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*Supplement of*

## **Quantification of the depletion of ozone in the plume of Mount Etna**

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# 1 Initial conditions for HSC calculations

Species	low Br/S	high Br/S
H <sub>2</sub> O	8.33-01	8.17e-01
O <sub>2</sub>	9.12e-03	1.05e-02
CO <sub>2</sub>	9.28e-02	9.09e-02
N <sub>2</sub>	3.90e-02	3.90e-02
NO	1.66e-05	2.00e-05
NO <sub>2</sub>	4.31e-08	4.19e-08
NO <sub>3</sub>	2.72e-15	2.80e-15
HNO <sub>3</sub>	7.16e-13	6.02e-13
HONO	1.95e-09	2.00e-09
NH <sub>3</sub>	8.36e-15	2.08e-14
SO <sub>2</sub>	1.60e-02	2.82e-02
H <sub>2</sub> SO <sub>4</sub>	2.95e-07	2.96e-07
SO <sub>3</sub>	2.32e-04	2.99e-05
H <sub>2</sub> S	0.00e+00	0.00e+00
SO	7.40e-10	3.10e-09
O <sub>3</sub>	3.43e-13	4.24e-13
CH <sub>4</sub>	1.03e-30	1.05e-29
CO	8.23e-08	1.82e-07
HCOOH	4.47e-14	9.17e-14
OH	9.81e-06	1.46e-05
HO <sub>2</sub>	2.10e-08	2.65e-08
H <sub>2</sub> O <sub>2</sub>	3.51e-09	4.34e-09
HCl	6.52e-03	1.36e-02
HOCl	1.56e-07	3.21e-07
Cl <sub>2</sub>	3.62e-07	1.29e-06
Cl	5.74e-06	1.51e-05
ClO	1.73e-08	4.05e-08
HBr	7.32e-06	5.08e-05
HOBr	1.75e-10	1.20e-09
Br <sub>2</sub>	8.97e-10	2.85e-08
BrCl	3.85e-08	4.08e-07
Br	2.14e-06	1.57e-05
BrO	2.80e-10	2.02e-09

Table S1: Initial conditions (in mol/mol) from HSC calculations for the subset of species that have been used in the model runs presented in this paper. Note that HOBr is not included in HSC. It has been calculated from HBr using the HOCl/HCl ratio as determined by HSC. H<sub>2</sub>S has been set to 0., as there are strong indications from field measurements (Aiuppa et al., 2007) that equilibrium models and kinetic models do not capture the chemistry of H<sub>2</sub>S. On the times scales considered here, H<sub>2</sub>S is considered inert. “Low Br/S” refers to volcanic volatile measurements taken in June 2012, “high Br/S” refers to the composition used in von Glasow, 2010. For more details see text.

## 2 Plots relating to “High Br/S” initialisation

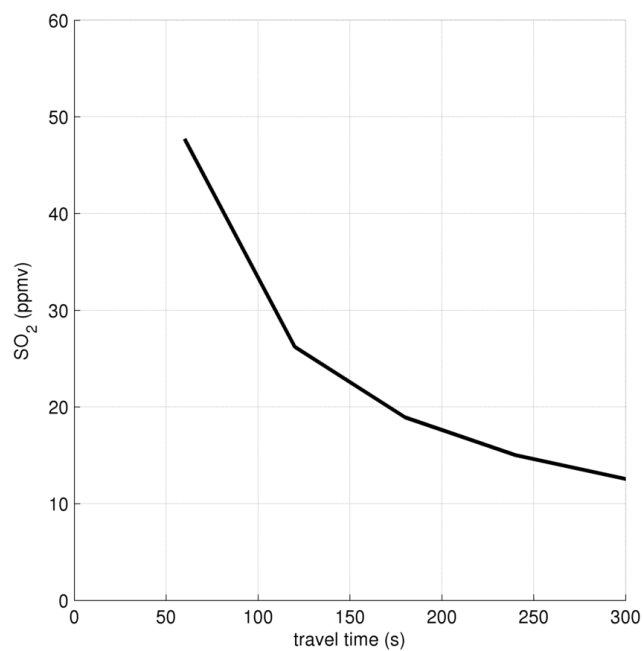


Figure S1: Modelled SO<sub>2</sub> mixing ratio in the plume core (“high Br/S” initialisation).

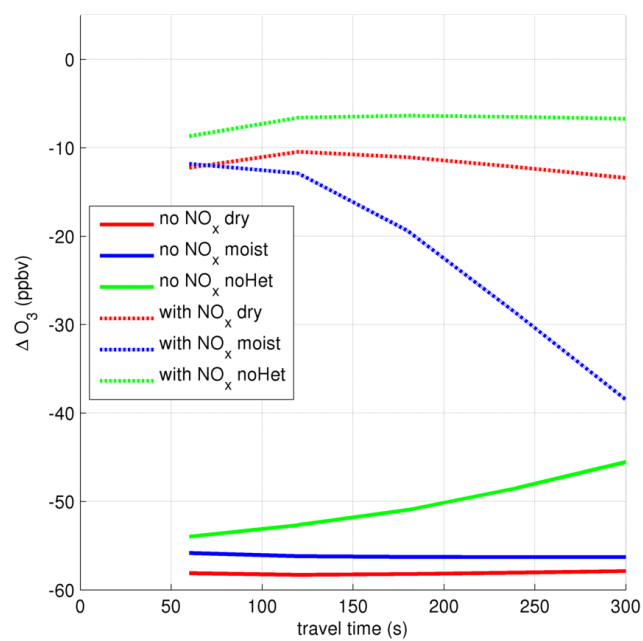


Figure S2: Evolution of ΔO<sub>3</sub> in the core of the plume for the six model scenarios discussed in the text (“high Br/S” initialisation).

