Formation and composition of organic aerosols from the uptake of glyoxal on natural mineral dust aerosols: a laboratory study

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Abstract

The uptake of glyoxal on realistic submicron mineral dust aerosol particles from a natural soil (Gobi Desert) is investigated during experiments in a large simulation chamber, under variable experimental conditions of relative humidity (RH), irradiation, and ozone concentrations. The uptake of glyoxal on the dust particles starts as soon as the glyoxal is injected into the chamber. At 80% RH, the measured uptake coefficient of glyoxal on mineral dust is $\gamma = (9 \pm 5) \times 10^{-3}$. The totality of the mass of reacting glyoxal is transformed into organic matter on the surface of the dust particles. The uptake of glyoxal is accompanied by the appearance of marker peaks in the organic mass spectra and a persistent growth in the volume concentration of the dust particles. While the mass of the organic matter on the dust rapidly reverts to values prior to uptake, the organic composition of the dust is modified irreversibly. Glycolic and other organic acids, but also oligomers, are detected on the dust. At 80% RH, compounds ranging from C_4 to C_{10} are observed as oligomerization products of glyoxal mono- and di-hydrate forms. The study suggests that dust aerosols could play a very substantial role in the formation of organic aerosols at high RH, but also that the

42 reaction could have potentially important implications for the dust optical and
43 hygroscopic properties, including their pH.

1. Introduction

45 Mineral dust originates naturally from the wind erosion of arid or semi-arid soils,
46 resulting in the suspension of particles with diameters from fractions to hundreds of
47 microns, which can be transported over thousands of kilometres whilst in the
48 atmosphere (Adebiyi et al., 2023; Mahowald et al., 2014). The total global mass of
49 mineral dust particles emitted annually in the atmosphere is of the order of 4600 Tg
50 yr^{-1}, accounting for approximately 40% of the total annual aerosol emissions (Knippertz
51 and Stuut, 2014; Kok et al., 2021). Major natural source areas of mineral dust are North
52 Africa (~50% of the global annual dust emissions), Asia (~40%), North America, and
53 the Southern Hemisphere (~10%; Kok et al., 2023). Anthropogenic emissions are
54 associated with soil erosion for agriculture, pasture, and deforestation (Tegen and
55 Fung, 1995; Webb and Pierre, 2018), but their contribution to the total annual dust
56 mass loading is uncertain, ranging from 5 to 60% (Chen et al., 2023). Mineral dust
57 significantly impacts the Earth's energy balance by absorbing and scattering radiation
58 in the solar and terrestrial spectra (Di Biagio et al., 2019; Kok et al., 2023) and by
59 influencing the lifetime and optical properties of mixed-phase and ice clouds (e.g.,
60 Atkinson et al., 2013; Harrison et al., 2001; Steinke et al., 2016). Current estimates of
61 the effective radiative forcing (sum of direct and indirect) of natural mineral dust are in
62 the range of -0.07 ± 0.18 W m^{-2} (Kok et al., 2023), owing to large uncertainties in the
63 atmospheric mass loading and properties of dust at emission and during transport
64 (Castellanos et al., 2024; Li et al., 2021).

65 Gas-particle interactions along the dust lifecycle contribute to these uncertainties.
66 Numerous laboratory and field studies show that mineral dust is capable to adsorb
67 various reactive gaseous compounds, which may modify its chemical composition, and
68 in turn to alter optical properties, hygroscopicity and ice nucleation activity but also may
69 affect the oxidative capacity of the atmosphere (Bauer et al., 2007; Chirizzi, 2017;
70 Crowley et al., 2010; Joshi et al., 2017; Liu et al., 2013; Ooki and Uematsu, 2005;
71 Romanias et al., 2012; Seisel et al., 2004; Tang et al., 2017; Turpin and Huntzicker,
72 1995; Usher et al., 2003; Wagner et al., 2008). Dust aerosol may promote
73 photocatalytic reactions of inorganic gases such as SO_2 and NO_2, initiating nucleation
74 events (Dupart et al., 2012; Nie et al., 2014).

The uptake of volatile organic compounds (VOCs) on mineral dust, such as limonene, toluene (Romanías et al., 2016), isoprene (Zeineddine et al., 2017), phenol (Hettiarachchi and Grassian, 2024), and dicarboxylic acids (Ponczek et al., 2019), is also documented. These reactions may alter the VOC budget in the atmosphere and lead to the formation of secondary organic aerosols (SOA) (Li et al., 2019; Tang et al., 2017; Usher et al., 2003; Xu et al., 2023; Zeineddine et al., 2023), one of the key players of atmospheric chemistry (Shrivastava et al., 2017).

Glyoxal (CHOCHO) is one of the most abundant VOCs in the troposphere (Lewis et al., 2020). It is produced through the oxidation of aromatic compounds like benzene, toluene, and p-xylene (Volkamer et al., 2001) as well as by the photochemical oxidation of isoprene (Chan et al., 2017). The global atmospheric concentrations of glyoxal have been evaluated in the range of 10 – 100 pptv by Fu et al. (2008). However, case studies show sometimes higher concentrations. During a field study in Shanghai in the summer of 2018, Guo et al. (2021) reported an average glyoxal concentration of 164 ± 73 pptv, due to daytime photochemistry. Local concentrations of up to 400 pptv have been documented in regions influenced by aromatic pollution (Li et al., 2022). Satellite measurements of glyoxal show that the highest concentrations in tropical and sub-tropical regions are found during warm, dry periods influenced by biogenic emissions and vegetation fires, but also anthropogenic pollution (Vrekoussis et al., 2009). Elevated glyoxal concentrations have been observed in aged biomass burning plumes and tropical ocean regions, revealing model under-predictions in high-emission areas due to missing complex organic compound sources (Kluge et al., 2023). Field measurements in the northeast Atlantic Ocean reveal that models generally underestimate glyoxal concentrations due to missing contributions from acetaldehyde and other chemical precursors, and a potential glyoxal source from the ocean surface organic microlayer, particularly significant at night (Walker et al., 2022).

Glyoxal is a very soluble molecule which readily oligomerises in water, leading to the formation of larger molecules (Kalberer et al., 2004; Shapiro et al., 2009). Several previous studies have revealed that glyoxal canhas the ability to be uptaken up onto aerosol particles, potentially serving as a significant source of organic aerosols (e.g., Liggio et al., 2005b,; Carlton et al., 2007; Ervens and Volkamer, 2010; Galloway et al., 2009; Knote et al., 2014). The uptake of glyoxal on ammonium sulphate particles can lead to the formation of carbon-nitrogen compounds (such as imidazole derivatives),

108 oligomers, and organic acids (Galloway et al., 2009), that has been observed to cause
109 browning (De Haan et al., 2020). The light-absorbing imidazole derivatives formed by
110 glyoxal have been found to act as a photosensitizer, initiating radical chemistry under
111 realistic irradiation conditions in the aerosol phase and initiating aerosol growth in the
112 presence of limonene (Rossignol et al., 2014).

113 The study by Shen et al. (2016) revealed that glyoxal can also uptake onto synthetic
114 minerals proxies of natural mineral dust, forming oligomers, organo-sulphates, formic
115 acid, and glycolic acid, henceforth suggesting a potential significant mechanism for
116 organic aerosol formation and modification of the optical and hygroscopic properties of
117 mineral dust. Recently, Zogka et al. (2024) highlighted the dependence of the soil
118 composition and size for the uptake of glyoxal on soils and soil surrogates (synthetic
119 mineral).

120 Following up from those studies, in this paper we present novel laboratory experiments
121 using a large-scale simulation chamber to investigate the formation of organic aerosol
122 from the uptake of glyoxal on realistic airborne mineral dust particles, in
123 atmospherically-relevant conditions. Dust aerosols are generated from a natural parent
124 soil from the Gobi Desert, one of the most important sources of tropospheric dust and
125 representative of an area where this interaction could take place (Wang et al., 2015).

126 This paper has two major objectives. First, it provides experimental observations of the
127 uptake of glyoxal on mineral dust aerosol, leading to the formation of organic aerosol
128 mass upon interaction and measuring glyoxal uptake coefficient of mineral dust.
129 Secondly, it presents the chemical composition of the mixed organic-dust aerosols, in
130 terms of ~~its~~ their oxidation state, molecular composition and the evolution of SOA
131 content from glyoxal.

2. Experiments and methods

133 This study uses the CESAM atmospheric simulation chamber (CESAM – Chambre de
134 Simulation Atmosphérique Multiphasique, which translates to Experimental
135 Multiphasic Atmospheric Simulation Chamber in English), a 4.2 m^3 cylindrical
136 stainless-steel reactor initially described by Wang et al. (2011). CESAM was
137 specifically designed to study multiphase processes involving aerosol particles, gas-
138 phase compounds and water, both in the vapour and liquid phases (Brégonzio-Rozier
139 et al., 2016; Denjean et al., 2014; Giorio et al., 2017). CESAM is equipped with three

6.5 kW high-pressure arc xenon lamps (model EX-170GM3-E, IREM SpA, Borgone, Italy) and 6 mm Pyrex plate filters to mimic the solar radiation. A 50 cm stainless-steel four-blade fan located at the bottom of the chamber ensures a mixing time of about 1 minute for the gas phase and the homogeneity of the internal composition.

2.1. Injection and cleaning protocols

Dust aerosols were generated and injected into the chamber according to the protocol detailed in Battaglia et al. (2025) using a natural soil sample from the Gobi Desert (107.48°N; 36.49°E). Prior to use, the soil was sieved at 1000 μm and dried at 100°C for less than one hour to remove adsorbed water and contamination from volatile gases. A quantity ranging from 30 and 50 g was placed in a 1 L Büchner flask and shaken at 100 Hz using a sieve shaker (Retsch® AS200) to simulate the saltation and sandblasting mechanisms through which wind erosion generates airborne dust in the real atmosphere (Di Biagio et al., 2017). An Aerodynamic Aerosol Classifier (AAC, Cambustion®) was placed between the dust generator and the chamber to inject mono-modal dust centred between 300 and 400 nm in geometric diameter.

Glyoxal was prepared by heating a mixture of equal amounts of its trimer hydrate (Fluka® Analytical) and P_2O_5 (Sigma – Aldrich ReagentPlus®, 99%) at 150°C (Horowitz et al., 2001). The trimer decomposition occurs inside a vial connected to a vacuum gas manifold. Glyoxal was collected as yellow crystals in a second vial immersed in an ethanol – liquid nitrogen cold trap at around -90°C and then vaporised in a 2.1 L glass bulb to a controlled pressure. This vial was connected to the simulation chamber to inject the glyoxal through a pure nitrogen flow.

