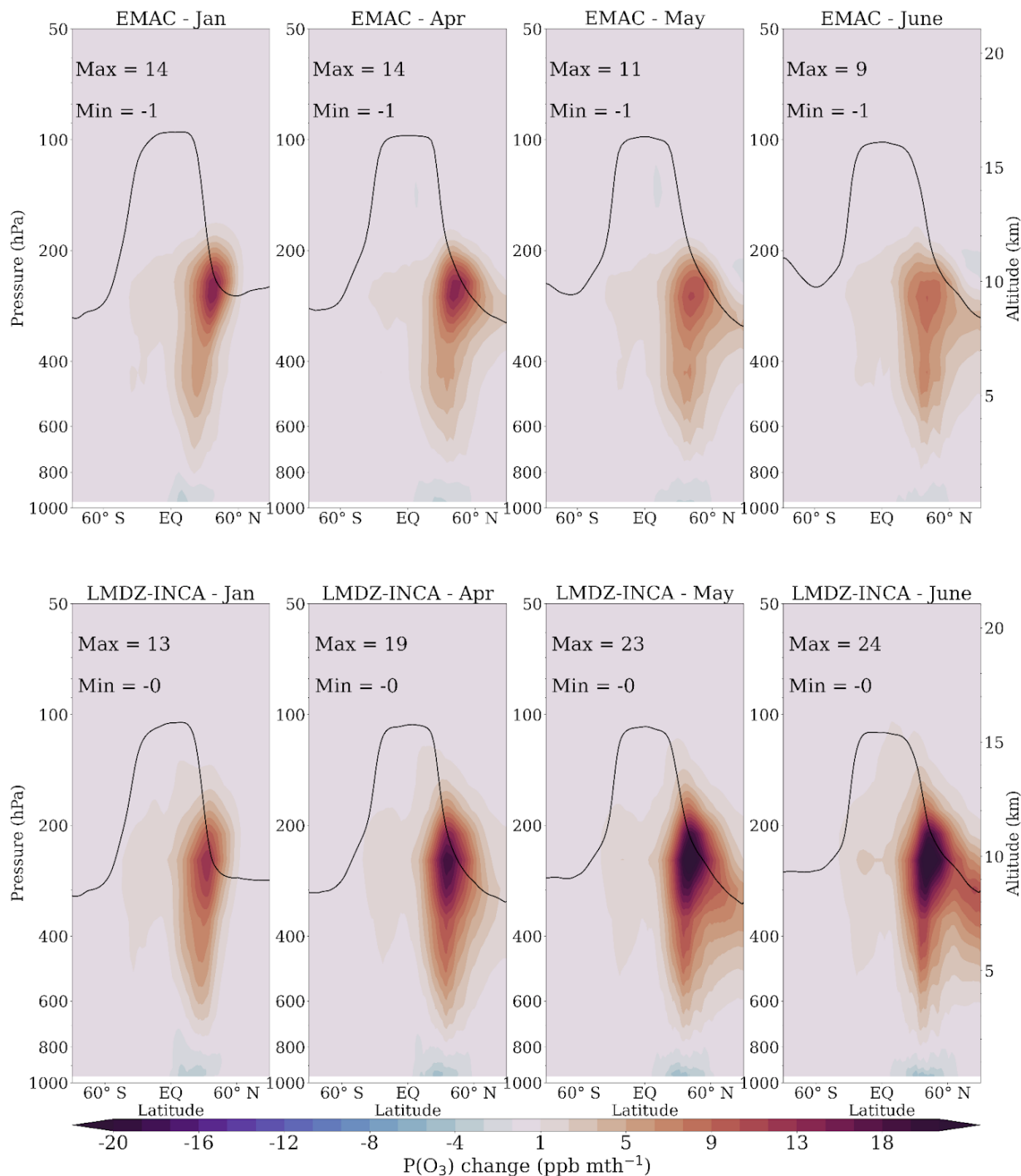


# Supplementary material

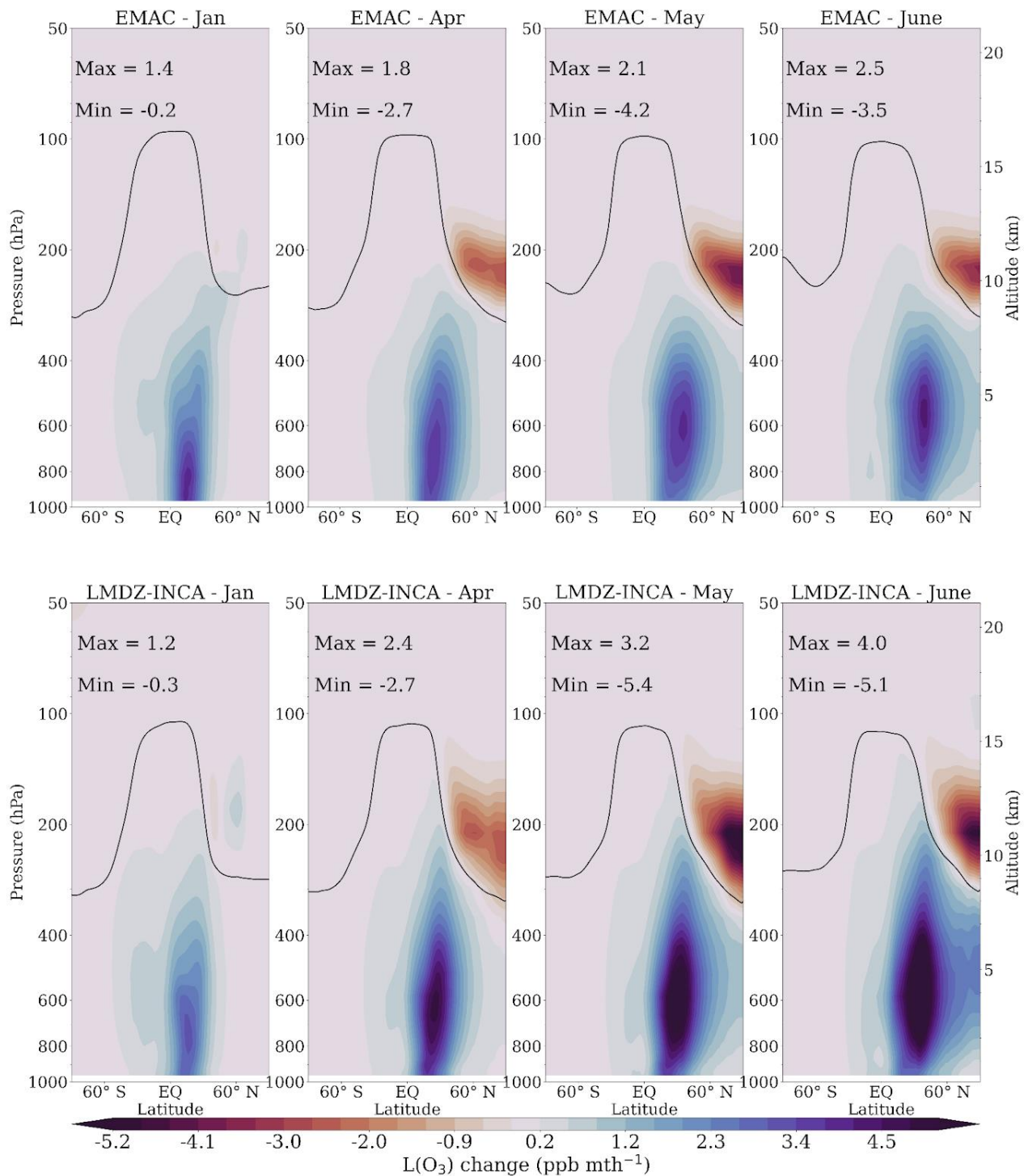
## S1. Chemical background in the UTLS

Table S1: Backgrounds expressed as global mass burdens, between 150 and 300 hPa, annually. The black carbon lifetime is written in brackets below the BC burden. It is worth noting that it is derived from the total BC burden, and not only that in the UTLS.

