

1 **Table S1.** Prediction skill metrics of PMCAMx-2008 against AMS hourly ground  
 2 measurements during the EUCAARI summer campaign\*.

PM <sub>1</sub>	Mean Observed ( $\mu\text{g m}^{-3}$ )	Mean Predicted ( $\mu\text{g m}^{-3}$ )	MB <sup>#</sup> ( $\mu\text{g m}^{-3}$ )	MAGE ( $\mu\text{g m}^{-3}$ )	FBIAS	FERROR	Percent within a factor of 2
<u>Cabauw</u>							
OA	4.1	4.0	-0.1	1.1	0.005	0.3	89
Sulfate	1.5	2.2	0.7	1.1	0.3	0.5	60
Nitrate	2.5	3.2	0.8	1.7	0.3	0.7	53
Ammonium	1.7	1.9	0.2	0.7	0.1	0.4	76
<u>Finokalia</u>							
OA	2.5	2.1	-0.4	0.7	-0.1	0.3	92
Sulfate	5.2	4.7	-0.5	2.0	-0.005	0.4	83
Nitrate	0.08	0.09	0.02	0.1	-	-	-
Ammonium	1.5	1.3	-0.2	0.5	-0.1	0.4	83
<u>MaceHead</u>							
OA	2.3	2.0	-0.4	0.8	-0.2	0.4	79
Sulfate	1.8	1.7	-0.1	0.7	-0.1	0.4	74
Nitrate	0.8	3.2	2.4	2.6	1.1	1.2	16
Ammonium	1.0	2.5	1.5	1.6	0.8	0.9	23
<u>Melpitz</u>							
OA	5.1	3.7	-1.4	1.6	-0.3	0.3	81
Sulfate	2.3	2.1	-0.2	1.1	-0.1	0.4	65
Nitrate	0.7	0.5	-0.1	0.4	-0.6	0.8	43
Ammonium	1.0	0.9	-0.01	0.4	-0.02	0.4	70
<u>Overall</u>							
OA	3.3	3.0	-0.4	1.0	-0.1	0.3	87
Sulfate	2.8	2.9	0.1	1.3	0.1	0.4	70
Nitrate	1.7	2.8	1.1	1.8	0.4	0.8	41
Ammonium	1.5	1.7	0.3	0.8	0.2	0.5	67

3 \*Number of data points; Cabauw: 640, Finokalia: 490, Mace Head: 330, Melpitz: 160.

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$$\# \quad MB = \frac{1}{n} \sum_{i=1}^n (P_i - O_i)$$

$$MAGE = \frac{1}{n} \sum_{i=1}^n |P_i - O_i|$$

$$FBIAS = \frac{2}{n} \sum_{i=1}^n \frac{(P_i - O_i)}{(P_i + O_i)}$$

$$FERROR = \frac{2}{n} \sum_{i=1}^n \frac{|P_i - O_i|}{(P_i + O_i)}$$

1 **Table S2.** Prediction skill metrics of PMCAMx-2008 against AMS airborne measurements  
 2 during the EUCAARI-LONGREX summer campaign\*.

	Mean Observed	Mean Predicted	MB	MAGE	FBIAS	FERROR	Percent within a factor of 2
PM <sub>1</sub>	( $\mu\text{g m}^{-3}$ )	( $\mu\text{g m}^{-3}$ )	( $\mu\text{g m}^{-3}$ )	( $\mu\text{g m}^{-3}$ )			
OA	2.6	2.2	-0.4	1.1	-0.2	0.6	66
Sulfate	1.6	1.6	-0.1	0.8	0.2	0.6	62
Nitrate	1.6	1.4	-0.2	1.1	-0.3	1.0	38
Ammonium	1.2	1.3	0.01	0.7	-0.08	0.7	53

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4 \* 7010 data points.

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1 **Table S3.** Prediction skill metrics of PMCAMx-2008 against AMS hourly ground  
 2 measurements during the EUCAARI winter campaign\*.