Ozone was generated by a Corona discharge in pure O_2 using a commercial dielectric ozone generator (MBT 802N, Messtechnik GmbH, Stahnsdorf, Germany). Water vapour was generated by heating ultrapure water (Milli-Q IQ 7000, Merk™) inside a pressurised stainless-steel vessel, previously rinsed at least three times. The total organic carbon (TOC) content of the ultrapure water was monitored to evaluate the influence on the production of organic particles, which was found to be minor (see **Text S1 and Figures S1 and S2** in the Supplementary Material). The relative humidity (RH) inside the chamber was measured by a HMP234 Vaisala® humidity and temperature transmitter. Before each experiment, the chamber was evacuated down to 10^{-4} mbar and then filled with a mixture of 80% N_2 (Messer, purity > 99.995%) and 20% O_2 (Linde

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172 5.0, purity 99.999%) to an internal pressure exceeding by about 5 to 10 mbar the room

173 atmospheric pressure, to prevent accidental contamination during the experiments.

174 **2.2. Instrumentation**

175 The CESAM chamber was equipped with a suite of standard instrumentation for the

176 detection of the aerosol and the gas- phase, whose details are reported in **Table S1**.

177 Gas-phase glyoxal was measured by a combination of in-situ long-path Fourier

178 Transform Infra-Red (FTIR) spectrometry (Bruker Tensor 37) in the 2720-2930 cm^{-1}

179 absorption band corresponding to the C–H bonds, by a Cavity Attenuated Phase Shift

180 (CAPS) NO_2 analyser (Model T500U, from Teledyne API) and a Proton Transfer

181 Reaction-Time Of Flight-Mass Spectrometer (PTR-ToF-MS) (KORE Technology[®],

182 second generation) operated in H_3O^+ ionization mode for VOC detection. The FTIR

183 spectrometer also provided~~provided also with~~ the concentrations of formic acid

184 (HCOOH), measured in the C-O bond vibration band centred at 1105 cm^{-1}; ozone (O_3),

185 measured in the asymmetric stretching of the absorption band centred at about 1043

186 cm^{-1}; and carbon monoxide (CO), detected at about 2143 cm^{-1}. The standard infrared

187 absorption spectra of the compounds used for quantification, along with the specific

188 absorption bands integrated for their quantification, are shown in **Figure ~~S4~~S3**. CO

189 and carbon dioxide (CO_2) were additionally measured by an APEE ProCeas[®] analyser.

190 Nitrogen oxides (NO_x) were monitored by the APNA-370 analyser by Horiba[®].

191 The aerosol total number concentration above 2.5 nm was measured by a

192 Condensation Particle Counter (TSI[®] model 3075). The aerosol number size

193 distribution in the submicron fraction was measured by a combination of a Scanning

194 Mobility Particle Sizer (SMPS) consisting of a Differential Mobility Analyser (TSI[®],

195 model 3080) coupled with a Condensation Particle Counter (TSI[®] model 3072) and an

196 Optical Particle Counter (sky-GRIMM[®] OPC model 1.109).

197 The aerosol chemical composition was measured by a combination of online and

198 offline methods. A Time-of-flight Aerosol Chemical Speciation Monitor (ToF–ACSM,

199 Aerodyne Research Inc.) equipped with a standard vaporiser provided quantitative

200 unitary mass resolution spectra between 40 nm and 1 µm vacuum aerodynamic

201 diameter (Fröhlich et al., 2013). The instrument was operated through a Nafion

202 membrane dryer (model PD-50T-12). The organic mass concentration (m_{org}) was

203 obtained considering a unitary collection efficiency (CE = 1) and a relative ionization
204 efficiency (~~REI~~RIE) of 1.4 (Nault et al., 2023).

205 The glyoxal fragment CH_2O^+ at m/z 30 has an isobaric interference with the NO^+
206 fragment from nitrate. Following Galloway et al. (2009), the contribution of nitrate to
207 the total signal at m/z 30 was calculated as 1.7 times the intensity of the nitrate signal
208 at m/z 46, which corresponds to the 30/46 signal ratio measured during nitrate
209 calibration. The contribution to m/z 30 of glyoxal was then calculated from the total
210 signal by subtracting the contribution of the nitrate and the contribution of air. The
211 elemental ratios of the organic fraction O/C and H/C are calculated from the measured
212 f_{44} and f_{43}, respectively, following the parametrizations proposed by Aiken et al. (2008)
213 and Ng et al. (2011), respectively.

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214 Filter samples were collected on PTFE filters (Zefluor, 47 mm diameter, 2 μm pore
215 size, Pall Life Sciences) and quartz fiber membranes (Tissuquartz 2500 QATUP, 47
216 mm diameter, Pall Life Sciences) using a series of custom-made stainless-steel holder
217 of 6-mm diameter operated at 10 L min^{-1}, and preceded by an denuder filled with active
218 charcoal paper to remove ozone and VOCs. The sampling time ranged from 30
219 minutes to 3 hours. The filter holders and PTFE filters were pre-cleaned with
220 dichloromethane (99.8 %, HPLC grade) in an ultrasonic bath, while the quartz filters
221 were pyrolyzed at 550°C for approximately 8 hours. After sampling, filters were folded
222 and placed in an aluminium paper envelope previously pyrolyzed (same protocol
223 as~~than~~ the filters)~~,~~ and stored in a refrigerator at -18°C. Chamber blanks were collected
224 by sampling for about 20 min from the chamber only filled with N_2 and O_2. Analytical
225 blanks were also collected.

226 **2.3. Filter analysis**
227 The analysis of the filter samples was conducted by a combination of three techniques
228 to provide ~~with~~ a comprehensive view of the chemical composition of the organic
229 fraction formed on the dust particles as the result of the processing by glyoxal. The full
230 details of the analysers and analytical protocols are reported in as detailed in **Text S2**
231 in the supplementary material. Those include

232 1/   the Supercritical fluid extraction coupled with gas chromatography mass
233   spectrometry (SFE/GC-MS; Chiappini et al. (2006)) for target analysis of low-
234   weight mass fragments derived from molecules linked to glyoxal reactivity (see

Table S2.1 in the Supplementary Material). such as the m/z 131 fragment (Glyoxylic acid TMS derivatized – CH$_3$), and the m/z 205 fragment (Glyoxal monohydrate – CH$_3$);

2/ Electrospray ionization (ESI) high-resolution mass spectrometry (Kourtchev et al., 2015) was used to obtain high-resolution mass spectra and formulae assignments for low-volatility compounds in the ranges m/z 50–500 and m/z 150–1000 (Zielinski et al., 2018). These were processed to provide ~~with~~ parameters describing the carbon oxidation, through O/C and H/C bulk ratios, and the identification of molecular groups such as CHO, CHON, CHOS, CHNS, and CHONS. ESI-HR-MS was also used for a screening- analysis of molecules resulting from the glyoxal transformation due to hydration, oxidation, and oligomerization. These included formulas for mono- and dihydration products (C$_2$H$_4$O$_3$ and C$_2$H$_6$O$_4$, respectively), oxidation products (formic acid (CH$_2$O$_2$), glycolic acid (C$_2$H$_4$O$_3$), glyoxylic acid (C$_2$H$_2$O$_3$), and oxalic acid (C$_2$H$_2$O$_2$)), and oligomers formed by the hydrolysis of hydrated glyoxal formulas (linear oligomers with generic stoichiometric formula C$_{2n+2m}$H$_{4n+6m-2(n+m-1)}$O$_{3n+4m-(n+m-1)}$, and ring oligomers with generic formulae C$_{2n+2m}$H$_{4n+6m-2(n+m)}$O$_{3n+4m-(n+m)}$).

3/ X-ray photoelectron spectrometry (XPS) was used following –Denjean et al. (2015) to quantify the elemental O/C ratio of the particle surface (O/C$_{surf}$) to a depth of less than 10 nm. The O/C$_{surf}$ was calculated ~~as,~~ as explained in Text S2 in the Supplementary Material. The XPS measurement on a filter collected during one ageing experiment ~~are~~ is shown as an example in **Figure S~~2.1~~4** in the Supplementary Material.

2.4. Calculation of the aerosol size distribution

The procedure for combining the measurements of the SMPS and the sky-GRIMM® OPC is based on Baldo et al. (2023) and ~~it is~~ described in detail in Battaglia et al. (2025). The number size distributions, expressed in dN/dlog$D$ (cm^{-3}), are used to evaluate the total particle surface S (μm^2 cm^{-3}) and volume V (μm^3 cm^{-3}) by assuming spherical particles as

$$S = \int \pi D^2 \frac{\mathrm{d}N}{\mathrm{dlog}D} \, \mathrm{dlog}D \qquad (2)$$

$$V = \int \frac{\pi}{6} D^3 \frac{dN}{dlogD} \, dlogD \qquad (3)$$

2.5. Calculation of the glyoxal uptake coefficient and rate of particle formation

The uptake coefficient (γ) is defined as the probability of the gas to be taken up on the aerosol surface. It is a unit-less parameter expressed by the ratio between the number of molecules taken up on a surface and the total number of collisions of the gas on the surface as

$$\gamma = \frac{number\ of\ total\ molecules\ taken\ up}{total\ number\ of\ collisions} \qquad (4)$$

The gas-phase uptake coefficient (γ) was estimated from the first-order heterogeneous loss rate of glyoxal (k_{het}, s^{-1}) as

$$\gamma = \frac{k_{het}}{\omega} \qquad (5)$$

The rate of collisions (collision frequency, ω) is the defined as

$$\omega = \frac{cA_s}{4} \qquad (6)$$

where:

- $c = 146 \times \sqrt{\frac{T}{MW}}$ is the mean molecular speed (m s^{-1}), where T is the air temperature (here 298 K) and MW the molecular weight of the compound of interest (in the case of glyoxal MW = 58 g mol^{-1}).
- A_s is the total aerosol surface concentration (m^2 m^{-3}).

The total aerosol surface concentration (A_s) is calculated from the aerosol size distribution recorded at the end of the dust injection.

