Supplement of Atmos. Chem. Phys., 22, 11987–12023, 2022
https://doi.org/10.5194/acp-22-11987-2022-supplement

Atmospheric Chemistry and Physics
Open Access
EGU

Supplement of

Impact of present and future aircraft NO_x and aerosol emissions on atmospheric composition and associated direct radiative forcing of climate

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Supplementary information of

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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_2	Tg yr^{-1}	560	672	594	715
NO_2	TgN yr^{-1}	0.71	0.84	0.81	0.88
BC	Gg yr^{-1}	4.0	5.0	5.96	6.0
SO_x	GgS yr^{-1}	73	88	113	110

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_2	Tg yr^{-1}	2257	1367	986	1301	421	2852	1625
NO_2	TgN yr^{-1}	3.3	1.0	0.69	1.76	0.87	3.95	1.57
BC	Gg yr^{-1}	16	8.9	6.4	19.7	10.3	29.0	16.6
SO_x	GgS yr^{-1}	289	175	124	257	141	541	308

Table S3. Radiative forcing of ozone (F_{O3}), methane total forcing (F_{CH4}) and total of the two forcings calculated for the different simulations (mW/m^2). The radiative forcing of methane is calculated based on Etminan et al. (2016) (F_{CH4}) or on Myhre et al. (1998) (F_{CH4}^{old}).

Scenario	F_{O3}	F_{CH4}	Total	F_{CH4}^{old}	$Total^{old}$
REACT4C_2006	15.87	-14.69	**1.18**	-12.61	**3.26**
REACT4C_2006 NOx Only	15.90	-14.72	**1.19**	-12.63	**3.27**
QUANTIFY_2000	17.19	-16.69	**0.50**	-14.32	**2.87**
REACT4C_PLUS	17.72	-14.80	**2.92**	-12.71	**5.01**
REACT4C_MINUS	14.28	-14.68	**-0.40**	-12.60	**1.68**
QUANTIFY_A1	70.56	-70.84	**-0.28**	-60.86	**9.70**
QUANTIFY_A1_LowNOx	39.29	-39.45	**-0.16**	-33.91	**5.38**
QUANTIFY_A1_Desulfurized	70.08	-70.65	**-0.57**	-60.69	**9.39**
QUANTIFY_B1	27.57	-26.43	**1.14**	-22.73	**4.84**
QUANTIFY_B1_ACARE	18.74	-17.90	**0.84**	-15.40	**3.34**

Table S4. Factors governing the climate impact of aviation NO_x emissions and comparison with previous factor decompositions. The decomposition of the NO_x forcing F_{NOx}, following the methodology proposed by Holmes et al. (2011), is given by:

$$F = F_{O3} + F_{CH4}$$

where :

$$F_{O3} = \left(\frac{\partial[O_3]}{\partial E}\right)\left(\frac{\partial F}{\partial[O_3]}\right)$$

and

$$F_{CH4} = q_{CH4}\, f \left(\frac{\partial L_{CH4}}{\partial E}\right)\left(\frac{\partial F}{\partial q_{CH4}} + \frac{\partial[O_3]}{\partial q_{CH4}}\frac{\partial F}{\partial[O_3]} + \frac{\partial[H_2O]}{\partial q_{CH4}}\frac{\partial F}{\partial[H_2O]} + \frac{\partial q_{CO2}}{\partial q_{CH4}}\frac{\partial F}{\partial q_{CO2}}\right)$$

and where $[O_3]$ is the tropospheric ozone burden (DU), $[H_2O]$ the stratospheric wator vapor burden (Tg), E the aircraft NO_x emissions (TgN), q_{CH4} the methane mixing ratio (ppmv), q_{CO2} the carbon dioxide mixing ratio (ppmv), and L_{CH4} the methane loss rate through reaction with OH.

The NO_x forcing is extended in order to include the forcings involving aerosols:

$$F = F_{O3} + F_{CH4\text{-}OH} + F_{CH4\text{-}O3} + F_{CH4\text{-}SWV} + F_{CH4\text{-}CO2} + F_{SO4} + F_{NO3}$$

