ITINERIS

Particulate matter in the atmosphere: size distribution and chemical composition

Mario Commodo – mario.commodo@stems.cnr.it
081 7682256

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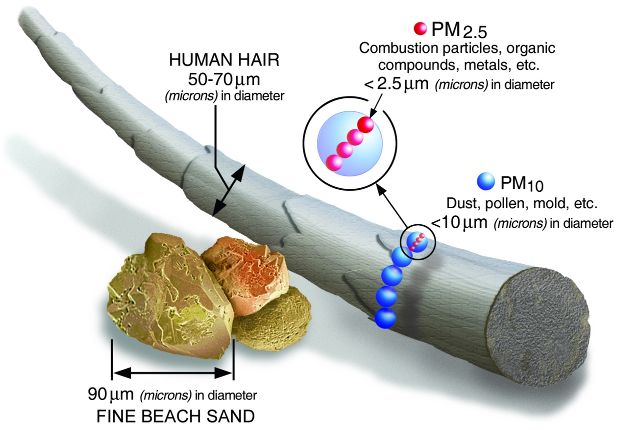
Ministero dell'Università e della Ricerca

Italiadomani

Particulate Matter Size Distribution

The atmosphere is a mixture of gaseous and aerosols components.
Particulate matter (PM) indicates **solid and/or liquid particles suspended in the air** (excluding rain droplets or ice crystals) with diameters ranging from few nm up to 100 µm

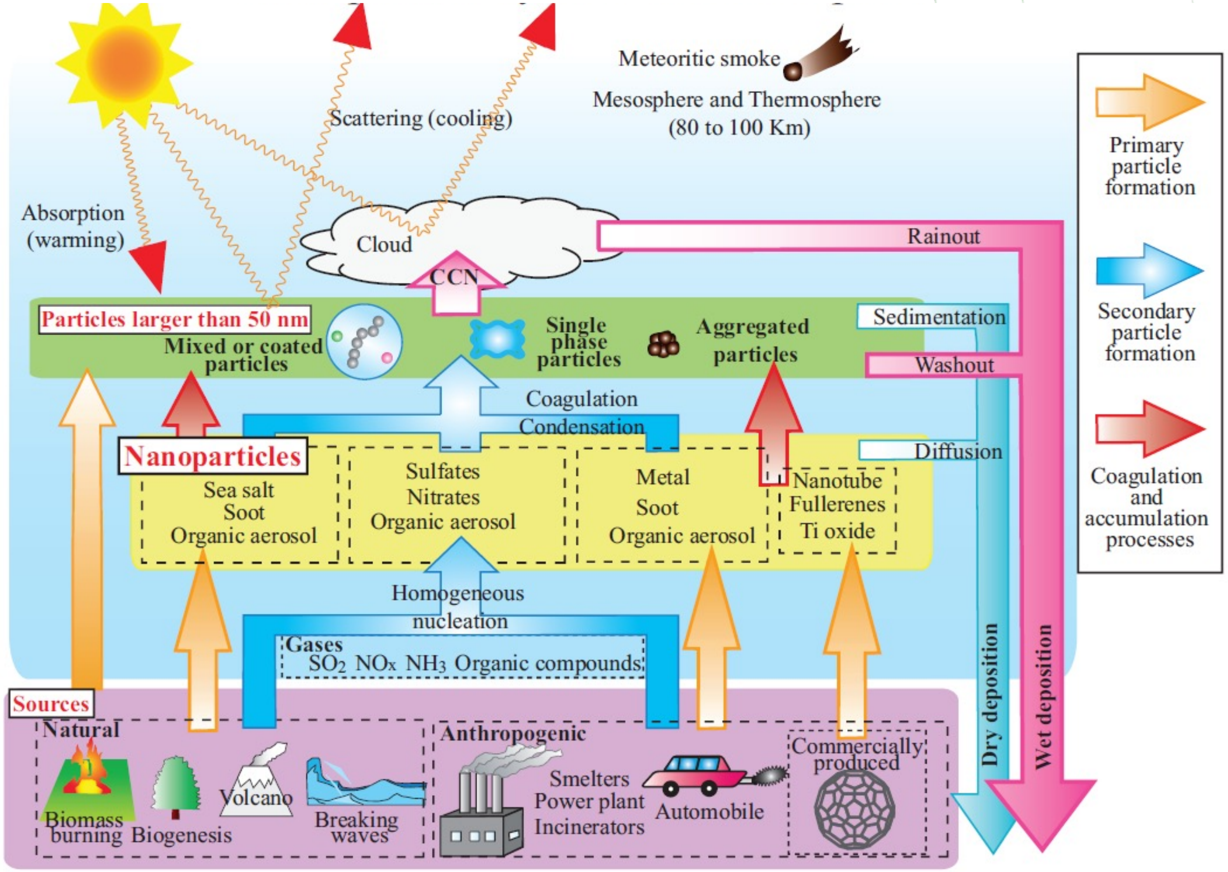
Any aerosol (including the atmospheric aerosol) can be classified according to **origin, size, source and chemical composition** of the particles



K.H. Kim et al. Environ. Int. 74 (2015)
R. Guaita et al. Int. J. Environ. Heal. R 21 (2011)

E. Mészáros. Fundamentals of Atmospheric Aerosol Chemistry (1999)

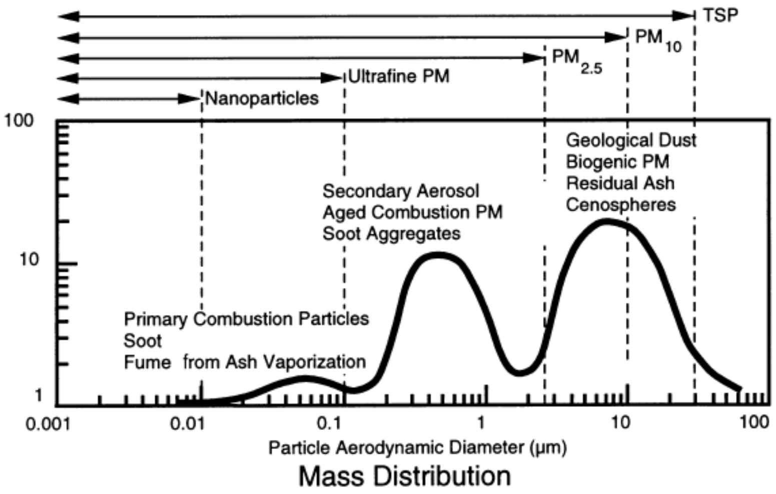
Atmospheric Aerosol Size Distribution

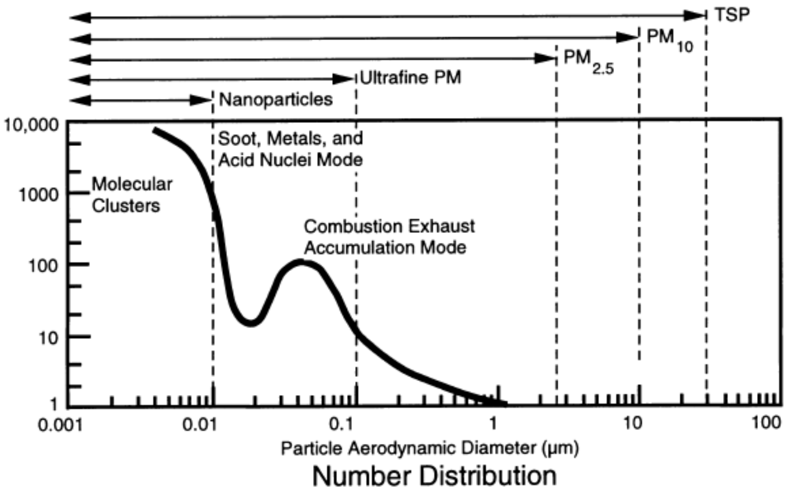


P. R. Buseck and K. Adachi *Elements* **4** (2008)

Atmospheric Aerosol Size Distribution

Typical PM mass and number size distributions
showing the **multimodal nature of atmospheric aerosol**





J.H. Seinfeld and S.N. Pandis. Atmospheric Chemistry and Physics – From Air Pollution to Climate Change (1998)
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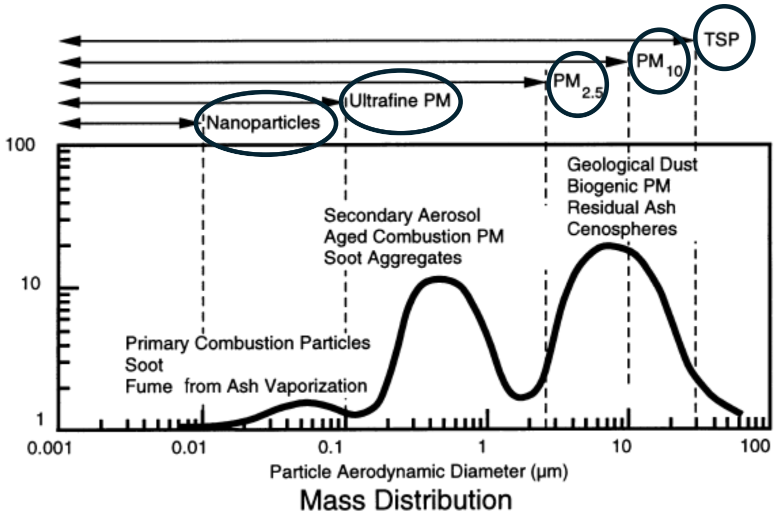