PM <sub>1</sub>	Mean Observed ( $\mu\text{g m}^{-3}$ )	Mean Predicted ( $\mu\text{g m}^{-3}$ )	MB ( $\mu\text{g m}^{-3}$ )	MAGE ( $\mu\text{g m}^{-3}$ )	FBIAS	FERROR	Percent within a factor of 2
<u>Cabauw</u>							
OA	1.4	1.6	0.2	0.8	0.2	0.6	55
Sulfate	1.1	0.8	-0.3	0.8	-0.02	0.8	45
Nitrate	2.7	2.7	-0.04	1.8	0.2	0.8	50
Ammonium	1.2	2.1	0.9	1.4	0.6	0.9	38
<u>Finokalia</u>							
OA	1.4	1.2	-0.2	0.5	-0.2	0.5	72
Sulfate	1.3	1.6	0.3	0.8	0.1	0.6	60
Nitrate	0.05	0.2	0.15	0.2	-	-	-
Ammonium	0.5	0.8	0.3	0.4	0.4	0.7	48
<u>MaceHead</u>							
OA	0.9	0.7	-0.2	0.7	0.6	1.0	30
Sulfate	0.4	0.9	0.5	0.6	0.9	1.1	23
Nitrate	0.6	1.0	0.4	0.5	-0.4	1.3	20
Ammonium	0.3	1.2	0.9	0.9	1.4	1.4	10
<u>Melpitz</u>							
OA	2.2	0.7	-1.5	1.6	-0.9	1.0	29
Sulfate	1.5	0.5	-1.0	1.1	-0.8	1.0	31
Nitrate	3.7	1.7	-2	2.8	-0.5	1.0	36
Ammonium	1.7	0.9	-0.8	1.3	-0.5	1.0	33
<u>Helsinki</u>							
OA	2.6	2.2	-0.4	0.8	-0.05	0.3	87
Sulfate	2.4	1.6	-0.8	0.9	-0.2	0.5	72
Nitrate	1.0	3.2	2.2	2.4	0.9	1.1	20
Ammonium	0.7	2.1	1.4	1.4	1.0	1.0	21

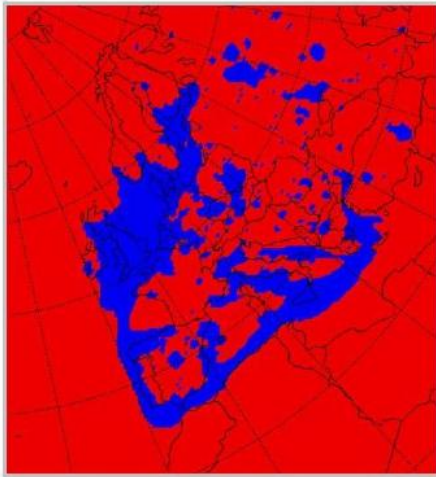
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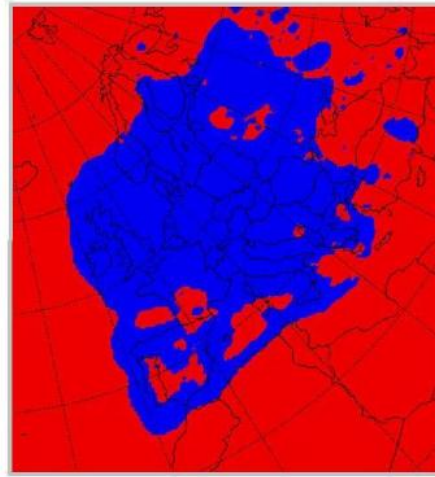
PM <sub>1</sub>	Mean Observed ( $\mu\text{g m}^{-3}$ )	Mean Predicted ( $\mu\text{g m}^{-3}$ )	MB ( $\mu\text{g m}^{-3}$ )	MAGE ( $\mu\text{g m}^{-3}$ )	FBIAS	FERROR	Percent within a factor of 2
<u>Payerne</u>							
OA	4.3	1.0	-3.3	3.3	-1.1	1.1	15
Sulfate	1.1	0.6	-0.5	0.6	-0.5	0.7	53
Nitrate	4.0	1.9	-2.1	2.7	-0.5	0.9	39
Ammonium	1.7	0.9	-0.8	1.1	-0.6	0.9	39
<u>Puy de Dome</u>							
OA	0.6	0.4	-0.2	0.5	0.3	1.5	32
Sulfate	0.7	0.7	0.05	0.5	0.5	1.0	42
Nitrate	0.9	0.02	-0.9	0.9	-1.6	1.8	6
Ammonium	0.8	0.4	-0.4	0.6	-0.2	1.0	35
<u>Vavihill</u>							
OA	4.1	0.9	-3.2	3.2	-1.0	1.1	29
Sulfate	0.2	0.7	0.5	0.6	1.0	1.2	19
Nitrate	0.8	1.4	0.6	1.3	0.3	1.2	21
Ammonium	0.4	1.3	0.9	1.1	1.0	1.3	17
<u>Overall</u>							
OA	2.3	1.1	-1.2	1.5	-0.3	0.9	44
Sulfate	1.0	0.9	-0.1	0.8	0.1	0.9	42
Nitrate	2.1	1.8	-0.3	1.8	-0.2	1.1	30
Ammonium	0.9	1.2	0.3	1.0	0.4	1.0	30

