Supporting information

Biodegradation of phenol and catechol in cloud water: Comparison to chemical oxidation in the atmospheric multiphase system

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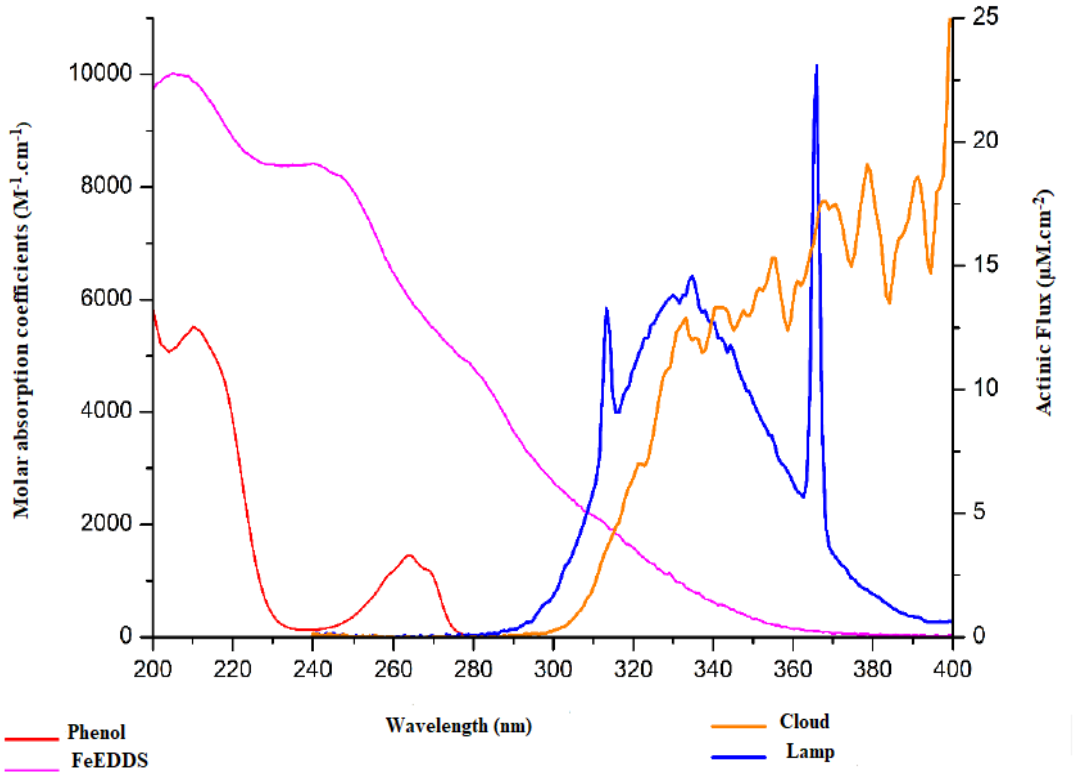

Figure S- 1 : *Comparison of the actinic fluxes of the lamps used and the emission of the solar spectrum measured in-cloud at the puy de Dôme station. The green line represents the actinic flux of the lamp; the brown line corresponds to the actinic flux of the solar emission spectrum. The red line represents the molar absorption coefficient of the Fe-EDDS complex. The blue line represents the molar absorption coefficient of phenol.*

Section S-1 Calculation of the biodegradation rates for the *Pseudomonas* strains