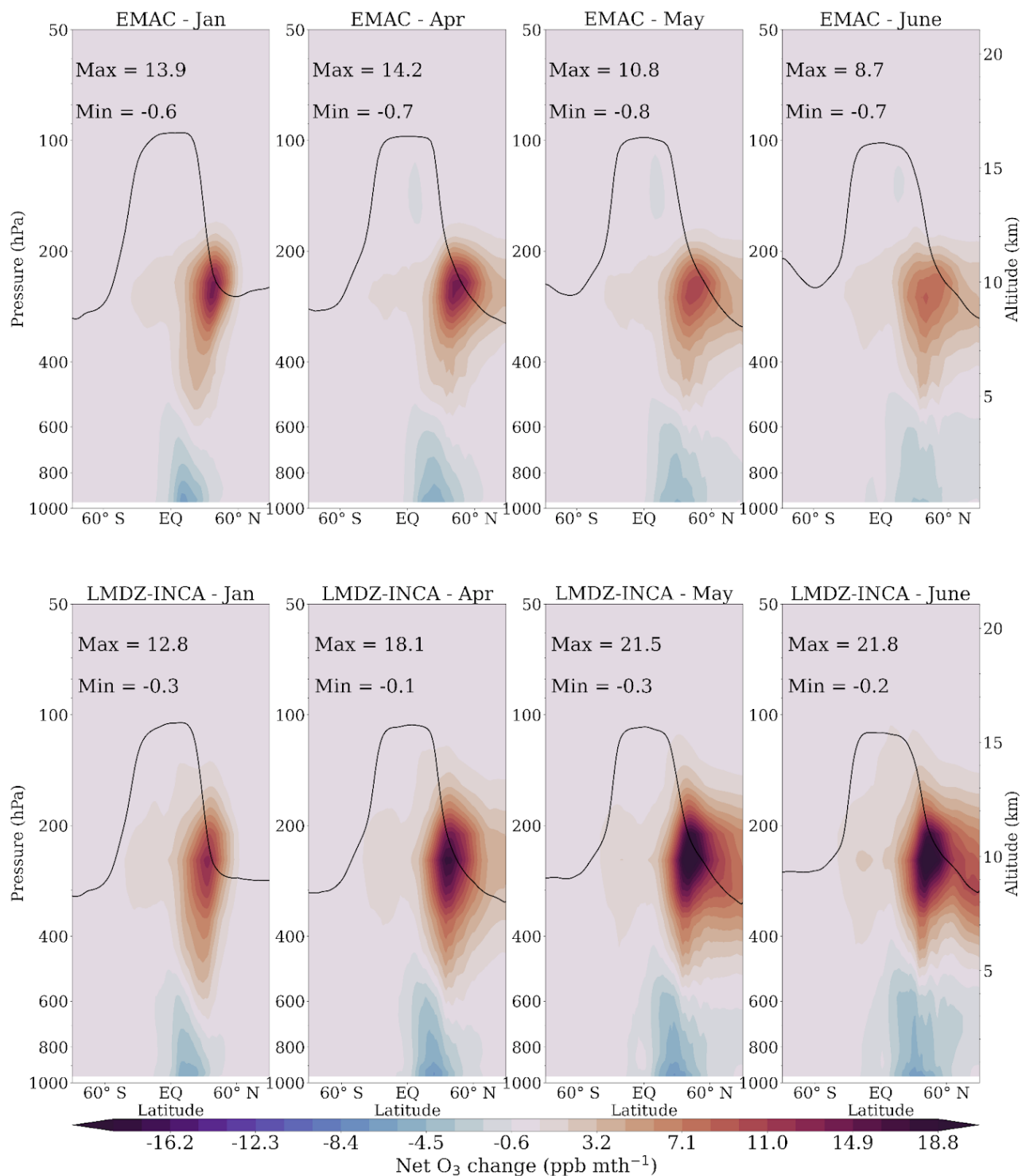
| Model                              | EMAC-NO <sub>x</sub>  | LMDZ-INCA             | MOZART3               | OsloCTM3              | GEOS-Chem             |
|------------------------------------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|
| Period                             | 2014–2018             | 2014–2018             | 2014–2018             | 2014–2017             | 2019                  |
| O <sub>3</sub> (TgO <sub>3</sub> ) | 199                   | 218                   | 231                   | 172                   | 139                   |
| NO <sub>y</sub> (TgN)              | 0.318                 | 0.292                 | 0.230                 | 0.317                 | 0.167                 |
| NO <sub>x</sub> (TgN)              | 6.76 10 <sup>-2</sup> | 3.42 10 <sup>-2</sup> | 4.64 10 <sup>-2</sup> | 6.95 10 <sup>-2</sup> | 2.53 10 <sup>-2</sup> |
| HNO <sub>3</sub> (TgN)             | 0.152                 | 0.152                 | 0.125                 | 0.224                 | 0.107                 |
| OH (Tg)                            | 8.47 10 <sup>-5</sup> | 7.02 10 <sup>-5</sup> | 7.31 10 <sup>-5</sup> | 8.26 10 <sup>-5</sup> | 5.88 10 <sup>-5</sup> |
| CH <sub>4</sub> (TgC)              | 641                   | 744                   | 651                   | 614                   | 546                   |
| NH <sub>3</sub> (TgN)              | 3.06 10 <sup>-2</sup> | 7.31 10 <sup>-3</sup> | -                     | 3.09 10 <sup>-4</sup> | 1.22 10 <sup>-3</sup> |
| SO <sub>2</sub> (TgS)              | 2.33 10 <sup>-2</sup> | 4.91 10 <sup>-2</sup> | -                     | 7.53 10 <sup>-3</sup> | 1.95 10 <sup>-2</sup> |
| Aerosols                           | EMAC-aer              |                       |                       |                       |                       |
| BC (Gg)<br>[lifetime]              | 3.95<br>[7.7 days]    | 38.0<br>[8.0 days]    | -                     | 2.93<br>[4.6 days]    | 1.55<br>[5.1 days]    |
| SO <sub>4</sub> (GgS)              | 116                   | 473                   | -                     | 384                   | 59.6                  |
| NO <sub>3</sub> (GgN)              | 56.2                  | 239                   | -                     | 226                   | 33.6                  |



