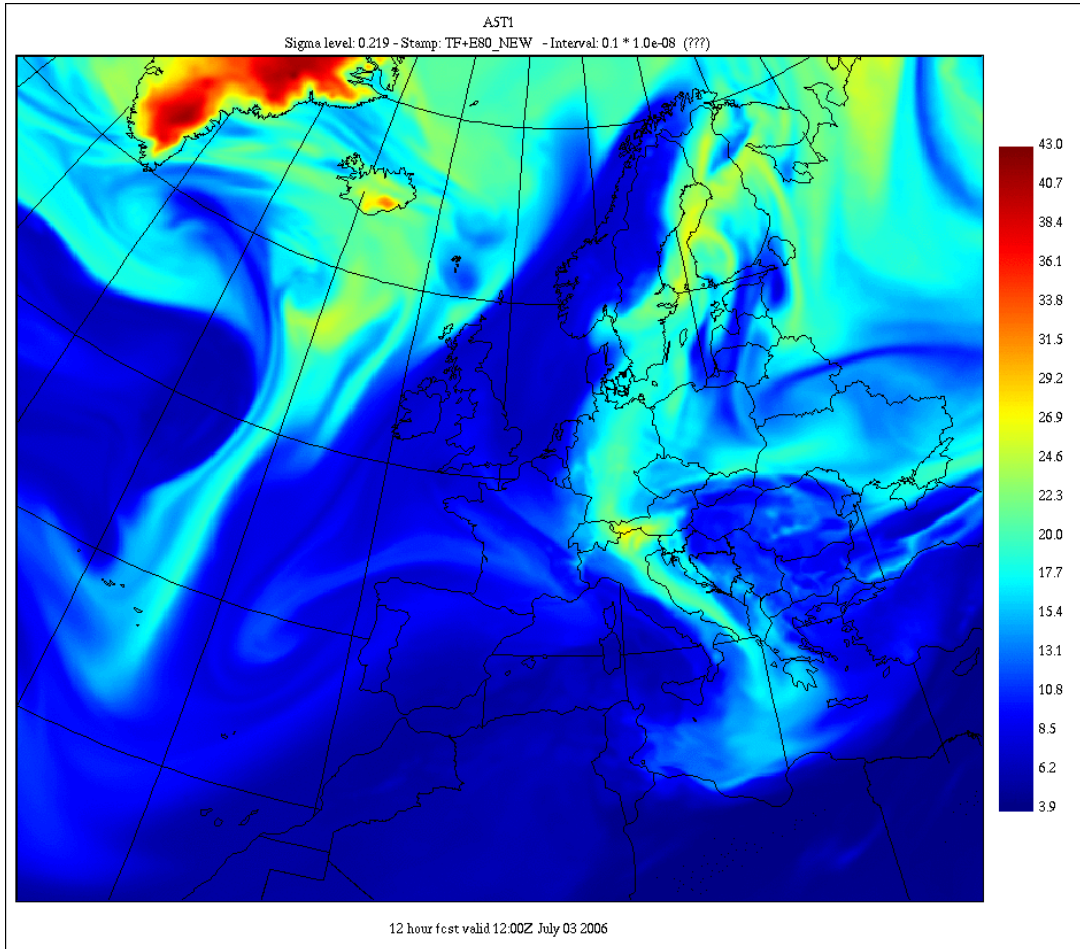
- 1 Figure 3(a) NO distribution at ~ 220 mb over eastern Atlantic and Western
- 2 Europe calculated using GEM-AQ (Kaminski et al., 2007) at  $15 \times 15^2$  km
- 3 resolution
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Chemical and dynamical processes: revised final

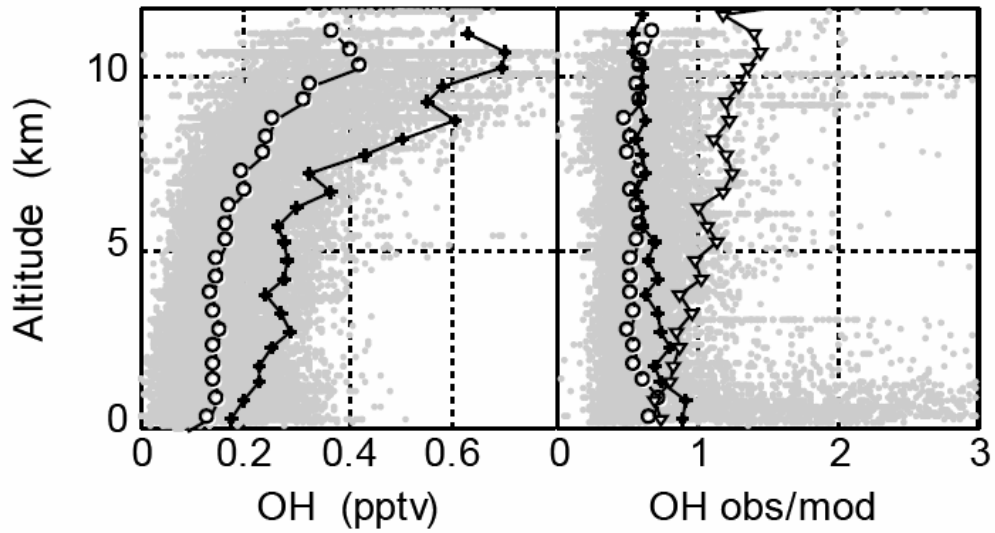
- 1 Figure 3(b) O<sub>3</sub> distribution at ~ 220 mb over eastern Atlantic and Western
- 2 Europe calculated using GEM-AQ (Kaminski et al., 2007) at 15x15<sup>2</sup> km
- 3 resolution
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- 1 Figure 4: Comparison of the vertical profiles of (left) measured (circles) and
- 2 modeled (stars) OH and (right) measured-to-modeled OH ratios during in
- 3 INTEX-NA (circles), TRACE-P (stars) and PEM-TB (triangles). Individual
- 4 INTEX-NA 1-minute measurements are shown (gray dots). (Ren et al., 2006)
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## 1 **Annex I: Models with comprehensive Tropospheric and** 2 **Stratospheric chemistry**