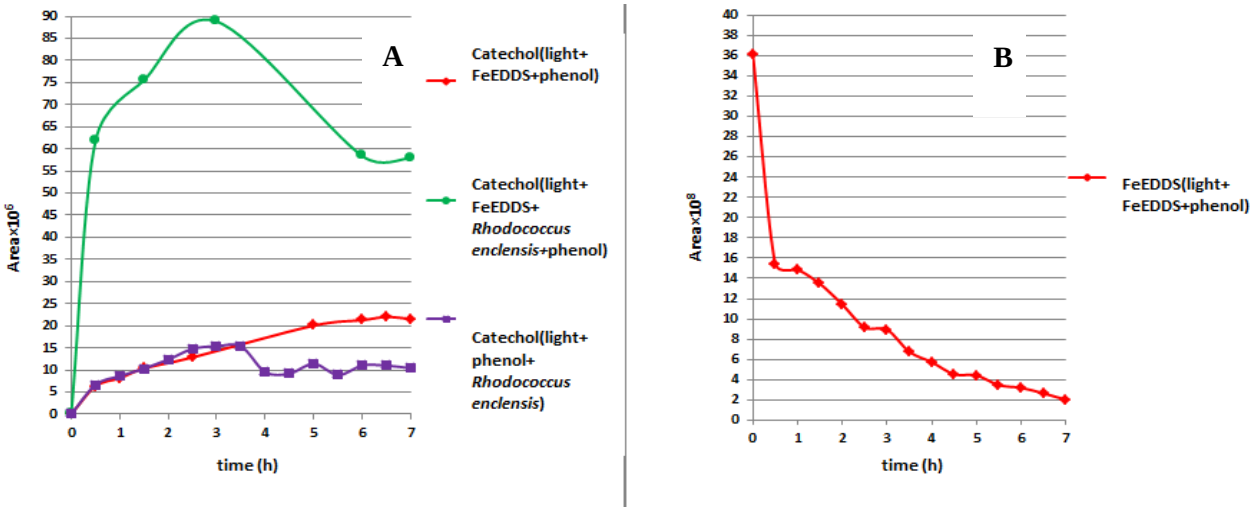

Figure S- 2 : *A) time dependence of the integral of catechol signal (m/z= 110.03678) detected in mass spectra of incubations with Fe(EDDS)+light and Phenol (red), Fe(EDDS)+ light, Phenol and R. enclensis (green), light + Phenol and* R. enclensis *without Fe(EDDS)(violet). B) Time dependence of the integral of Fe(EDDS) signal (m/z = 346.0086) detected in the mass spectrum, recorded during the incubation with Fe(EDDS)+ light and Phenol.*

S-1.1 Pseudomonas putida **EKII**

To calculate the biodegradation rate of phenol and catechol by *Pseudomonas putida* EKII, we used the following data from Hinteregger et al. (1992):

Phenol: Biodegradation of 654 μmol L^{-1} h^{-1}, number of cells: 3.3 ·10^9 cell L^{-1}

Biodegradation rate of phenol: 1.98·10^{-17} mol $cell^{-1}$ h^{-1}

Catechol: Biodegradation rate of catechol is twelve times higher than of phenol (ratio = 2.4μmol min^{-1} mg^{-1} / 0.2 μmol min^{-1} mg^{-1}, expressed per mg of cells)

Biodegradation of catechol: 1.98·10^{-17} ·12 = 23.78 ·10^{-17} mol $cell^{-1}$ h^{-1}

S-1.2 Pseudomonas aeruginosa

To calculate the biodegradation rate of phenol and catechol by *Pseusomonas aeriginosa,* we used the following data from Razika et al. (2010):

Phenol: Biodegradation of 10 mg L^{-1} during 96 hours, number concentration of cells: 4.7. 10^9 cell L^{-1}

Biodegradation rate of phenol: 23.49·10^{-17} mol $cell^{-1}$ h^{-1}

Biodegradation of 50 mg L^{-1} during 120 hours, number concentration of cells: 4.7·10^9 cell L^{-1}

Biodegradation rate of phenol: 94.31·10^{-17} mol $cell^{-1}$ h^{-1}

Biodegradation rate of phenol (average value) taken into account: 58.9·10^{-17} mol $cell^{-1}$ h^{-1}

Catechol: No information is available in Razika et al (2010), so we multiplied the biodegradation rates of phenol with a factor of twelve as it is within the same order of magnitude of what we found in our study (Factor ~ 10)

Biodegradation rate of catechol (average value): 58.9·10^{-17} mol $cell^{-1}$ h^{-1}·12= 706.8 10^{-17} mol $cell^{-1}$.