The heterogeneous loss rate of glyoxal (k_{het}) due to its uptake on dust particles can be determined as the difference betweenof the loss rate of glyoxal measured during the uptake experiments (k_{obs}) and the glyoxal loss rate on the chamber walls (k_{loss}) as

294

$$k_{het} = k_{obs} - k_{loss} \tag{7}$$

296

The glyoxal wall loss is represented by a partition equilibrium described by two first-order reactions: one for the adsorption of gas-phase~~gas phase~~ molecules onto the chamber walls, and one for the reverse process. The rate constants for both processes have been obtained experimentally through control experiments with only glyoxal in the chamber and at different relative humidity conditions, as detailed in **Text S3** and Figures S5 to S9 in the supplementary material.

If the uptake reaction is of the first rate, k_{het} is henceforth calculated as

$$k_{het} = \frac{ln\left(\frac{[Gly]_0}{[Gly]_{obs}}\right) - ln\left(\frac{[Gly]_0}{[Gly]_{loss}}\right)}{t} \tag{8}$$

306

where $[Gly]_0$ is the initial concentration of glyoxal, $[Gly]_{obs}$ represents the observed evolution of glyoxal concentration in time, resulting from the sum of uptake and wall loss, and $[Gly]_{loss}$ represents the estimated glyoxal concentration resulting from the wall loss.

The rate of formation of the particulate organic matter (POM; k_{F-POM}) due to the uptake of glyoxal on the dust can be calculated as

$$k_{F-POM} = \frac{ln\left(\frac{[POM]_t}{[POM]_0}\right)}{t} \tag{9}$$

315

where $[POM]_0$ represents the initial POM concentration in the particle phase, and $[POM]_t$ represents the concentration of the POM formed at a given time.

In the hypothesis that the POM formation is solely due to the uptake of glyoxal, γ can also be~~be also~~ evaluated as

$$\gamma = \frac{k_{F-POM}}{\omega} \tag{10}$$

321

3. Results and discussion

3.1. Overview of experiments

The ageing experiments of monodispersed mineral dust and glyoxal described in this paper are summarised in **Table 1**. All the aerosol data are corrected for dilution, wall loss, and particle loss through the tubing systems as detailed in **Text S4** and Figures S10 and S11 in the Supplementary Material. Gas phase concentrations are corrected for dilution only.

329

Table 1. Listing and initial conditions of the experiments considered in this study, including experiments with glyoxal only (experiment type GL), ammonium sulphate and glyoxal (AS + GL), dust only (D) and dust with glyoxal (D + GL). The glyoxal and ozone gas phase concentrations correspond to the maximum value measured by FTIR after the respective injections. V_{seed} indicates the maximum volume concentrations of seed particles (either dust or ammonium sulphate) measured after the particle injection. The notation "dark/light" indicates experiments when filter samples were collected both in the dark and with irradiation.

Experiment type	Reagents	Date	Experiment number	RH, %	Light	$[O_3]$, ppb_v	Temp, K	$[GL]$, ppb_v	V_{seed}, μm^3 cm^{-3}
Control	GL	29/04/2021	G_1	< 5	dark	---	292	1130	---
		11/02/2022	G_2	77	light	1440	291	627	---
	AS+GL	21/02/2023	AS_1	38	dark	---	298	527	50.1
		23/02/2023a	AS_2	35	dark	---	298	516	48.3
		23/02/2023b	AS_3	32	light	---	298	445	64.8
		07/09/2023	AS_4	81	light	---	301	779	304.1
		08/09/2023	AS_5	83	light	---	300	430	161.2
Uptake	D	31/01/2022	D_1	< 5	dark/light	---	292	---	31.5
		03/02/2022	D_2	75%	dark/light	---	293	---	55.4
	D+GL	04/02/2022	D_3	< 5	dark/light	---	293	690	35.6
		08/02/2023	D_4	32	dark	---	294	940	21.5
		09/02/2023	D_5	31	light	---	295	1050	52.7
		10/02/2023	D_6	35	dark	---	294	809	37.4
		13/02/2023	D_7	34	light	---	296	850	51.3
		30/04/2021	D_8	76	light	---	289	759	28.3
		03/05/2021	D_9	79	light	---	290	607	38.7
		04/05/2021	D_{10}	81	light	---	290	371	31.5
		05/05/2021	D_{11}	78	dark	---	291	805	30.1
		06/05/2021	D_{12}	82	dark	---	292	432	21.1
		08/02/2022	D_{13}	81	dark/light	1270	293	555	64.0
		09/02/2022	D_{14}	78	dark/light	1450	293	756	79.8
		10/02/2022	D_{15}	75	dark/light	---	295	600	68.4
		14/02/2023	D_{16}	83	dark	---	296	661	35.8
		15/02/2023	D_{17}	75	light	---	298	444	41.0

337

338 **Table 1** also lists the few control experiments using ammonium sulphate as seed
339 particles, described in detail in **Text S5** and Figures 12 to 14 in the Supplementary
340 Material. No POM formation was observed during control experiments with dust or
341 glyoxal only, both dry and humid conditions and with and without irradiation.

342 **3.1.1. Timeline of particle concentration and composition**

343 The ageing experiments lasted up to five hours. For the experiments carried out in wet
344 conditions, the injection of water vapour preceded the injection of dust. The injection
345 of glyoxal (nominal concentration of 1 ppmv) was conducted at least after 30 minutes
346 after the dust to ensure that the dust particles were homogenously distributed in the
347 reactor. Irradiation was started within one hour after the glyoxal uptake onto the
348 particles. InOn a few experiments, ozone was injected before glyoxal to verify the
349 sensitivity of the reactions to the presence of an oxidant.

350 The typical timelines of the particle concentrations (number and volume) and the non-
351 refractory composition measured in dry conditions and at 30% and 80% relative
352 humidity are shown in **Figure 1**.

353
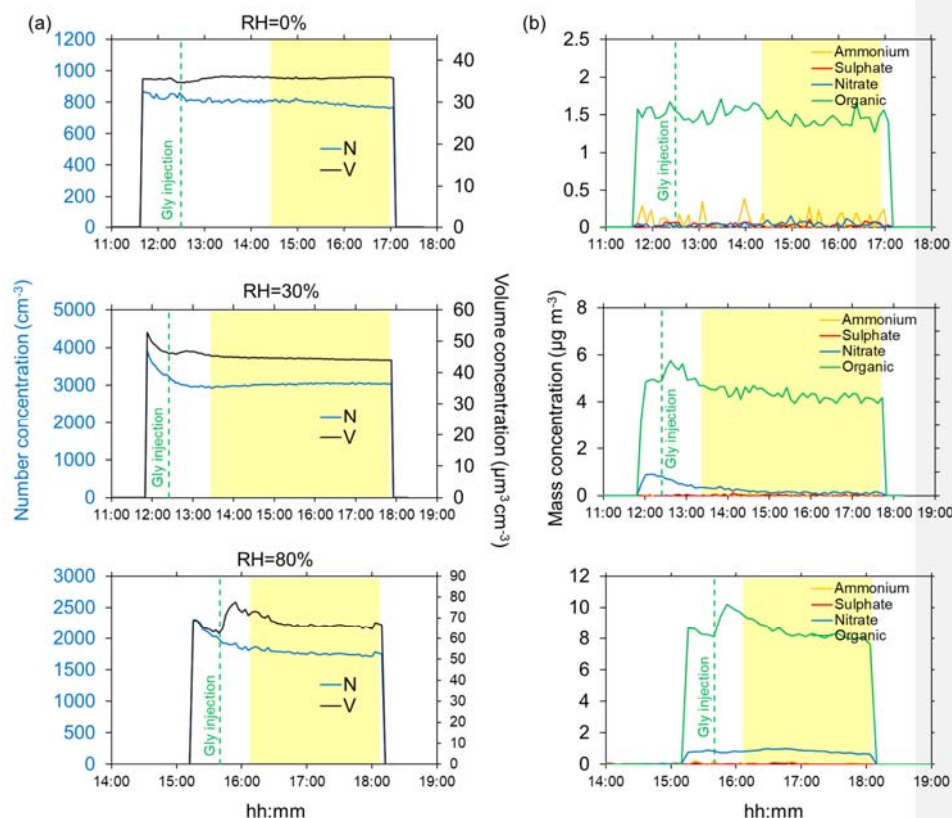
Figure 1. Timeline of ageing experiments of submicron dust with gas-phase glyoxal in dry conditions (top, experiment D_3), 30% (middle, experiment D_7), and 80% RH (bottom, experiment D_{15}). Left (a): aerosol total number (N) and volume (V) concentrations (blue and black lines, respectively) calculated from the measured dust size distributions. Right (b): mass concentrations of ammonium, sulphate, nitrate and organic (yellow, red, blue and green lines, respectively) measured by the ACSM. The yellow-highlighted portion of the graph indicates the interval where irradiation takes place, while the green vertical dashed lines indicate the injection of glyoxal in the chamber. The dust injection corresponds to the time of the initial increase of the number and volume concentrations. Aerosol time series are corrected for dilution, wall loss and particle loss through the tubings.

Figure 1 shows that in dry conditions, there is no significant variation of either the aerosol number or the volume concentrations, nor the chemical composition (including organics) following the glyoxal injection.

At 30% RH, a small increase in the total volume concentration (approximately 5 μm^3 cm^{-3}) is observed for about 30 minutes after the injection of glyoxal. This corresponds to an increase of the POM of about 1 $\mu g\ m^{-3}$, approximately 20% more with respect to

the value measured before the uptake. On the other hand, the particle number concentration shows an apparent decrease at the beginning of the experiments, possibly because the particle loss correction model of Lai and Nazaroff (2000) does not fully apply to dust particles and humid conditions (see discussion in Battaglia et al. (2025). After that, and through the duration of the experiment, however, it remains constant, indicating that the increase in the particle volume occurs on the dust particles and not because of new particle formation.

At 80% RH, the increase in both the total volume and the POM concentrations is more pronounced, approximately 10-15 μm^3 cm^{-3} and 2 μg m^{-3}, respectively. As for 30% RH, both total particle volume and the POM concentrations return to values observed prior to the injection of glyoxal, within approximately 30 minutes from their maximum values, likely due to evaporation. A similar behaviour is observed in the presence of ozone (**Figure ~~S2~~ S15** in the Supplementary Material). As for 30% RH, the particle number concentration slightly decreases in time at the beginning of the experiment, but then remains constant, again excluding the formation of new particles but rather confirming the formation of organic matter on pre-existing particles. This is also supported by the fact that the rate of increase of POM and particle volume is the same (slope 3.2 × 10^{-4} s^{-1} and 3.2 × 10^{-4} s^{-1}, respectively, for POM and total volume), as shown by **Figure ~~S3~~ S16** in the Supplementary Material.