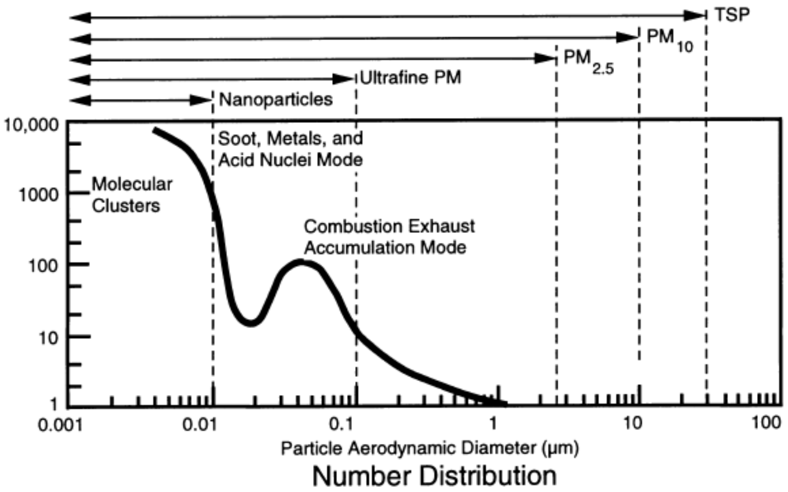

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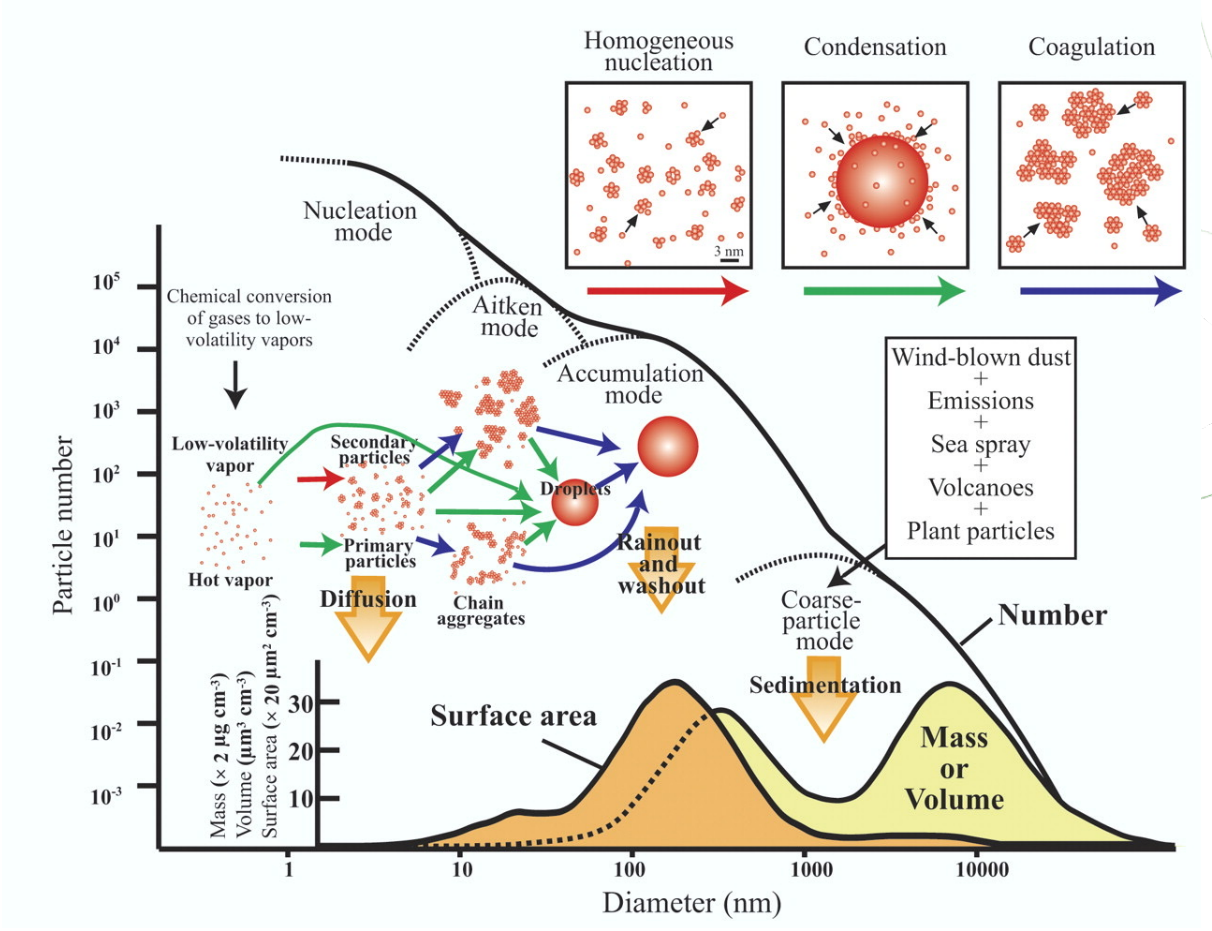
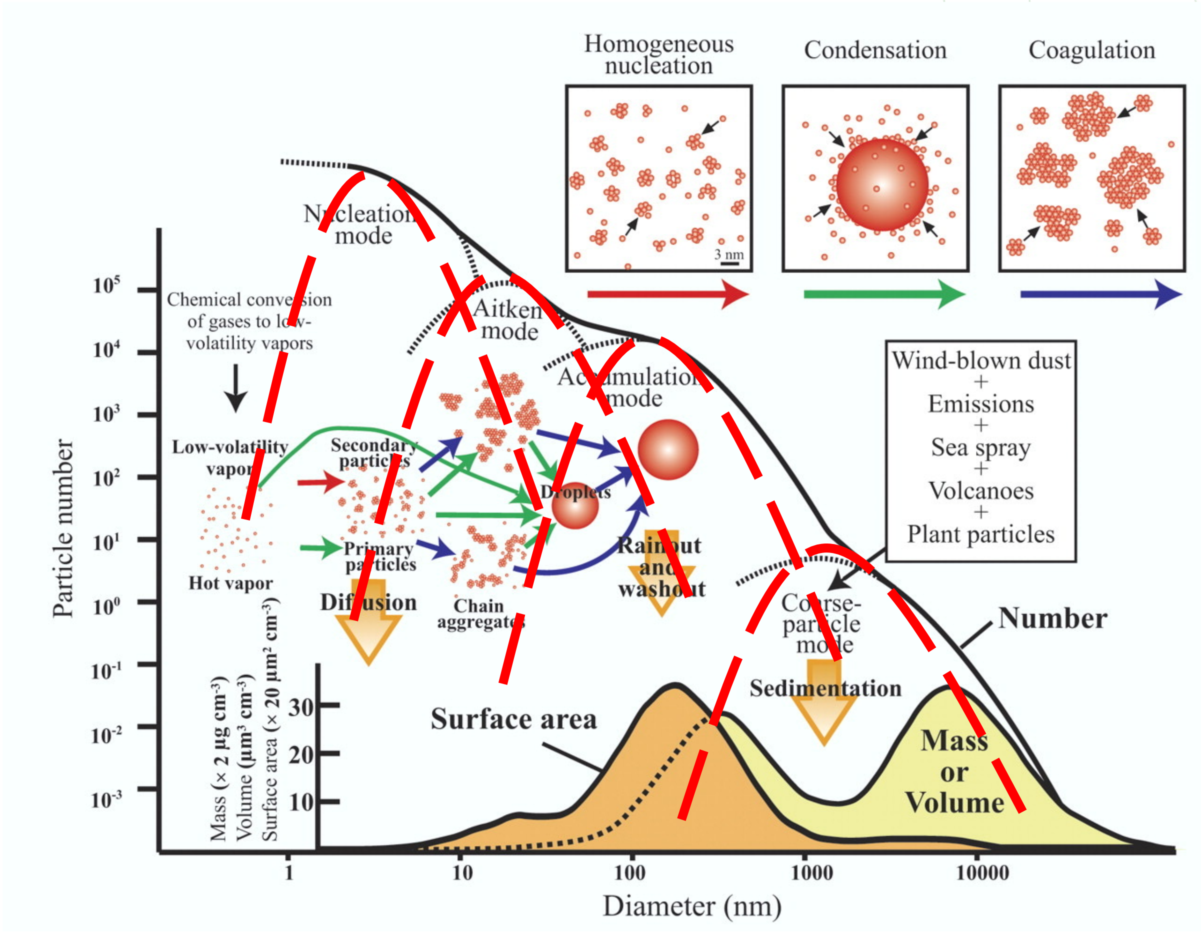


P. R. Buseck and K. Adachi *Elements* **4** (2008)

Atmospheric Aerosol Size Distribution



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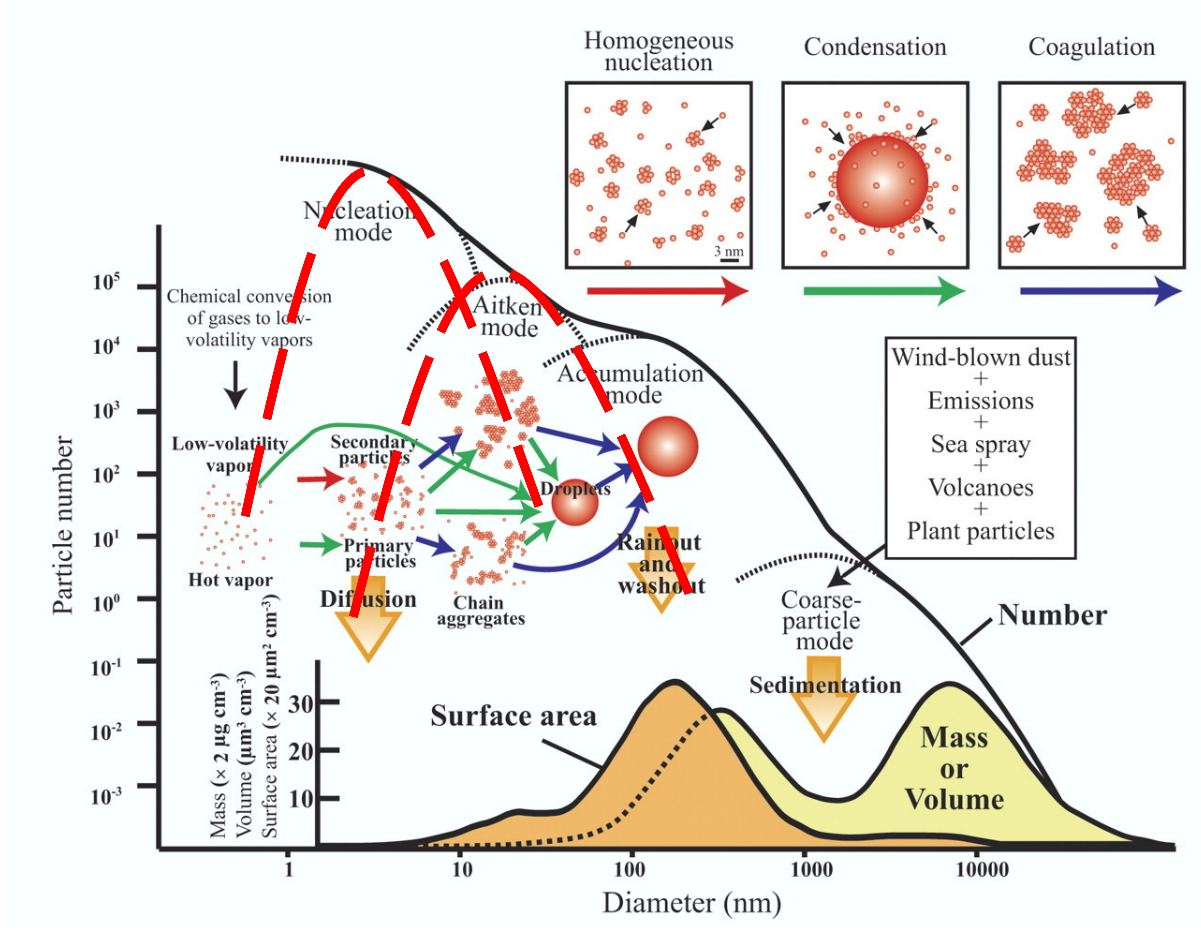

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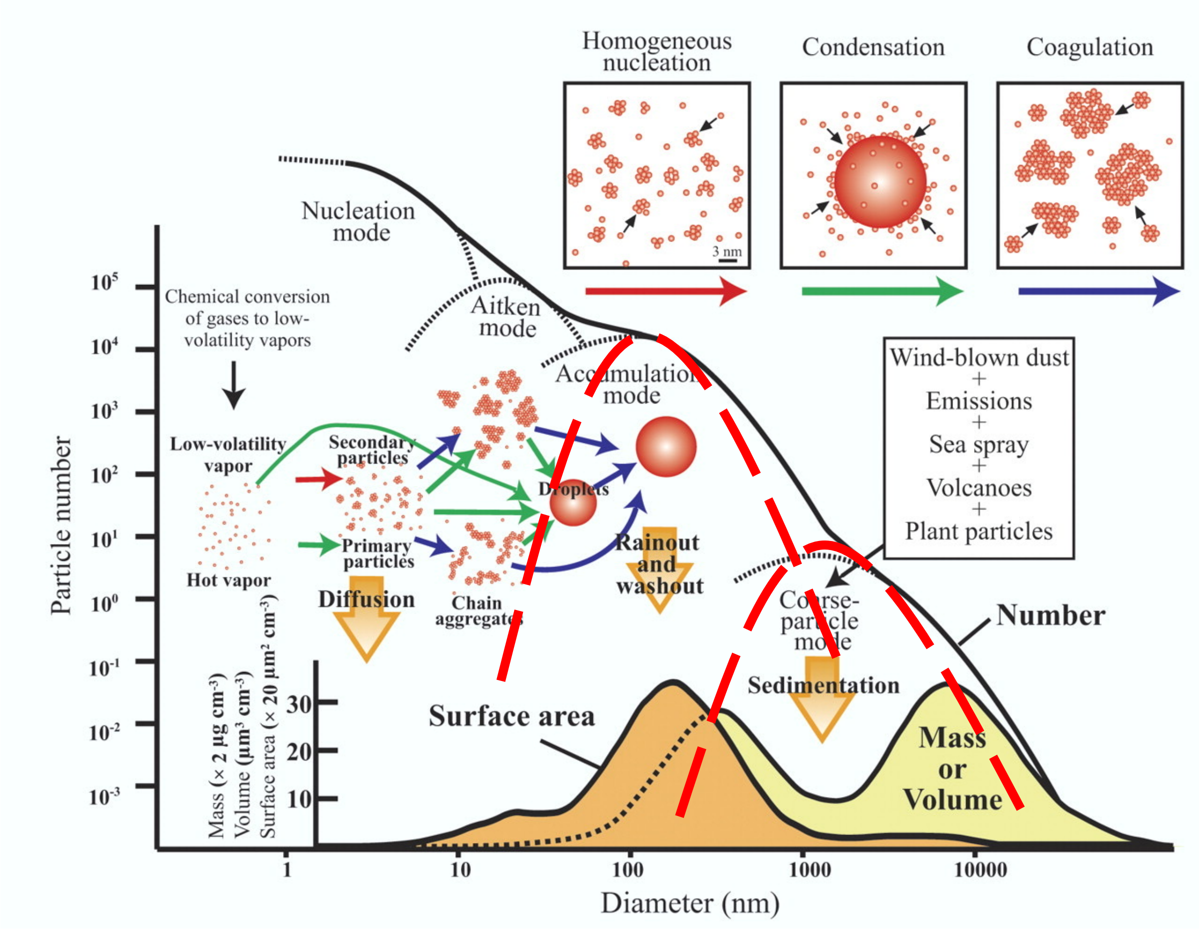