Section S-2: Calculation of photolysis rate j(Fe(EDDS)) and resulting OH concentration in the experiments

$$j = \int_{250}^{400} I_{0,\lambda} \cdot \varepsilon_\lambda \cdot \phi_\lambda \cdot d\lambda \; \frac{photons}{cm^2\ s\ nm} \frac{cm^3}{molec\ cm} \frac{nm}{} \qquad [s^{-1}]$$

$I_{0,\lambda}$ = spectral actinic flux [photons cm^{-2} s^{-1} nm^{-1}]

ε_λ = extinction coefficient [cm^3 $molec^{-1}$ cm^{-1}]

ϕ_λ = Quantum yield [dimension less]

Experimental data

Irradiance E(λ) [μW cm^{-2}] ; convert into SI units E'[W/m^2] = E·10^{-6} W/μW ·10^4 cm^2/m^2 = E·0.01

Convert irradiance E(λ) [μW cm^{-2}] to actinic flux I [photons cm^{-2} s^{-1}]:

Actinic flux $I' = \frac{E\prime\,\lambda}{h \cdot c} \left[\frac{W\ m}{m^2} \frac{s}{J\ s\ m}\right] = \frac{E\ \lambda}{h \cdot c} \left[\frac{kg\ m^2}{s^3 m^2} \frac{m}{} \frac{s^2}{kg\ m^2 s} \frac{s}{m}\right] = \frac{photons}{m^2\ s}$

Spectral actinic flux $I_\lambda = \frac{I'}{\lambda} \cdot\ 10^{-4} = \frac{E(\lambda)}{h \cdot c} \left[\frac{W}{m^2} \frac{s}{J\ s\ m}\right] = \frac{E\ \lambda}{h \cdot c} \left[\frac{kg\ m^2}{s^3 m^2} \frac{m}{} \frac{s^2}{kg\ m^2 s} \frac{s}{m}\right] = \frac{photons}{cm^2\ s\ nm}$

h = 6.62606×10^{-34} J s

c = 3·10^8 m/s

ε' molar absorption coefficient (L mol^{-1} cm^{-1}) = extinction coefficient

$\varepsilon_\lambda =$ ε' $\frac{L}{mol\ cm} \cdot \frac{1000\ cm^3}{L} \cdot \frac{mol}{6.022e23\ molec} = \varepsilon' \cdot 1000/N_A$ [cm^3 $molec^{-1}$ cm^{-1}]

Quantum yield: ϕ_λ = 0.025 (at 290 < λ < 400 nm)

$$j = \int_{250}^{400} I_\lambda \cdot \varepsilon_\lambda \cdot \phi_\lambda \cdot d\lambda \; \frac{photons}{cm^2\ s\ nm} \frac{cm^3}{molec\ cm} \frac{nm}{} = \mathbf{0.001388\ s^{-1}}$$

Calculation of steady-state References: •OH(aq) concentration

<u>OH formation:</u>

[Fe-EDDS] + hν → •OH + products j = 1.388e-3 s^{-1} $[Fe(EDDS)]_0$ = 0.5 mM

$$-\frac{\mathrm{d[Fe(EDDS)]}}{\mathrm{dt}} = \frac{\mathrm{d[OH]}}{\mathrm{dt}} = \mathrm{j\ [Fe(EDDS)]}$$

<u>•OH loss</u>

•OH + Phenol → Products k = 8.41e9 M^{-1} s^{-1} $[Phenol]_0$ = 0.1 mM

$$\frac{\mathrm{d[OH]}}{\mathrm{dt}} = -\ \mathrm{k\ [OH][Phenol]}$$

→ Steady-state OH concentration at the beginning of experiment

k [•OH] [Phenol] = j [Fe(EDDS)]

$$[\mathrm{OH}] = \frac{\mathrm{j\ [Fe(EDDS)]}}{\mathrm{k\ [Phenol]}} = \frac{1.388\mathrm{e}-3\ \mathrm{s^{-1}}\ 5\mathrm{e}-4\ \mathrm{M}}{8.41\mathrm{e}9\ \mathrm{M^{-1}s^{-1}}\ \ 1\mathrm{e}-4\ \mathrm{M}} = 8.3\mathrm{e}-13\ \mathrm{M}$$