3.1.2. Timeline of gas-phase concentration

Figure 1 also shows~~shows also~~ that, while sulphate and ammonium are never detected, a background concentration of nitrate up to 1 μg m^{-3} is measured by the ACSM as soon as the dust particles are injected in the presence of water. We attribute it to the heterogeneous interaction between NO_2 and the dust particles (Goodman et al., 1999), as indeed, a background concentration of a few ppb of NO_2 is present in the chamber as a result of the procedure used to reduce the TOC content in the injected water (see **Figure S17~~4~~** in the Supplementary Material). However, since the contribution of nitrate represents at maximum 1% of the injected dust mass and ~~whether~~ it decreases or remains constant throughout the experiment, its contribution to the particle growth and overall ageing of the mineral dust should be negligible.

Figure 2 shows the time series of the gas-phase compounds detected during the same experiment (D_{15}) at 80% RH.

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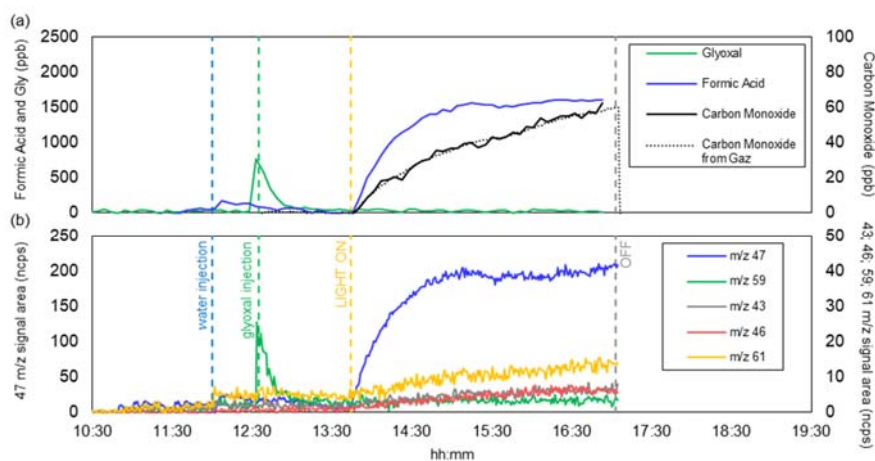
Figure 2. Time series of the gas-phase composition observed during experiment D_{15}: (a) concentrations of carbon monoxide, glyoxal and formic acid measured by FTIR (for CO, the ~~measured~~ measurements of the online analyser are also shown); (b) various VOC ions (m/z 47, 59, 43, 46 and 61) measured by the PTR-MS. Ion signals measured by PTR-MS are normalized by signals of reagent ions (i.e. H_3O^+ and $H_3O^+(H_2O)$) and therefore expressed in normalized counts (ncps). The blue vertical dashed lines indicate the injection of water in the chamber; the green vertical dashed lines indicate the injection of glyoxal in the chamber, the yellow dashed lines indicate the beginning of irradiation, and the grey dashed lines indicate the end of irradiation.

The measured glyoxal concentration after the injection (**Figure 2a**) is lower than the nominal concentration of 1 ppm and goes to zero within minutes due to the rapid interactions with the walls of the chamber, water vapour, and the dust particles. Upon irradiation, formic acid and carbon monoxide are formed, as expected by the photolysis of glyoxal (De Haan et al., 2020). Fragments m/z = 46 and 47 are observed during water injection and photolysis, which could originate from the deprotonated and protonated form of formic acid, respectively. This suggests that a minor fraction of the formic acid could result from the desorption of compounds (including glyoxal) from the chamber walls. Fragments m/z = 43 and m/z = 61, and occasionally m/z = 45 (not seen during experiment D_{15} and therefore not shown in **Figure 2b**), are observed at a normalised intensity two orders of magnitude lower than that of formic acid, but not attributed. The quantification with both PTR-MS and FTIR in our experimental RH conditions is complex due to water presence, which reduces the sensitivity of PTR-MS and can interfere with the absorption of various organic compounds, making their quantification less accurate.

3.2. Evaluation of glyoxal uptake coefficient and rate of formation of organic
aerosols

Figure 3 shows an example of the temporal evolution of the natural logarithm of the glyoxal concentration measured by the FTIR, compared to that measured during a typical blank experiment without dust particles (top panel) and the variation of the aerosol organic fraction measured by the ToF-ACSM during the same time period (lower panel).
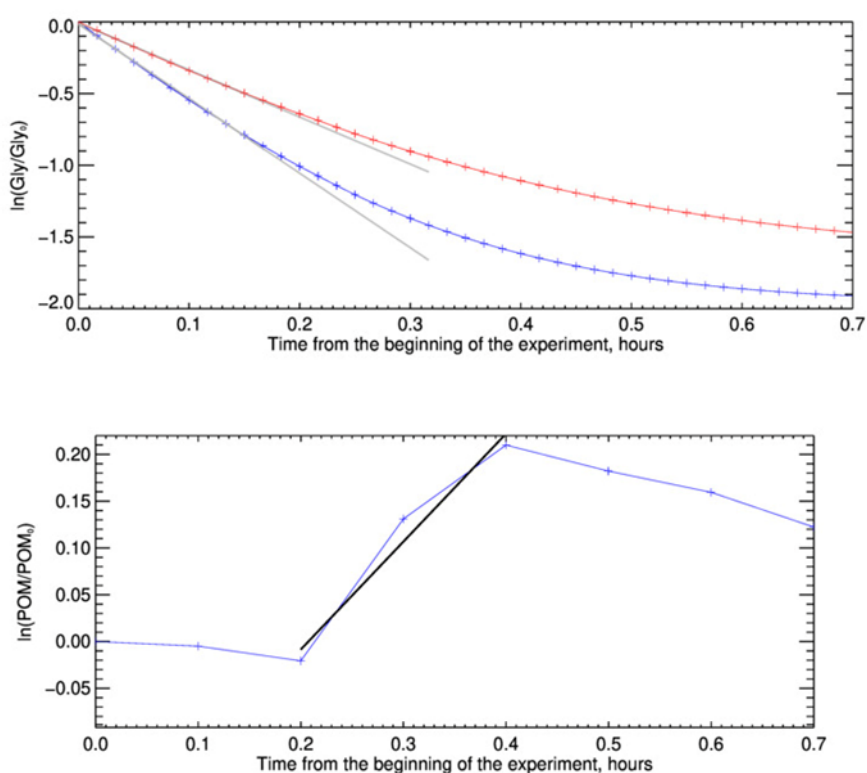
Figure 3. Measurement of glyoxal uptake coefficient on dust for the experiment D_{15}, conducted at 80% RH. The figure compares the two methods for measuring the uptake coefficient. In the top image, results are shown for the method based on monitoring the decay of gas-phase glyoxal. The red and blue curves represent the logarithm of the ratio between the calculated decay of gas-phase glyoxal in the absence and presence of dust aerosols, respectively. The black lines represent the linear fit whose slope provides the heterogeneous kinetic constants of the two processes. The image at the bottom displays the result of the uptake coefficient measurement for the same experiment, obtained from the organic formation on the dust aerosol monitored by the ToF-ACSM. The blue time series shows the logarithm of the ratio between the measured organic concentration divided by the initial organic on dust aerosol, while the black line is the linear fit representing the kinetics of organic formation.

Within the first 10 minutes after the injection of glyoxal, the decrease of the natural logarithm concentrations ratio with time in the presence of dust is linear (that is, the rate is constant). After that, the loss slightly deviates from linearity. The difference from linearity is more evident for the blank experiment, when it occurs earlier than when the dust is present. These observations indicate that, within the first 10 minutes, the uptake of glyoxal on the dust particles can be considered to follow a first-order kinetics and its rate represents an initial uptake coefficient. In the following 20 minutes approximately, the uptake slows down, possibly because all the sites available on the particle surface become occupied, but also because that desorption from the particle surface could reinject glyoxal into the reactive mixture. On the particle phase, the natural logarithm of the organic concentration, normalised by its initial value, increases rapidly and linearly, almost on the same time scale as that of the loss of glyoxal, but then decreases to return to its initial value within approximately one hour. These observations confirm that the uptake of glyoxal results in the formation of OA on the dust particles, but that this process is reversible.

The uptake coefficients calculated as the linear fit of the glyoxal and particle organic concentration are presented in **Table 2**.

Table 2. Uptake coefficients for glyoxal on mineral dust and ammonium sulphate calculated from the loss of gas-phase glyoxal ($\gamma_{Gly\text{-}Dust\text{-}gas}$) and the rate of OA formation ($\gamma_{Gly\text{-}Dust\text{-}gas}$) for the experiments conducted at 80% RH. The initial glyoxal concentration is reported. The aerosol surface concentration (A_s) corresponds to the value preceding the glyoxal injection. Ozone concentration is the maximum concentration measured by FTIR spectroscopy after the injection. For ammonium sulphate, only the γ values calculated from the loss of gas phase glyoxal are presented, as the ACSM collection efficiency (CE) for ammonium sulphate varies significantly during OA formation (Matthew et al., 2008).

