Aviation NOx-induced CH_4 effect: fixed mixing ratio boundary conditions versus flux boundary conditions

Arezoo Khodayari[a,*], Seth C Olsen[b], Donald J Wuebbles[b], and Daniel B Phoenix[b]

[a]Department of Civil and Environmental Engineering, University of Illinois at Urbana-Champaign, Urbana, IL 61801

[b]Department of Atmospheric Sciences, University of Illinois at Urbana-Champaign, Urbana, IL 61801,USA

*Corresponding author address: Arezoo Khodayari, Department of Atmospheric Sciences, University of Illinois at Urbana-Champaign, 105 S. Gregory St., Urbana, IL, 61801. E-mail address: akhoday2@illinois.edu

Keywords: Aviation NOx emissions, CH_4 lifetime, feedbacks

Abstract

Atmospheric chemistry-climate models are often used to calculate the effect of aviation NOx emissions on atmospheric ozone (O_3) and methane (CH_4). Due to the long (~10 yr) atmospheric lifetime of methane, model simulations must be run for long time periods, typically for more than 40 simulation years, to reach steady-state if using CH_4 emission fluxes. Because of the computational expense of such long runs, studies have traditionally used specified CH_4 mixing ratio lower boundary conditions (BCs) and then applied a simple parameterization based on the change in CH_4 lifetime between the control and NOx-perturbed simulations to estimate the change in CH_4 concentration induced by NOx emissions. In this parameterization a feedback factor (typically a value of 1.4) is used to account for the feedback of CH_4 concentrations on its lifetime. Modeling studies comparing simulations using CH_4 surface fluxes and fixed mixing ratio BCs are used to examine the validity of this parameterization. The latest version of the Community Earth System Model (CESM), with the CAM5 atmospheric model, was used for this study. Aviation NOx emissions for 2006 were obtained from the AEDT (Aviation Environmental

© 2015. This manuscript version is made available under the Elsevier user license
http://www.elsevier.com/open-access/userlicense/1.0/

Design Tool) global commercial aircraft emissions. Results show a 31.4 ppb change in CH_4 concentration when estimated using the parameterization and a 1.4 feedback factor, and a 28.9 ppb change when the concentration was directly calculated in the CH_4 flux simulations. The model calculated value for CH_4 feedback on its own lifetime agrees well with the 1.4 feedback factor. Systematic comparisons between the separate runs indicated that the parameterization technique overestimates the CH_4 concentration by 8.6%. Therefore, it is concluded that the estimation technique is good to within ~10% and decreases the computational requirements in our simulations by nearly a factor of 8.

1. Introduction

Methane (CH_4) plays an important role in atmospheric chemistry and as a greenhouse gas that can affect climate. Since pre-industrial times (~1750), atmospheric CH_4 has increased about 150% and it accounts for 20% of the change in radiative forcing on climate from all long-lived greenhouse gases from 1750 to 2000, excluding water vapor (IPCC, 2013). NOx emissions affect atmospheric concentrations of CH_4 as an indirect result of their effect on concentrations of ozone (O_3) (Wild et al., 2001). Resolving the resulting effects of aviation NOx emissions on ozone and methane remains an important issue towards understanding the effects of aviation emissions on climate. Many studies have shown that aviation NOx emissions at cruise altitudes (~200 – 300 hPa in the UTLS) increase the production of ozone in this region (Derwent et al., 1999; Fuglestvedt et al., 1999; Stevenson et al., 2004; Köhler et al., 2008; Hoor et al., 2009; Koffi et al., 2010; Hodnebrog et al., 2011; Khodayari et al., 2014; and others). Additionally, these studies show that increased concentrations of O_3 will increase the atmospheric concentrations of hydroxyl (OH). OH is a major sink for CH_4 in the troposphere and stratosphere, and thus decreases its concentration, while also affecting the feedback of CH_4 on its own lifetime (Prather, 1994; Wild et al., 2001; IPCC, 2007).

The lifetime of methane has been evaluated in many studies. IPCC (2001, 2007) reported a methane lifetime of 9.6 years against reaction with OH as evaluated with contemporary chemistry-transport models (CTMs) (Ehhalt et al., 2001). Due to the near decadal lifetime of methane, in order to directly calculate the changes in its atmospheric concentration and response

time, three-dimensional (3-D) models using CH_4 surface fluxes must run for more than 40 simulated years to reach steady-state. Simulations of this length are, although computationally feasible (Hsu and Prather, 2010; Prather and Hsu, 2010), often too computationally expensive; therefore, past studies have used fixed mixing ratio lower BCs for CH_4 and then applied a parameterization to estimate the change in CH4 concentrations that would have occurred if CH_4 surface fluxes had been used. The parameterization equation (Equation 1), is derived from a one-box model assumption that assumes the change in CH_4 concentration is proportional to the change in CH_4 lifetime obtained from simulations with and without perturbations (e.g., aviation emissions), using fixed mixing ration BCs for CH_4 (Fuglestvedt et al. 1999). The parameterization coefficient is derived from a CTM study (Fuglestvedt et al. 1999).