Section S-3: Input data to the multiphase box model

S-3.1: Multiphase processes

Table S-1: *Chemical and microbial processes in the multiphase model*

Gas Phase		
	Chemical rate constant [cm^3 s^{-1}]	**Reference**
$^{\bullet}OH$ + Phenol → 0.5 Catechol + 0.5 Prod [a)]	$2.81 \cdot 10^{-11}$	(Berndt and Böge, 2001)
$NO_3^{\bullet}$ + Phenol → Products	$5.8 \cdot 10^{-12}$	(Bolzacchini et al., 2001)
$^{\bullet}OH$ + Catechol → Products	$1.1 \cdot 10^{-10}$	(Olariu et al., 2000)
$NO_3^{\bullet}$ + Catechol → Products	$9.8 \cdot 10^{-11}$	(Olariu et al., 2004)
Aqueous phase		
	Chemical rate constant [M^{-1} s^{-1}]	
$^{\bullet}OH$ + Phenol → 0.5 Catechol + 0.5 Prod [b)]	$8.41 \cdot 10^{9}$	(Raghavan and Steenken, 1980)
$NO_3^{\bullet}$ + Phenol → Products	$1.9 \cdot 10^{9}$	(Umschlag et al., 2002)
$^{\bullet}OH$ + Catechol → Products	$4.7 \cdot 10^{9}$	(Hoffmann et al., 2018)
$NO_3^{\bullet}$ + Catechol → Products	$1.9 \cdot 10^{9}$	(Hoffmann et al., 2018)
$^{\bullet}OH$ + WSOC → Products	$2 \cdot 10^{6}$ s^{-1}	Based on (Arakaki et al., 2013), assuming [WSOC] = 5 mM
NO_3 → Products	10^{5} s^{-1}	Based on (Exner et al., 1992; Zellner and Herrmann, 1994); assuming 1 mM Cl^-, 0.01 mM Br^-
The following three reactions are only considered in sensitivity simulations, Figure S-4		
O_3 + Phenol → Products	1300	(Hoigné and Bader, 1983)
O_3 + Catechol → Products	$3.1 \cdot 10^{5}$	(Gurol and Nekouinaini, 1984)
$HO_2^{\bullet}/O_2^{\bullet-}$ + Catechol → Products	$7.8 \cdot 10^{4}$	Rate constant for HO_2/O_2^- ratio at pH = 4 ($pK_a(HO_2^{\bullet})$ = 4.8) calculated based on k_{HO2}, k_{O2}- by (Bielski et al., 1985)

	Microbial rate constant [L $cell^{-1}$ s^{-1}]	
Rhodococcus + Phenol → Catechol	$1.8 \cdot 10^{-13}$	[d)]
Rhodococcus + Catechol → Products	$1.5 \cdot 10^{-12}$	[d)]
Pseudomonas + Phenol → Catechol	$1 \cdot 10^{-13}$	[d)]
Pseudomonas + Catechol → Prod	$1.2 \cdot 10^{-12}$	[d)]

Phase transfer processes				
	K_H [M atm^{-1}]	**Reference**	**α [c)]**	**D_g [cm^2 s^{-1}][c)]**
$^{\bullet}OH(aq) \leftrightarrow {}^{\bullet}OH(gas)$	25	(Kläning et al., 1985)	0.05	0.15
$NO_3^{\bullet}(aq) \leftrightarrow NO_3^{\bullet}(gas)$	0.6	(Rudich et al., 1996)	0.1	0.1
Phenol(aq) ↔ Phenol(gas)	647	(Feigenbrugel et al., 2004)	0.027	0.09
Catechol(aq) ↔ Catechol(gas)	$8.31 \cdot 10^{5}$	(Sander, 2015)	0.1	0.08

[a)] Catechol yield likely represents an upper estimate for the total of all dihydroxybenzene compounds [b)] Initial formation of the phenoxy radical and the subsequent reaction with O_2 are lumped here, leading to 0.5 catechol into one step since the second reaction is diffusion controlled; [c)] These values were taken from CAPRAM (Ervens et al., 2003; Hoffmann et al., 2018) [d)] See calculation of values in Section S-3.2