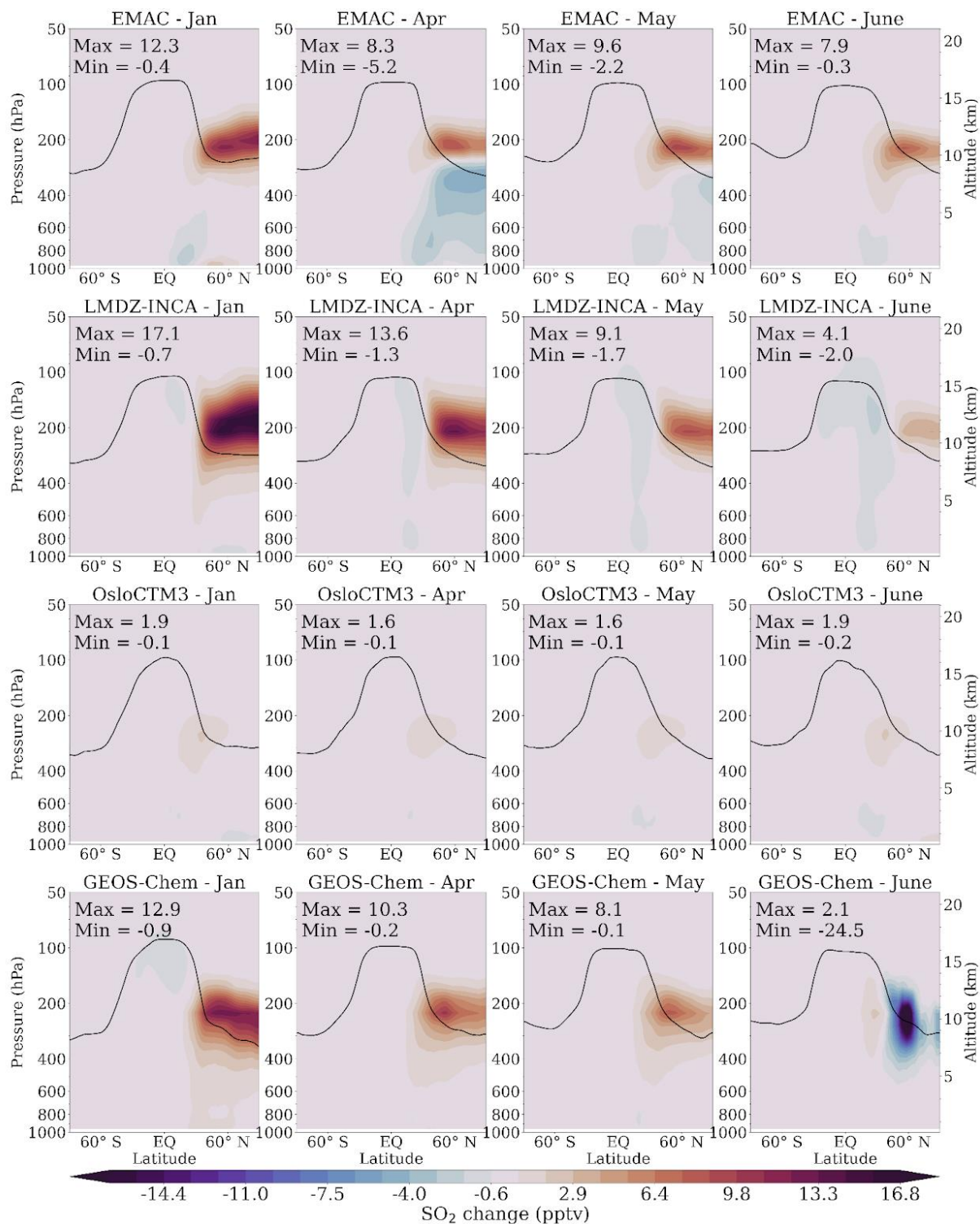
**Figure S1: Zonal cross sections in the perturbation of ozone production  $P(O_3)$ , for the EMAC and LMDZ-INCA models (top and bottom), during January, April, May, and June (from left to right).**