Date	Experiment ID	RH%	$[GL]_0$, ppb$_v$	Ozone (ppb)	A_s (m² m⁻³)	ω (s⁻¹)	$\gamma_{Gly\text{-}Dust\text{-}gas}$	$\gamma_{Gly\text{-}Dust\text{-}OA}$
30/04/2021	D_8	76	759	---	4.8×10^{-4}	3.9×10^{-2}	6.0×10^{-3}	1.0×10^{-3}
03/05/2021	D_9	79	607	---	6.1×10^{-4}	5.0×10^{-2}	1.5×10^{-2}	1.5×10^{-2}
04/05/2021	D_{10}	81	371	---	5.1×10^{-4}	4.2×10^{-2}	1.7×10^{-2}	9.0×10^{-3}
05/05/2021	D_{11}	78	805	---	4.6×10^{-4}	3.8×10^{-2}	8.0×10^{-3}	5.0×10^{-3}
06/05/2021	D_{12}	82	432	---	3.5×10^{-4}	2.9×10^{-2}	1.2×10^{-2}	2.3×10^{-2}
08/02/2022	D_{13}	81	555	1270	7.1×10^{-4}	5.8×10^{-2}	4.0×10^{-3}	4.0×10^{-3}
09/02/2022	D_{14}	78	756	1450	8.5×10^{-4}	7.0×10^{-2}	4.0×10^{-3}	5.0×10^{-3}
10/02/2022	D_{15}	75	600	---	8.4×10^{-4}	6.9×10^{-2}	1.0×10^{-2}	5.0×10^{-3}
14/02/2023	D_{16}	83	661	---	6.0×10^{-4}	4.9×10^{-2}	4.0×10^{-3}	1.5×10^{-2}
07/09/2023	AS_4	81	779	---	6.3×10^{-3}	5.2×10^{-1}	9.8×10^{-4}	---
08/09/2023	AS_5	83	430	---	2.0×10^{-3}	1.7×10^{-1}	1.2×10^{-3}	---
Average dust							$9\ (\pm 5) \times 10^{-3}$	$9\ (\pm 7) \times 10^{-3}$
Average AS							$1.1\ (\pm 0.2) \times 10^{-4}$	

The average uptake coefficients for glyoxal on the Gobi mineral dust calculated at 80% RH from the gas-phase uptake and the particle formation are $\gamma_{Gly-Dust-gas}$ = 9 × 10^{-3} (standard deviation ± 5) and $\gamma_{Gly-Dust-OA}$ = 9 × 10^{-3} (standard deviation ± 7), respectively. The two average values agree. This suggests that every glyoxal molecule in the gas phase is taken up by the airborne dust particles. This also suggestssuggests also that the uptake occurs on airborne particles only, as expected, as the dust particles are selected in the submicron range and that minimal deposition of dust particles is observed in the first 30 minutes after injection. The primary mechanism of particle loss during this period is dilution, which does not interfere with uptake. The standard deviations of the mean values are large, being attributed to the fact that the state of the chamber walls and the dust size distribution vary from one experiment to the other, and that the aerosol/chamber walls surface ratio is very low (0.08-1.5 × 10^{-3}).

The presence of ozone appears to be uninfluential. Hanish and Crowley (2003a) investigated the combined uptake of O_3 and HNO_3 onto dust, and showed that the uptake of HNO_3 on dust was not influenced by the presence of O_3 (and conversely, the uptake of O_3 was not influenced by the presence of HNO_3). These authors attributed these observations to the fact that in their experiments, O_3 concentrations were in excess by more than three orders of magnitude with respect to those of HNO_3, compensating for the fact that the uptake coefficients for O_3 is approximately four orders of magnitude lower than for HNO_3 (Hanish and Croley 2003b; Chang et al., 2005). In these conditions, which are not generally met in the atmosphere, the presence of O_3 could result in the modification of surface chemical characteristics, or to competition for adsorption sites of the two species. These considerations are applicable to our experiments, as the uptake coefficients of O_3 and glyoxal are of comparable magnitudes, and we use comparable concentrations.

3.3. Evidence of irreversible particle growth

Figure 1 shows that, even in the more favourable conditions (RH=80%), the ratio between the observed increase of the POM (2 µg m^{-3}) and that of the particle volume concentration (20 µm^3 cm^{-3}) corresponds to an estimated mass density of the order of 0.1 g cm^{-3}, which is about 10 times lower than the value of 1 g cm^{-3} expected for glyoxal.

505   This is partially attributed to the fact that only part of the organic matter formed on dust
506   is detected by the ACSM. On the other hand, **Figure ~~S3~~ S16** shows that, after reaching
507   its maximum value, the volume concentration decreases at a lower rate than the POM
508   (slope 3.9×10^{-5} s^{-1} and 6.1×10^{-5} s^{-1}, respectively). This suggests that an additional
509   process could contribute to the particle volume concentration, partially compensating
510   for the loss of organic matter on the dust particles.

511   This is confirmed by **Figure 4,** which shows the variation in time of the normalised
512   particle volume distributions, at four steps of the experiments performed at 30% RH
513   and 80% RH. Those include prior and glyoxal injection, POM maximum peak value,
514   and the end of the experiment.

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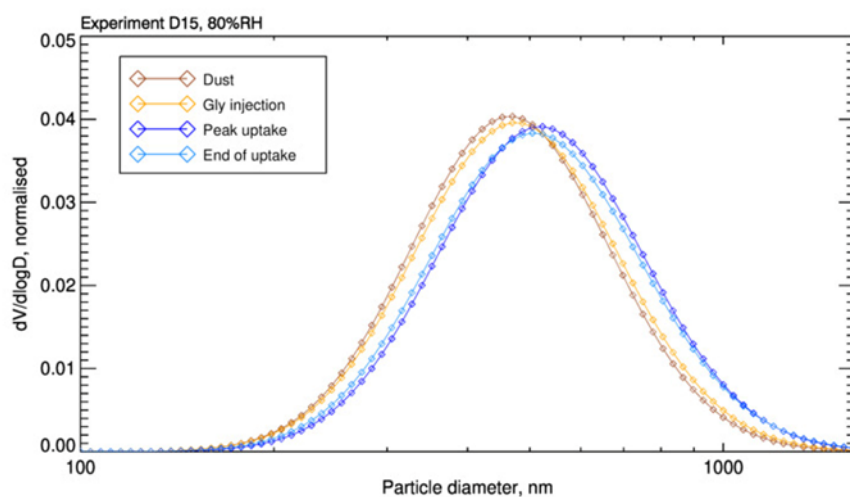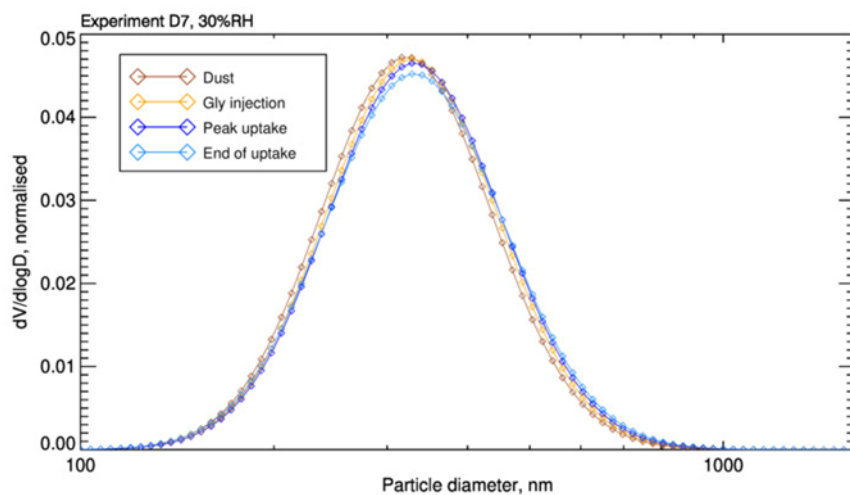
518 **Figure 4.** Evolution of volume-size distributions for two glyoxal uptake experiments in different relative
519 humidity conditions. The images illustrate the progression of volume-size distributions recorded at four
520 key moments during the experiments. The first distribution (orange) is recorded after the dust is injected
521 into the simulation chamber. The second distribution (yellow) is recorded at the moment of glyoxal
522 injection. The third distribution (blue) corresponds to the peak uptake of glyoxal on the aerosol, and the
523 fourth (light blue) is recorded at the end of the glyoxal uptake process. The left image depicts the
524 evolution for the experiment D7 conducted at 30% RH, while the image on the right shows the
525 distributions for the experiment D15 conducted at 80% RH. The results highlight that the distributions
526 grow more significantly at 80% RH, indicating a higher glyoxal uptake and organic formation at elevated
527 humidity levels.
528

529   All distributions have a single mode. However, after the injection of glyoxal, the
530   geometric mean volume diameter, measured at the maximum POM concentration,
531   increases by up to 10% (from 310 to 340 nm) at 30% RH, and up to 20% (from 450 to
532   540 nm) at 80% RH. Interestingly, even at the end of the experiment, when the POM
533   concentration returns to its initial value, the increase in geometric mean diameter of
534   the aerosol is irreversible. We hypothesize that the uptake of glyoxal enhances the
535   dust hygroscopicity. Therefore, the difference in total volume between the beginning
536   and end of the experiment is due to formed organic aerosol mass, resulting from water
537   uptake.

3.4. Chemical composition of the particulate organic matter

539   This section discusses the chemical composition of the particulate organic matter
540   formed on the mineral dust following the interaction with glyoxal. The list and conditions
541   of the filter samples analysed by SFE/GC-MS and ESI-Orbitrap are reported in **Table
542   S2** in the Supplementary Material. Details of the organic composition of the native dust
543   are provided in **Text S6 and Figures S18 to S20**.

3.4.1. Timeline of chemical evolution

545   **Figure 5** shows the time evolution of the intensity of the organic fragments detected
546   by the ToF-ACSM at 80% RH (experiment D_{15}) at four moments of the experiment: 1)
547   before the injection of glyoxal; 2) during the uptake and the formation of the organic
548   matter; 3) after the organic matter has reached its maximum concentration; and 4) at
549   the end of the experiment, when the organic matter returned to its initial concentration.