$$\Delta[CH_4] = [CH_4]_{bg} \times \left(-1.4\left(\frac{\Delta\tau}{\tau_{bg}}\right)\right) \quad (1)$$

$[CH_4]_{bg}$ and τ_{bg} represent the CH_4 concentration and lifetime in the control simulation. $\Delta[CH_4]$ and $\Delta\tau$ are the changes in CH_4 concentration and lifetime due to the aviation NOx emissions, and 1.4 represents the CH_4 feedback on its own lifetime.

The feedback of CH_4 on its own lifetime lengthens the duration of a CH_4 perturbation (CH_4 response time) to 12 years (Prather, 1994; IPCC, 1995). This can be illustrated as follows: the initial perturbation in nitrogen oxides (NOx) (ΔE_{NO_x}) affects the amount of ozone, which produces an effect on OH ($\Delta[OH]_0$) that then affects CH_4 ($\Delta[CH_4]_0$) (e. g. Prather et al., 1999; Wuebbles and Hayhoe, 2000; and Stevenson et al., 2004). The change in CH_4 then leads to further effects on OH and CH_4 ($\Delta[OH]_1, \Delta[CH_4]_1, \Delta[OH]_2, \Delta[CH_4]_2, etc$) until eventually the

system reaches steady-state (the feedback effect). The entire process can be illustrated as the following sequence of feedbacks (Fuglestvedt et al., 1999):

$$\Delta E_{NO_x} \rightarrow \Delta[OH]_0 \rightarrow \Delta[CH_4]_0 \rightarrow \Delta[OH]_1 \rightarrow \Delta[CH_4]_1 \rightarrow \Delta[OH]_2 \rightarrow etc.$$

The feedback effect due to the initial sequence of feedbacks can then be calculated from Equation 2.

$$R_1 = \frac{\Delta[CH_4]_1 / \Delta[CH_4]_0}{\Delta\tau_1 / \Delta\tau_0} \quad (2)$$

Here, R_1 corresponds to the initial feedback effect. $\Delta[CH_4]_0$ and $\Delta\tau_0$ are the initial changes in CH_4 concentration and lifetime and $\Delta[CH_4]_1$ and $\Delta\tau_1$ are the changes in CH_4 concentration and lifetime after the first sequence of feedbacks. The initial feedback (R_1) can be calculated by performing two simulations, first a NOx-perturbed simulation with fixed CH_4 mixing ratio BCs and then a similar NOx-perturbed simulation, but with a small increase in CH_4 mixing ratio at the boundary layer (i.e., $\Delta[CH_4]_1 / \Delta[CH_4]_0$ = 0.20 when there is a 20% increase in CH_4 mixing ratio at the boundary layer). The corresponding CH_4 lifetime is calculated in each simulation.

Assuming that the feedback strength does not vary significantly with the strength of the initial perturbation, the feedback factor due to the whole sequence can be derived from Equation 3 (Fuglestvedt et al., 1999; Karlsdottir and Isaksen, 2000; IPCC, 2013):

$$f = \frac{1}{1-s} = \frac{1}{1-\frac{1}{R_1}}; \text{ where } s = \frac{1}{R_1} \quad (3)$$

IPCC (2001) calculated a feedback factor with a range of 1.33 – 1.45 (with an average value of 1.4). This feedback factor of 1.4 has been widely accepted and used (e.g., IPCC, 2001; Holmes et al., 2011).

In this study we used the latest version of the atmospheric component of the Community Earth System Model (CESM), CAM5, to evaluate the parameterization commonly used to correct for assuming fixed CH_4 mixing ratio BCs and also to evaluate the validity of the 1.4 feedback factor for the case of aviation perturbations. Estimates of the methane feedback on its own lifetime have been evaluated for aviation perturbations previously, e.g., feedback factors of 1.3 and 1.52 were calculated in Köhler et al. (2008) and Holmes et al. (2011), respectively, and in this study we re-evaluate this feedback factor using CAM5. However, there has been no previous evaluation of the validity of the aforementioned parameterization (Equation 1) for the case of aviation perturbations.

The remainder of this paper is organized as follows: section 2 describes the data and model used in the study, section 3 describes the model configuration, section 4 describes the model simulations done to test the accuracy of Equation 1, section 5 will discuss the results, and section 6 provides a concluding argument.