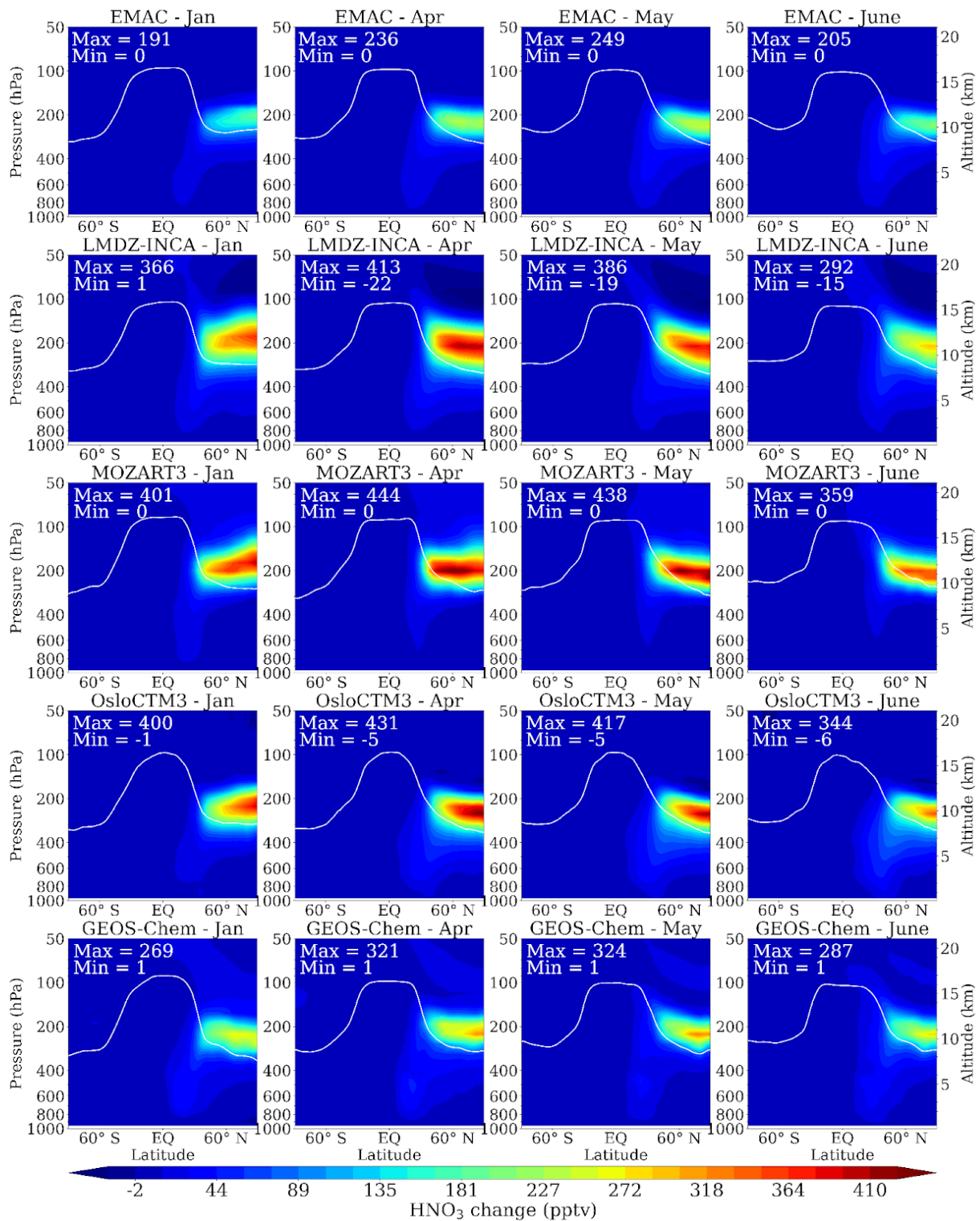
15 **Figure S2:** Same as Fig. S1 for ozone chemical loss  $L(O_3)$ . Note that the colors are reversed.