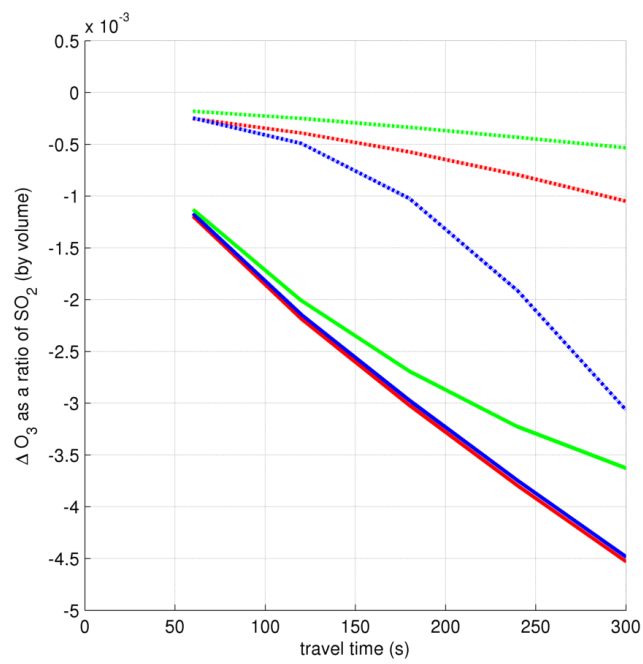


Figure S3: Evolution of  $\Delta O_3/SO_2$  in the core of the plume for the six model scenarios discussed in the text (“high Br/S” initialisation). The colour code is the same as in Figure S2.