S-3.2 Calculation of microbial rate constants from experimentally derived rates

Experimentally-derived rates R of microbial activity towards phenol and catechol are summarized in Table 2 of the main part of the manuscript, together with the bacteria type (*Rhodococcus*, *Pseudomonas putida*, *Pseudomonas aeruginosa*) and aqueous phase concentrations of substrate (phenol, catechol) and bacteria cells. Strictly, the measured rates might be only valid for the same substrate-to-cell ratio as the substrate availability determines the cell activity. Since these concentrations differ greatly, we derive the first-order rate constant k' [h^{-1}]

$$k' = R\,[Cell] / [Substrate] \quad \text{(S-1)}$$

Ambient cell concentrations in cloud water are on the order of $10^6 – 10^8$ cell L^{-1}. We assume a total cell concentration of $6.8 \cdot 10^7$ cell L^{-1} of which 3.6% are *Rhodococcus* ($C_{Rh,cloud} = 2.7 \cdot 10^6$ cell L^{-1}) and 19.5% *Pseudomonas* ($C_{Ps,cloud} = 1.3 \cdot 10^7$ cell L^{-1}). Phenol concentrations in cloud water are in the range of 5.5 - 7.7 nM (Lebedev et al., 2018). Using the lower value of this range yields phenol-to-cell ratios in cloud water of $2 \cdot 10^{-15}$ mol $cell^{-1}$ and $4.2 \cdot 10^{-16}$ mol $cell^{-1}$ for *Rhodococcus* and *Pseudomonas*, respectively, which is within two orders of magnitude of the ratios as used in the experiments. Corresponding cloud water measurements for catechol are not available.

In the multiphase model, we describe the microbial processes analogous to chemical reactions, i.e. with a formal second-order rate constant in units of L $cell^{-1}$ s^{-1} using the constant cell concentrations in the aqueous phase.

$$k_{2nd}\,[L\ cell^{-1}\ s^{-1}] = k' \ / \ [Cell]_{cloud} \ / 3600\ s\ h^{-1} \quad \text{(S-2)}$$

The resulting k_{2nd} are then used in the model studies for the assumed (constant) cell concentrations in cloud water

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Table S-2: *Summary of literature data on microbial activity towards phenol and catechol by Rhodococcus and Pseudomonas. For the estimates of unknown rates, refer to Section 3.2 (Comparison to literature data) in the main part of the manuscript*

Substrate	**Bacteria type**	**R** / **mol cell^{-1} h^{-1}**	**[Substrate]** / **M**	**[Cell]$_{experiment}$** / **(cell L^{-1})**	**Ref**	**[Substrate]/ [Cell]** / **mol cell^{-1}**	**k'** / **h^{-1}**	**[Cell]$_{cloud}$**	**k$_{2nd}$** / **L cell^{-1} s^{-1}**
Phenol	*Rhodococcus*	$1.76\cdot10^{-16}$	10^{-4}	10^{9}	a	10^{-13}	$1.76\cdot10^{-3}$	$2.7\cdot10^{6}$	$1.8\cdot10^{-13}$
Catechol	*Rhodococcus*	$1.5\cdot10^{-15}$	10^{-4}	10^{9}	b	10^{-13}	$1.5\cdot10^{-2}$	$2.7\cdot10^{6}$	$1.5\cdot10^{-12}$
Phenol	*Pseudomonas putida*	$1.99\cdot10^{-17}$	$6.54\cdot10^{-4}$	$3.3\cdot10^{9}$	c	$2\cdot10^{-13}$	$1\cdot10^{-4}$		
Catechol	*Pseudomonas putida*	$2.39\cdot10^{-16}$			c		$2.4\cdot10^{-3}$		
Phenol	*Pseudomonas*	$2.35\cdot10^{-16}$	$1.06\cdot10^{-4}$	$4.7\cdot10^{9}$	d	$2.3\cdot10^{-14}$	$1\cdot10^{-2}$		
	aeruginosa	$9.43\cdot10^{-16}$	$5.31\cdot10^{-4}$	$4.7\cdot10^{9}$		$1.1\cdot10^{-13}$	$8.3\cdot10^{-3}$		
Catechol	*Pseudomonas aeruginosa*				e		0.11		
Phenol	*Pseudomonas (Average)*						$5\cdot10^{-3}$	$1.3\cdot10^{7}$	$1\cdot10^{-13}$
Catechol	*Pseudomonas (Average)*							$1.3\cdot10^{7}$	$1.2\cdot10^{-12}$