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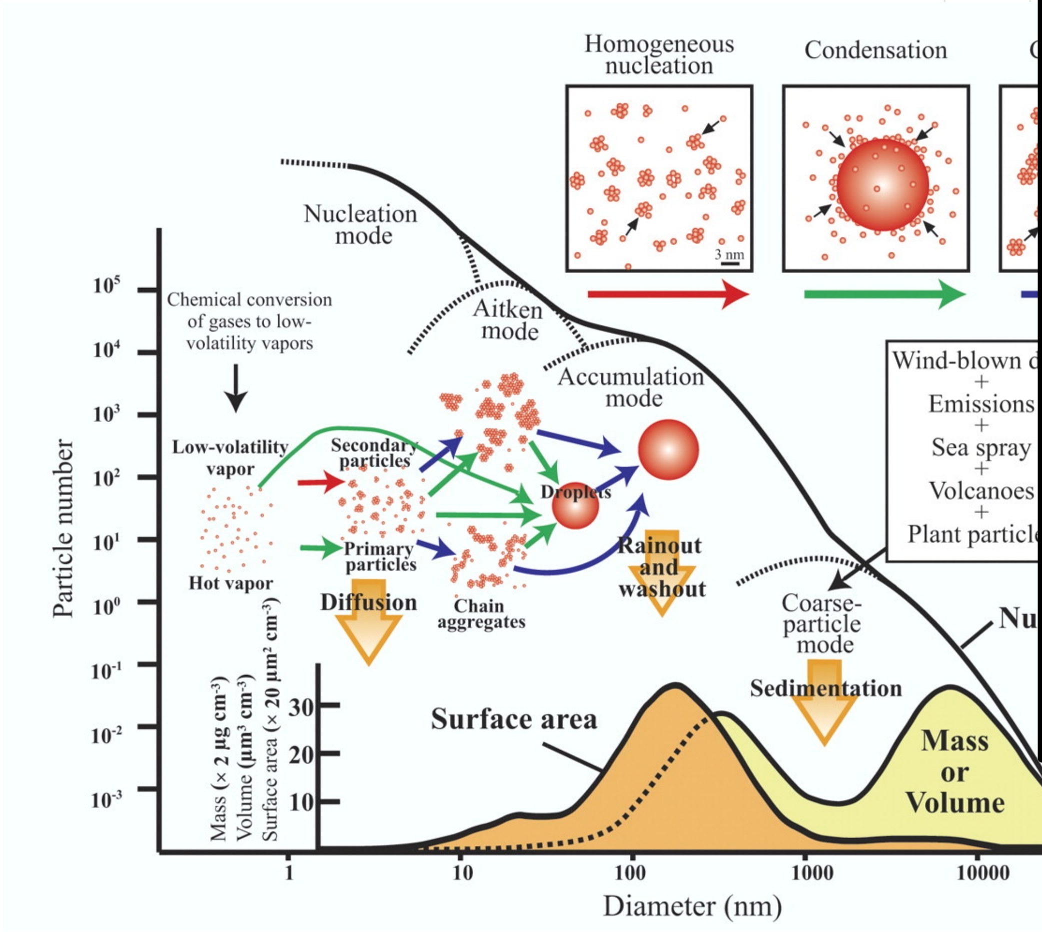
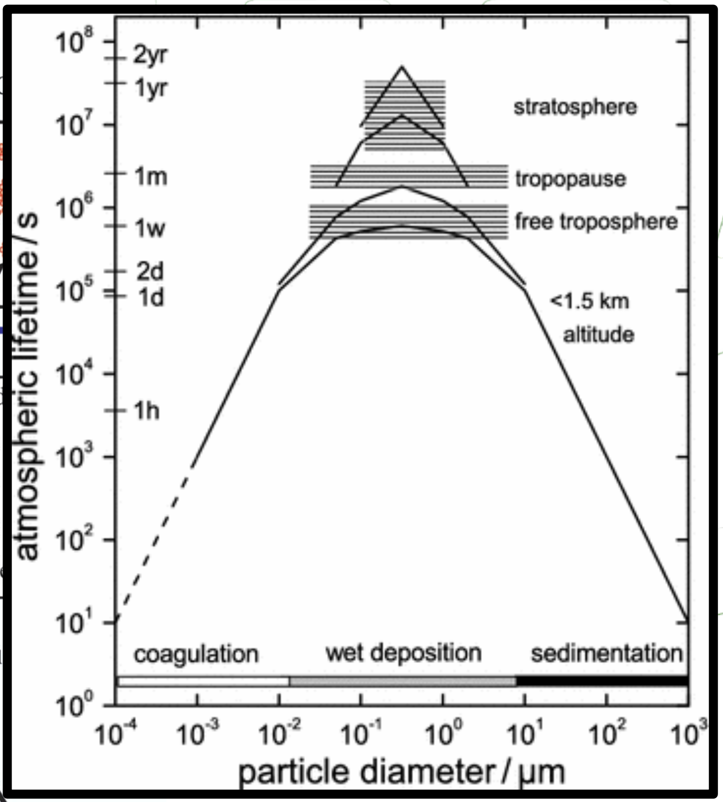
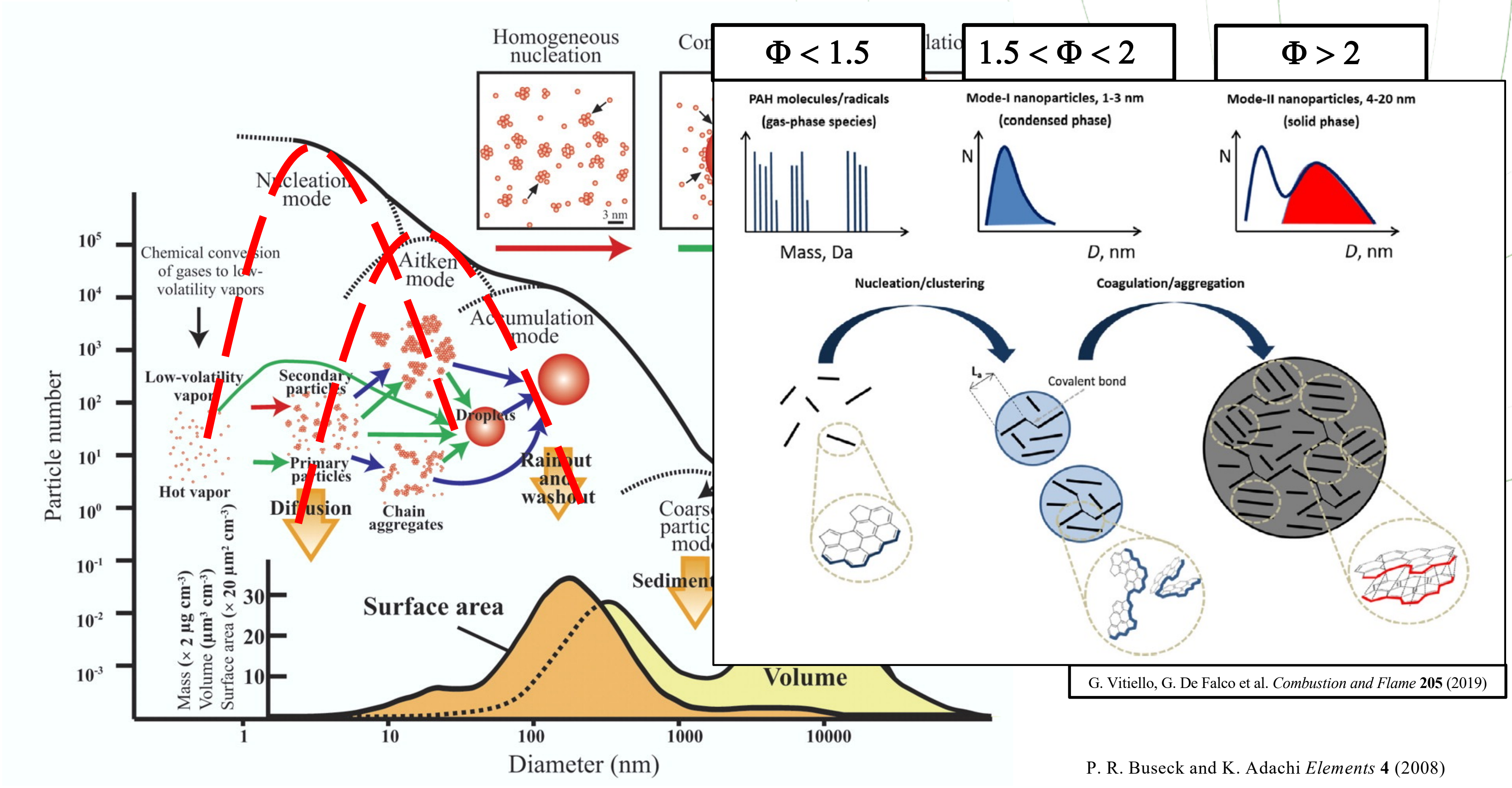




A. Petzold and B. Kärcher. Aerosols in the Atmosphere (2012)

P. R. Buseck and K. Adachi *Elements* **4** (2008)

Atmospheric Aerosol Size Distribution



G. Vitiello, G. De Falco et al. *Combustion and Flame* **205** (2019)

P. R. Buseck and K. Adachi *Elements* **4** (2008)

Atmospheric particle nucleation: an example

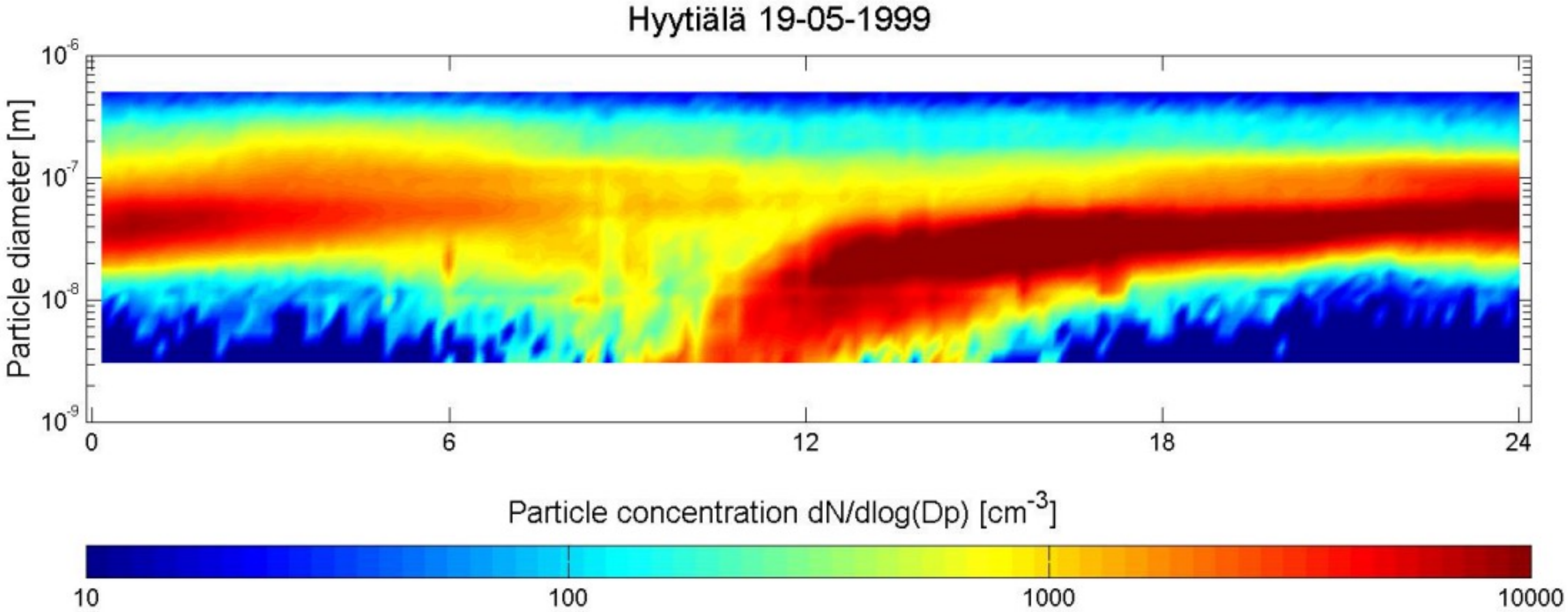
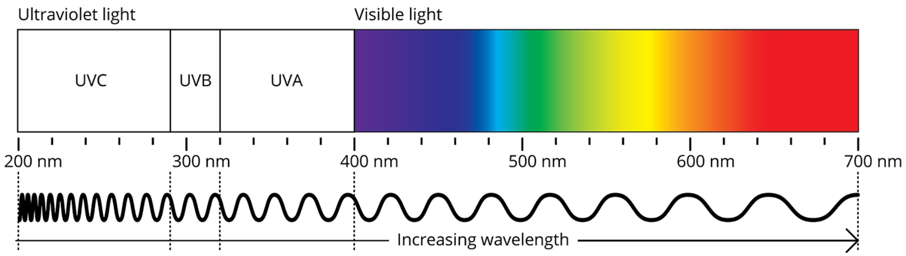


Fig. 1. A typical particle nucleation event is detected when plotting the particle concentration as a function of time and particle size for measurement observed in a boreal forest at Hyytiälä, Finland, for 19 May 1999. Between 9:00 and 12:30 numerous freshly nucleated particles appear at the smallest measurable sizes (>3 nm) and grow within hours to sizes of around 50 nm. From [16].

M. Boy and M. Kulmala Atmos. Chem. Phys. 2 (2002)

Atmospheric particle nucleation: an example



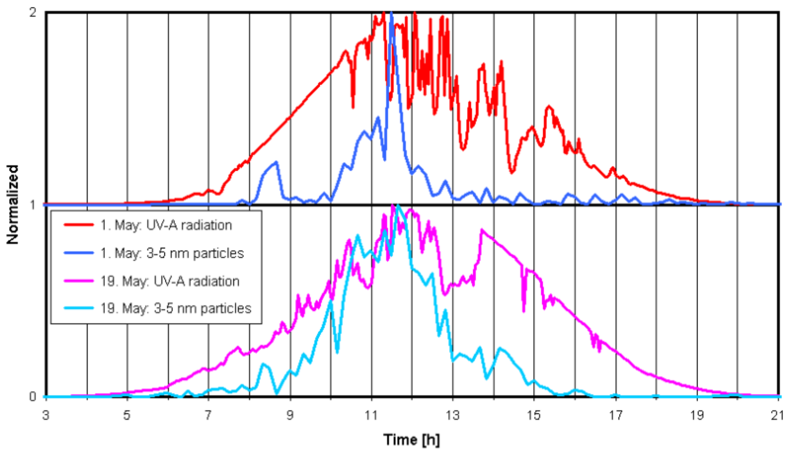


Fig. 4. Normalised values of UV-A solar radiation (3 minutes time interval) and the concentration of 3–5 nm particles (10 minutes time interval).