**Figure S3: Same as Fig. S1 for net ozone production  $P(O_3) - L(O_3)$ .**



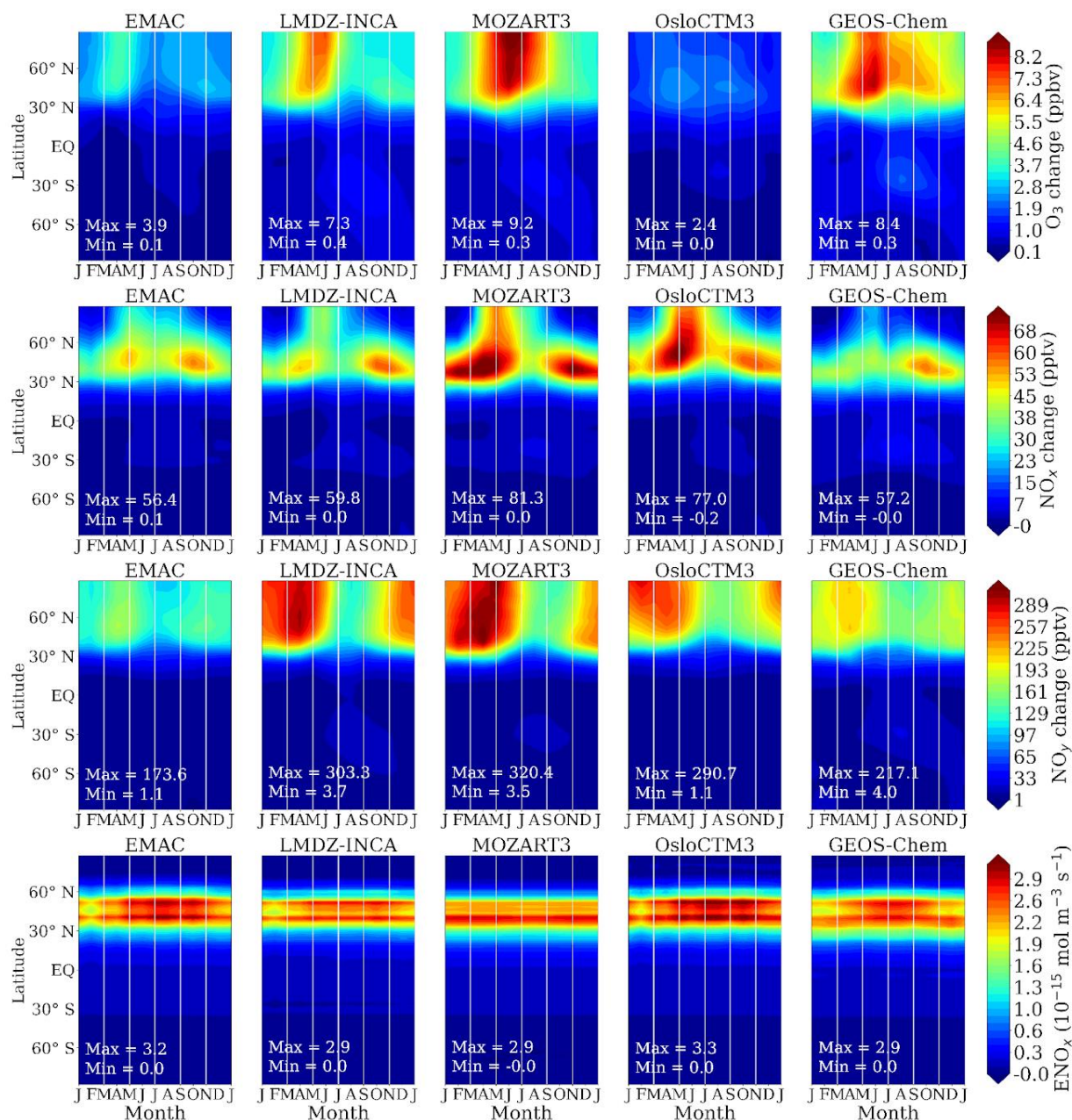
**Figure S4: Zonal cross sections in the perturbation sulfate dioxide ( $\text{SO}_2$ ), for the EMAC and LMDZ-INCA models (top and bottom), during January, April, May, and June (from left to right).**



