Simulation of the Global Sulfate Cycle with the ECHAM/GRANTOUR model: Coupling oxidant and sulfate chemistry

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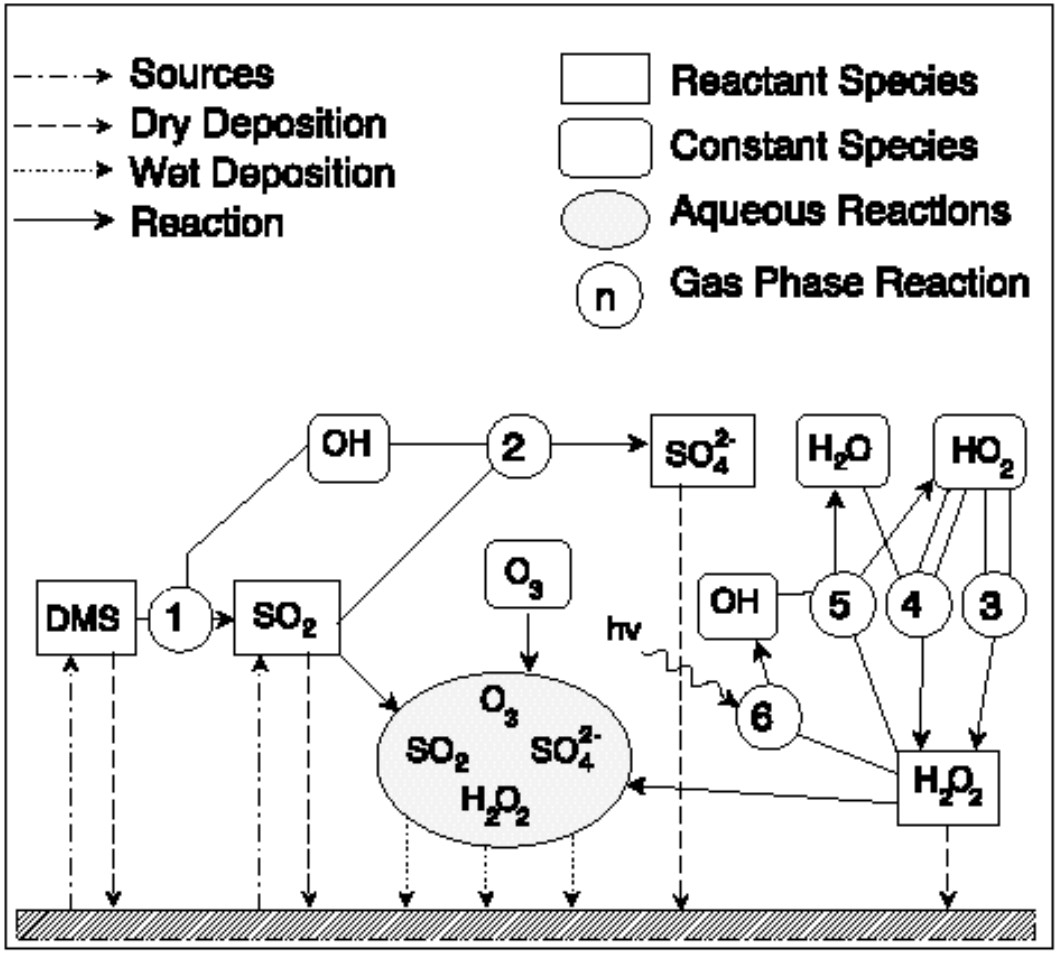
The oxidant and aerosol budgets are coupled through the sulfate system:



Oxidation of H_2O_2 by SO_2 is not very large in the H_2O_2 budget (Tg / mo.):

	January	July	Global Average
Sources:			
HO_2+HO_2	20.5	27.7	24.1
$2*HO_2+H_2O$	12.8	16.7	14.8
Sinks:			
$H_2O_2+h\nu$	11.0	14.0	12.5
H_2O_2+OH	10.6	14.6	12.6
$H_2O_2+SO_2$	1.6	2.5	2.0
Dry deposition	4.0	5.4	4.8
Wet deposition	5.4	7.1	6.2
Total	32.6	43.6	38.1
Atmospheric Burden	1.15	1.5	1.33
Lifetime (days)	1.1	1.1	1.1

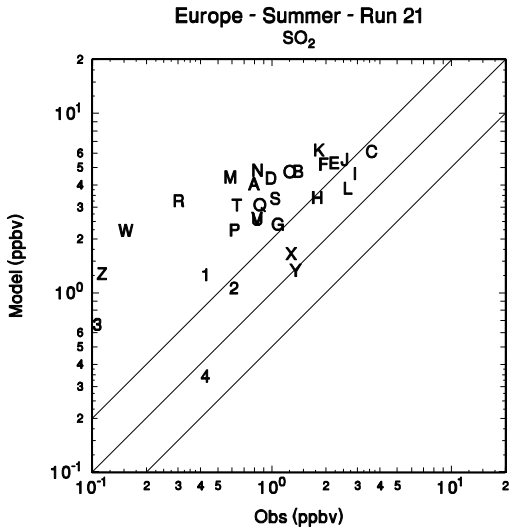
But this reaction is very significant in the SO_2 budget (Tg S/ mo.):