2. Model Description and Configuration

CAM5 is the atmospheric component for the Community Earth System Model (CESM) (http://www.cesm.ucar.edu/). The previous version of the Community Atmosphere Model (CAM) and CAM5 have been documented and discussed in other studies (e.g. Neale et al., 2011, Gent et al., 2011, Lamarque et al., 2012). CAM5 was successfully coupled to the full chemical mechanism and released in CESM 1_2_0 and versions thereafter. We used the development version of CAM5 (Community Atmosphere Model version 5, cesm1_1_alpha17b_modal) in this

114 study since the coupling of aerosols and chemistry in CAM5 has not been released at the time
115 model runs were completed.

116 CAM5 includes tropospheric and stratospheric chemistry with 133 species and 330
117 photochemical reactions as described in Lamarque et al. (2012). A major improvement in
118 CAM5 over CAM4 is a new modal aerosol module (MAM) (Liu et al., 2012) with two different
119 versions available: a version with seven lognormal modes (MAM7), and a version with three
120 lognormal modes—Aitken, accumulation, and coarse (MAM3) (Liu et al., 2012). For this study,
121 CAM5 was configured with MAM3. MAM simulates the size distribution of aerosols for both
122 internal and external mixing, the chemical and optical properties of aerosols, as well as various
123 other complicated aerosol processes (Liu et al., 2012). Further details on CAM5 can be found on
124 the NCAR website (http://www.cesm.ucar.edu/models/cesm1.0/cam/).

125 CAM5 was run with a horizontal resolution of 2° latitude x 2.5° longitude with 30 vertical
126 levels from the surface up to ~2 hPa. To reduce the year-to-year climate variability in the model
127 simulation and to be able to detect the aviation NOx-induced signal, specified dynamics ("off-
128 line" mode) simulations were performed. The model was driven with the 2005 meteorology from
129 a coupled CAM5 simulation. The aviation NOx emissions are from the AEDT aviation emissions
130 scenarios for the year 2006 (Roof et al., 2007; Wilkerson et al., 2010; Olsen et al., 2012). The
131 NOx emissions used as the input to the model had a horizontal resolution of 1° latitude x 1°
132 longitude, vertical resolution of 150 m, and hourly temporal resolution. The total NO_X emissions
133 amounted to 2.7 Tg (NO_2)/yr. The NOx emissions were then regridded to the model resolution.
134 Background emissions of non-aviation short-lived species (e.g., NOx, volatile organic
135 compounds (VOCs)) and longer-lived species (e.g., CO_2, chlorofluorocarbons (CFCs), and
136 nitrous oxide (N_2O)), which were specified as mixing ratio lower BCs, are from the IPCC
137 RCP4.5 scenario (van Vuuren et al., 2011). Biogenic emissions were from the Model of
138 Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006).

139

140

3. Experimental Design

We evaluated the CH_4 feedback factor and the parameterization commonly used to calculate the change in CH_4 concentration based on the changes in its lifetime by performing three sets of simulations: Set 1- a control (1c) and an aviation NOx-perturbed (1p) run with fixed mixing ratio BCs for zonal mean CH_4 that had a monthly temporal resolution and were interpolated to the model resolution, Set 2- an aviation NOx-perturbed (2p) run with a 20% increase in CH_4 mixing ratio at the boundary layer, and Set 3- a control (3c) and a NOx-perturbed (3p) run with CH_4 surface fluxes. The changes induced by aviation NOx were then obtained by taking the difference between the control and the NOx-perturbed simulation in each set of simulation.

The purpose of Set 1 was to calculate the changes in CH_4 lifetime with fixed CH_4 mixing ratio at the boundary layer. After running these simulations for 7 years, the steady-state condition was reached. Data from the 7^{th} year was used to calculate the steady state change in CH_4 lifetime due to its reaction with tropospheric OH. The parameterization described in Equation 1 (Fuglestvedt et al., 1999) was then used to calculate the steady state change in CH_4 concentration using the changes in its lifetime and the 1.4 feedback factor.

The purpose of Set 2 was to calculate the CH_4 feedback on its lifetime. By using the CH_4 lifetime obtained in this run (2p) and the CH_4 lifetime obtained in the NOx-perturbed run from Set 1 (1p) with Equations 3 and 4, the CH_4 feedback factor was calculated.

