

**Supplementary information of**

# **Impact of aircraft NO<sub>x</sub> and aerosol emissions on atmospheric composition and associated direct radiative forcing of climate**

**Terrenoire et al.**

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10 **Table S1.** Total global aircraft emissions corresponding to the baseline REACT4C\_2006 and QUANTIFY\_2000 inventories and comparison with ACCRI\_2006 (AEDT) (Brasseur et al., 2016) and CEDS\_2006 (Hoesly et al., 2018) inventories.

Emission	Units	REACT4C_2006	QUANTIFY_2000	ACCRI_2006	CEDS_2006
CO <sub>2</sub>	Tg yr <sup>-1</sup>	560	672	594	715
NO <sub>2</sub>	TgN yr <sup>-1</sup>	0.71	0.84	0.81	0.88
BC	Gg yr <sup>-1</sup>	4.0	5.0	5.96	6.0
SO <sub>x</sub>	GgS yr <sup>-1</sup>	73	88	113	110

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20 **Table S2.** Total global aircraft emissions corresponding to 2050 QUANTIFY\_A1B, QUANTIFY\_B1 and QUANTIFY\_B1\_ACARE inventories and comparison with ACCRI\_2050\_Base, ACCRI\_2050\_S1 (AEDT) (Brasseur et al., 2016) and SSP3.70 and SSP1.26 (Gidden et al., 2019) inventories.

Units	A1	B1	B1_ACARE	SSP3.70	SSP1.26	ACCRI_2050-Base	ACCRI_2050-S1
CO <sub>2</sub>	Tg yr <sup>-1</sup>	2257	1367	986	1301	421	2852
NO <sub>2</sub>	TgN yr <sup>-1</sup>	3.3	1.0	0.69	1.76	0.87	3.95
BC	Gg yr <sup>-1</sup>	16	8.9	6.4	19.7	10.3	29.0
SO <sub>x</sub>	GgS yr <sup>-1</sup>	289	175	124	257	141	541

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**Table S3.** Radiative forcing of ozone (F<sub>O<sub>3</sub></sub>), methane total forcing (F<sub>CH<sub>4</sub></sub>) and total of the two forcings calculated for the different simulations (mW/m<sup>2</sup>). The radiative forcing of methane is calculated based on Etminan et al. (2016) (F<sub>CH<sub>4</sub></sub>) or on Myhre et al. (1998) (F<sub>CH<sub>4</sub>old</sub>).

Scenario	F <sub>O<sub>3</sub></sub>	F <sub>CH<sub>4</sub></sub>	Total	F <sub>CH<sub>4</sub>old</sub>	Total <sup>old</sup>
REACT4C_2006	15.87	-14.69	<b>1.18</b>	-12.61	<b>3.26</b>
REACT4C_2006 NOx Only	15.90	-14.72	<b>1.19</b>	-12.63	<b>3.27</b>
QUANTIFY_2000	17.19	-16.69	<b>0.50</b>	-14.32	<b>2.87</b>
REACT4C_PLUS	17.72	-14.80	<b>2.92</b>	-12.71	<b>5.01</b>
REACT4C_MINUS	14.28	-14.68	<b>-0.40</b>	-12.60	<b>1.68</b>
QUANTIFY_A1	70.56	-70.84	<b>-0.28</b>	-60.86	<b>9.70</b>
QUANTIFY_A1_LowNOx	39.29	-39.45	<b>-0.16</b>	-33.91	<b>5.38</b>
QUANTIFY_A1_Desulfurized	70.08	-70.65	<b>-0.57</b>	-60.69	<b>9.39</b>
QUANTIFY_B1	27.57	-26.43	<b>1.14</b>	-22.73	<b>4.84</b>
QUANTIFY_B1_ACARE	18.74	-17.90	<b>0.84</b>	-15.40	<b>3.34</b>

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**Table S4.** Factors governing the climate impact of aviation NO<sub>x</sub> emissions and comparison with previous factor decompositions. The decomposition of the NO<sub>x</sub> forcing F<sub>NO<sub>x</sub></sub>, following the methodology proposed by Holmes et al. (2011), is given by:

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$$F = F_{O_3} + F_{CH_4}$$

where :

$$40 \quad F_{O_3} = \left( \frac{\partial [O_3]}{\partial E} \right) \left( \frac{\partial F}{\partial [O_3]} \right)$$

and

$$45 \quad F_{CH_4} = q_{CH_4} f \left( \frac{\partial L_{CH_4}}{\partial E} \right) \left( \frac{\partial F}{\partial q_{CH_4}} + \frac{\partial [O_3]}{\partial q_{CH_4}} \frac{\partial F}{\partial [O_3]} + \frac{\partial [H_2O]}{\partial q_{CH_4}} \frac{\partial F}{\partial [H_2O]} + \frac{\partial q_{CO_2}}{\partial q_{CH_4}} \frac{\partial F}{\partial q_{CO_2}} \right)$$

and where [O<sub>3</sub>] is the tropospheric ozone burden (DU), [H<sub>2</sub>O] the stratospheric water vapor burden (Tg), E the aircraft NO<sub>x</sub> emissions (TgN), q<sub>CH<sub>4</sub></sub> the methane mixing ratio (ppmv), q<sub>CO<sub>2</sub></sub> the carbon dioxide mixing ratio (ppmv), and L<sub>CH<sub>4</sub></sub> the methane loss rate through reaction with OH.

50 The NO<sub>x</sub> forcing is extended in order to include the forcings involving aerosols:

$$F = F_{O_3} + F_{CH_4-OH} + F_{CH_4-O_3} + F_{CH_4-SWV} + F_{CH_4-CO_2} + F_{SO_4} + F_{NO_3}$$

55 where F<sub>CH<sub>4</sub>-OH</sub>, F<sub>CH<sub>4</sub>-O<sub>3</sub></sub>, F<sub>CH<sub>4</sub>-SWV</sub>, and F<sub>CH<sub>4</sub>-CO<sub>2</sub></sub> are the methane forcing individual components, F<sub>SO<sub>4</sub></sub> the forcing associated with sulfate particles and F<sub>NO<sub>3</sub></sub> the forcing associated with nitrate particles. The NO<sub>x</sub> forcing components, normalized by the NO<sub>x</sub> emissions, for the REACT4C\_2006\_NOx\_Only simulation and based on **Table 3** are also provided in the Table. The methane forcing F<sub>CH<sub>4</sub>-OH</sub> is calculated based on Etminan et al. (2016) or on Myhre et al. (1998) (under parenthesis).