### 3 4 CTMs

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### 24 Climate models

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## Annex II. Suggested structure for each SSWP

1. Introduction and Background specific to your theme area
2. Review of specific theme
  - a. Current state of science
  - b. Critical role of the specific theme
  - c. Advancements since the IPCC 1999 report
  - d. Present state of measurements and data analysis
  - e. Present state of modeling capability/best approaches
  - f. Current estimates of climate impacts and uncertainties
  - g. Interconnectivity with other SSWP theme areas
3. Outstanding limitations, gaps and issues that need improvement
  - a. Science
  - b. Measurements and analysis
  - c. Modeling capability
  - d. Interconnectivity with other SSWP theme areas
4. Prioritization for tackling outstanding issues based on their
  - a. Impact
  - b. Ability to improve the climate impacts estimates with reduced uncertainties
  - c. Practical use (e.g. model improvement, sensitivity analysis, metric development etc.)
  - d. Achievability
  - e. Estimated cost
  - f. Timeline
5. Recommendations for best use of current tools for modeling and data analysis
  - a. Options
  - b. Supporting rationale
  - c. How to best integrate best available options?
6. Summary

## 1 **Annex III: Questions to be considered in report**

2  
3 Please note that each of the SSWP is meant to provide recommendations on:

- 4 \* Improvement needed to advance the state of science and modeling
- 5 capability; and
- 6 \* best use of the present state of science and modeling capability to
- 7 + better quantify magnitude of climate impacts of aviation and
- 8 associated uncertainties for present and future condition
- 9 + develop metrics to measure these impacts on all relevant scales

10  
11 It is expected that each SSWP will also address the following common questions

12 to the best extent possible:

- 13 \* What are the key science questions/issues specific to aviation induced
- 14 climate change impacts for the present and future conditions?
- 15 \* What are the state of science, present modeling capability, and
- 16 observation databases available to answer these key questions?
- 17 \* What are the controlling factors: scientific knowledge vs. modeling
- 18 capability vs. computational resources given that aviation-induced
- 19 atmospheric perturbations range from plume to global scales?
- 20 \* What are the presently available best options among existing models and
- 21 their individual modules to isolate and estimate atmospheric and radiative
- 22 perturbations due to aviation emissions? How well these models perform to
- 23 simulate the state of the background atmosphere due to all non-aviation
- 24 sources?
- 25
- 26 \* How to best integrate available modeling options to simulate atmospheric
- 27 perturbations due to aviation and how to evaluate the model performance to
- 28 characterize the aviation-induced perturbations?
- 29 \* What are the gaps and uncertainties in science? What are the limitations
- 30 in observations and modeling tools to answer the key questions?
- 31 \* With no further scientific knowledge, how and with what level of
- 32 uncertainties, can the key questions be answered today using the best
- 33 available modeling tools?
- 34 \* If the gaps were to be addressed, would the ability to answer the key
- 35 questions get any better? If so, to what possible extent and within what
- 36 possible timeframe?

37  
38  
39  
40 . The review panel has made some suggestions that are general and applicable

41 to all selected proposals:

- 42
- 43 \* Within the sphere of your own SSWP, reach beyond the issues that were
- 44 listed in the solicitation. Our stated questions/issues were merely the
- 45 sample to provide some guidance as well as relevance and they were not

## Chemical and dynamical processes: revised final

- 1 intended to limit your scope of work.
- 2 \* Be comprehensive in preparing the SSWP by including the review on the
- 3 state of science, gaps and uncertainties, modeling capabilities and current state
- 4 of relevant measurements and analysis of the existing data with a focus on
- 5 present and future climate impacts due to aviation.
- 6 \* Do not limit the scope of SSWP to your own activities. Outreach the latest
- 7 efforts of the entire scientific community as a whole.
- 8
- 9 \* Wherever possible, maintain the interconnectivity among themes of all
- 10 other SSWPs. Study of climate impacts needs to be carried out within the
- 11 one-atmosphere framework through interrelated processes irrespective of
- 12 how they are distinctly classified as dynamical, transport, chemical,
- 13 microphysical, optical and/or radiation.
- 14 \* Identify the best modeling and analysis options presently available and
- 15 provide the supporting rationale.
- 16 \* Address the gaps and uncertainties (in the state of science, modeling
- 17 capabilities and their practical applications), identify the key areas of
- 18 improvements and prioritize them based on their practical achievability as
- 19 well as associated timelines.
- 20