M. Boy and M. Kulmala Atmos. Chem. Phys. 2 (2002)

Binary Homogenous Nucleation: an example

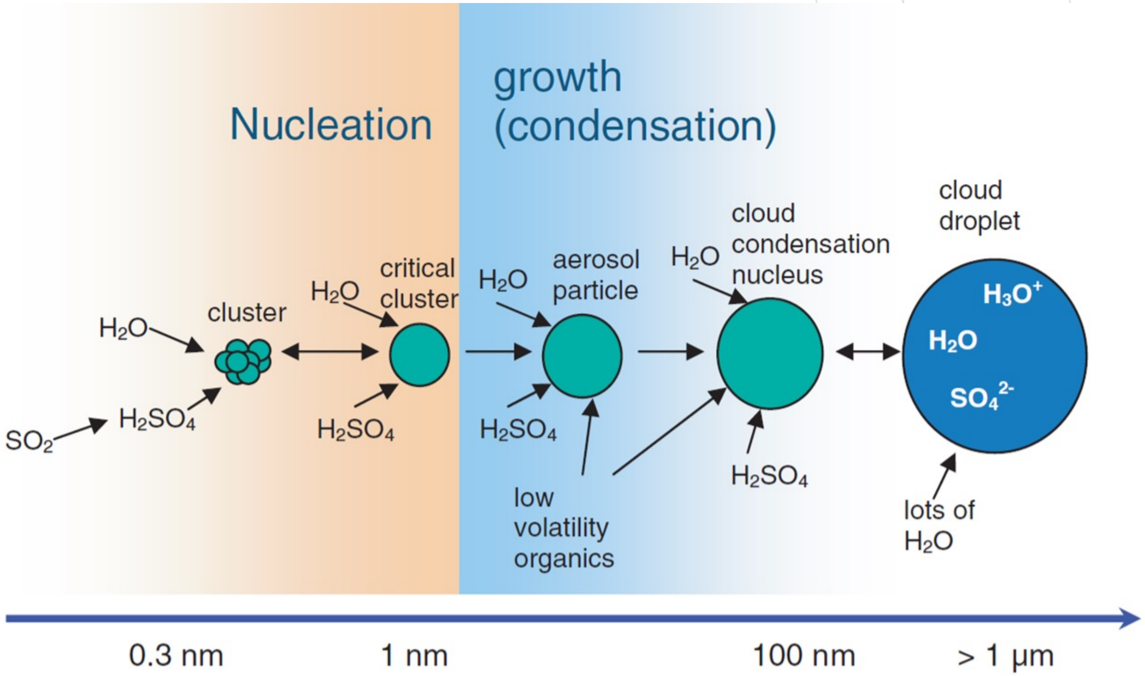
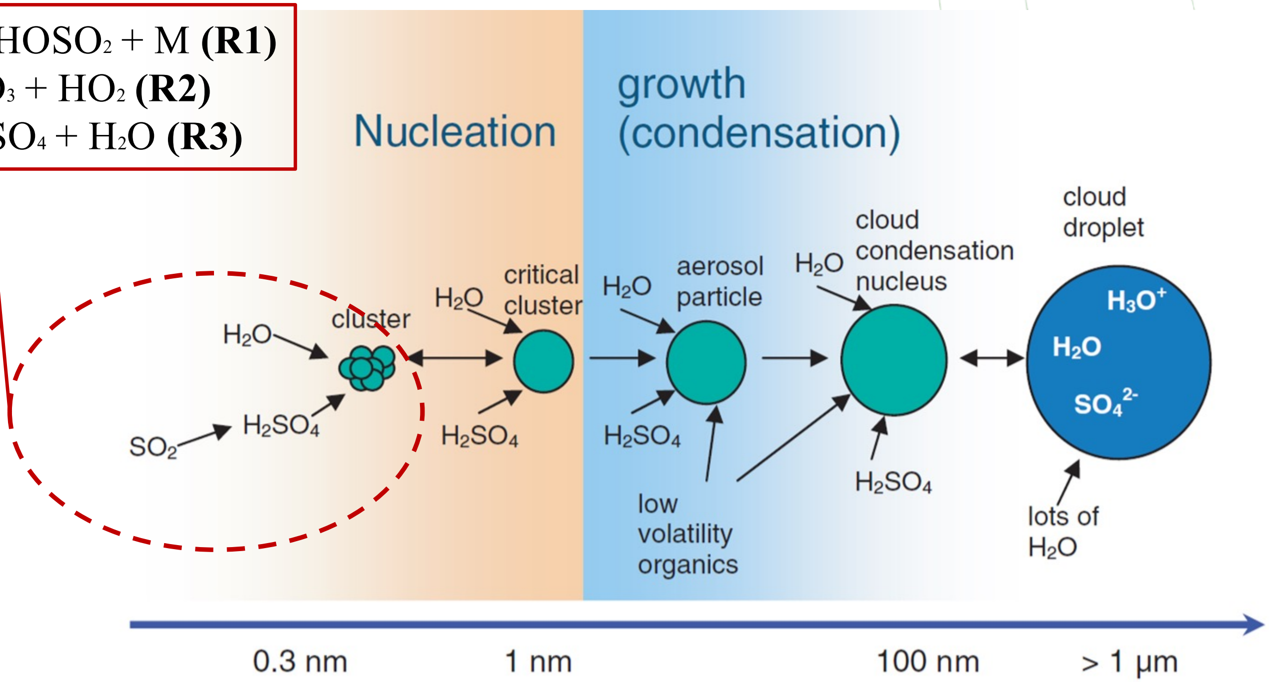


Fig. 2. Schematic representation of the nucleation and subsequent growth process for atmospheric binary homogeneous nucleation of H_2SO_4 and H_2O. Once thermodynamically stable clusters larger than the so-called critical cluster have formed, also other substances such as low-volatility organics can take part in the growth process. Particles may grow further to sizes large enough to act as cloud condensation nuclei on which cloud droplets may form eventually. From [14].

Binary Homogenous Nucleation: an example

$SO_2 + OH + M \rightarrow HOSO_2 + M$ **(R1)**
$HOSO_2 + O_2 \rightarrow SO_3 + HO_2$ **(R2)**
$SO_3 + 2H_2O \rightarrow H_2SO_4 + H_2O$ **(R3)**



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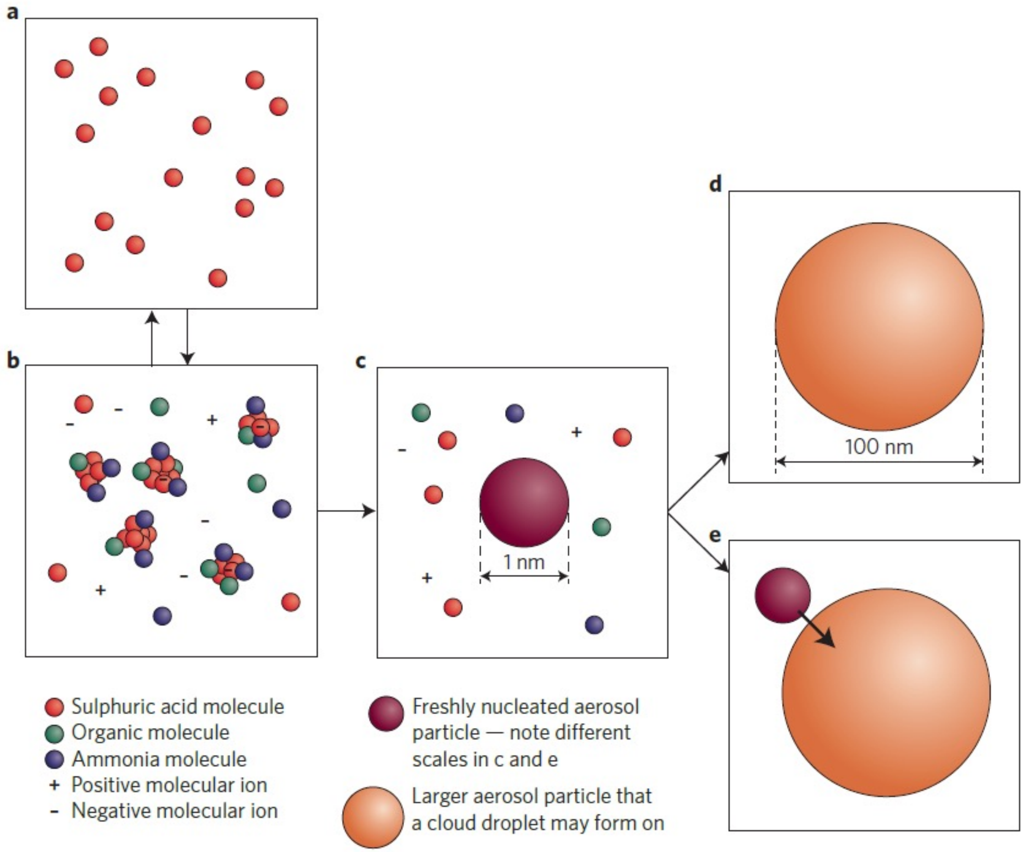
Ternary Homogenous Nucleation: an example

Theory suggests that nucleation of H_2SO_4 aerosols, even with the help of water vapor, should not occur under conditions near the Earth's surface.
NH_3 and organic molecules can pair up with H_2SO_4 early in the nucleation process, preventing the small clusters of molecules from evaporating and promoting the subsequent nucleation

J. Kirkby et al. Nature 476 (2011)



J. Pierce. Nature Geoscience 4 (2011)

Atmospheric Aerosol

Primary Aerosols
when emitted directly by sources into the atmosphere

Elemental Carbon EC or Black Carbon BC
Organic Carbon OC
Inorganic Aerosol

Secondary Aerosols
when formed in the atmosphere by physicochemical processes from precursor gases or preexisting aerosols.

Secondary Organic Aerosols SOA
Secondary Inorganic Aerosols SIA

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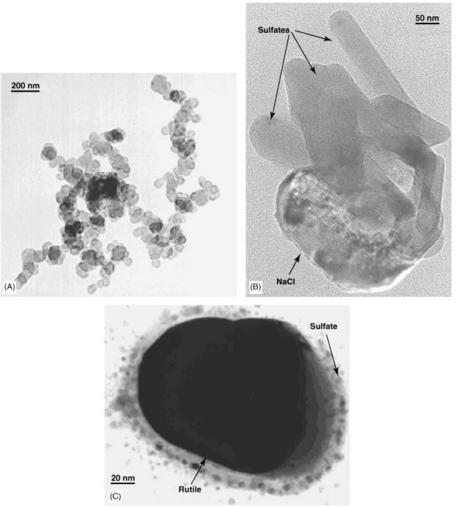

Photographs of atmospheric particles obtained by Transmission Electron Microscopy TEM:

(A) Chain agglomerate soot particles
(B) Sulfate attached to a sodium chloride particle
(C) Mineral dust particle coated with sulfate

P..H. McMurry, AEROSOLS | Observations and Measurements In Encyclopedia of Atmospheric Sciences (2003)

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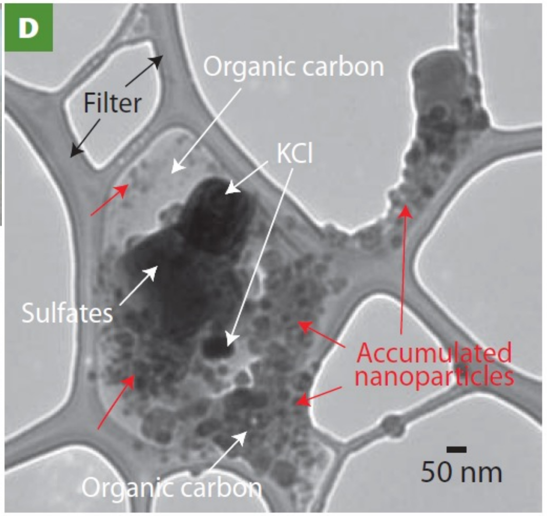

High-magnification TEM image of biomass-burning particles collected on a filter of lacey carbon

Secondary aerosols may present an internal mixing state between SOA and SIA, such as a shell of SOA around a core of ammonium sulfate

Buseck, P. R., and Adachi, K. Elements 4 (2008)

Atmospheric Aerosol

Primary Aerosols

when emitted directly by sources into the atmosphere

main NATURAL sources:

- Mineral dust (soil, rock debris,…)
- Sea spray
- Biogenic aerosols (plant debris, microbial particles,…)
- Forest fire smoke (i.e., biomass burning)
- Volcanic emissions

C. Tomasi and A. Lupi. Primary and Secondary Sources of Atmospheric Aerosol (2016)

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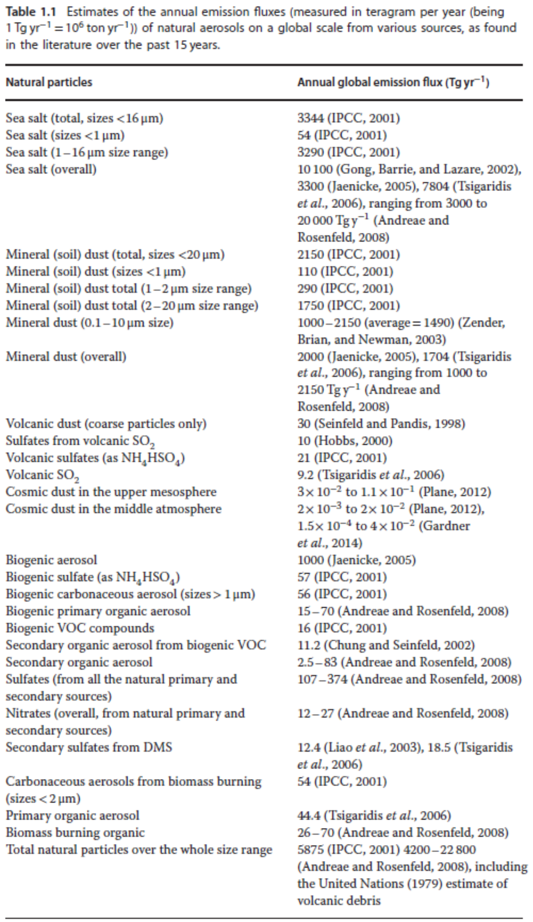

Table 1.1 Estimates of the annual emission fluxes (measured in teragram per year (being $1\ \mathrm{Tg\,yr^{-1}} = 10^6\ \mathrm{ton\,yr^{-1}}$)) of natural aerosols on a global scale from various sources, as found in the literature over the past 15 years.

