Size-resolved aerosol pH over Europe during summer

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EGU 2020 – session AS2.14 Atmospheric Acidity, Air-sea Chemical Fluxes and their Impacts

Based on work in review: Zakoura et al., ACPD, 2020: https://doi.org/10.5194/acp-2019-1146

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European Research Council
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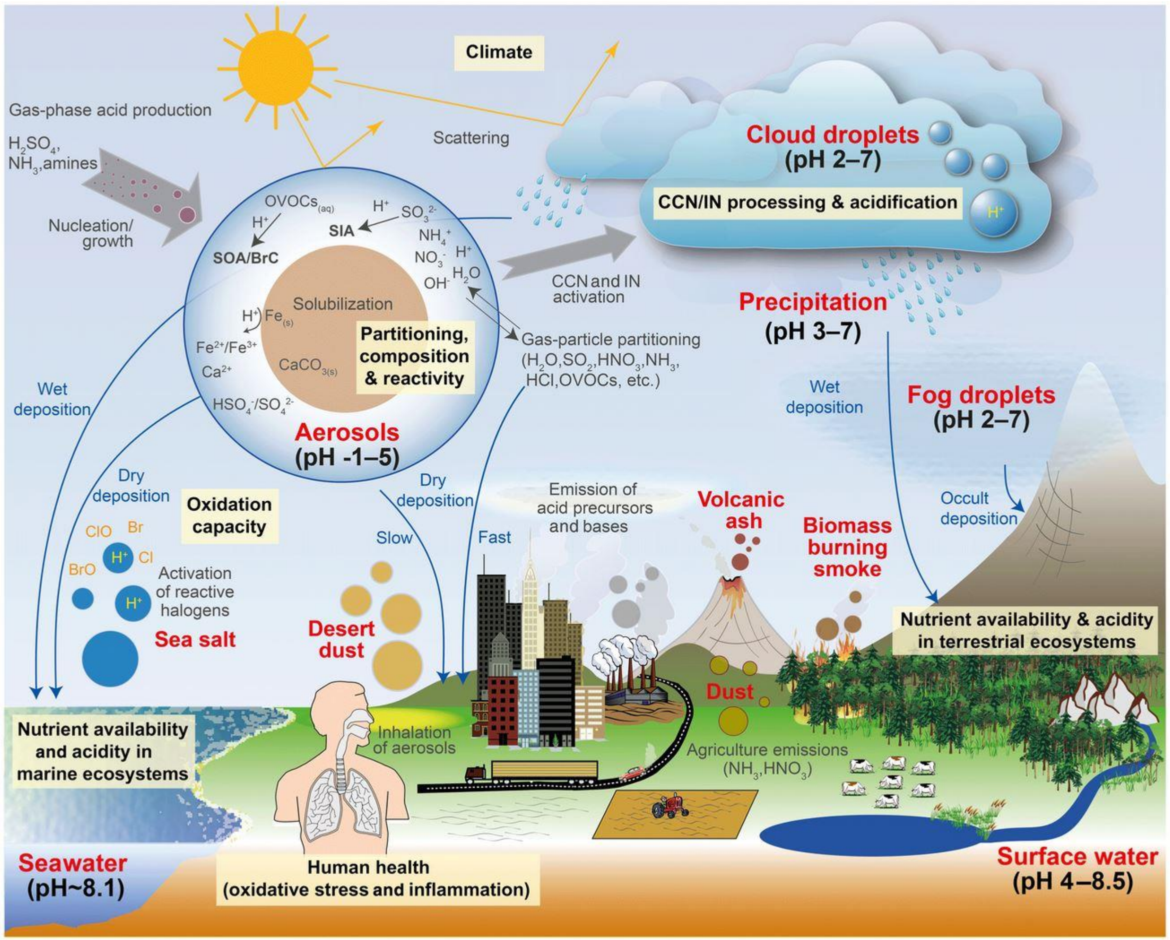
Motivation

- Aerosol Acidity (pH) is a critical property for atmospheric processes.
- Chemical transport models do not simulate properly or evaluate aerosol acidity.
- These studies focus mainly on PM_1 or $PM_{2.5}$ pH, not on pH of all size ranges.