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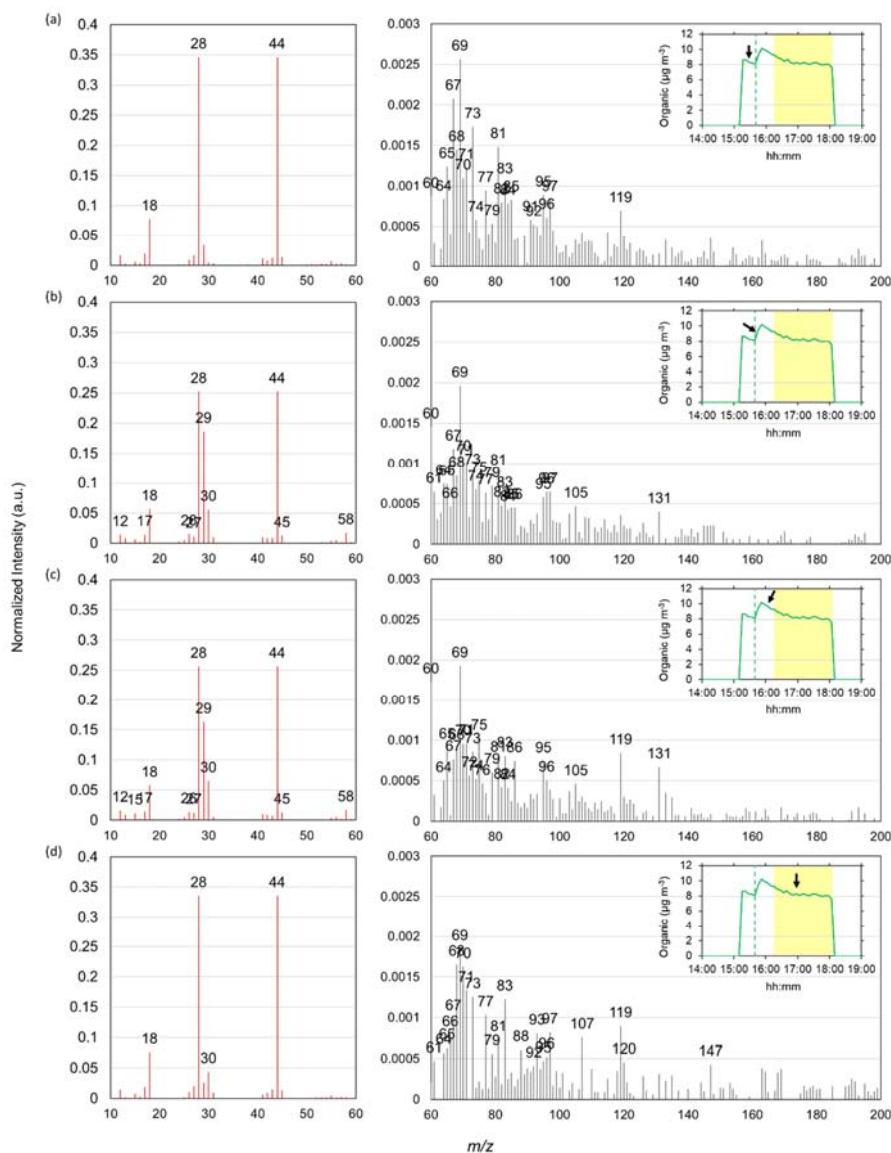
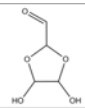
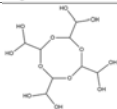
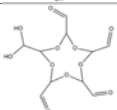

551 **Figure 25.** ACSM organic mass spectra (intensities normalized to the total organic concentration) recorded during
552 the experiment D_{15}: (a) before glyoxal uptake (dust organic fraction composition), (b) during glyoxal uptake, (c) after
553 reaching the maximum uptake on the particles and (d) 1h later under irradiation. Panels on the left show the mass
554 spectra ranging from m/z 10 to 60, while the panels on the right represent fragments from m/z 60 to 200 (their
555 intensity is approximately one hundred times lower). The inserts display the time series of organic concentrations
556 measured by the ToF-ACSM. A black arrow indicates the time corresponding to the mass spectrum shown. The
557 yellow-highlighted shaded area indicates the interval where irradiation takes place, while the green vertical dashed
558 lines indicate the moment of glyoxal injection in the chamber.

559 **Figure 5** reveals that the organic matter formed as a result of the interaction between

560 mineral dust and glyoxal is composed of oxidized organic fragments, organic acids,

561 and oligomers. Oxidized organic fragments such as CO^+ (m/z 28) and CO_2^+ (m/z 44)

562 are observed through all phases of the experiments. Fragments such as CHO^+ (m/z

563 29), CH_2O^+ (m/z 30), and $C_2H_2O_2^+$ (m/z 58), appear during the uptake of glyoxal, but

564 diminish over time, due to oligomerization or further oxidation processes. The most

565 intense fragment above m/z 60 is m/z 69, which appears unrelated to glyoxal reactivity

566 but could be attributed to nitrogen-containing organic species such as $C_3H_3NO^+$,

567 originating from glyoxal–ammonia reactions (Galloway et al., 2009) or plant and soil

568 residues (Sun et al., 2010; Nieder et al., 2018). Fragments at m/z 105 and 131,

569 observed consistently during the uptake and the photolysis, could be attributed to

570 condensed glyoxal hydrates (Liggio et al., 2005; Carlton et al., 2007). Fragments at

571 m/z 119 and 120, associated with oxidized products from isoprene-derived aerosols

572 (Safi Shalamzari et al., 2013), increased after glyoxal uptake and under irradiation,

573 suggesting the formation of oxidized oligomers. Upon irradiation, the appearance of

574 m/z 147 and 165 – products of aqueous glyoxal oxidation and oligomer condensation

575 (Lim et al., 2010) – suggests photo-chemically enhanced transformation, though their

576 presence in native dust also points to reversible adsorption processes.

577 **3.4.2. Molecular identification**

578 The list and conditions of the samples analysed by SFE/GC-MS and ESI-Orbitrap are

579 reported in **Text S7** and Figures S21 and S22 in the Supplementary Material, which

580 also provides with examples of analysis. The summary of the organic molecules

581 detected by those analysis is presented in **Table 3**.

582

583 **Table 3.** Summary of observed compounds identified by SFE/GC-MS analysis and glyoxal-related
584 formulas observed with ESI-Orbitrap, along with the suggested structures under the different
585 experimental conditions tested.

Molecular formula	Name	Tentative Structure	Technique	Experimental conditions
$C_2H_2O_3$	Glyoxylic acid		ESI-Orbitrap	Dust+Gly, 80%, Dark, O_3
$C_2H_4O_3$	Glycolic acid		ESI-Orbitrap SFE/GC-MS	Dust+Gly, 30%, Dark Dust+Gly, 30%, Light Dust+Gly, 80%, Light Dust+Gly, 80%, Light, O_3
$C_2H_2O_4$	Oxalic acid		ESI-Orbitrap	Dust+Gly, 80%, Light

$C_2H_4O_4$	Glyoxylic acid monohydrate		ESI-Orbitrap SFE/GC-MS	Dust+Gly, 80%, Light
$C_4H_4O_4$	Glycolic acid dimer		ESI-Orbitrap	Dust+Gly, 80%, Light
$C_4H_6O_5$	Glyoxal oligomer		ESI-Orbitrap	Dust+Gly, 80%, Light
$C_6H_6O_6$	Glyoxal oligomer		ESI-Orbitrap	Dust+Gly, 80%, Light
$C_8H_{16}O_{12}$	Glyoxal oligomer		ESI-Orbitrap	Dust+Gly, 80%, Light
$C_{10}H_{12}O_{11}$	Glyoxal oligomer		ESI-Orbitrap	Dust+Gly, 80%, Dark

586

587 Oxidized organic compounds such as glycolic acid ($C_2H_4O_3$), oxalic acid ($C_2H_2O_4$),
588 and a possible dimer of glycolic acid ($C_4H_4O_4$) were mainly observed under irradiated
589 conditions, while their hydrated forms also appeared in dark conditions. Glyoxylic acid
590 was detected in dark conditions in the presence of ozone. Glycolic and glyoxylic acids
591 consistently form under humid conditions, indicating that their pathways may be less
592 sensitive to water competition or that their precursors interact more strongly with dust
593 surfaces. Light but also ozone tends to favor the formation of glycolic acid from glyoxal
594 at high RH (**Figure S7.1**), suggesting two possible oxidative pathways. Glycolic acid is
595 also detected at 30% RH (not showed), with and without irradiation, in agreement with
596 the experiments on dust by Shen et al. (2016), but differently than reported by Galloway
597 et al. (2009) on ammonium sulphate. Monohydrated glyoxylic acid is found in one
598 sample at 80% RH under irradiated conditions, likely due to the known pattern of
599 oxidation of glyoxal and glycolic acid with OH radicals (Buxton et al., 1997). Unlike
600 previous studies (Galloway et al., 2009; Rubasinghege et al., 2013; Shen et al., 2016),
601 formic acid was not detected on the particle phase despite high concentrations
602 observed in the gas phase (**Figure 2**), possibly due to high RH suppressing its
603 formation by limiting access to reactive surface sites, as suggested by Shen et al.
604 (2016). It can be hypothesized that formic acid was not detected in the particle phase
605 due to the low amount of organic material on the filter samples and the unsuitability of

606 the method used (due to interferences at the retention time where formic acid is
607 expected).

608 Oligomerization products (compounds from C4 to C10) of the glyoxal mono- and di-
609 hydrate forms, are observed only at 80% RH. These are: $C_4H_6O_5$ (1 monohydrated
610 glyoxal + 1 dehydrated glyoxal forming a 5-atom ring), $C_8H_{16}O_{12}$ (4 dehydrated glyoxal
611 molecules forming an 8-membered ring), and $C_{10}H_{12}O_{11}$ (4 monohydrated glyoxal
612 molecules + 1 dehydrated glyoxal molecule forming a ring structure). The oligomer
613 $C_6H_6O_6$ (3 molecules of monohydrated glyoxal forming a 6-membered ring) is detected
614 only under dark conditions. $C_8H_{16}O_{12}$, can correspond to an oligomer previously
615 observed by Shen et al. (2016) on mineral dust.

616 The persistence of low-volatility, heavy compounds, such as oligomers at the surface
617 of mineral dust has implications for its oxidation state, which is modified in an
618 irreversible way its surface composition, as already shown by previous studies on
619 ammonium sulphate seeds (Kroll et al., 2005; Galloway et al., 2009; De Haan et al.,
620 2020; Hu et al., 2022). This is illustrated by the van Krevelen diagrams obtained from
621 the ESI-Orbitrap analysis in **Figure 6**.