Natural particles	Annual global emission flux ($\mathrm{Tg\,yr^{-1}}$)
Sea salt (total, sizes <16 µm)	3344 (IPCC, 2001)
Sea salt (sizes <1 µm)	54 (IPCC, 2001)
Sea salt (1–16 µm size range)	3290 (IPCC, 2001)
Sea salt (overall)	10 100 (Gong, Barrie, and Lazare, 2002), 3300 (Jaenicke, 2005), 7804 (Tsigaridis *et al.*, 2006), ranging from 3000 to 20 000 $\mathrm{Tg\,y^{-1}}$ (Andreae and Rosenfeld, 2008)
Mineral (soil) dust (total, sizes <20 µm)	2150 (IPCC, 2001)
Mineral (soil) dust (sizes <1 µm)	110 (IPCC, 2001)
Mineral (soil) dust total (1–2 µm size range)	290 (IPCC, 2001)
Mineral (soil) dust total (2–20 µm size range)	1750 (IPCC, 2001)
Mineral dust (0.1–10 µm size)	1000–2150 (average = 1490) (Zender, Brian, and Newman, 2003)
Mineral dust (overall)	2000 (Jaenicke, 2005), 1704 (Tsigaridis *et al.*, 2006), ranging from 1000 to 2150 $\mathrm{Tg\,y^{-1}}$ (Andreae and Rosenfeld, 2008)
Volcanic dust (coarse particles only)	30 (Seinfeld and Pandis, 1998)
Sulfates from volcanic SO_2	10 (Hobbs, 2000)
Volcanic sulfates (as NH_4HSO_4)	21 (IPCC, 2001)
Volcanic SO_2	9.2 (Tsigaridis *et al.*, 2006)
Cosmic dust in the upper mesosphere	3×10^{-2} to 1.1×10^{-1} (Plane, 2012)
Cosmic dust in the middle atmosphere	2×10^{-3} to 2×10^{-2} (Plane, 2012), 1.5×10^{-4} to 4×10^{-2} (Gardner *et al.*, 2014)
Biogenic aerosol	1000 (Jaenicke, 2005)
Biogenic sulfate (as NH_4HSO_4)	57 (IPCC, 2001)
Biogenic carbonaceous aerosol (sizes > 1 µm)	56 (IPCC, 2001)
Biogenic primary organic aerosol	15–70 (Andreae and Rosenfeld, 2008)
Biogenic VOC compounds	16 (IPCC, 2001)
Secondary organic aerosol from biogenic VOC	11.2 (Chung and Seinfeld, 2002)
Secondary organic aerosol	2.5–83 (Andreae and Rosenfeld, 2008)
Sulfates (from all the natural primary and secondary sources)	107–374 (Andreae and Rosenfeld, 2008)
Nitrates (overall, from natural primary and secondary sources)	12–27 (Andreae and Rosenfeld, 2008)
Secondary sulfates from DMS	12.4 (Liao *et al.*, 2003), 18.5 (Tsigaridis *et al.*, 2006)
Carbonaceous aerosols from biomass burning (sizes < 2 µm)	54 (IPCC, 2001)
Primary organic aerosol	44.4 (Tsigaridis *et al.*, 2006)
Biomass burning organic	26–70 (Andreae and Rosenfeld, 2008)
Total natural particles over the whole size range	5875 (IPCC, 2001) 4200–22 800 (Andreae and Rosenfeld, 2008), including the United Nations (1979) estimate of volcanic debris

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Atmospheric Aerosol

Primary Aerosols

when emitted directly by sources into the atmosphere

Secondary Aerosols

when formed in the atmosphere by physicochemical processes from precursor gases or preexisting aerosols.

main NATURAL sources:

- Gas-to-particle conversion of sulfur-containing and nitrogen-containing gases from natural emissions

C. Tomasi and A. Lupi. Primary and Secondary Sources of Atmospheric Aerosol (2016)

Atmospheric Aerosol

Primary Aerosols

when emitted directly by sources into the atmosphere

main ANTHROPOGENIC sources:

- Fuel combustion
- Industrial processes
- Fugitive emissions and agriculture
- Road and non-road transportation

Primary anthropogenic aerosols contain **sulfate, ammonium, nitrate, trace metals, carbonaceous material and water**. The carbonaceous fraction of this particulate matter consists of both EC and OC

C. Tomasi and A. Lupi. Primary and Secondary Sources of Atmospheric Aerosol (2016)

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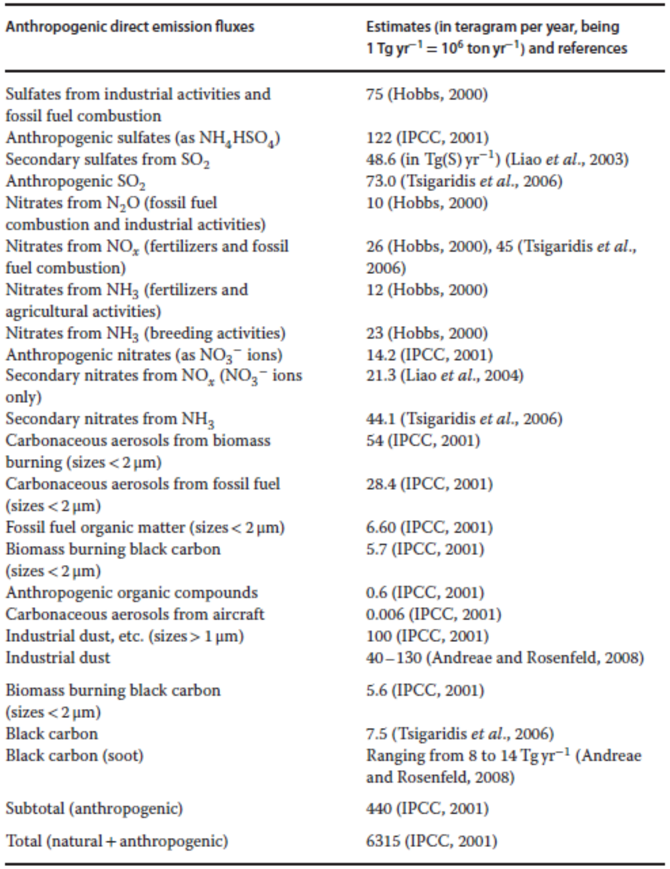


Anthropogenic direct emission fluxes	Estimates (in teragram per year, being 1 Tg $yr^{-1} = 10^6$ ton yr^{-1}) and references
Sulfates from industrial activities and fossil fuel combustion	75 (Hobbs, 2000)
Anthropogenic sulfates (as NH_4HSO_4)	122 (IPCC, 2001)
Secondary sulfates from SO_2	48.6 (in Tg(S) yr^{-1}) (Liao *et al.*, 2003)
Anthropogenic SO_2	73.0 (Tsigaridis *et al.*, 2006)
Nitrates from N_2O (fossil fuel combustion and industrial activities)	10 (Hobbs, 2000)
Nitrates from NO_x (fertilizers and fossil fuel combustion)	26 (Hobbs, 2000), 45 (Tsigaridis *et al.*, 2006)
Nitrates from NH_3 (fertilizers and agricultural activities)	12 (Hobbs, 2000)
Nitrates from NH_3 (breeding activities)	23 (Hobbs, 2000)
Anthropogenic nitrates (as NO_3^- ions)	14.2 (IPCC, 2001)
Secondary nitrates from NO_x (NO_3^- ions only)	21.3 (Liao *et al.*, 2004)
Secondary nitrates from NH_3	44.1 (Tsigaridis *et al.*, 2006)
Carbonaceous aerosols from biomass burning (sizes $< 2\,\mu m$)	54 (IPCC, 2001)
Carbonaceous aerosols from fossil fuel (sizes $< 2\,\mu m$)	28.4 (IPCC, 2001)
Fossil fuel organic matter (sizes $< 2\,\mu m$)	6.60 (IPCC, 2001)
Biomass burning black carbon (sizes $< 2\,\mu m$)	5.7 (IPCC, 2001)
Anthropogenic organic compounds	0.6 (IPCC, 2001)
Carbonaceous aerosols from aircraft	0.006 (IPCC, 2001)
Industrial dust, etc. (sizes $> 1\,\mu m$)	100 (IPCC, 2001)
Industrial dust	40–130 (Andreae and Rosenfeld, 2008)
Biomass burning black carbon (sizes $< 2\,\mu m$)	5.6 (IPCC, 2001)
Black carbon	7.5 (Tsigaridis *et al.*, 2006)
Black carbon (soot)	Ranging from 8 to 14 Tg yr^{-1} (Andreae and Rosenfeld, 2008)
Subtotal (anthropogenic)	440 (IPCC, 2001)
Total (natural + anthropogenic)	6315 (IPCC, 2001)

C. Tomasi and A. Lupi. Primary and Secondary Sources of Atmospheric Aerosol (2016)

Atmospheric Aerosol

Primary Aerosols

when emitted directly by sources into the atmosphere

Secondary Aerosols

when formed in the atmosphere by physicochemical processes from precursor gases or preexisting aerosols.

Some examples

$$SO_2 + OH \rightarrow SO_3 \rightarrow SO_3 + H_2O \rightarrow H_2SO_{4,(v)}$$

$$NO + O_2 + H \rightarrow HNO_{3,(v)}$$

$H_2SO_{4,(v)} + H_2O \rightarrow$ homogeneous bimol. nucleation

$H_2SO_{4,(v)} + H_2O + HNO_{3,(v)} \rightarrow$ homogeneous trimol. nucleation

$H_2SO_{4,(v)} + H_2O + HNO_{3,(v)} \rightarrow$ heterogeneous trimol. nucleation

main ANTHROPOGENIC sources:

- Fuel combustion
- Industrial processes
- Fugitive emissions and agriculture
- Road and non-road transportation

Secondary anthropogenic aerosols contain
secondary particles from SO_2
secondary particles from NO_X
secondary organic carbon SOC

C. Tomasi and A. Lupi. Primary and Secondary Sources of Atmospheric Aerosol (2016)

Atmospheric Aerosol

Primary Aerosols

when emitted directly by sources into the atmosphere

Secondary Aerosols

when formed in the atmosphere by physicochemical processes from precursor gases or preexisting aerosols.

Under atmospheric conditions, VOCs may undergo atmospheric oxidation with O3, OH, etc. to form less-volatile products which undergo further reactions to the condensed phase. Also, absorption and condensation of semi-volatile organic compounds on existing particles can occur.