Pye et al., ACP, 2020

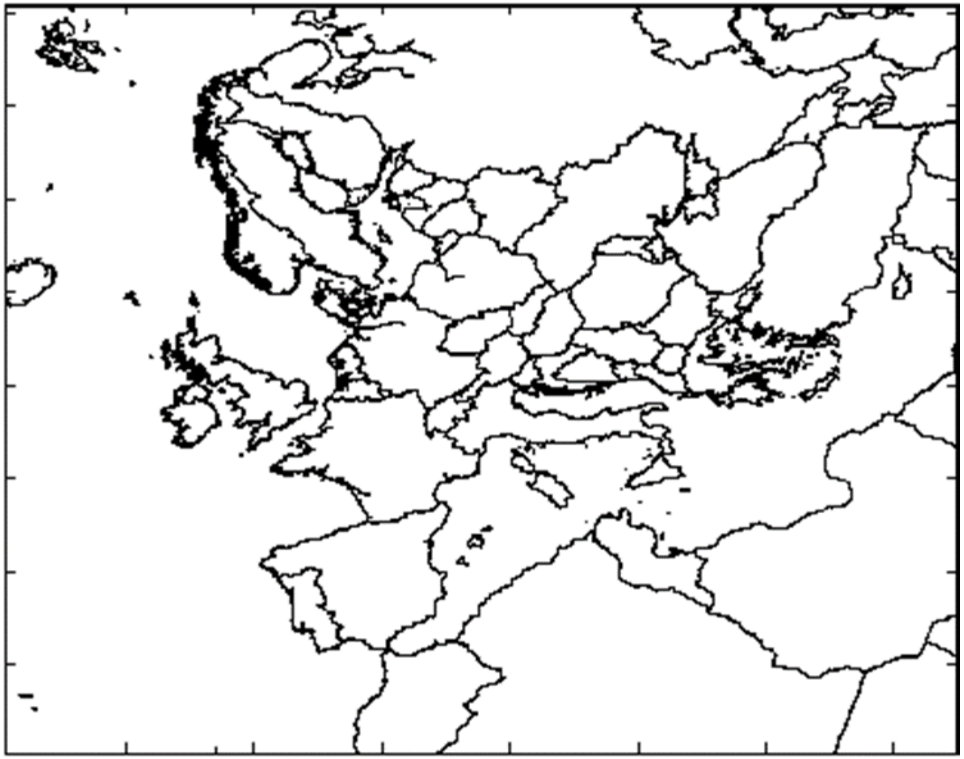
- Most of the studies are for the US, almost no information about Europe.
- One (observational) study exploring the dependence of aerosol acidity on altitude.
- **This study is the first to explore the size-and-altitude dependent acidity for aerosol in Europe.**
- **Implications for nitrate and other processes**

PMCAMx application over Europe

- 5400 x 5832 km^2 domain
- 36x36 km grid resolution

  x=150 and y=162 cells

  14 vertical layers (up to 6 km)
- Simulation period: 1-29 May 2008
- Aerosol thermodynamics simulated with the ISORROPIA-II model (Fountoukis and Nenes, 2007).