where $F_{CH4\text{-}OH}$, $F_{CH4\text{-}O3}$, $F_{CH4\text{-}SWV}$, and $F_{CH4\text{-}CO2}$ are the methane forcing individual components, F_{SO4} the forcing associated with sulfate particles and F_{NO3} the forcing associated with nitrate particles. The NO_x forcing components, normalized by the NO_x emissions, for the REACT4C_2006_NOx_Only simulation and based on **Table 3** are also provided in the Table. The methane forcing $F_{CH4\text{-}OH}$ 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**
∂[O3]/ ∂E [DU/TgN]	0.6±0.15		0.60
∂F/∂[O3] [mW/m^2/DU]	36±8		37.3
$\partial L_{CH4}/\partial E$ [%/TgN]	-1.7±0.35		-1.4
∂[O3]/∂q_{CH4} [DU/ppmv]	3.5±1.0		3.5
∂[H2O]/∂q_{CH4} [Tg/ppmv]			87.6
$\partial q_{CO2}/\partial q_{CH4}$ [ppmv/ppmv]			0.6
∂F/∂q_{CH4} [mW/m^2/ppmv]	370		454.1 (368.3)
∂F/∂[O3] [mW/m^2/DU]	36±8		33.2
∂F/∂[H2O] [mW/m^2/Tg]			0.26
∂F/∂q_{CO2} [mW/m^2/ppmv]			13.7
F_{O3} (mW/m^2/TgN)	21.6±7.2	25.1±7.2	22.3
$F_{CH4\text{-}OH}$ (mW/m^2/TgN)	-15.7±3.6	-13.4±4.5	-15.5 (-12.6)
$F_{CH4\text{-}O3}$ (mW/m^2/TgN)	-5.3±2.2	-6.7±2.3	-3.98
$F_{CH4\text{-}swv}$ (mW/m^2/TgN)	-	-2.0±0.7	-0.92
$F_{CH4\text{-}CO2}$ (mW/m^2/TgN)	-	-	-0.28
F_{SO4} (mW/m^2/TgN)	-	-	-2.81
F_{NO3} (mW/m^2/TgN)	-	-	-0.17
Total ($F_{O3} + F_{CH4\text{-}OH} + F_{CH4\text{-}O3}$)	0.6±8.3	5.0±8.8	2.9 (2.0)
Total ($F_{O3} + F_{CH4\text{-}OH} + F_{CH4\text{-}O3} + F_{CH4\text{-}swv}$)	-	4.0±5.8	1.9 (4.9)
Total (all)	-	-	-1.3 (1.6)

Table S5. Radiative forcing of ozone (F_{O3}), methane total forcing (F_{CH4}) and total of the two forcings calculated for the different simulations (mW/m^2). The radiative forcing of methane is calculated without (F_{CH4}) or with a transient correction factor (F_{CH4}^{tr}).

Scenario	**F_{O3}**	**F_{CH4}**	**Total**	**F_{CH4}^{tr}**	**Total^{tr}**
REACT4C_2006	15.87	-14.69	**1.18**	-11.02	**4.85**
REACT4C_2006 NOx Only	15.90	-14.72	**1.19**	-11.04	**4.86**
QUANTIFY_2000	17.19	-16.69	**0.50**	-12.18	**5.01**
REACT4C_PLUS	17.72	-14.80	**2.92**	-11.10	**6.62**
REACT4C_MINUS	14.28	-14.68	**-0.40**	-11.01	**3.27**
QUANTIFY_A1	70.56	-70.84	**-0.28**	-52.42	**18.14**
QUANTIFY_A1_LowNOx	39.29	-39.45	**-0.16**	-29.16	**10.10**
QUANTIFY_A1_Desulfurized	70.08	-70.65	**-0.57**	-52.28	**17.80**
QUANTIFY_B1	27.57	-26.43	**1.14**	-26.43	**1.14**
QUANTIFY_B1_ACARE	18.74	-17.90	**0.84**	-20.58	**-1.84**