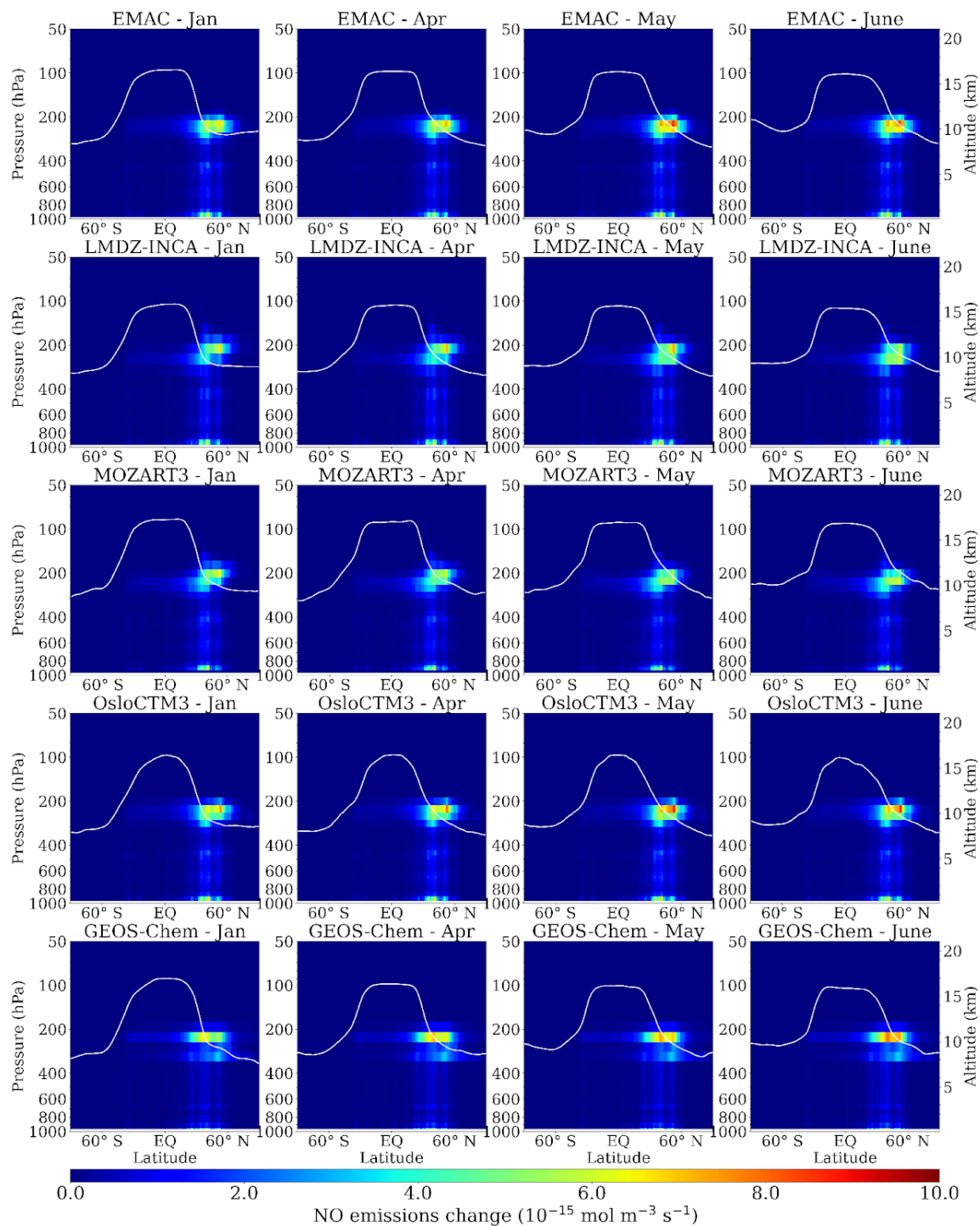
**Figure S5:** Same as Fig. S4 for nitric acid ( $\text{HNO}_3$ ).