<u>pH is calculated for particles</u>: smaller than 1 μm

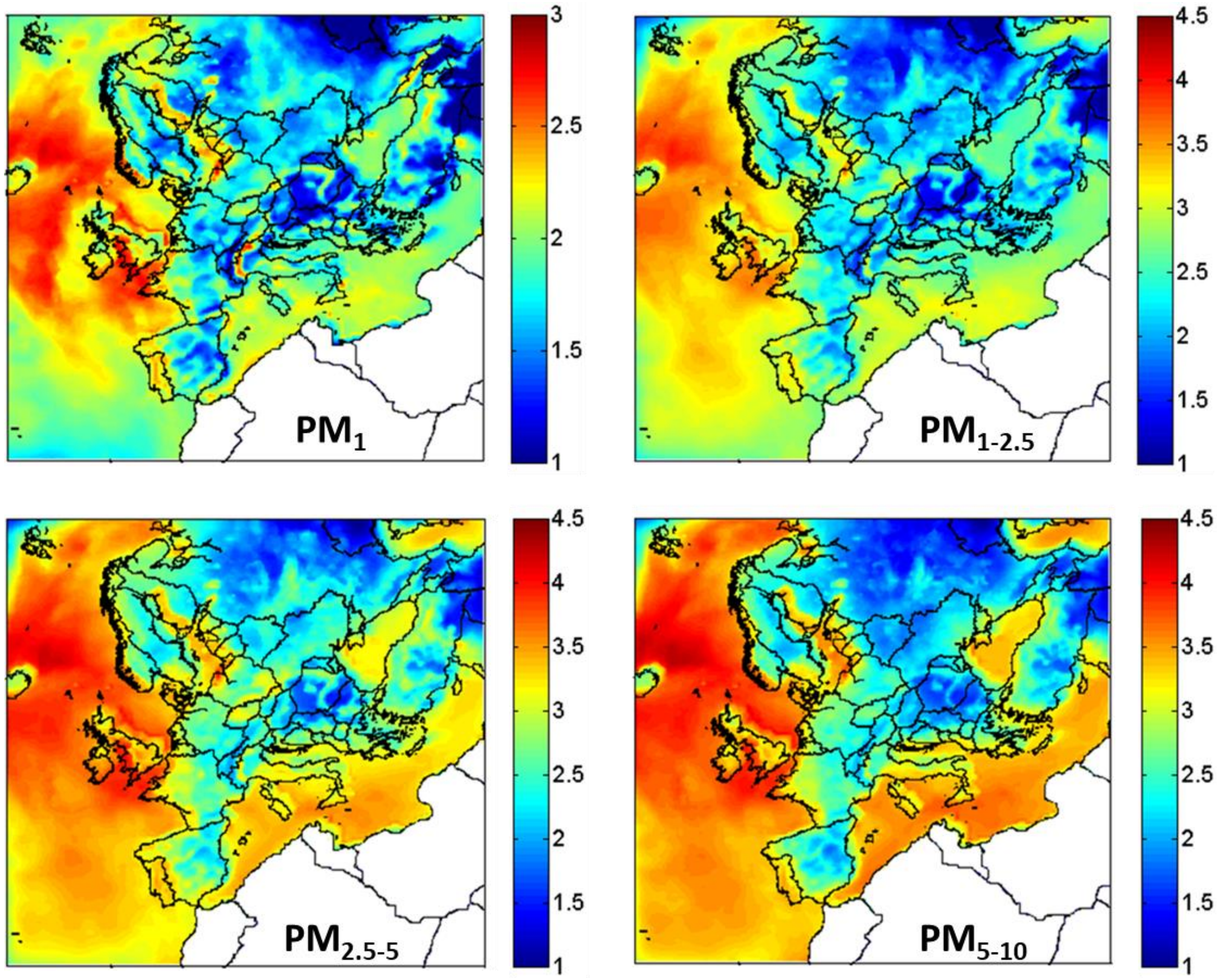
1-2.5 μm

2.5-5 μm

5-10 μm.