The purpose of Set 3 was to directly calculate the changes in CH_4 concentration caused by aviation NOx emissions. A control run and a NOx-perturbed run with CH_4 surface fluxes were done to calculate the actual changes in CH_4 response time and concentration. The CH_4 fluxes used in Set 3 were obtained using the global annual mean steady-state loss of CH_4 from the control run of Set 1 (1c) since at steady-state, production is equal to loss. This resulted in a global annual mean CH_4 flux of 453 Tg that is well within the IPCC (2007) reported range (428-507 Tg) and close to the most recent reported range of 554±56 Tg/yr (Prather, 2012; IPCC, 2013). Next, the CH_4 fluxes from IPCC RCP 4.5, that had a horizontal resolution of 2.8° latitude x 2.8° longitude and monthly temporal resolution, were scaled to match the global annual mean

CH_4 flux calculated from Set 1 (453 Tg). This was done to produce a regional and temporal distribution of CH_4 input flux that matched the background atmosphere of Set 1, for Set 3. The two simulations were then run until steady-state conditions were achieved. By comparing the result from Set 1 to the direct changes in CH_4 concentration calculated from Set 3, using CH4 fluxes, the accuracy of Equation 1 was evaluated.

4. Results and Discussion

Based on our three sets of simulations the accuracy of the parameterization used in Equation 1 has been evaluated and the feedback factor used to describe CH_4 feedback on its lifetime has been calculated. Set 1 resulted in a global annual mean background CH_4 concentration of 1713.0 ppb in both the control and the NOx-perturbed simulations, since both simulations were run with the same CH_4 mixing ratio BCs. The calculated CH_4 lifetime for the control run and the NOx-perturbed simulations using the global burden and the global loss of CH_4 were 10.74 and 10.58 years, respectively. The global losses do not include losses to soils, the stratosphere, or Cl-atom losses. The reduction in CH_4 lifetime is about 1.3% (1.60%/[TgN/yr]), which falls within the -1.4±0.40 (%/[TgN/yr]) to -1.6±0.37 (%/[TgN/yr]) range reported by Hodnebrog et al. (2011) using an ensemble of six different atmospheric chemistry models with different emissions. A global mean aviation NOx-induced CH_4 change of 31.4 ppb was calculated by using the CH_4 lifetimes and the global annual mean background CH_4 concentration in Equation 1.

Based on Set 2, the CH_4 feedback on its lifetime was calculated. The CAM5 calculated feedback factor was 1.39, very close to the average feedback factor of 1.4 adopted in most studies.

Based on Set 3, the change in CH_4 concentration was calculated directly. As discussed earlier, the runs in this set used CH_4 fluxes to produce the actual change in CH_4 concentration. Both the NOx-perturbed run and the control run were run until reached steady-state.

Since the surface CH_4 fluxes used in Set 3 were calculated based on the steady-state loss of CH_4 in the control simulation of Set 1, the background CH_4 concentration calculated in the

control simulation of Set 3 should be very similar to the background CH_4 concentration in the control simulation of Set 1. A comparison of the background concentration in the two sets indicates that the CH_4 concentration in the background atmospheres between the two sets were essentially the same (± 0.10% difference at a global annual level and ± 0.35% difference (with a max difference of 3%) at a monthly grid box level).

The aviation NOx-induced global mean changes in CH_4 concentration calculated from the Set 3 simulations was 28.9 ppb which is about 8.6% lower than the change calculated from simulations in Set 1.

Four approaches (lifetime approaches) were used to calculate the atmospheric concentration of CH_4 and its lifetime as all these approaches were used alternatively in previous studies (e.g., Dentener et al., 2003; Stevenson et al., 2004; Stevenson et al., 2006). The most representative lifetime approach was the global annual mean, which was calculated by using the global annual mean burden and the global annual mean loss of CH_4. The other three lifetime approaches differed by: (1) using the tropospheric CH_4 burden and loss, with the tropopause marked by the pressure level of the tropopause, (2) using the tropospheric CH_4 burden and loss, with the tropopause marked by the altitude where the ozone concentration dropped below 150 ppb, and (3) using the global CH_4 burden and tropospheric CH_4 loss with the tropopause marked by the altitude where the ozone concentration dropped below 150 ppb. Using these four different approaches, a range for the values of interest is presented in the third column of table 1. Table 1 shows the results from the three sets of simulations using the aforementioned approaches. Columns 2 and 3 show the values of the quantities of interest using the global mean lifetime approach and the range of values obtained using other defined lifetime approaches (approaches 1-4).

The most representative and thereby accurate lifetime method was the global mean approach (global burden and loss of CH_4). The feedback factor using this lifetime approach was 1.39, very close to the IPCC reported value of 1.40 (0.4% lower). For the other lifetime approaches, the feedback factor ranges from 1.35 to 1.43 (differs from the IPCC adopted value of 1.4 by -3.7% to 2.2%). The CH_4 concentration change calculated from the Set 1 simulations using Equation 1 is

about 8.6% higher than the actual CH_4 concentration change calculated from the simulations in Set 3. For the other lifetime approaches, the change in CH_4 concentration calculated from the Set 1 simulations using Equation 1 is about 12.1% to 20.0% higher than the actual change in CH_4 concentration calculated from the Set 3 simulations. Thus, the parameterization used in Equation 1 is good to within ~10% when using the global mean CH_4 lifetime and decreases the computational requirements by nearly a factor of 8.