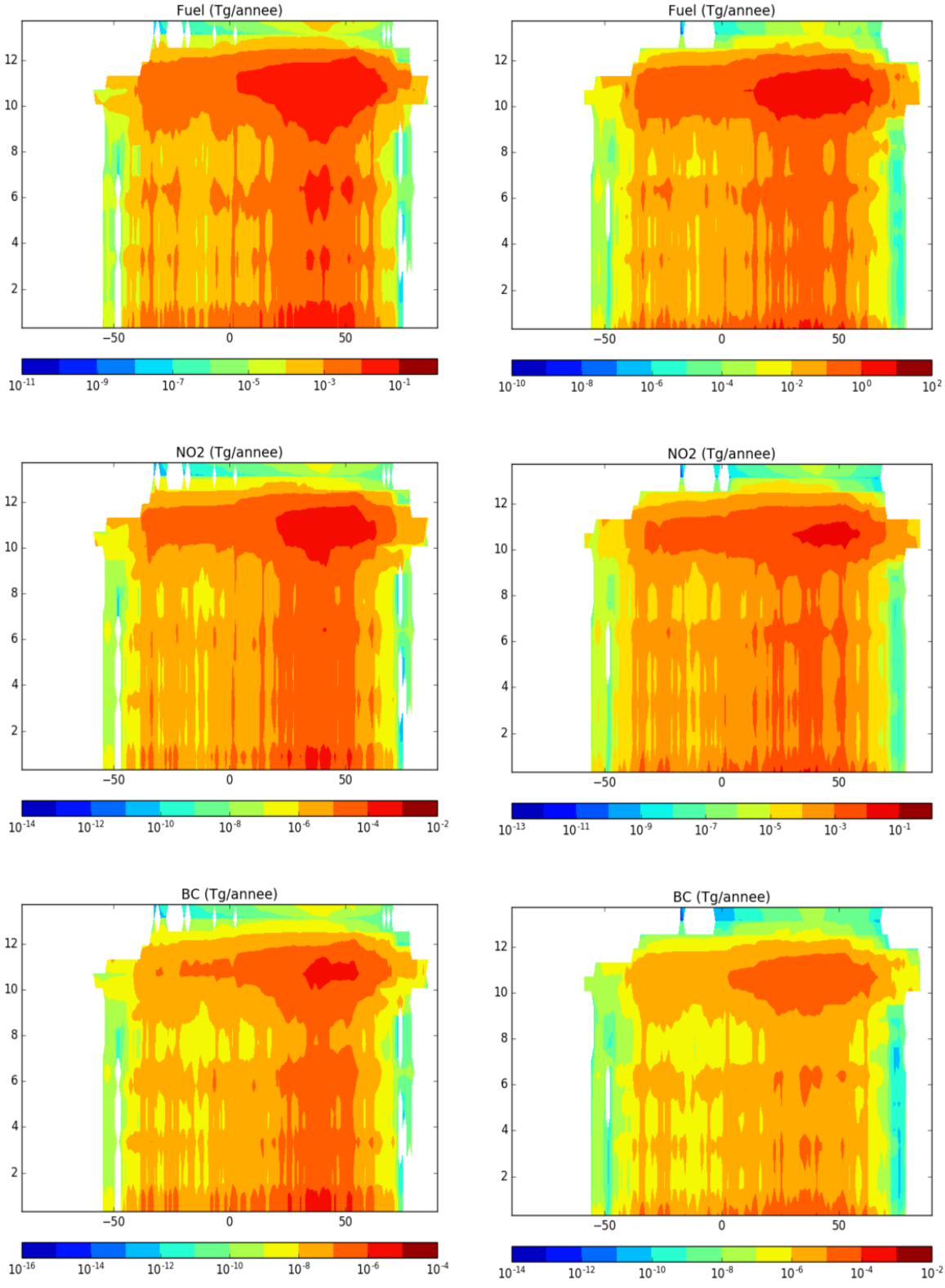

Figure S1. Zonal and annual mean fuel consumption (top row), NO_2 (middle row) and BC (bottom row) emissions for the REACT4C_2006 (left) and QUANTIFY_A1_2050 inventories (Tg/gridcell/yr). Please note the different color scales for the "present" and "future" inventories.

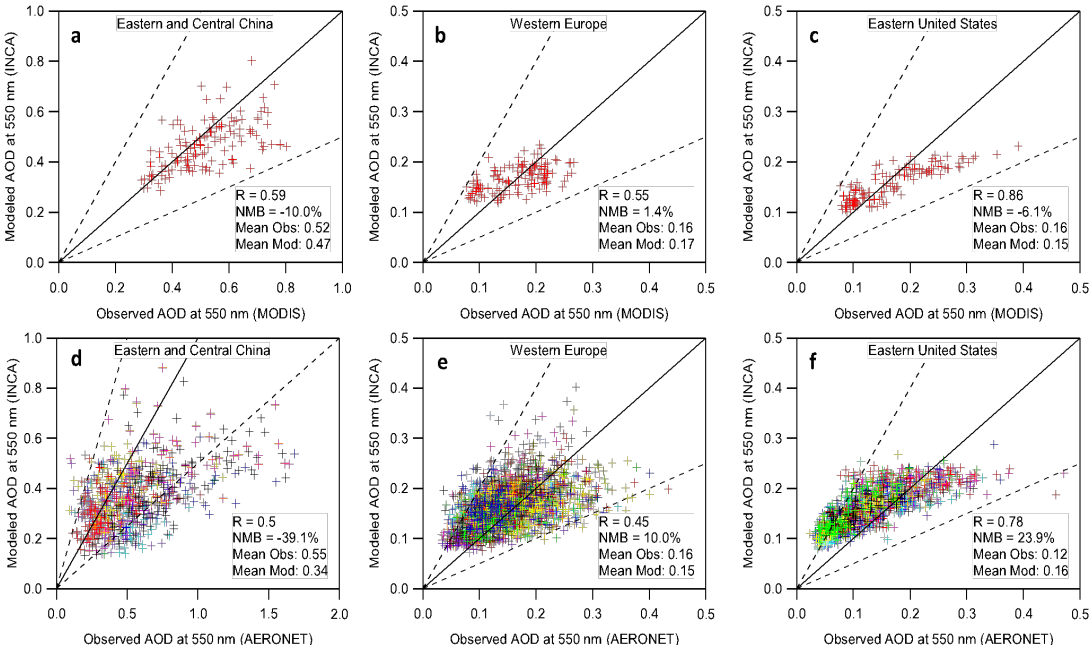

Figure S2. Simulated total aerosol optical depth at 550 nm for the year 2007-2017 compared to the MODIS data (top row) and the AERONET data worldwide (all available stations in different colors), over Eastern and Central China, Western Europe, , and the Eastern United States. Please note the different scale for the Eastern and Central China region. Dashed lines indicate 1:2 and 2:1 ratios.