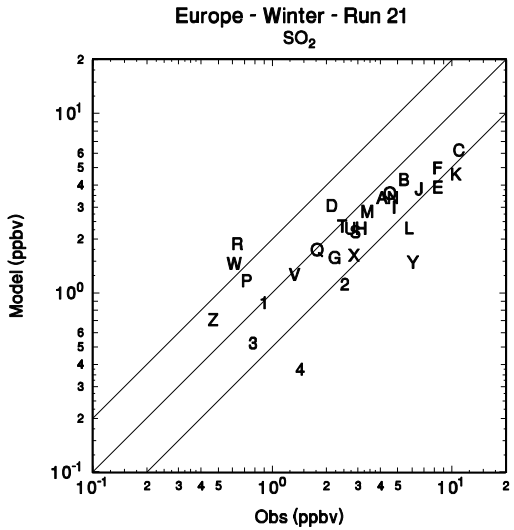
	January	July	Global Average
Sources:			
SO_2 source	6.4	6.4	6.4
DMS+OH	2.3	2.0	2.2
Sinks:			
SO_2+OH	0.9	2.0	1.4
SO_2+O_3	1.1	0.3	0.7
$SO_2+H_2O_2$	1.5	2.4	1.9
Dry deposition	3.7	3.0	3.3
Wet deposition	1.4	0.7	1.1
Total	8.6	8.4	8.4
Atmospheric Burden (TgS)	0.72	0.31	0.33
Lifetime (days)	1.2	1.2	1.2

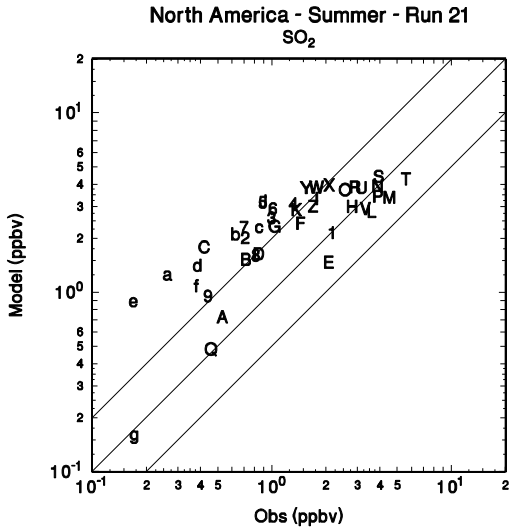
This reaction is also very significant in the H_2SO_4 budget (Tg S/ mo.):

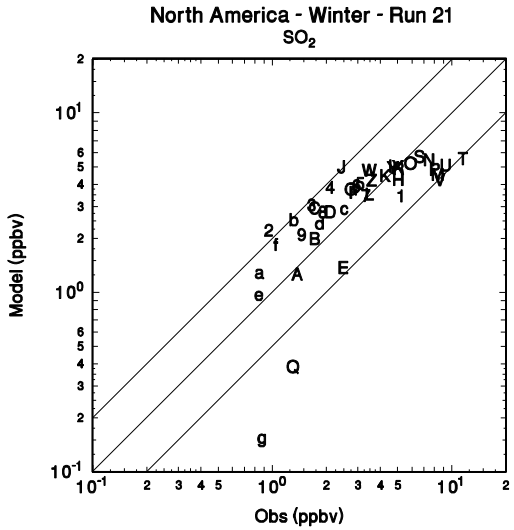
	January	July	Global Average
Sources:			
SO_2+OH	0.9	2.0	1.4
SO_2+O_3	1.1	0.3	0.7
SO_2+H_2O_2	1.5	2.4	1.9
Total	3.5	4.7	4.0
Sinks:			
Dry deposition	0.3	0.1	0.5
Wet deposition	3.2	3.8	3.5
Atmospheric Burden (TgS)	0.31	0.57	0.43
Lifetime (days)	2.8	4.1	3.3

These simulations overestimate SO_2 in NH summer:

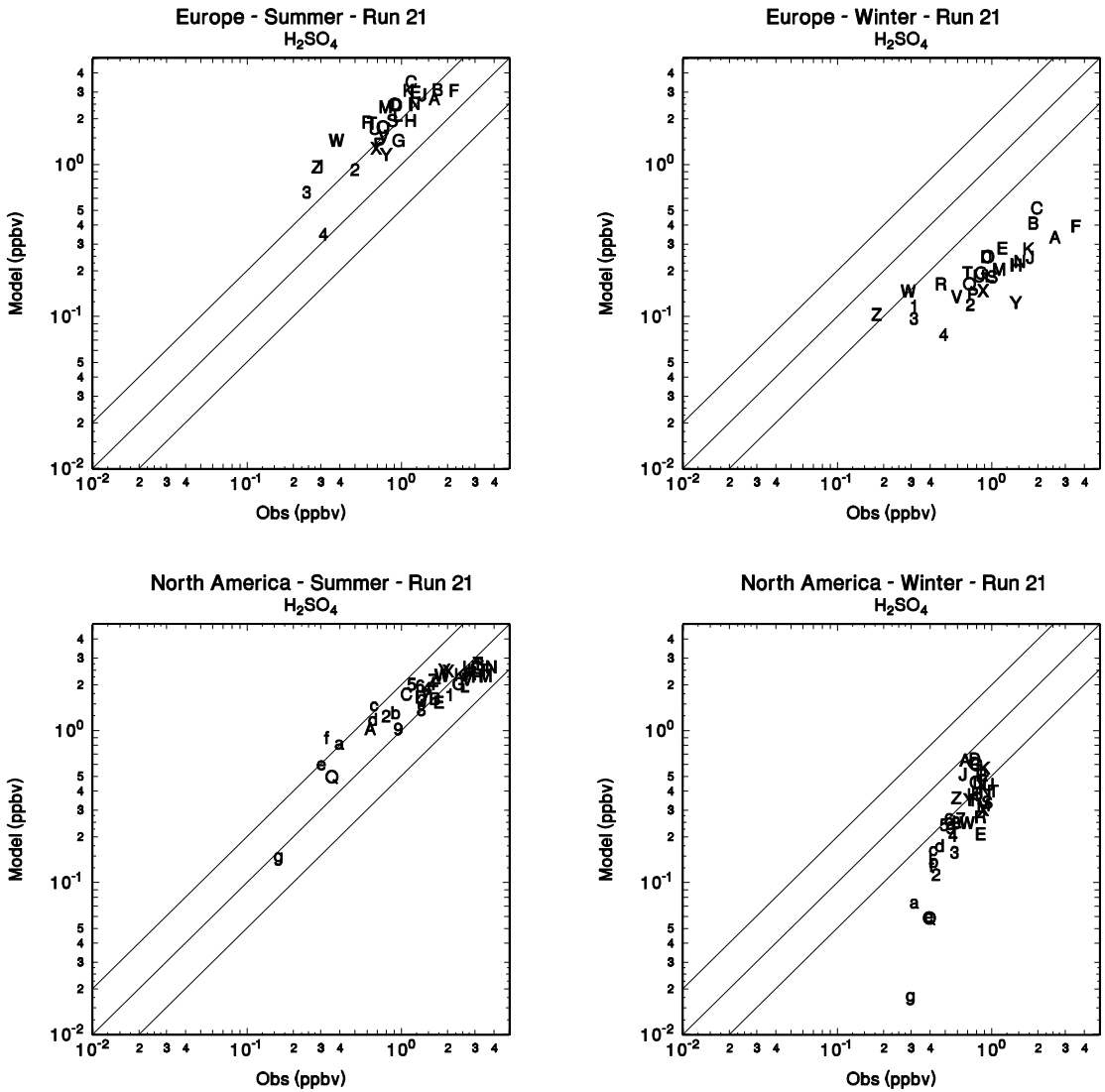








These simulations underestimate H_2SO_4 in NH winter:



Sensitivity studies to evaluate effects of uncertainties in role of H_2O_2+SO_2 for loss of SO_2 and production of H_2SO_4:

	Base: Tg S or Tg H_2O_2	Ph=3.5 (% change from base)	Ph=5.5	Cloud lifetime = 1h	Double HO_2	Reduce Scavenging of SO_2	R(SO_2+ H_2O_2)= 0.	R(SO_2+ H_2O_2)= 0.; R(SO_2+ O_3)=0.	R(SO_2+ O_3)=0.
SO_2									
Jan	0.72	+28.5	-33.7	-11.2	-5.6	22.2	22.9	78.4	29.7
Jul	0.62	8.3	-12.2	-19.1	-9.7	9.1	32.4	86.4	8.8
H_2SO_4									
Jan	0.94	-5.2	+15.0	8.2	4.5	22.1	-13.0	-29.6	-6.2
Jul	1.70	-0.8	0.7	6.5	0.8	16.0	-7.5	-16.7	-0.9
H_2O_2									
Jan	1.15	-0.5	+3.1	-1.3	312.3	-0.6	4.8	4.8	-0.6
Jul	1.53	-0.5	2.9	-1.5	312.0	-0.7	4.8	4.8	-0.5

Conclusions:

- **Uncertainties in oxidation rate of SO_2 to H_2SO_4 cannot explain the overestimation of SO_2 and underestimation of H_2SO_4**

- **Additional pathways for formation of H_2O_2 are unlikely to lead to improvement in the model simulation**

- **The budget for H_2SO_4 and SO_2 are not helpful in quantifying global concentrations of H_2O_2**

- **Improvements in the sulfur budget are likely to require improvements in the representation of clouds and cloud frequency in the parent GCM**