5. Conclusion

We evaluated the accuracy of the parameterization widely used in estimating the changes in CH_4 concentration based on the changes in its lifetime and calculated the feedback factor that describes CH_4 feedback on its own lifetime. Results show a 1.3% decrease in CH_4 lifetime due to total aviation NOx emissions from the year 2006 in the simulations with fixed CH_4 mixing ratio as the boundary condition. The CAM5 calculated global mean CH_4 feedback factor of 1.39 compared well with the IPCC adopted feedback factor of 1.4. It was concluded that for the simulations with fixed CH_4 at the lower boundary condition, the parameterizing technique using the global mean lifetime approach overestimates the change in CH_4 by 8.6% compared to the change calculated directly from the model using CH_4 surface emissions. The overestimation is 12.1% to 20.0% if using other lifetime approaches. Thus, for these simulations, the parameterization technique is good to within ~10% when using the global mean lifetime approach and decreases the computational requirements by nearly a factor of 8.

Acknowledgements

The authors would like to thank the Federal Aviation Administration, Aviation Climate Change Research Initiative (ACCRI) for support under Contract #: 10-C-NE-UI amendment 001 and The Partnership for AiR Transportation Noise and Emissions Reduction (PARTNER). The opinions, findings, and conclusions or recommendations expressed in this material are those of the authors and do not necessarily reflect the views of ACCRI, PARTNER, or the FAA. We would like to acknowledge high-performance computing support from Yellowstone

(ark:/85065/d7wd3xhc) provided by NCAR's Computational and Information Systems Laboratory, sponsored by the National Science Foundation.

References

Dentener, F., W. Peters, M. Krol, M. van Weele, P. Bergamaschi, and J. Lelieveld, J., 2003: Interannual variability and trend of CH4 lifetime as a measure for OH changes in the 1979–1993 time period. *J. Geophys. Res.,* 108, 4442, doi:10.1029/2002JD002916.

Derwent, R., R. Friedl, I. L. Karol, H Kelder, V. W. J. H. Kirchoff, T. Ogawa, M. J. Rossi, and P. Wennberg, 1999: Impacts of aircraft emissions on atmospheric ozone. In: *IPCC Special Report on Aviation and the Global Atmosphere.* [Penner, J. E., D. H. Lister, D. J. Griggs, D. J. Dokken, and M. McFarland (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, New York, USA, 373pp.

Ehhalt, D., M. Prather, F. Dentener, R. Derwent, E. Dlugokencky, E. Holland, I. Isaksen, J. Katima, V. Kirchhoff, P. Matson, P. Midgley, M. Wang, 2001: Atmospheric chemistry and greenhouse gases. In: *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change* [Houghton, J.T., Y. Ding, D.J. Griggs, M. Noguer, P.J. van der Linden, X. Dai, K. Maskell, and C.A. Johnson (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 996 pp.

Fuglestvedt J. S., T. K. Berntsen, I. S. A. Isaksen, H. Mao, X. Z. Liang and W. C. Wang, 1999: Climatic forcing of nitrogen oxides through changes in tropospheric ozone and methane global 3D model studies. *Atmos. Environ.,* **33**, 961-77, doi: 10.1016/S1352-2310(98)00217-9.

Gent, P. R., D. Gokhan, J. D. Leo, M. M. Holland, E. C. Hunke, S. R. Jayne, D. M. Lawrence, R. B. Neale, P. J. Rasch, M. Vertenstein, P. H. Worley, Z-L. Yang, and M. Zhang, 2011: The community climate system model version 4. *J. Climate*, **24**, 4973–91, doi: http://dx.doi.org/10.1175/2011JCLI4083.1.

277 Guenther, A., T. Karl, P. Harley, C. Wiedinmyer, P. I. Palmer, and C. Geron, 2006:
278 Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases
279 and Aerosols from Nature), *Atmos. Chem. Phys*., 6, 3181-3210, doi:10.5194/acp-6-3181-2006.

280 Hodnebrog, Ø.,, T. K. Berntsen, O. Dessens, M. Gauss, V. Grewe, I. S. A. Isaksen, B. Koffi,
281 G. Myhre, D. Olivie, M. J. Prather, J. A. Pyle, F. Stordal, S. Szopa, Q. Tang, P. van Velthoven, J.
282 E. Williams, and K. Ødemark, 2011: Future impact of non-land based traffic emissions on
283 atmospheric ozone and OH – an optimistic scenario and a possible mitigation strategy. *Atmos.*
284 *Chem. Phys.*, **11,** 11293–317, doi:10.5194/acp-11-11293-2011.

