



## Supplement of

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Figure S1. Map of the two domains of this study. The outer area, marked with the red colour, refers to the European domain while the inner area, marked with the blue colour, refers to the Greek domain.



Figure S2. Aggregated anthropogenic and biogenic NO emissions in Greece for June-December 2018 (left) and biogenic NO emissions (right) for the same period, in logarithmic scale.



**Figure S3.** NO<sub>2</sub> diurnal cycle for three air quality stations during the period June to December 2018, AUTH/Malakopi, Peristeri and Geoponiki, from top to bottom. The in situ measurements are shown in black and the LOTOS-EUROS simulations in red (shaded areas denote the standard deviation of the temporal average.)



**Figure S4.** Scatter plot between daily mean LOTOS-EUROS and MAX-DOAS NO<sub>2</sub> columns in AUTH for July (upper) and December (lower). The regression equation and the correlation coefficient between the model and the observed data are given in the top left of each plot. The horizontal error bars refer to the standard deviation of averaged MAX-DOAS observations while the vertical error bars refer to the standard deviations.



Figure S5. Time series of LOTOS-EUROS (red) and MAX-DOAS (green) NO<sub>2</sub> columns over Athens for July (upper) and December (lower) 2018 in rural direction.



**Figure S6.** Average diurnal cycle of the MAX-DOAS in Athens (green line), LOTOS-EUROS full-profile (red line) NO<sub>2</sub> columns, LOTOS-EUROS partial NO<sub>2</sub> columns (blue line) and boundary layer height (black line) during July (upper) and December (lower) for rural direction (bottom). The error bars refer to the standard deviations of the averaged measurements and simulations.



**Figure S7.** Scatter plots between daily LOTOS-EUROS integrated column above 424 m with MAX-DOAS NO<sub>2</sub> columns in Athens for July (upper) and December 2018 (lower) for the urban direction. The horizontal error bars refer to the standard deviation of averaged MAX-DOAS observations while the vertical error bars refer to the standard deviation of averaged LOTOS-EUROS simulations. The linear regression equation and the correlation coefficient between the model and the observed data are given in the top right of each plot.



**Figure S8.** Scatter plots between daily LOTOS-EUROS integrated column above 424 m with MAX-DOAS NO<sub>2</sub> columns in Athens for July (upper) and December 2018 (lower) for the rural viewing direction. The horizontal error bars refer to the standard deviation of averaged MAX-DOAS observations while the vertical error bars refer to the standard deviation of averaged LOTOS-EUROS simulations. The linear regression equation and the correlation coefficient between the model and the observed data are given in the top right of each plot.



Figure S9. Bar plots of the relative biases between the LOTOS-EUROS partial column and the MAX-DOAS measurements during July for the urban direction. The y-axis refers to the percentage of occurrences of biases.



**Figure S10.** Average diurnal cycle of NO<sub>2</sub> (black line) and the standard deviations (black shaded area) of the suburban industrial station "Geoponiki" in Athens and LOTOS-EUROS NO<sub>2</sub> surface simulations (red line) and standard deviation (red shaded area) during July (top) and December (bottom).

## Geoponiki in summer



Figure S11. Average diurnal cycle of NO<sub>2</sub> (black line) and the standard deviations (black shaded area) of the urban background stations "AUTH/Malakopi" in Thessaloniki and LOTOS-EUROS NO2 surface simulations (red line) and standard deviation (red shaded area) during July (top) and December (bottom).

## AUTHMALAKOPI in summer



**Figure S12.** Time series of average TROPOMI (blue lines) and LOTOS-EUROS (red lines) NO<sub>2</sub> columns over the 3 distinct domains at the TROPOMI overpasses: the domain over Greece (top), the domain over Athens (middle) and the domain over Thessaloniki (bottom). July is shown in the left column and December in the right. The blue shaded area refers to the precision of the TROPOMI product.