1 \*Number of data points; Cabauw: 460, Finokalia: 570, Mace Head: 580, Melpitz: 450,  
2 Helsinki: 400, Payerne: 540, Puy de Dome: 210, Vavihill: 590.

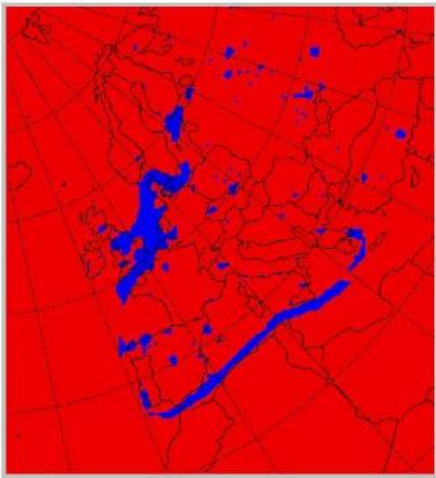
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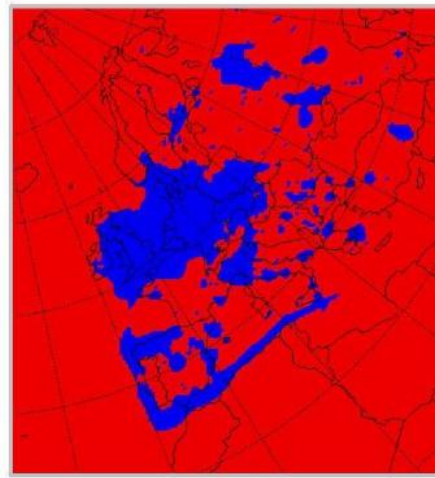
(a) Base Case - Summer



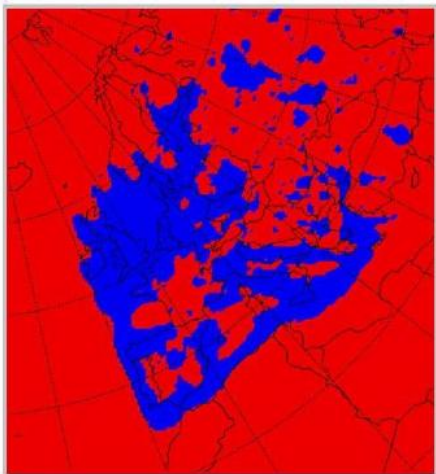
(b) Base Case - Winter



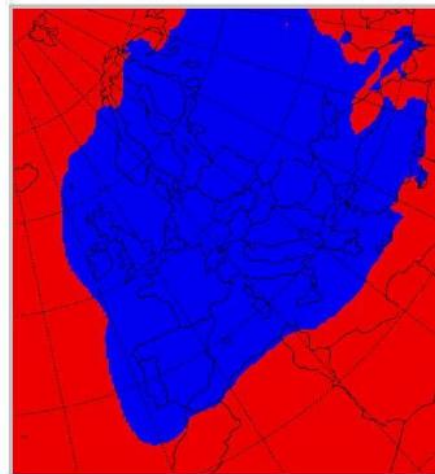
(c) -50% NO<sub>x</sub> - Summer



(d) -50% NO<sub>x</sub> - Winter



(e) -50% VOCs - Summer



(f) -50% VOCs - Winter

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2 **Figure S1.** Predicted base case VOC/NO<sub>x</sub> ground-level concentration ratio (a-b), and

3 predicted VOC/NO<sub>x</sub> ground-level concentration ratio after a 50% reduction of NO<sub>x</sub> (c-d), and

1 anthropogenic VOCs emissions (e-f) during the modeled summer and the modeled winter  
2 period. The red colour indicates ratio values higher than 5.5:1 (NO<sub>x</sub>-limited areas), whereas  
3 the blue colour indicates ratio values lower than 5.5:1 (NO<sub>x</sub>-saturated areas).

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