285 Holmes, C. D., Q. Tang, and M. J. Prather, 2011: Uncertainties in climate assessment for the
286 case of aviation NO. *Proceedings of the National Academy of Sciences*, USA, **108**, 10997–
287 11002.

288 Hoor, P., J. Borken-Kleefeld, D. Caro, O. Dessens, O. Endresen, M. Gauss, V. Grewe, D.
289 Hauglustaine, I. S. A. Isaksen, P. Jockel, J. Lelieveld, G. Myhre, E. Meijer, D. Olivie, M.
290 Prather, C. Schnadt Poberaj, K. P. Shine, J. Staehelin, Q. Tang, J. van Aardenne, P. van
291 Velthoven, and R. Sausen, 2009: The Impact of Traffic Emissions on Atmospheric Ozone and
292 OH: results from QUANTIFY. *Atmos. Chem. Phys.*, **9**, 3113-36, doi:10.5194/acp-9-3113-2009.

293 Hsu, J. and M. J. Prather, 2010: Global long-lived chemical modes exited in a 3-D chemistry
294 transport model: Stratospheric N2O, NOy, O3, and CH4 chemistry. *Geophys. Res. Lett.*, **37**,
295 L07805, doi:10.1029/2009GL042243.

296 IPCC, 1999: *Aviation and the global atmosphere: A Special Report of IPCC Working*
297 *Groups I and III. Intergovernmental Panel on Climate Change*. [Penner, J. E., D. H. Lister, D. J.
298 Griggs, D. J. Dokken, and M. McFarland (eds.)]. Cambridge University Press, Cambridge,
299 United Kingdom and New York, New York, USA, 373pp.

300 IPCC, 2001: *Climate Change 2001: The Scientific Basis. Contribution of Working Group I*
301 *to the Third Assessment Report of the Intergovernmental Panel on Climate Change* [Houghton,
302 J.T., Y. Ding, D.J. Griggs, M. Noguer, P.J. van der Linden, X. Dai, K. Maskell, and C.A.

303 Johnson (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY,
304 USA, 996 pp.

305 IPCC, 2007: *Climate change 2007: The physical science basis. Contribution of Working*
306 *Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*.
307 [Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averyt, M. Tignor and H. L.
308 Miller (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY,
309 USA, 881pp.

310 IPCC, 2013: Climate change 2013: The physical science basis. Contribution of Working
311 Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change.
312 [Stocker, T.F., D. Qin, G. K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V.
313 Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, UK.

314 Karlsdottir, S., and I. S. A., Isaksen, 2000: Changing methane lifetime: Possible cause for
315 reduced growth. *Geophys. Res. Lett.*, **27**, 1, 93-6, doi: 10.1029/1999GL010860.

316 Khodayari, A., Tilmes, S., Olsen, S. C., Phoenix, D. B., Wuebbles, D. J., Lamarque, J.-F.,
317 and Chen, C., 2014: Aviation 2006 NOx-induced effects on atmospheric ozone and OH in
318 Community Earth System Model (CESM), *Atmos. Chem. Phys.*, 14, 9925–9939.
319 doi:10.5194/acp-14-9925-2014.

320 Köhler, M. O., G. Radel, O. Dessens, K. P. Shine, H. L. Rogers, O. Wild, and J. A. Pyle,
321 2008: Impact of perturbations to nitrogen oxide emissions from global aviation. *J. Geophys. Res.*,
322 **113**, D11305, doi: 10.1029/2007JD009140.

323 Koffi, B., S. Szopa, A. Cozic, D. Hauglustaine, and P. van Velthoven, 2010: Present and
324 future impact of aircraft, road traffic and shipping emissions on global tropospheric ozone.
325 *Atmos. Chem. Phys.*, **10**, 11681–705, doi:10.5194/acp-10-11681-2010.

326 Lamarque, J.-F., L. K. Emmons, P. G. Hess, D. E. Kinnison, S. Tilmes, F. Vitt, C. L. Heald,
327 E. A. Holland, P. H. Lauritzen, J. Neu, J. J. Orlando, P. J. Rasch, and G. K. Tyndall, , 2012:

CAM-chem: description and evaluation of interactive atmospheric chemistry in CESM. *Geosci. Model Dev. Discuss.*, **5**, 369 – 411, doi:10.5194/gmd-5-369-2012.

Liu, X., R. C. Easter, S. J. Ghan, R. Zaveri, P. Rasch, X. Shi, J.-F. Lamarque, A. Gettelman, H. Morrison, F. Vitt, A. Conley, S. Park, R. Neale, C. Hannay, A. Ekman, P. Hess, N. Mahowald, W. Collins, M. Iacono, C. Bretherton, and M. Flanner, 2012: Toward a minimal representation of aerosols in climate models: description and evaluation in the Community Atmosphere Model CAM5. *Geosci. Model Dev.*, **5**, 709-739.