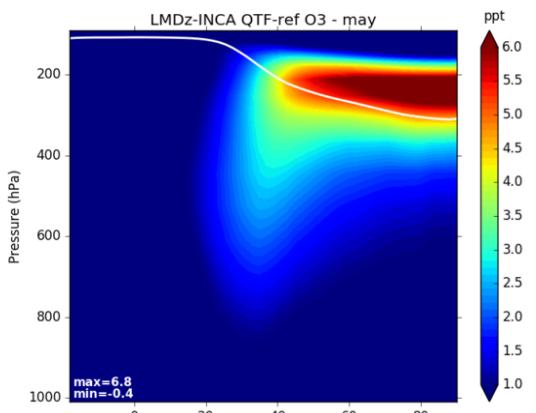
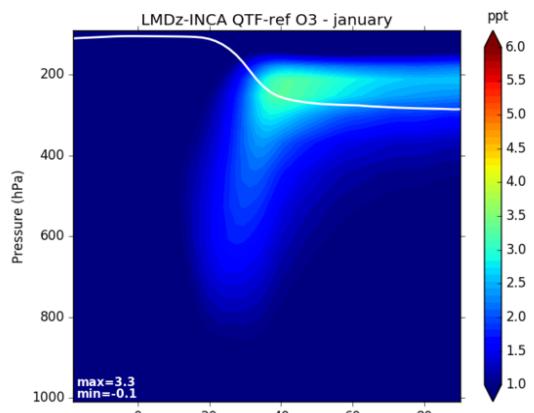
	Holmes et al. (2011)	Lee et al. (2021)	This work
$\partial [O_3]/\partial E$ [DU/TgN]	0.6±0.15		0.60
$\partial F/\partial [O_3]$ [mW/m <sup>2</sup> /DU]	36±8		37.3
$\partial L_{CH_4}/\partial E$ [%/TgN]	-1.7±0.35		-1.4
$\partial [O_3]/\partial q_{CH_4}$ [DU/ppmv]	3.5±1.0		3.5
$\partial [H_2O]/\partial q_{CH_4}$ [Tg/ppmv]			87.6
$\partial q_{CO_2}/\partial q_{CH_4}$ [ppmv/ppmv]			0.6
$\partial F/\partial q_{CH_4}$ [mW/m <sup>2</sup> /ppmv]	370		454.1 (368.3)
$\partial F/\partial [O_3]$ [mW/m <sup>2</sup> /DU]	36±8		33.2
$\partial F/\partial [H_2O]$ [mW/m <sup>2</sup> /Tg]			0.26
$\partial F/\partial q_{CO_2}$ [mW/m <sup>2</sup> /ppmv]			13.7
F <sub>O<sub>3</sub></sub> (mW/m <sup>2</sup> /TgN)	21.6±7.2	25.1±7.2	22.3
F <sub>CH<sub>4</sub>-OH</sub> (mW/m <sup>2</sup> /TgN)	-15.7±3.6	-13.4±4.5	-15.5 (-12.6)
F <sub>CH<sub>4</sub>-O<sub>3</sub></sub> (mW/m <sup>2</sup> /TgN)	-5.3±2.2	-6.7±2.3	-3.98
F <sub>CH<sub>4</sub>-swv</sub> (mW/m <sup>2</sup> /TgN)	-	-2.0±0.7	-0.92
F <sub>CH<sub>4</sub>-CO<sub>2</sub></sub> (mW/m <sup>2</sup> /TgN)	-	-	-0.28
F <sub>SO<sub>4</sub></sub> (mW/m <sup>2</sup> /TgN)	-	-	-2.81
F <sub>NO<sub>3</sub></sub> (mW/m <sup>2</sup> /TgN)	-	-	-0.17
Total (F <sub>O<sub>3</sub></sub> + F <sub>CH<sub>4</sub>-OH</sub> + F <sub>CH<sub>4</sub>-O<sub>3</sub></sub> )	0.6±8.3	5.0±8.8	2.9 (2.0)
Total (F <sub>O<sub>3</sub></sub> + F <sub>CH<sub>4</sub>-OH</sub> + F <sub>CH<sub>4</sub>-O<sub>3</sub></sub> + F <sub>CH<sub>4</sub>-swv</sub> )	-	4.0±5.8	1.9 (4.9)
Total (all)	-	-	-1.3 (1.6)

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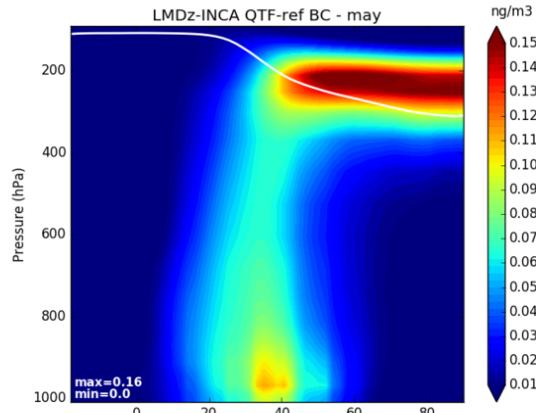
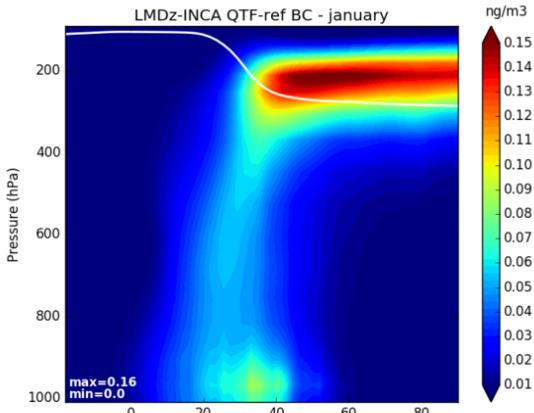
**Table S5.** Radiative forcing of ozone ( $F_{O_3}$ ), methane total forcing ( $F_{CH_4}$ ) and total of the two forcings calculated for the different simulations ( $mW/m^2$ ). The radiative forcing of methane is calculated without ( $F_{CH_4}$ ) or with a transient correction factor ( $F_{CH_4}^{tr}$ ).