[a)] (Lallement et al., 2018), [b)] This study, [c)] (Hinteregger et al., 1992) [d)] (Razika et al., 2010), [e)] Scaled up from data for phenol by reference [d)] using the same ratio of activities to phenol and catechol (12) as for the average value for *Pseudomonas putida*

Section S-4: Model sensitivity study including the aqueous phase reactions of phenol with ozone and of catechol with ozone and $HO_2^\bullet/O_2^{\bullet-}$

In a recent model study by (Hoffmann et al., 2018), it was suggested that catechol (and other dihydroxybenzenes) are efficiently oxidized not only by $^\bullet$OH but also by ozone and the hydroperoxy ($HO_2^\bullet/O_2^{\bullet-}$) radical. Also the reaction of phenol with ozone was included in this model study. In that latter model study, a rate constant of k(O_3 + Catechol) = $5.2 \cdot 10^5$ M^{-1} s^{-1} was estimated. This rate constant is similar to an experimentally-derived value of k(O_3 + Catechol) = $3.1 \cdot 10^5$ M^{-1} s^{-1} (Gurol and Nekouinaini, 1984). This latter study was performed at very acidic conditions (pH = 1.5) and a strong pH dependence of the rate constant was pointed out leading to a decreasing rate constant with increasing pH and resulting in the predominance of the $^\bullet$OH reaction at atmospherically-relevant pH values (~5).

Since the exact pH dependence is not available, we show in the following model results from a sensitivity studies including the $HO_2^\bullet$ and O_3 reactions in order to provide an upper estimate of their role in the multiphase system. Initial concentrations of 0.1 ppt $HO_2^\bullet$ and 40 ppb ozone in the gas phase are assumed and held constant throughout the simulation. In agreement with the model results by Hoffmann et al. (2018), we find large contributions of the ozone reactions in the aqueous phase to the total loss. The relative contributions of the ozone (57 – 68%) and $HO_2^\bullet/O_2^{\bullet-}$ (16 – 19%) reactions with catechol predicted here are also similar as predicted in the previous model study.

S-4.2 Model results

All model results [relative contribution to total loss [%]) are summarized in Table S-3. The upper part of the table contains results for the base simulations as shown in Figure 4 (microbial aqueous phase processes and $^\bullet$OH and $NO_3^\bullet$ reactions in gas and aqueous phases); the bottom part of the table includes results for the sensitivity simulations that also include $HO_2^\bullet$(aq) and O_3(aq) reactions (Figure S-3).

Table S-3: *Model results of base case and sensitivity simulations: Relative contributions to total loss of phenol and catechol, respectively*

		$^\bullet$OH(g)	$NO_3^\bullet$(g)	$^\bullet$OH(aq)	$NO_3^\bullet$(aq)	O_3(aq)	$HO_2^\bullet$(aq)	*Rhodo-coccus*	*Pseudo-monas*
Base case									
Day	Phenol	99.8	0	0.22	0	-	-	0.01	0.006
	Catechol	69.3	0	14	0	-	-	9.3	7.4
Night	Phenol	0	99.8	0	0.18	-	-	0.0005	0.00027
	Catechol	0	97.5	0	2.2	-	-	0.18	0.14
Sensitivity simulation including aqueous phase reactions of O_3 (phenol, catechol) and HO_2/O_2^- (catechol)									
Day	Phenol	99.7	0	0.22	0	0.02	0	0.01	0.0056
	Catechol	6.1	0	1.2	0	58.9	33	0.81	0.65
Night	Phenol	0	99.8	0	0.18	0.0007	0	0.0005	0.00027
	Catechol	0	86.4	0	2	11.4	0	0.16	0.13