[Zakoura et al., ACPD, 2020](#)

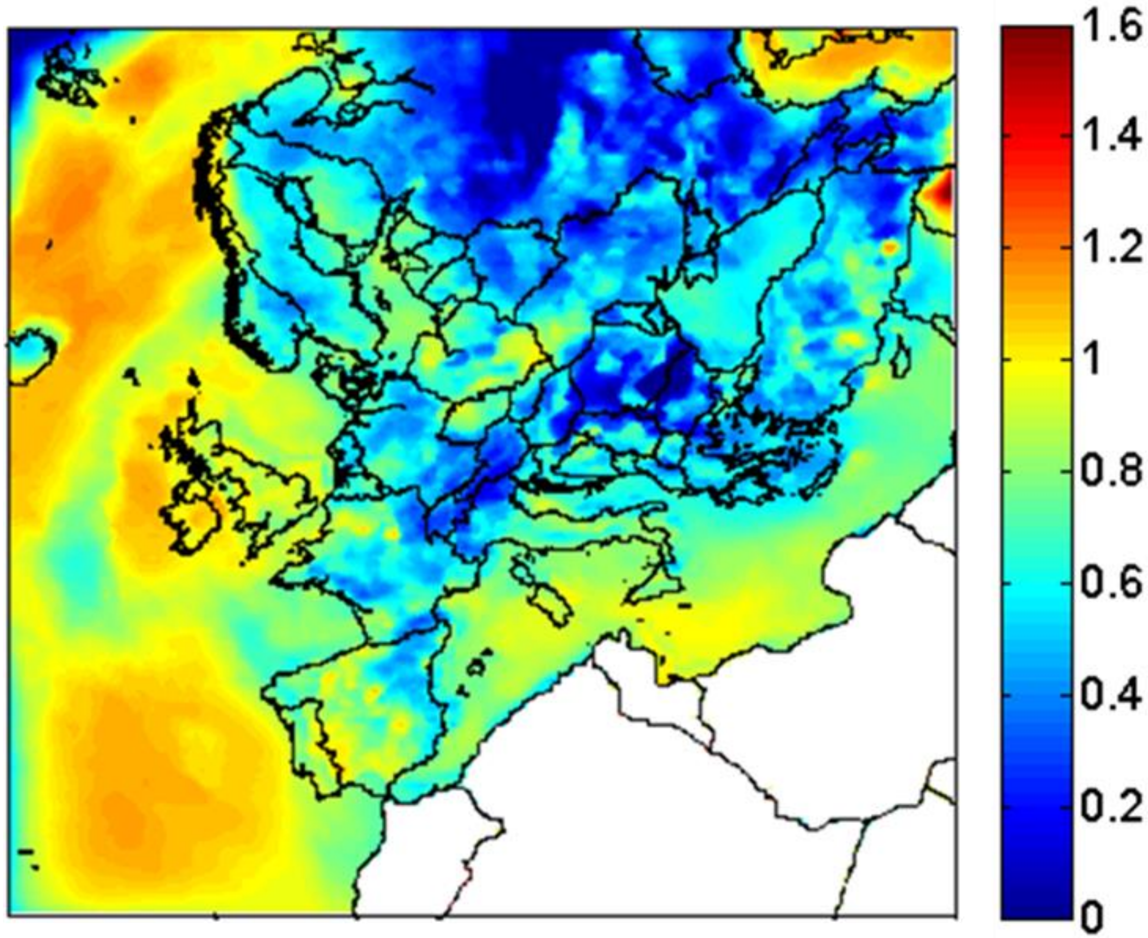
pH at ground level over Europe for May 2008



- Fine mode aerosol is more acidic (lower pH) than larger particles.
- Large contrast in pH between marine and continental sites (owing to the presence of non-volatile cations from seasalt and dust).
- The acidity contrast tends to increase as particles become larger.