### S3. NO<sub>x</sub> emissions



25 **Figure S6: Hovmöller diagrams in NO<sub>x</sub> emissions for each model, between 150 and 350 hPa.**



**Figure S7: Zonal cross sections in  $\text{NO}_x$  emissions for each model (from top to bottom) during Jan, April, May, and June (from left to right).**



S4. Linearity of the responses with respect to NO<sub>x</sub> emissions

30 Table S2: Ratio between the 100% perturbation and the 20% perturbation rescaled up to 100%, for each model (columns) and gaseous species (row). TCH<sub>4</sub> refers to methane lifetime.

| Ratio 100% / 20% | LMDZ-INCA | MOZART3 | OsloCTM3 |
|------------------|-----------|---------|----------|
| O <sub>3</sub>   | 1.13      | 1.18    | 1.09     |
| NO <sub>y</sub>  | 1.08      | 0.97    | 1.04     |
| NO <sub>x</sub>  | 0.94      | 0.92    | 1.04     |
| HNO <sub>3</sub> | 1.00      | 0.97    | 1.03     |
| PAN              | 1.12      | 1.11    | 1.14     |
| CO               | 1.10      | 1.12    | 1.08     |
| TCH <sub>4</sub> | 1.09      | 1.09    | 1.05     |
| OH               | 1.11      | 1.14    | 1.08     |

S5. Radiative forcing terms

35 Table S3: Effective radiative forcings from the different terms linked to NO<sub>x</sub> emissions, as in Fig. 12a. The value in bracket shown for the short-term ozone forcing in LMDZ-INCA is calculated from an offline version of the LMDZ GCM radiative code, as described in Terrenoire et al. (2022).

| ERF (mW m <sup>-2</sup> )      | EMAC-NO <sub>x</sub> | LMDZ-INCA   | MOZART3 | OsloCTM3 | GEOS-Chem |
|--------------------------------|----------------------|-------------|---------|----------|-----------|
| Short-term ozone               | 27.7                 | 43.0 [40.6] | 42.0    | 34.0     | 56.0      |
| CH <sub>4</sub> direct effect  | -13.2                | -17.1       | -12.9   | -12.9    | -24.8     |
| Long-term ozone                | -5.37                | -6.96       | -5.24   | -5.24    | -10.1     |
| Stratospheric H <sub>2</sub> O | -1.74                | -2.25       | -1.70   | -1.70    | -3.26     |
| Total CH <sub>4</sub>          | -20.4                | -26.4       | -19.9   | -19.9    | -38.2     |
| Total                          | 7.37                 | 16.7        | 22.2    | 14.1     | 17.9      |

**Table S4: Effective direct radiative forcings of the different aerosol species, as in Fig. 12b.**

| Aerosol-radiation interactions<br>(mW m <sup>-2</sup> ) | EMAC-aer<br>(2015) | LMDZ-INCA<br>(2014–2018) | OsloCTM3<br>(2014–2017) | GEOS-Chem<br>(2019) |
|---|--------------------|--------------------------|-------------------------|---------------------|
| BC  | 6.37               | 2.08                     | 0.82                    | 2.65                |
| OC  | -1.16              | -0.09                    | -0.05                   | -0.19               |
| SO <sub>4</sub>   | -11.2              | -6.85                    | -5.10                   | -8.09               |
| NO <sub>3</sub>   | -3.82              | -1.62                    | -13.5                   | -6.36               |
| Total Aerosol   | -10.2              | -6.47                    | -17.8                   | -12.0               |

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