S.N. Pandis et al. Atmospheric Environment 26 (1992)
J.H. Kroll and J.H. Seinfeld. Atmospheric Environment 42 (2008)

main ANTHROPOGENIC sources:

- Fuel combustion
- Industrial processes
- Fugitive emissions and agriculture
- Road and non-road transportation

Secondary anthropogenic aerosols contain
secondary particles from SO_2
secondary particles from NO_X
secondary organic carbon SOC

C. Tomasi and A. Lupi. Primary and Secondary Sources of Atmospheric Aerosol (2016)

Atmospheric Aerosol

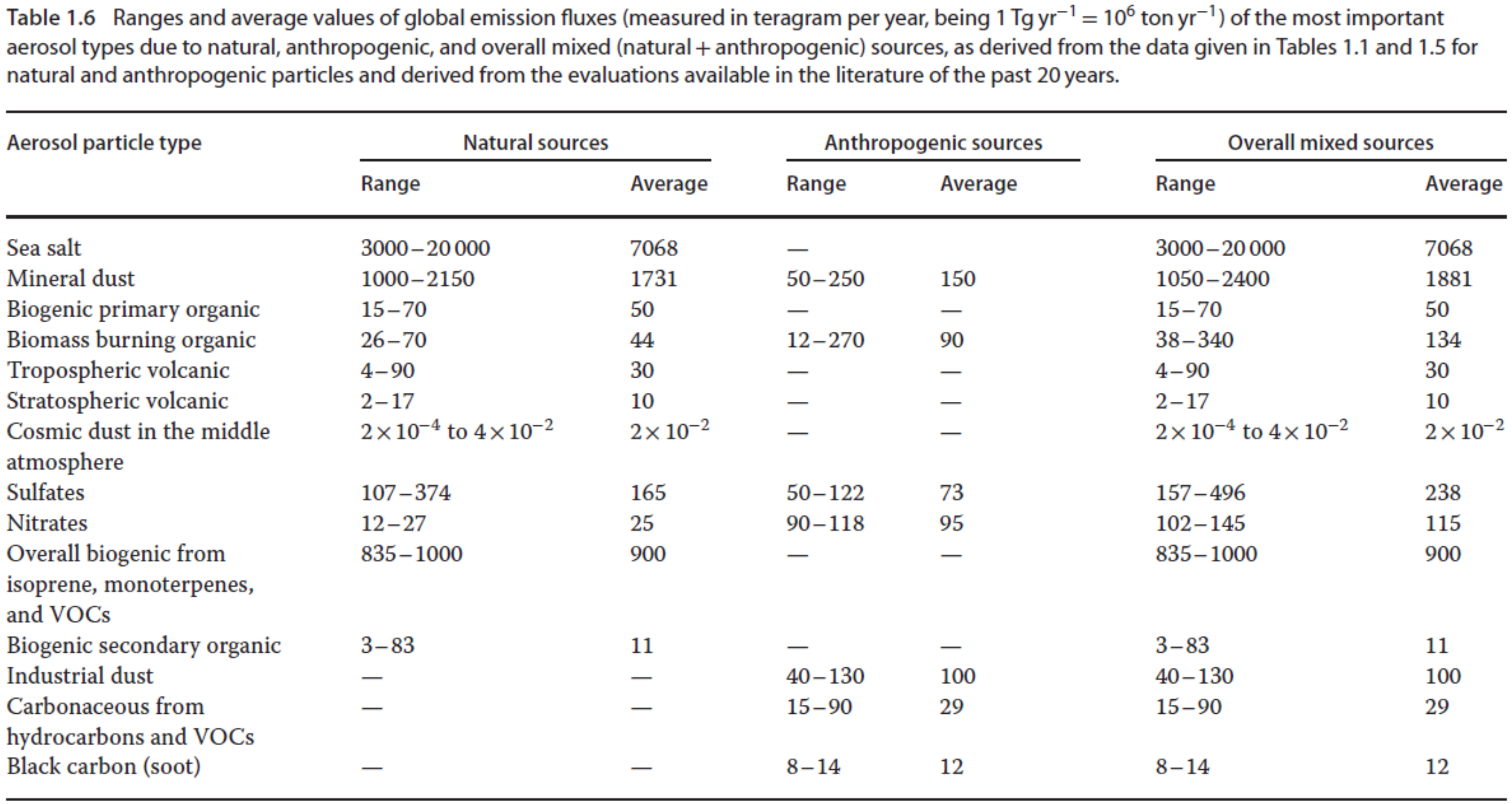


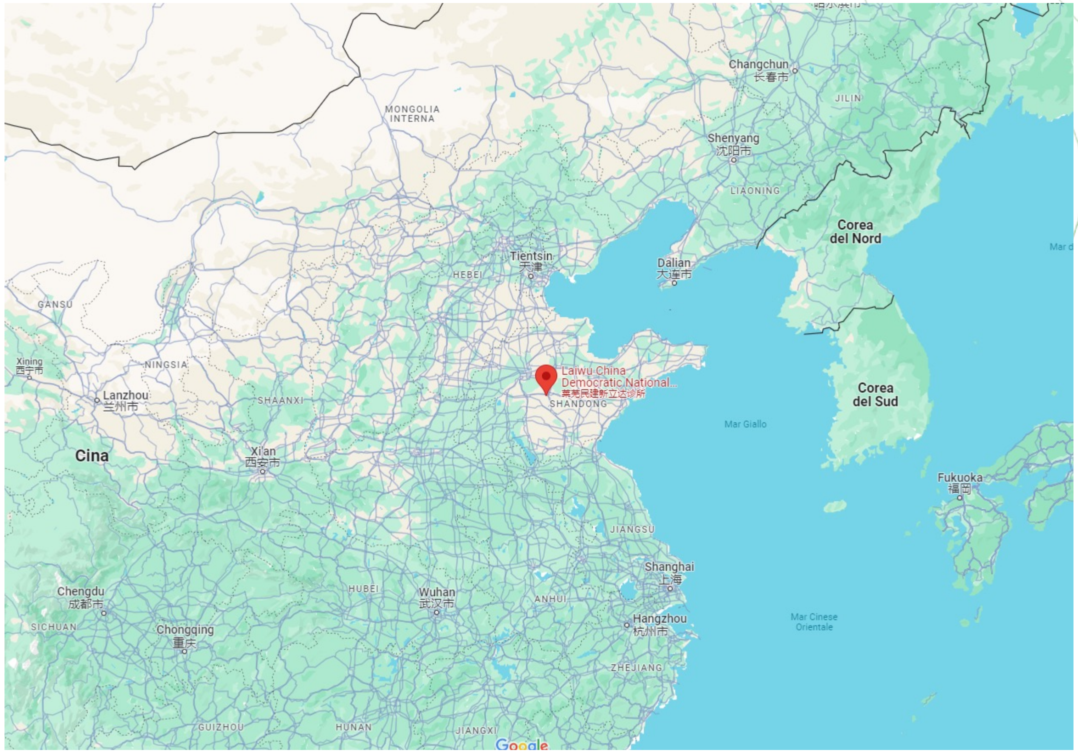
Table 1.6 Ranges and average values of global emission fluxes (measured in teragram per year, being 1 Tg $yr^{-1} = 10^6$ ton yr^{-1}) of the most important aerosol types due to natural, anthropogenic, and overall mixed (natural + anthropogenic) sources, as derived from the data given in Tables 1.1 and 1.5 for natural and anthropogenic particles and derived from the evaluations available in the literature of the past 20 years.

Aerosol particle type	Natural sources		Anthropogenic sources		Overall mixed sources	
	Range	Average	Range	Average	Range	Average
Sea salt	3000–20 000	7068	—		3000–20 000	7068
Mineral dust	1000–2150	1731	50–250	150	1050–2400	1881
Biogenic primary organic	15–70	50	—	—	15–70	50
Biomass burning organic	26–70	44	12–270	90	38–340	134
Tropospheric volcanic	4–90	30	—	—	4–90	30
Stratospheric volcanic	2–17	10	—	—	2–17	10
Cosmic dust in the middle atmosphere	2×10^{-4} to 4×10^{-2}	2×10^{-2}	—	—	2×10^{-4} to 4×10^{-2}	2×10^{-2}
Sulfates	107–374	165	50–122	73	157–496	238
Nitrates	12–27	25	90–118	95	102–145	115
Overall biogenic from isoprene, monoterpenes, and VOCs	835–1000	900	—	—	835–1000	900
Biogenic secondary organic	3–83	11	—	—	3–83	11
Industrial dust	—	—	40–130	100	40–130	100
Carbonaceous from hydrocarbons and VOCs	—	—	15–90	29	15–90	29
Black carbon (soot)	—	—	8–14	12	8–14	12

C. Tomasi and A. Lupi. Primary and Secondary Sources of Atmospheric Aerosol (2016)

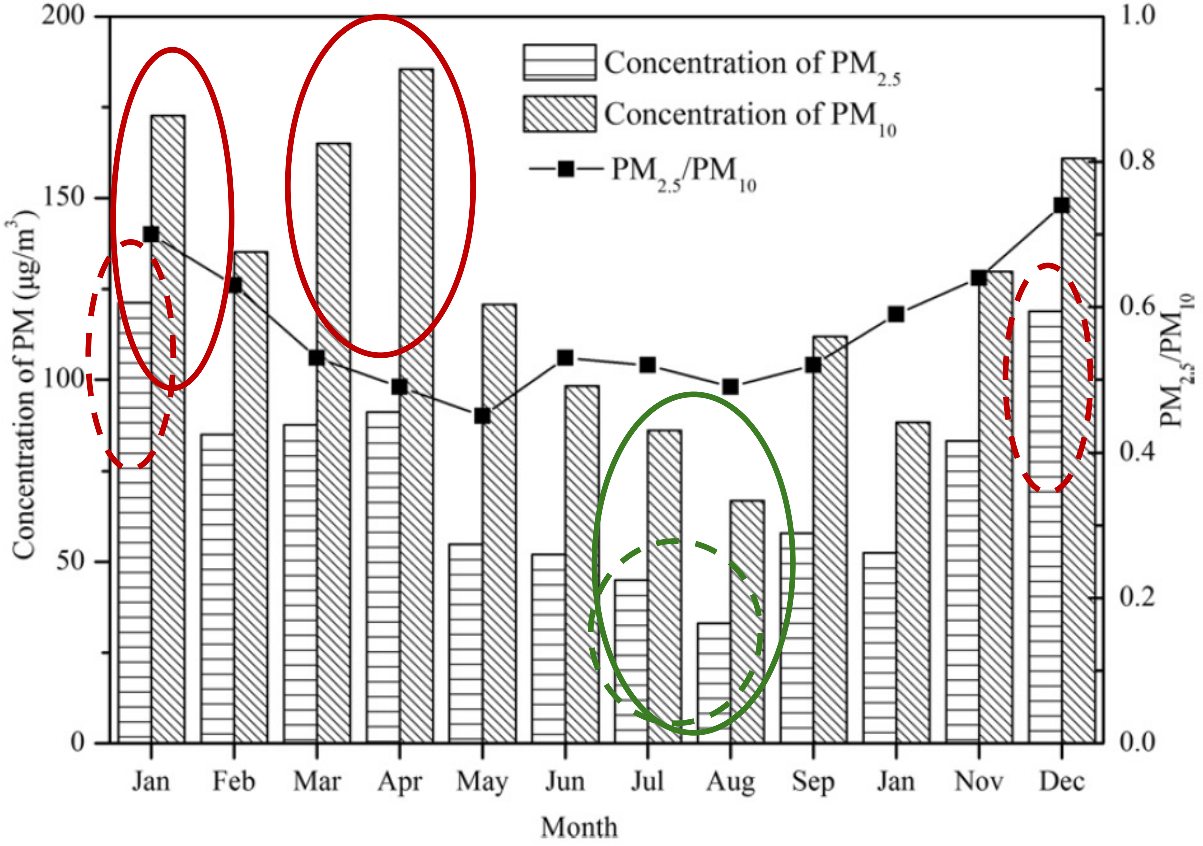
Atmospheric Aerosol: PM Composition and Sources Contribution at a Typical Industrial Urban Site

Online monitoring concentrations of PM from 01/01/2016 to 31/12/2016 in Laiwu, China



G. Zhang et al. Scientific Reports 10 (2020)

Atmospheric Aerosol: PM Composition and Sources Contribution at a Typical Industrial Urban Site