Scenario	$F_{O_3}$	$F_{CH_4}$	Total	$F_{CH_4}^{tr}$	Total <sup>tr</sup>
REACT4C_2006	15.87	-14.69	<b>1.18</b>	-11.02	<b>4.85</b>
REACT4C_2006 NOx Only	15.90	-14.72	<b>1.19</b>	-11.04	<b>4.86</b>
QUANTIFY_2000	17.19	-16.69	<b>0.50</b>	-12.18	<b>5.01</b>
REACT4C_PLUS	17.72	-14.80	<b>2.92</b>	-11.10	<b>6.62</b>
REACT4C_MINUS	14.28	-14.68	<b>-0.40</b>	-11.01	<b>3.27</b>
QUANTIFY_A1	70.56	-70.84	<b>-0.28</b>	-52.42	<b>18.14</b>
QUANTIFY_A1_LowNOx	39.29	-39.45	<b>-0.16</b>	-29.16	<b>10.10</b>
QUANTIFY_A1_Desulfurized	70.08	-70.65	<b>-0.57</b>	-52.28	<b>17.80</b>
QUANTIFY_B1	27.57	-26.43	<b>1.14</b>	-26.43	<b>1.14</b>
QUANTIFY_B1_ACARE	18.74	-17.90	<b>0.84</b>	-20.58	<b>-1.84</b>

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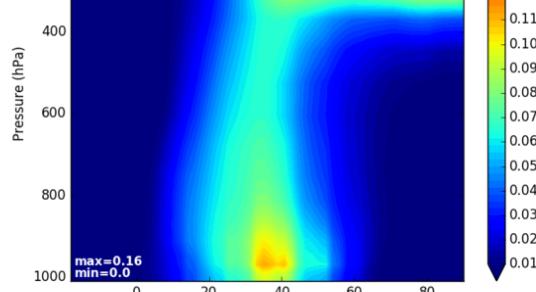
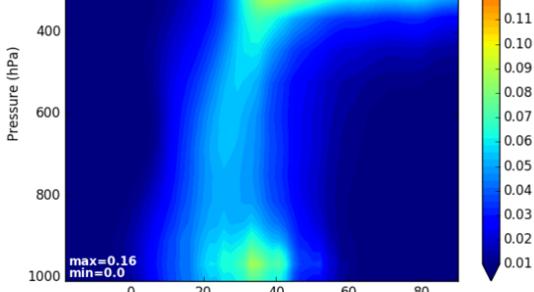


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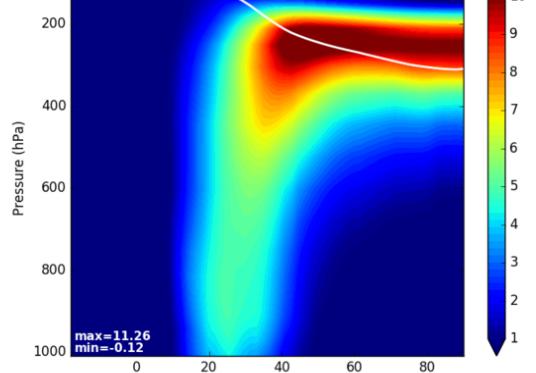
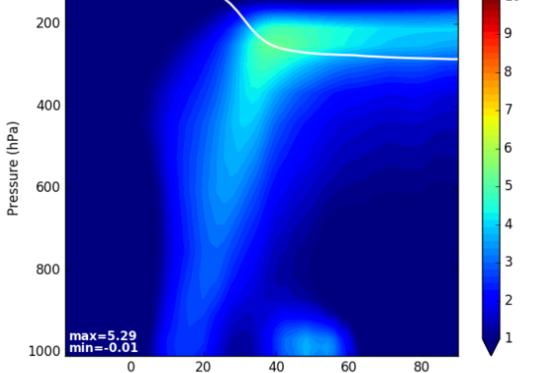
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**Figure S1.** Zonal mean perturbation due to aircraft emissions for January (left) and May (right) of O<sub>3</sub> (ppbv), BC (ng/m<sup>3</sup>), SO<sub>4</sub> (ng/m<sup>3</sup>) and NO<sub>3</sub> (ng/m<sup>3</sup>) for the QUANTIFY\_2000 inventory. The solid line represents the tropopause pressure.