Neale, R. B., J. Richter, S. Park, P. H. Lauritzen, S. J. Vavrus, P. J. Rasch, and M. Zhang, 2013: The mean climate of the Community Atmosphere Model (CAM4) in forced SST and fully coupled experiments. *J. Climate*, **26**, 5150–5168.
doi: http://dx.doi.org/10.1175/JCLI-D-12-00236.1.

Olsen, S. C., D. J. Wuebbles, and D. Owen, 2013: Comparison of global 3-D aviation emissions datasets. *Atmos. Chem. Phys.*, **13**, 429-41. doi:10.5194/acp-13-429-2013.

Prather, M. J., 1994: Lifetimes and eigenstates in atmospheric chemistry. *Geophys. Res. Lett.*, **21**, 801-4.

Prather, M. J. and J. Hsu, 2010: Coupling of nitrous oxide and methane by global atmospheric chemistry. *Science*, **330**, 952-954.

Prather, M., R. Sausen, A. S. Grossman, J. M. Haywood, D. Rind, and D. H. Subbaraya, 1999: Potential climate change from aviation. In: *IPCC Special Report on Aviation and the Global Atmosphere*. [Penner, J. E., D. H. Lister, D. J. Griggs, D. J. Dokken, and M. McFarland (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, New York, USA, 373pp.

Roof, C., Hansen, A., Fleming, G. G., Thrasher, T., Nguyen, A., Hall, C., Dinges, E., Bea, R., Grandi, F., Kim, B. Y., Usdrowski, S., and Hollingsworth, P.: Aviation Environmental Design Tool (AEDT) System Architecture AEDT-AD-01, 2007.

353 Stevenson, D. S., R. M. Doherty, M. G. Sanderson, W. J. Collins, C. E. Johnson, and R. G.
354 Derwent, 2004: Radiative forcing from aircraft NOx emissions mechanisms and seasonal
355 dependence. *Geophys. Res. Lett.*, **109**, D17307, doi: 10.1029/2004JD004759.

356 Stevenson, D. S., F. J. Dentener, M. G. Schultz, K. Ellingsen, T. P. C. van Noije, G. Zeng,
357 M. Amann, C. S. Atherton, N. Bell, D. J. Bergmann, I. Bey, T. Butler, J. Cofala, W. J. Collins,
358 R. G. Derwent, R. M. Doherty, J. Drevet, H. J. Eskes, A. M. Fiore, M. Gauss, D. A.
359 Hauglustaine, L. W. Horowitz, I. S. A. Isaksen, M. C. Krol, J-F. Lamarque, M. G. Lawrence, V.
360 Montanaro, J-F. Muller, G. Pitari, M. J. Prather, J. A. Pyle, S. Rast, J. M. Rodriguez, M. G.
361 Sanderson, N. H. Savage, D. T. Shindell, S. E. Strahan, K. Sudo, and S. Szopa, 2006:
362 Multimodel ensemble simulations of present-day and near-future tropospheric ozone, *J.*
363 *Geophys. Res.*, 111, D08301, doi:10.1029/2005JD006338.
364
365 UNEP, 2001: Annual Evaluation Report United Nations Environmental Programme,
366 Evaluation and Oversight Unit 2002. available from http://www.unep.org/eou/Portals/52/Reports
367 /AnnualEvalReportEnglish2001.pdf.

368 van Vuuren, D. P., J. Edmonds, M. L. T. Kainuma, K. Riahi, A. Thomson, T. Matsui, G.
369 Hurtt, J-F. Lamarque, M. Meinshausen, S. Smith, C. Grainer, S. Rose, K. A. Hibbard, N.
370 Nakicenovic, V. Krey, T. Kram , 2011: The representative concentration pathways: an overview.
371 *Climatic Change*, **109**, 5-31, doi: 10.1007/s10584-011-0148-z.

372 Wild, O., M. J. Prather, and H. Akimoto, 2001: Indirect long-term global cooling from NOx
373 emissions. *Geophys. Res. Lett.*, **28**, 1719-22, doi: 10.1029/2000GL012573.

374 Wilkerson, J. T., M. Z. Jacobson, A. Malwitz, S. Balasubramanian, R. Wayson, G. Fleming,
375 A. D. Naiman, and S. K. Lele, 2010: Analysis of emission data from global commercial aviation:
376 2004 and 2006. *Atmos. Chem. Phys.*, **10**, 6391–6408, doi:10.5194/acp-10-6391-2010.