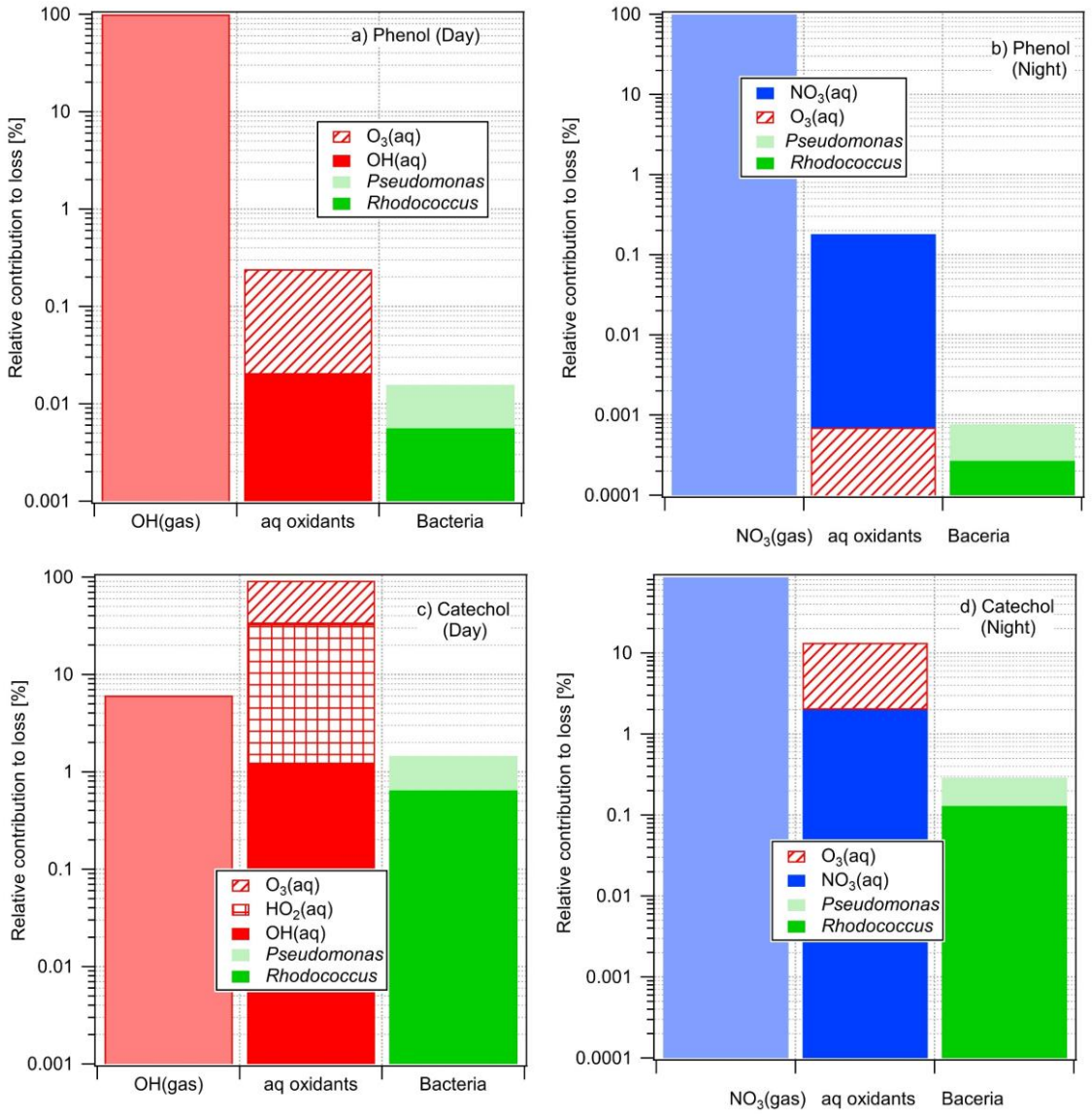

***Figure S- 3** : Relative contributions to total loss of phenol (a, b) and catechol (c, d) in the multiphase system including $HO_2^{\bullet}$ and O_3 reactions in the aqueous phase (Table S-3).*

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