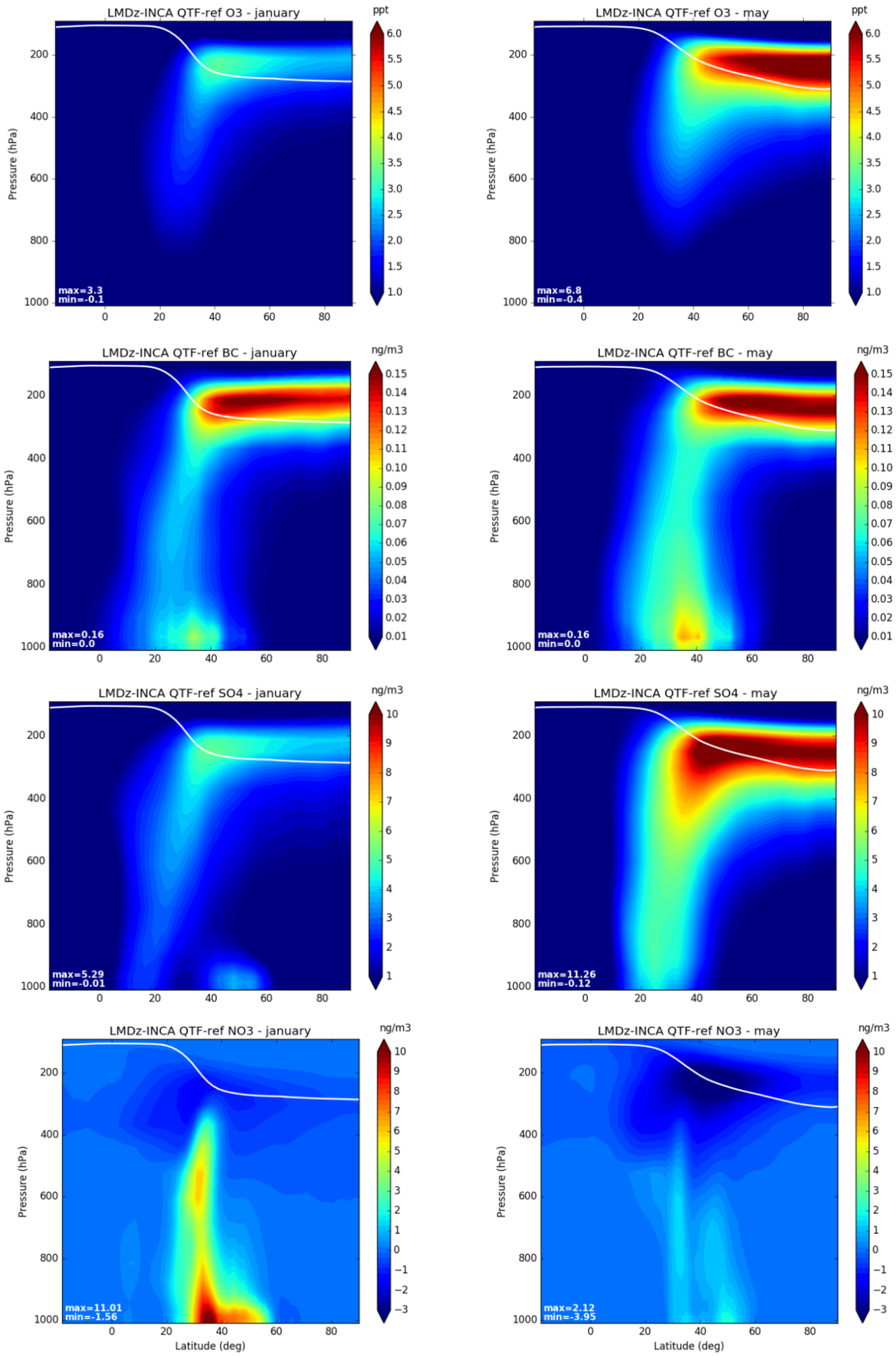

Figure S3. Zonal mean perturbation due to aircraft emissions for January (left) and May (right) of O_3 (ppbv), BC (ng/m^3), SO_4 (ng/m^3) and NO_3 (ng/m^3) for the QUANTIFY_2000 inventory. The solid line represents the tropopause pressure.