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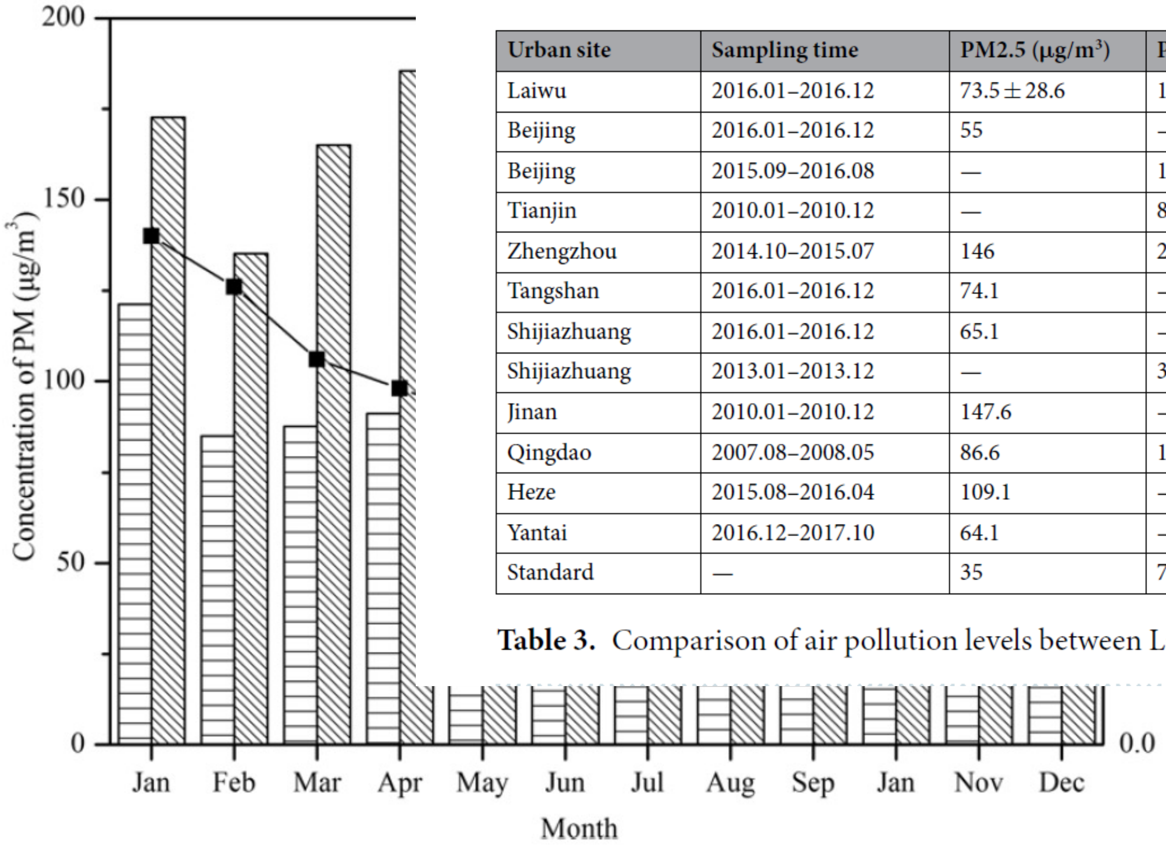
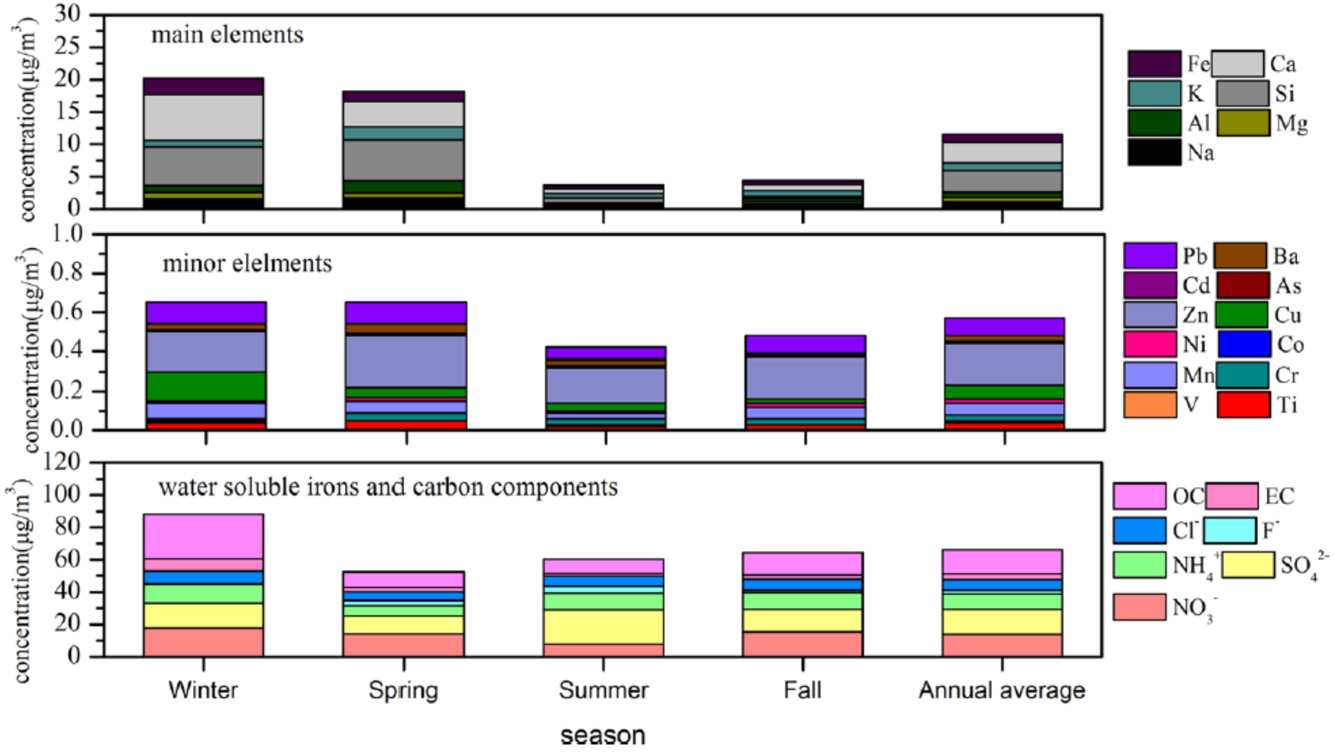


Urban site	Sampling time	PM2.5 (μg/m³)	PM10 (μg/m³)	References
Laiwu	2016.01–2016.12	73.5 ± 28.6	126.8 ± 38.2	This article
Beijing	2016.01–2016.12	55	—	[67]
Beijing	2015.09–2016.08	—	144.75	[68]
Tianjin	2010.01–2010.12	—	86.6	[69]
Zhengzhou	2014.10–2015.07	146	214	[70]
Tangshan	2016.01–2016.12	74.1	—	[67]
Shijiazhuang	2016.01–2016.12	65.1	—	[67]
Shijiazhuang	2013.01–2013.12	—	303	[71]
Jinan	2010.01–2010.12	147.6	—	[72]
Qingdao	2007.08–2008.05	86.6	120	[73]
Heze	2015.08–2016.04	109.1	—	[74]
Yantai	2016.12–2017.10	64.1	—	[75]
Standard	—	35	75	(GB 3095–2012)

Table 3. Comparison of air pollution levels between Laiwu and other cities.

G. Zhang et al. Scientific Reports 10 (2020)

Atmospheric Aerosol: PM Composition and Sources Contribution at a Typical Industrial Urban Site



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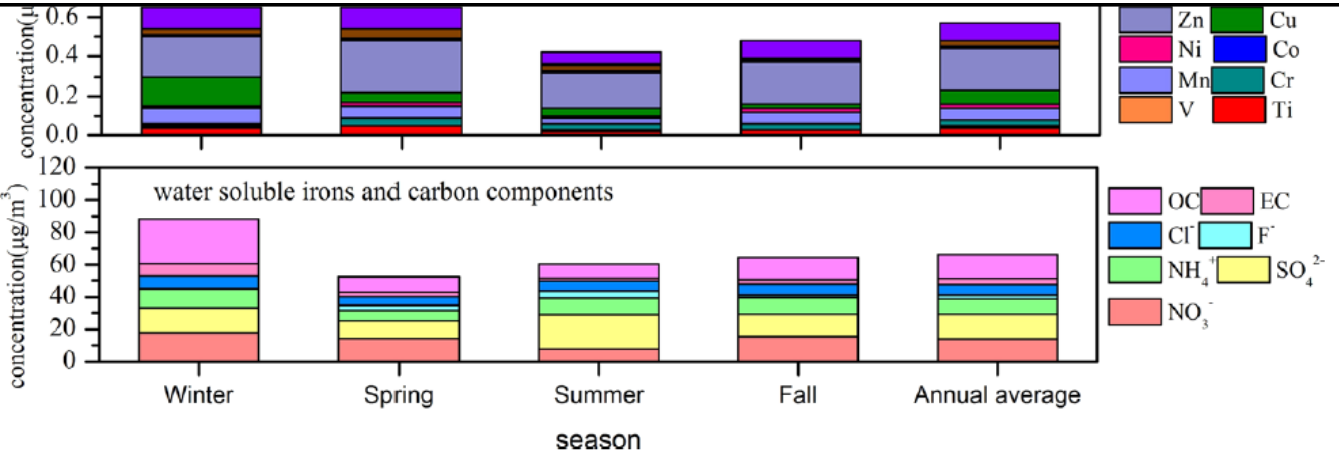
Atmospheric Aerosol: PM Composition and Sources Contribution at a Typical Industrial Urban Site

OC/EC indicates:

- vehicle exhaust in the range of 1.0 – 4.2
- fired sources (power plants, steel industries…) in the range 2.5 – 10.5
- cooking emission in the range 33 – 80

SOC is generated when OC/EC > 2.0 and is a function of OC/EC

B.J. Turpin and J.J. Huntzicker Atmospheric Environment 29 (1995)



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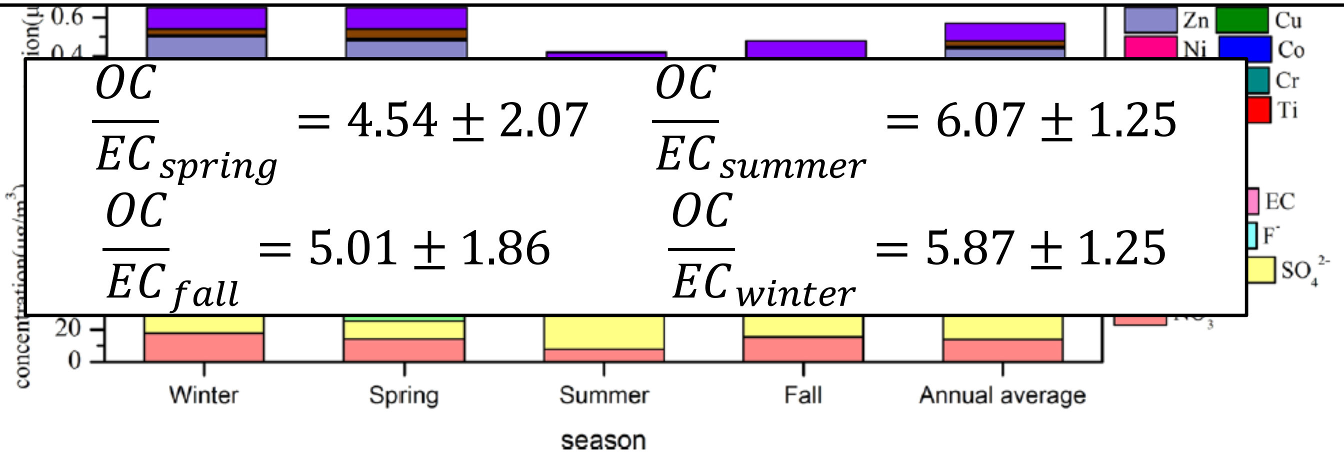
B.J. Turpin and J.J. Huntzicker Atmospheric Environment 29 (1995)

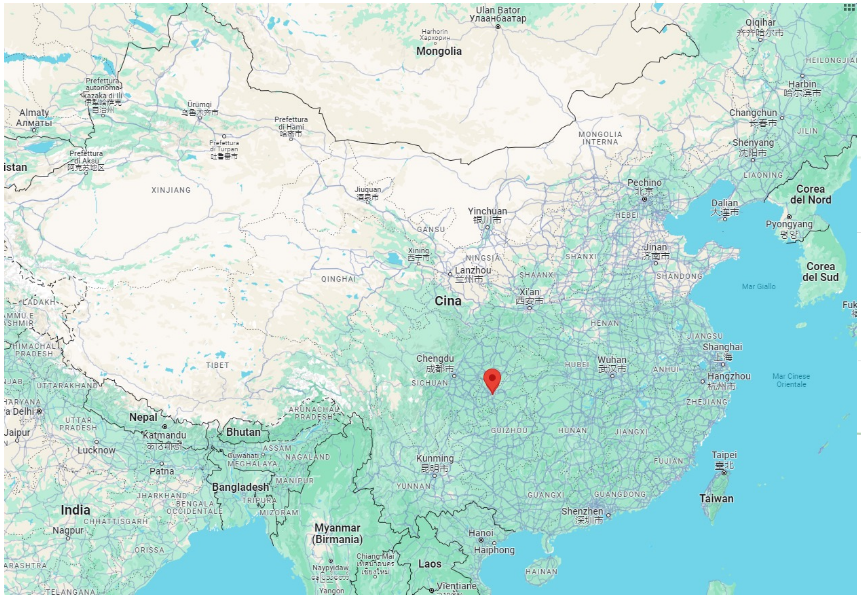
$$\frac{OC}{EC}_{spring} = 4.54 \pm 2.07 \qquad \frac{OC}{EC}_{summer} = 6.07 \pm 1.25$$

$$\frac{OC}{EC}_{fall} = 5.01 \pm 1.86 \qquad \frac{OC}{EC}_{winter} = 5.87 \pm 1.25$$



G. Zhang et al. Scientific Reports 10 (2020)

Atmospheric Aerosol: Characteristics of OC and EC in Atmospheric Aerosols of an Urban Area



Graded aerosol samples were continuously collected by cascade impactors over one year, from March 2014 to February 2015 in Beibei District, a suburb of Chongqing Municipal City

X. Peng et al. Aerosol and Air Quality Research 18 (2018)

Atmospheric Aerosol: Characteristics of OC and EC in Atmospheric Aerosols of an Urban Area

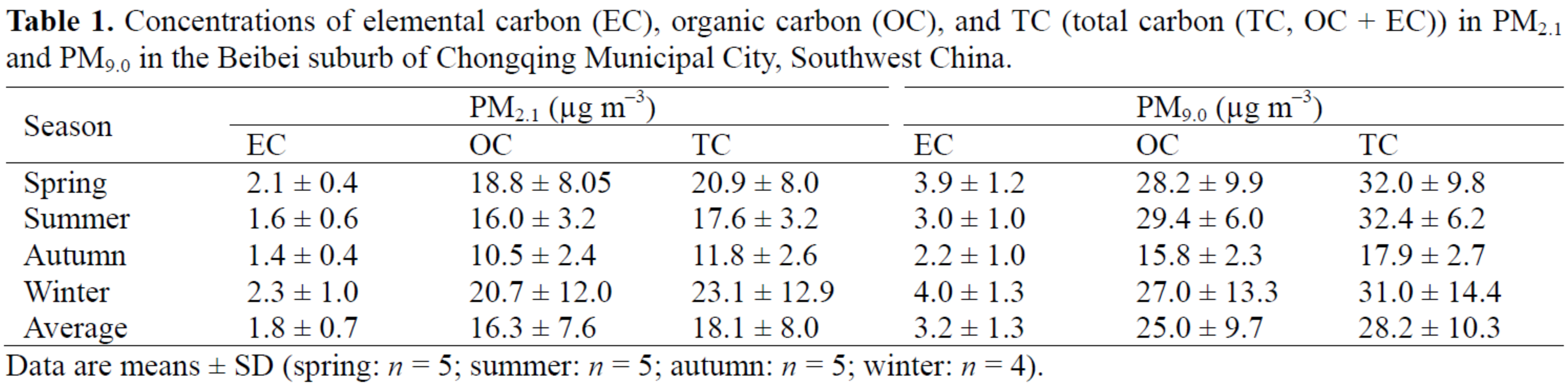


Table 1. Concentrations of elemental carbon (EC), organic carbon (OC), and TC (total carbon (TC, OC + EC)) in $PM_{2.1}$ and $PM_{9.0}$ in the Beibei suburb of Chongqing Municipal City, Southwest China.