### Speciation of bromine within the plume core

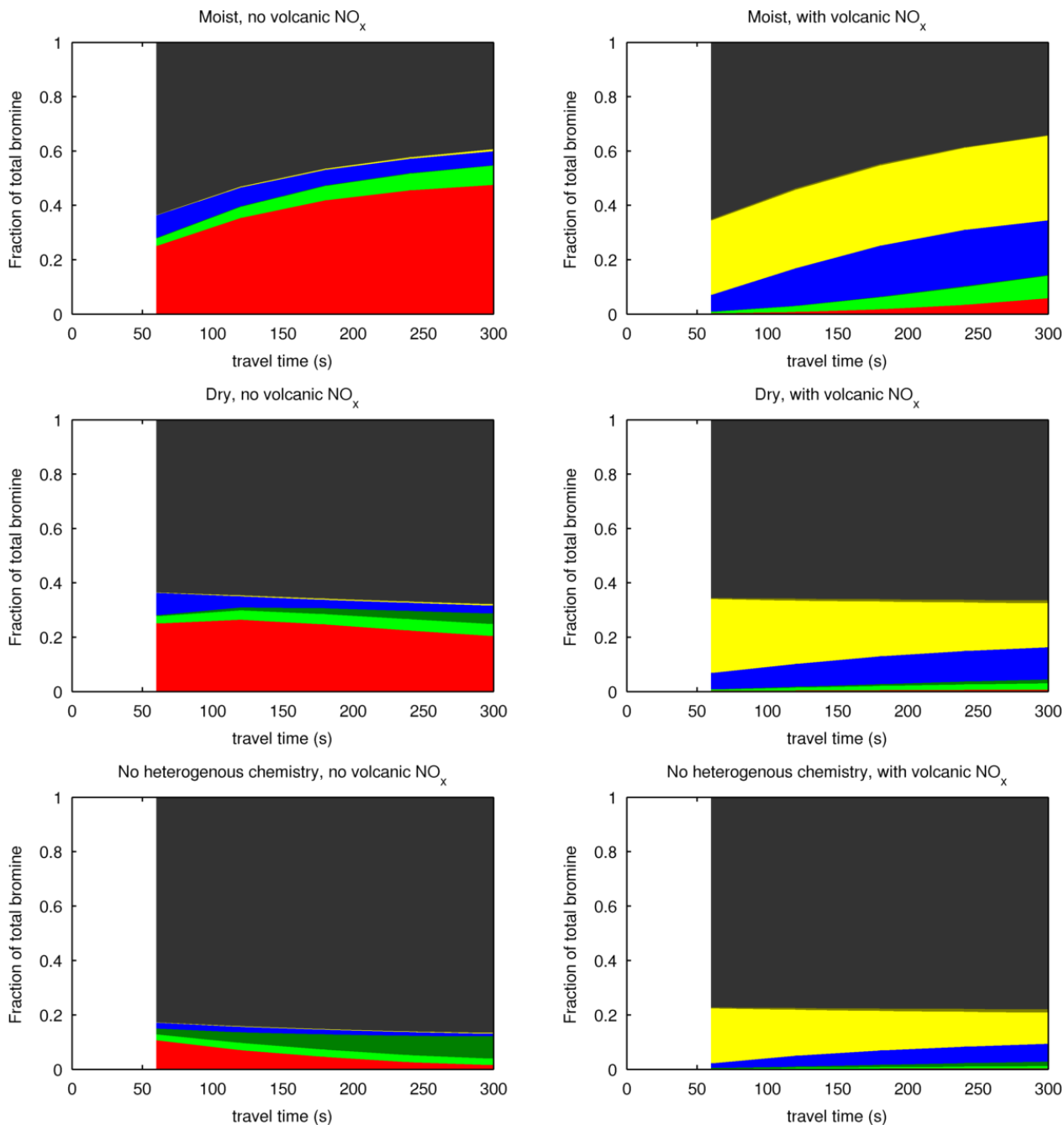


Figure S4: Speciation of bromine for the six model scenarios discussed in the text (“high Br/S” initialisation). Red - Br; Light Green - BrO; Dark Green - HOBr; Blue - Br<sub>2</sub>; Yellow - BrNO<sub>2</sub>; Dark Yellow - BrNO<sub>3</sub>; Grey - HBr.

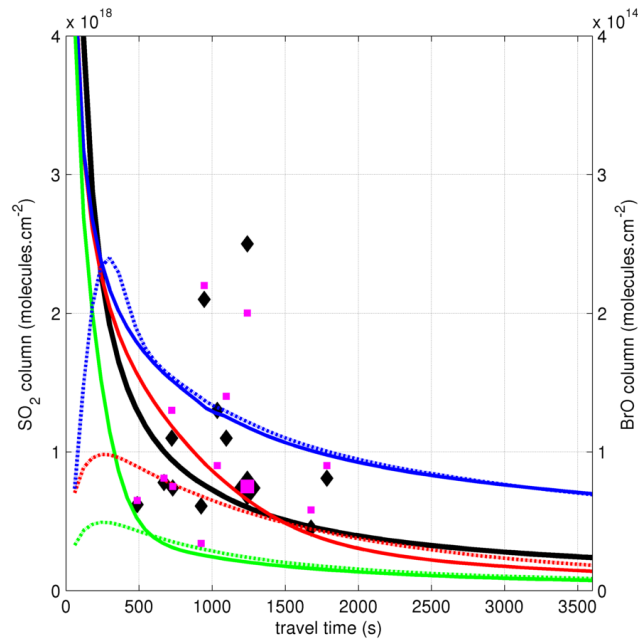


Figure S5: Evolution of vertical column SO<sub>2</sub> and BrO for the six model scenarios discussed in the text (“high Br/S” initialisation). SO<sub>2</sub> is plotted against the left axis scale in black (variation between the scenarios is minimal). The BrO columns are plotted against the right axis scale using the same colour code as in Figure S2. The black diamonds represent the spectroscopic measurements of SO<sub>2</sub>, the magenta squares represent the spectroscopic measurements of BrO. The larger markers represent the 2012-07-27 measurement contemporaneous with near-crater measurements of both O<sub>3</sub> and SO<sub>2</sub>.

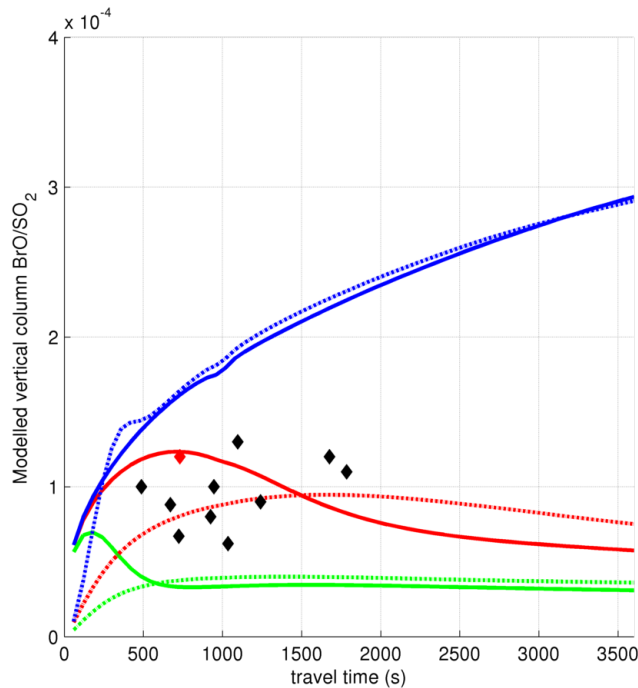


Figure S6: Evolution of vertical column BrO/SO<sub>2</sub> ratio for the six model scenarios discussed in the text (“high Br/S” initialisation). The colour code is the same as in Figure S2. The diamonds represent the spectroscopic measurements, the red diamond represents a measurement contemporaneous with near-crater measurements of both O<sub>3</sub> and SO<sub>2</sub>.