622

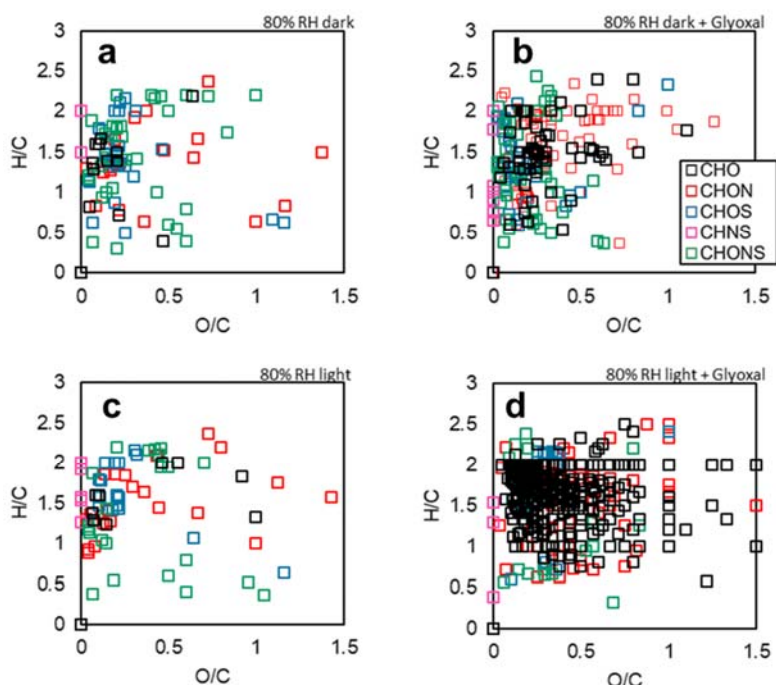



Figure 6. Van Krevelen diagrams recorded at 80% RH for: in the top line experiments in the dark for mineral dust only (control experiment D_2, left) and one ageing experiment of dust with glyoxal (experiment D10); bottom line: same with irradiation. Samples are equivalent in terms of load of particulate organic matter. These are 0.9 μg (sample a), 0.8 μg (sample b), 1.7 μg (sample c) and 0.6 μg (sample d). Despite an equivalent loading of particulate organic mass, the number of signals detected is significantly higher when the dust is exposed to glyoxal (86 and 102 peaks detected for dust only against 398 and 310 with glyoxal, and with and without light, respectively).

The processing by the glyoxal clearly has an effect on the oxidation state of the dust, particularly when lights are on, resulting in the appearance of signals with O/C ratio higher than 1, attributed to photo-oxidation. The predominant family in this case is that of CHO molecules, while the appearance of molecules for families CHON and CHONS is also observed.

4. Discussion

4.1. Comparison of uptake coefficients

The uptake of glyoxal on the dust particles occurs in humid conditions exceeding 30% RH. These observations agree with the results of Liggio et al. (2005a; 2005b) on the uptake of glyoxal on ammonium sulphate aerosols, observing the formation of organic matter only when RH exceeded 50%. Trainic et al. (2011) also observed that the uptake of glyoxal on glycine and ammonium sulphate particles occurred only when the relative humidity was above 35%. On the contrary, both Shen et al. (2016) and Zogka et al. (2024) demonstrated that the uptake can occur in dry conditions too, which was not observed in this work.

At 80% RH, the experimental average of the measured uptake coefficient of glyoxal on mineral dust is $\gamma = (9 \pm 5) \times 10^{-3}$. Our values are approximately one order of magnitude higher than those obtained by Shen et al. (2016), who investigated the uptake of glyoxal on mineral proxies, i.e. SiO_2, ~~and~~ $CaCO_3$ and $\alpha\text{-}Al_2O_3$ under various levels of RH. These authors determined the uptake coefficients after a long exposition of the surface to glyoxal (steady state uptakes) and found that the uptake coefficients decrease with increasing ~~the~~ gas phase concentration of glyoxal. At 1 ppb concentration and 60% RH, the uptake coefficient determined on suspended particles of calcite ($CaCO_3$) is 1.4 ($\pm$ 0.1) $\times 10^{-4}$ and 5.5 $\pm$ (0.1) $\times 10^{-5}$ on alumina ($\alpha\text{-}Al_2O_3AlO_3$).

Zogka et al. (2024) used a Knudsen cell to evaluate the initial and ~~steady~~ steady-state glyoxal uptake coefficient bulk soil samples of various origins. At low RH, these authors found that for Gobi soil sieved to less than 63 μm in diameter, the initial uptake coefficient using the geometric surface area was 0.18 (corresponding to an upper limit of the uptake), independent of glyoxal concentration. However, the steady state uptake coefficients determined after a long processing of the surface were found to decrease with increasing glyoxal concentration, due to aging of the surface.

Various reasons could induce those apparent differences. First, the differences in the experimental set up as in CESAM, the initial uptake coefficient was measured on dust aerosol particles in suspension in a large volume, compared to sieved soil (Zogka et al., 2024) and grinded mineral powders (Shen et al., 2016). The uptake coefficient is inversely proportional to the available particle surface. In our experiments, the dust available geometric surface density, calculated from the measured size distribution

669 assuming spherical particles, is in the range 0.35-6.3 × 10^{-3} m² m^{-3} (**Table 2**). As the

670 ~~the~~ CESAM chamber volume is equal to 4.2 m³, the total dust surface area available

671 for the reaction ranges from 2 to 2.6 × 10^{-3} m², several orders of magnitude lower than

672 in the experiments designed by Shen et al. (2016) and Zogka et al. (2024). Shen et al.

673 (2016) used model powders of various sizes between 35 nm and 5 µm, with BET

674 (define BET) surface areas ranging between 1.4 and 440 m² g^{-1}. In the conservative

675 assumption that only 5 mg of the model powder was used, the total reactive surface

676 area was up to 2.2 m². Zogka et al. (2024) used the soil from Gobi sieved to 63 µm

677 and a BET surface area of 10.5 ± 1.0 m² g^{-1}. Using even just 1 g would yield a surface

678 area of the order of 10.5 m². Differences in the results could also arise by differences

679 in the dust mineralogy, which are difficult to ascertain in the present study.

680 Our estimated uptake coefficient of glyoxal on mineral dust is also nearly two orders of

681 magnitude higher than for ammonium sulphate (γ_{gas-AS} = 1.1 (± 0.2) × 10^{-4}; our study

682 as well as Curry et al., 2018; De Haan et al., 2020; Galloway et al., 2009; Liggio et al.,

683 2005b, a; Trainic et al., 2011), but lower than γ_{gas-AS} = 2.9 × 10^{-3} at a lower RH (Liggio

684 et al., 2005). The difference could be due to the higher hygroscopicity of ammonium

685 sulphate, enhancing water's competition with glyoxal for adsorption sites at 80% RH,

686 when indeed ammonium sulphate is deliquescent. This suggests, nonetheless, that

687 dust aerosols could play a very substantial role in the formation of organic aerosols at

688 high RH compared to ammonium sulphate, which is often used as an aerosol proxy.

689

690 **4.2. Mechanism of chemical transformation**

691 **Figure 7** illustrates the suggested chemical mechanisms of the transformation of gas-

692 phase glyoxal on the mineral dust particles.

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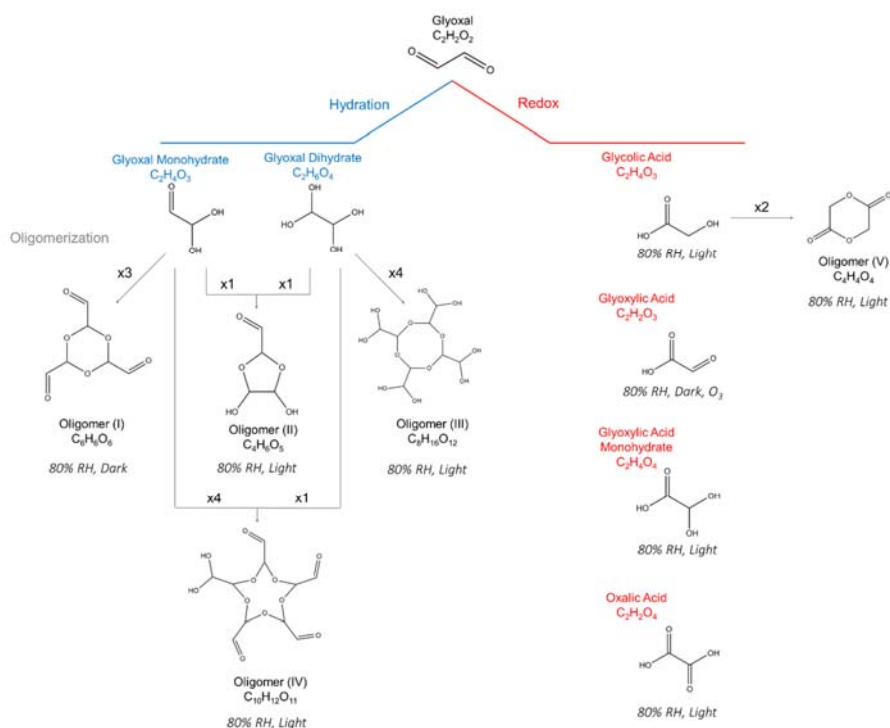
Figure 7. Proposed reaction scheme to explain the glyoxal-related molecular formulas detected through
ESI-Orbitrap mass spectrometry and SFE/GC-MS.

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The multiphase chemistry of glyoxal on mineral dust particles, leading to the formation
of low-volatility, particle-phase products, is complex, involving hydration, redox, and
oligomerization pathways. The chemical transformation of glyoxal upon uptake by
mineral dust particles primarily occurs through aqueous-phase reactions. The
mechanism is initiated by the hydration of glyoxal's aldehyde groups, forming mono-
and di-hydrated species. These hydrated intermediates undergo condensation
reactions, which involve nucleophilic attack and lead to the formation of cyclic acetal
structures, particularly from two to five-membered dioxolane rings. This mechanism is
well supported by prior studies (Loeffler et al., 2006; Kua et al., 2008; Hastings et al.,
2005), which have described similar pathways for glyoxal oligomerization in aqueous
environments.

709 This study provides new insight into ~~how~~ this chemistry on mineral surfaces. While
710 previous research has shown that glyoxal typically forms dimers and trimers in
711 aqueous solution (Liggio et al., 2005; Nozière et al., 2009), our mass spectral data
712 reveal the presence of oligomeric species containing up to 5 glyoxal units. This higher
713 degree of oligomerization - also observed by Shen et al. (2016) - suggests that the
714 particle-phase environment allows for extended oligomer growth. The likely
715 contributing factors include reduced water activity, surface confinement, and enhanced
716 proximity of reactants, all of which promote successive addition reactions that may be
717 hindered in bulk solution (Gomez et al., 2015; Avzianova & Brooks, 2013). Concerning
718 hydration reactions, Shen et al. (2016) showed that both glyoxal and water can
719 accumulate onto dust particle surfaces (as evidenced by FTIR spectra), promoting
720 hydration reactions through surface confinement of the reagents. Absorption of water
721 on dust surfaces can also promote oligomerisation, both directly (hydration is the first
722 step of the oligomerisation reaction) as well as indirectly by enhancing particles' ability
723 to absorb glyoxal.