Season	$PM_{2.1}$ ($\mu g\ m^{-3}$)			$PM_{9.0}$ ($\mu g\ m^{-3}$)		
	EC	OC	TC	EC	OC	TC
Spring	2.1 ± 0.4	18.8 ± 8.05	20.9 ± 8.0	3.9 ± 1.2	28.2 ± 9.9	32.0 ± 9.8
Summer	1.6 ± 0.6	16.0 ± 3.2	17.6 ± 3.2	3.0 ± 1.0	29.4 ± 6.0	32.4 ± 6.2
Autumn	1.4 ± 0.4	10.5 ± 2.4	11.8 ± 2.6	2.2 ± 1.0	15.8 ± 2.3	17.9 ± 2.7
Winter	2.3 ± 1.0	20.7 ± 12.0	23.1 ± 12.9	4.0 ± 1.3	27.0 ± 13.3	31.0 ± 14.4
Average	1.8 ± 0.7	16.3 ± 7.6	18.1 ± 8.0	3.2 ± 1.3	25.0 ± 9.7	28.2 ± 10.3

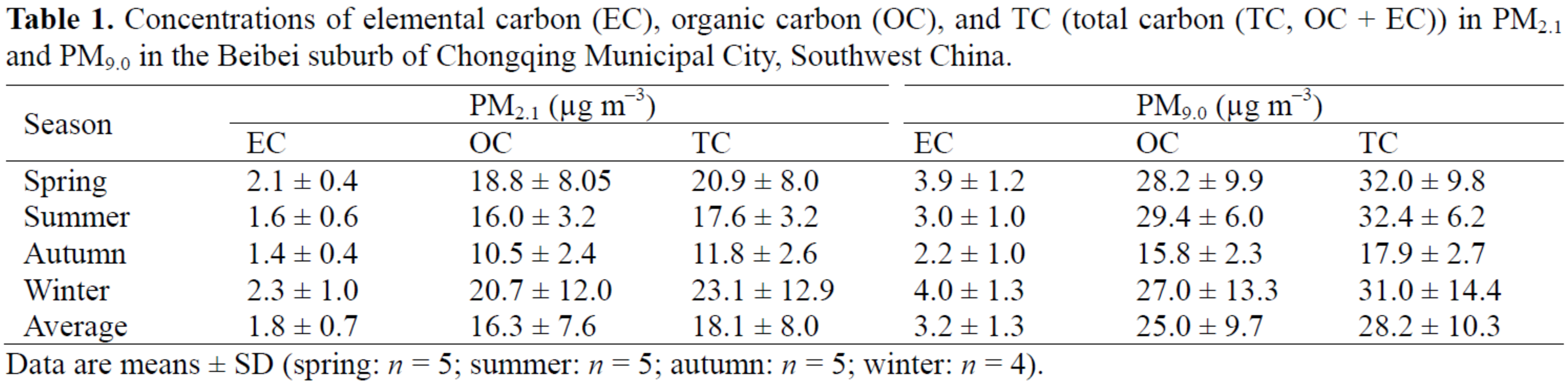
Data are means ± SD (spring: $n = 5$; summer: $n = 5$; autumn: $n = 5$; winter: $n = 4$).

$$\frac{OC}{EC}_{spring} \approx 9 \qquad \frac{OC}{EC}_{summer} \approx 10$$

$$\frac{OC}{EC}_{fall} \approx 7.5 \qquad \frac{OC}{EC}_{winter} \approx 9$$

X. Peng et al. Aerosol and Air Quality Research 18 (2018)

Atmospheric Aerosol: Characteristics of OC and EC in Atmospheric Aerosols of an Urban Area



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$$SOC = OC_{tot} - EC \cdot \left(\frac{OC}{EC}\right)_{pri} \approx OC_{tot} - EC \cdot \left(\frac{OC}{EC}\right)_{min}$$

Turpin, B. J. & Huntzicker, J. J.. Atmos. Environ. 29 (1995)
Chow, J. C., Watson, J. G. & Kuhns,. Chemosphere 54 (2004)

X. Peng et al. Aerosol and Air Quality Research 18 (2018)

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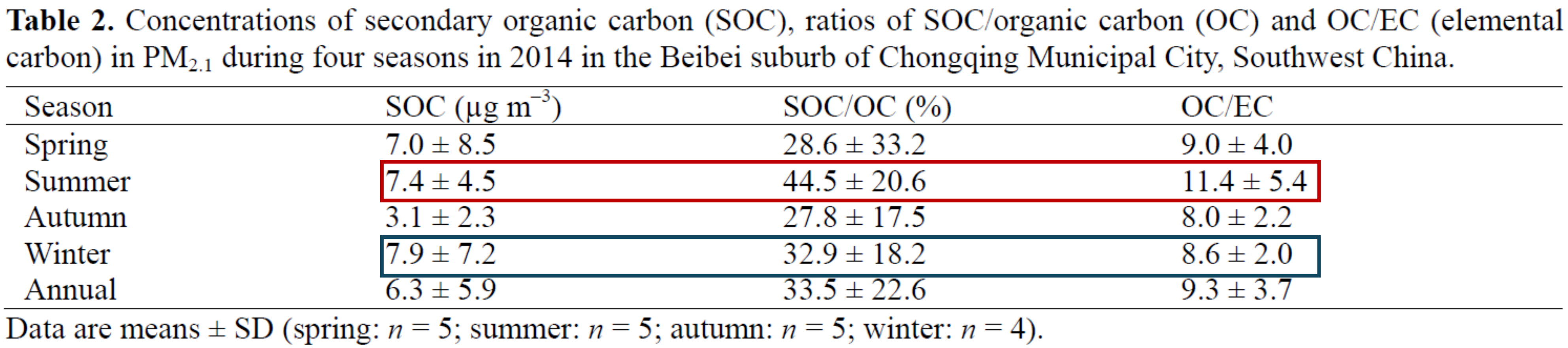


Table 2. Concentrations of secondary organic carbon (SOC), ratios of SOC/organic carbon (OC) and OC/EC (elemental carbon) in PM$_{2.1}$ during four seasons in 2014 in the Beibei suburb of Chongqing Municipal City, Southwest China.

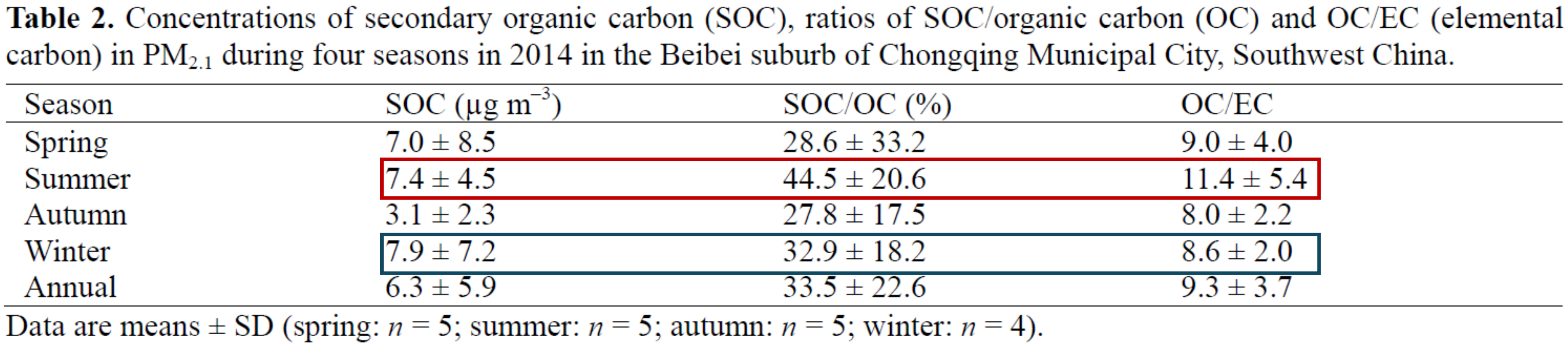
Season	SOC ($\mu g\ m^{-3}$)	SOC/OC (%)	OC/EC
Spring	7.0 ± 8.5	28.6 ± 33.2	9.0 ± 4.0
Summer	7.4 ± 4.5	44.5 ± 20.6	11.4 ± 5.4
Autumn	3.1 ± 2.3	27.8 ± 17.5	8.0 ± 2.2
Winter	7.9 ± 7.2	32.9 ± 18.2	8.6 ± 2.0
Annual	6.3 ± 5.9	33.5 ± 22.6	9.3 ± 3.7

Data are means ± SD (spring: n = 5; summer: n = 5; autumn: n = 5; winter: n = 4).

SUMMER → Formation of SOC due to high photochemical activity (intense solar radiation)

X. Peng et al. Aerosol and Air Quality Research 18 (2018)

Atmospheric Aerosol: Characteristics of OC and EC in Atmospheric Aerosols of an Urban Area



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Data are means ± SD (spring: *n* = 5; summer: *n* = 5; autumn: *n* = 5; winter: *n* = 4).

WINTER → lower photochemical activity, but lower temperature and high VOCs emissions

X. Peng et al. Aerosol and Air Quality Research 18 (2018)

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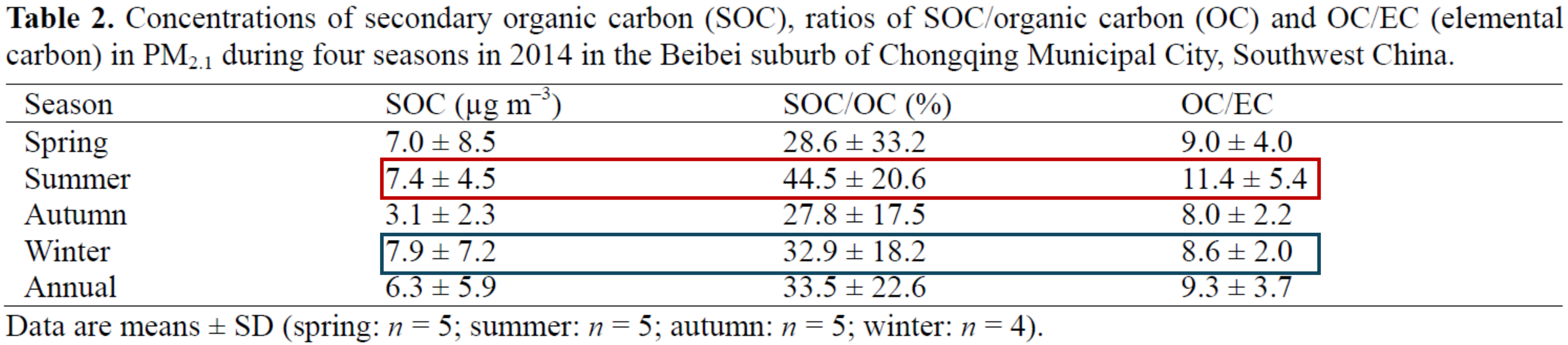

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WINTER → lower photochemical activity, but lower temperature and high VOCs emissions

optimal T for SOA formation, caused by the competing effects between

total secondary product increase with T due to change in reaction rate

VS

vapor pressure decrease and thus SOA increase with decreasing T by absorption and condensation of semi-volatile organic compounds on existing particles

R. Strader et al. Atmos. Environ. 33 (1999)

ITINERIS

THANKS!

IR0000032 – ITINERIS, Italian Integrated Environmental Research Infrastructures System (D.D. n. 130/2022 - CUP B53C22002150006) Funded by EU - Next Generation EU PNRR-Mission 4 “Education and Research” - Component 2: “From research to business” - Investment 3.1: “Fund for the realisation of an integrated system of research and innovation infrastructures”

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