377 Wuebbles, D. J., and K. Hayhoe, 2000: Atmospheric methane: trends and impacts. *Non-*
378 *CO2 Greenhouse Gases: Scientific Understanding, Control, and Implementation*, 1-44.

Table 1. Results from comparisons of the three sets of simulations, a control and a NOx-perturbed simulation with fixed mixing ratio boundary conditions for CH_4, a NOx-perturbed simulation with a 20% increase in CH_4 mixing ratio at the boundary layer, and a control and a NOx-perturbed simulation with CH_4 fluxes.

	Global annual mean method (Approach 1)	Range using other methods (Approach 2-4)
CH_4 lifetime (yr), control simulation, fixed CH_4 mixing ratio boundary condition (Set 1)	10.74	9.70 to 11.4
CH_4 lifetime (yr), NOx-perturbed simulation, fixed CH_4 mixing ratio boundary condition (Set 1)	10.58	9.52 to 11.21
Aviation-induced ΔCH_4 (ppb) using Eq. 1	31.4	32.5 to 35.3
Aviation-induced ΔCH_4 (ppb) using CH_4 fluxes (Set 3)	28.9	28.9 to 29.4
CAM5 calculated feedback factor (Set 2)	1.39	1.35 to 1.43
Percent difference in feedback factor	-0.4%	-3.5% to 2.2%
Percent difference in CH_4 concentration using CH_4 surface fluxes compared to using fixed mixing ratio BCs	8. 6%	12.1% to 20.0%

Parameterization used to calculate the change in CH4 concentration was evaluated.

Our CAM5 simulations show that the parameterization technique is good within 10%.

Methane feedback on its own lifetime was calculated.

Our model dependent feedback factor is well within the range reported by IPCC (2001).

Technical Report Documentation Page

1. Report No.	2. Government Accession No.	3. Recipient's Catalog No.
j.atmosenv.2015.04.070		

4. Title and Subtitle	5. Report Date
Aviation NOx-induced CH4 effect: Fixed mixing ratio boundary conditions versus flux boundary conditions	May 2015
	6. Performing Organization Code

7. Author(s)	8. Performing Organization Report No.
Arezoo Khodayari, Seth C. Olsen, Donald J. Wuebbles, Daniel B. Phoenix	

9. Performing Organization Name and Address	10. Work Unit No. (TRAIS)
Center of Excellence for: Alternative Jet Fuels and Environment University of Illinois 105 S. Gregory Street, Urbana, IL 61801	11. Contract or Grant No. 13-C-AJFE-UI

12. Sponsoring Agency Name and Address	13. Type of Report and Period Covered
Federal Aviation Administration Office of Environment and Energy	Journal Article
	14. Sponsoring Agency Code

15. Supplementary Notes

16. Abstract

Atmospheric chemistry-climate models are often used to calculate the effect of aviation NOx emissions on atmospheric ozone (O3) and methane (CH4). Due to the long (~10 yr) atmospheric lifetime of methane, model simulations must be run for long time periods, typically for more than 40 simulation years, to reach steady-state if using CH4 emission fluxes. Because of the computational expense of such long runs, studies have traditionally used specified CH4 mixing ratio lower boundary conditions (BCs) and then applied a simple parameterization based on the change in CH4 lifetime between the control and NOx-perturbed simulations to estimate the change in CH4 concentration induced by NOx emissions. In this parameterization a feedback factor (typically a value of 1.4) is used to account for the feedback of CH4 concentrations on its lifetime. Modeling studies comparing simulations using CH4 surface fluxes and fixed mixing ratio BCs are used to examine the validity of this parameterization. The latest version of the Community Earth System Model (CESM), with the CAM5 atmospheric model, was used for this study. Aviation NOx emissions for 2006 were obtained from the AEDT (Aviation Environmental Design Tool) global commercial aircraft emissions. Results show a 31.4 ppb change in CH4 concentration when estimated using the parameterization and a 1.4 feedback factor, and a 28.9 ppb change when the concentration was directly calculated in the CH4 flux simulations. The model calculated value for CH4 feedback on its own lifetime agrees well with the 1.4 feedback factor. Systematic comparisons between the separate runs indicated that the parameterization technique overestimates the CH4 concentration by 8.6%. Therefore, it is concluded that the estimation technique is good to within ~10% and decreases the computational requirements in our simulations by nearly a factor of 8.

17. Key Words	18. Distribution Statement
Aviation NOx emissions, CH4 lifetime, Feedbacks, PARTNER, ASCENT	© 2015. This manuscript version is made available under the Elsevier user license http://www.elsevier.com/open-access/userlicense/1.0/

19. Security Classif. (of this report)	20. Security Classif. (of this page)	21. No. of Pages	22. Price
Unclassified	Unclassified		