724 Concerning oxidation, leading to organic acid formation, the dust surface could
725 contribute directly to this mechanism by providing redox reactive sites (Shen et al.,
726 2016). Redox reactions ~~derive into~~promoted the formation of ~~oxidized species such as~~
727 glycolic, glyoxylic, and oxalic acids. These compounds not only represent aging
728 products but also participate in further condensation reactions, expanding the chemical
729 diversity of SOA. In particular, we detected the presence of oligomer V, which we
730 propose results not from direct glyoxal self-reactions but rather from the condensation
731 of two glycolic acid molecules. This suggests a broader heterogeneous transformation
732 process occurring on the dust surface, possibly involving oxidative pathways. Such
733 cross-reactions are consistent with mechanisms proposed in glyoxal–methylglyoxal
734 systems (Zhang et al., 2022), where mixed oligomers form through shared reaction
735 intermediates.

736 A key aspect of our findings is the irreversibility of glyoxal uptake on mineral dust. In
737 contrast to bulk aqueous systems, where glyoxal oligomerization can exist in
738 equilibrium with monomeric species, our results suggest that oligomer formation on
739 mineral particles is effectively irreversible. The persistence of high-molecular-weight
740 products following drying and thermal analysis confirms that these species are
741 chemically stable and remain in the condensed phase. This behaviour is consistent

742 with previous observations (Hastings et al., 2005; Ortiz-Montalvo et al., 2014), and

743 highlights the significance of particle-phase reactions in the formation of SOA.

744 The acidity of the particle surface could potentially play a role in catalysing glyoxal

745 oligomerization. It is plausible that localized acidity arises or is enhanced by the

746 accumulation of transformation products such as glycolic and oxalic acids, which were

747 detected in this study. This in-situ acidification may facilitate acid-catalysed reaction

748 pathways even under neutral or weakly acidic conditions. The importance of acidity in

749 glyoxal chemistry is supported by mechanistic proposals involving carbenium ion

750 intermediates or hydrogen-bond-assisted nucleophilic mechanisms, which can operate

751 effectively without requiring strongly acidic environments (Zhang et al., 2022; Gomez

752 et al., 2015). These pathways are known to promote the formation of low-volatility

753 oligomers, contributing to organic aerosol mass. Similarly, Ortiz-Montalvo et al. (2014)

754 demonstrated that even mildly acidic aerosols significantly enhance glyoxal

755 oligomerization and SOA formation.

756 Another critical factor is the presence of adsorbed water on mineral dust particles. Our

757 findings demonstrate, in fact, that the availability of surface water facilitates organic

758 aerosol formation from glyoxal. This agrees with chamber and field studies (Hastings

759 et al., 2005; Shen et al., 2016) that identified a RH threshold above which glyoxal

760 uptake dramatically increases. The presence of a thin aqueous film on the particle

761 surface creates a reactive medium where glyoxal can concentrate, hydrate, and

762 polymerize efficiently. Even after evaporation, the products formed are retained in the

763 particle phase, evidencing their low volatility and chemical resilience.

764 **4.3. Implications for the dust properties**

765 The irreversible transformation of the chemical composition of mineral dust following

766 the uptake of glyoxal could influence the physico-chemical properties of mineral dust

767 particles. The formation of organic acids (glycolic, glyoxylic and oxalic acids) should

768 change the dust pH, increasing its acidity and ability to dissolve transition metals like

769 iron and copper. This could alter their speciation, enhancing their bioavailability in

770 aqueous aerosol environments and potentially impacting atmospheric chemistry and

771 the reactivity of aerosol particles (Giorio et al., 2022). Changes in dust pH could also

772 affect its hygroscopic properties, influencing itstheir ability to adsorb water and grow in

773 size. This is consistent with our observation regarding the volume increase of dust

774 particles after the uptake and the subsequent growth that enhances particles

interaction with light and cloud droplets formation. Collectively, these processes highlight how aerosol acidity modulates not only chemical transformations but also key physical properties and atmospheric lifetimes of dust-glyoxal systems.

The newly formed organic matter from glyoxal on dust particles could also alter the aerosol's optical properties, affecting its ability to absorb solar radiation, as recently observed in aqueous solution (De Haan et al., 2023) or on ammonium sulphate aerosols (De Haan et al. 2020; Trainic et al. 2011). The presence of hydrated glyoxal oligomeric structures has already been observed to have UV radiation absorption properties (Kalberer et al., 2004; Shapiro et al., 2009). Glyoxal-derived oligomers could significantly influence the optical properties of mineral dust aerosols. Spectroscopic analyses revealed that these oligomers have broadened OH and C–O bands and enhanced Raman activity, indicative of their hydrated and cyclic structures (Avzianova and Brooks, 2013). They also have strong UV-visible absorption, particularly between 300–600 nm, with characteristic shoulders at ~280 and 345 nm attributed to acetal and aldol condensation products (Schwier et al., 2010; Shapiro et al., 2009). High RH conditions (> 80%) facilitate brown carbon formation, evidenced by the further appearance of UV-visible absorbance peaks and Raman background signals (De Haan et al., 2020; Zhang et al., 2022).

5. Conclusions

This paper presented a novel investigation of the interaction between gas-phase glyoxal and mineral dust. By taking advantage of the capabilities of the CESAM atmospheric simulation chamber to perform multiphase experiments on time scales relevant to atmospheric processes and dispersion, including aerosols, our experiments considered airborne submicron mineral dust particles generated from a natural soil (Gobi Desert) in realistic concentrations, composition and size distribution. While airborne, the dust aerosol was aged under variable conditions of RH, irradiation, and ozone concentrations. ~~Thanks to the realism of the tools and the experimental conditions, O~~our study investigates, for the first time, both the rate of uptake of glyoxal and the rate of formation of the organic aerosol from the gas-phase uptake, while providing ~~. We provide~~ the chemical composition of the ~~particle~~ organic matter formed on the dust particles, and ~~their~~ its implications on the particle microphysics. ~~, in atmospheric relevant conditions.~~

This study used a single and instantaneous injection of glyoxal, and not a constant steady state flux. Above 30% RH, upon injection, glyoxal is partitioned almost instantaneously between the gas phase, the chamber walls and the dust particles. This is an advantage to scale our results to ambient conditions. Indeed, Volkamer et al. (2005) estimated that the daytime lifetime of glyoxal is around 1.3 h. Alvarado et al. (2020) showed that the long-range transport of glyoxal produced from a point source (e.g., Canadian wildfires) may be possible only by invoking the progressive oxidation of its longer-lived precursors in the plume. In the scenario where dust aerosols interact with a glyoxal plume from a point source, one can expect an interaction time of a few minutes, compatible with that of this study. Furthermore, because the uptake follows a first-order kinetics, the measured uptake coefficient ($\gamma = (9 \pm 5) \times 10^{-3}$ at 80% RH) is independent of the glyoxal concentration and transferable to atmospheric conditions.

The uptake of glyoxal and the formation of organic matter start as soon as the glyoxal is injected into the chamber and last approximately 20 minutes. After this time, the dust surface seems saturated, likely because of the excessive glyoxal concentrations injected in the chamber. On the other hand, the uptake coefficient measured by the loss of gas-phase glyoxal molecules agrees very well with the rate of formation of the particulate organic mass on the dust, suggesting that the totality of the mass of reacting glyoxal is condensed on the dust particles. While some of the organic matter is lost again from the dust particles due to evaporation, oligomers and organic acids are detected on the dust even after the uptake has finished, indicating that the uptake of glyoxal modifies irreversibly both the composition and the physical properties of mineral dust.

This study reveals a significant quantitative transfer of gas-phase glyoxal molecules to mineral dust aerosol surfaces, occurring within a timescale of a few minutes, underscoring the important role of dust-glyoxal interactions in the atmosphere. Our results extend the current understanding of glyoxal chemistry in atmospheric systems. While oligomerization occurs through well-established aqueous-phase pathways, we suggest that mineral dust particles facilitate, in a unique way, higher-order oligomer formation, support irreversible uptake, and possibly promote alternative reaction channels such as glycolic acid condensation. These findings underscore the importance of mineral dust surfaces as active sites for multiphase SOA formation, with

implications for understanding aerosol growth, composition, and climate-relevant properties. This is also in agreement with the field observations conducted by Wang et al. (2015). Neglecting the uptake pathway on dust could result in an underestimation of glyoxal removal from the atmosphere, potentially leading to disparities between model predictions and observed gaseous concentrations of glyoxal (Kluge et al., 2023; Ling et al., 2020; Volkamer et al., 2007; Washenfelder et al., 2011). The findings of this study also have~~have also~~ important implications for the aerosol direct and indirect radiative effect and aerosol pH. The acidification and the oxidation of the dust aerosols by glyoxal should increase their hygroscopicity, especially under high RH conditions, affecting particle growth dynamics, phase behaviour, and cloud condensation nuclei (CCN) potential (Song and Osada, 2021). Further data analysis ~~are~~ is ongoing to address these aspects.

Data availability. The simulation chamber experiments that support the findings of this study are available through the Database of Atmospheric Simulation Chamber Studies (DASCS) of the EUROCHAMP Data Centre (https:// data.eurochamp.org/ data- access/ chamber- experiments/).

Code availability. The routine used for fitting the size distribution is available at https://doi.org/10.5281/zenodo.8135133 (Baldo and Lu, 2023). Note that in this study, we only used the size distribution measured by the OPC instrument, which was fitted with a lognormal function. The Tof-ACSM data processing (including mass calibration, peaks integration and air beam correction of ion intensities) was conducted with Tofware version 3_2_40209, the ACSM data analysis package for the software Igor Pro 7.08 (Wavemetrics, Inc., Portland, OR, USA). SFE-GC-MS data analysis was conducted using the proprietary software (TurboMass Version 6.1.0.1965 PerkinElmer®).

Author contributions. PF, JFD and FB conceptualized the study. PF and FB led the paper writing, with contributions from all the authors. JFD provided with expertise on multi-phase chemistry. CB analysed the aerosol size distribution data. VM supervised the analysis of PTR-MS data. FB, CG and DLP performed the ESI-Orbitrap analysis of filter samples. FB and JFB performed the analysis of ACSM observations. FB, TB and AG performed the SFE/CG-MS analysis of filter samples. FB, GN and SC performed the thermo-optical analysis of filter samples. FB, MC, AB, EP, VM, BPV and PF conducted the chamber experiments. MR provided with the soil sample and expertise on heterogeneous chemistry. PF provided with funding.

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