Zakoura et al., ACPD, 2020

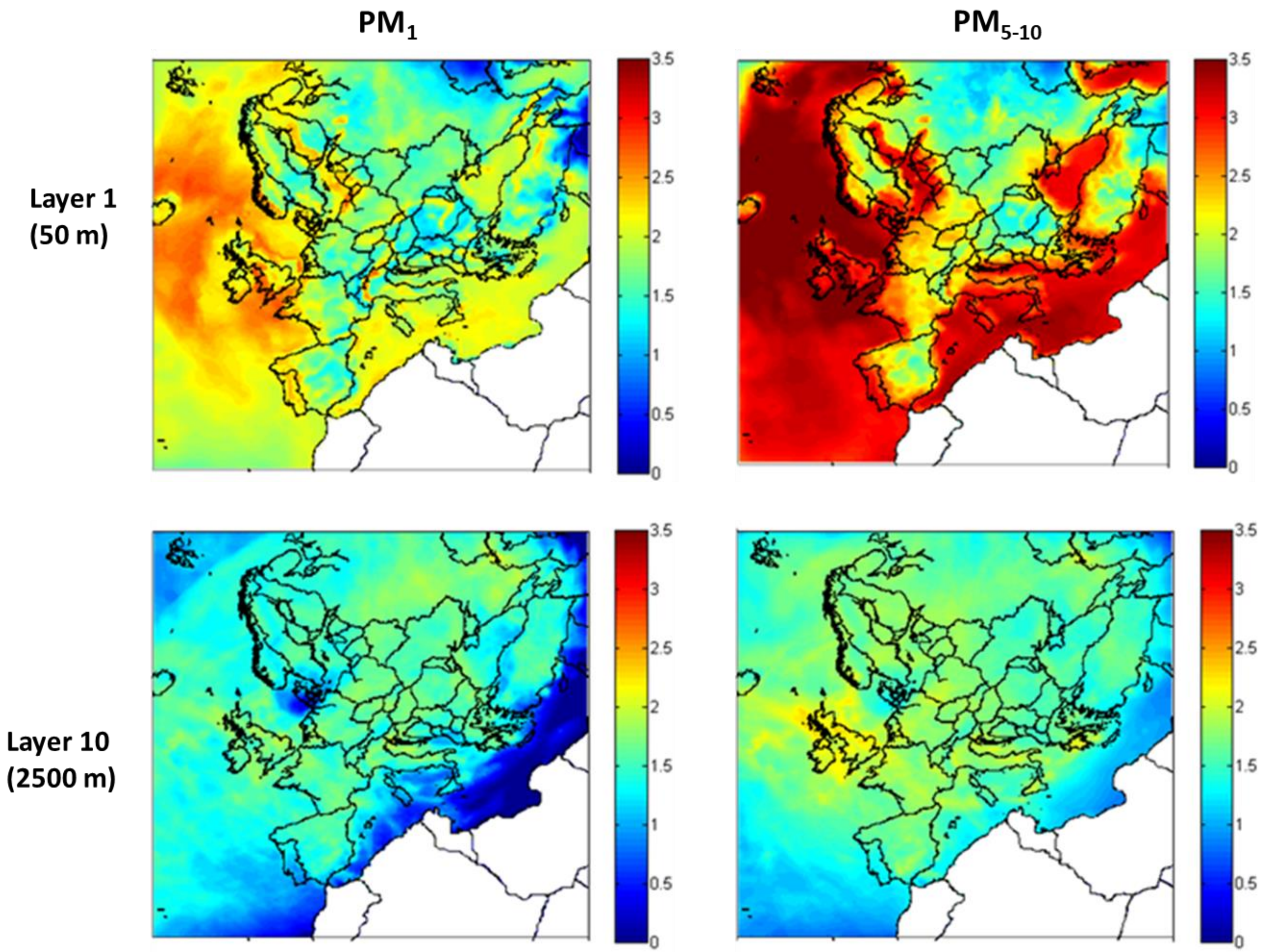
pH difference between $PM_{1-2.5}$ and PM_1 (ground level)



- Acidity gradient is low for continental aerosol far away from dust and marine influence (central/East Europe)
- Continental areas close to W.Mediterranean and marine evironments can have more than 1 pH unit difference.
- Important implications for PM formation and solubilization of metals and toxicity across size.

[Zakoura et al., ACPD, 2020]

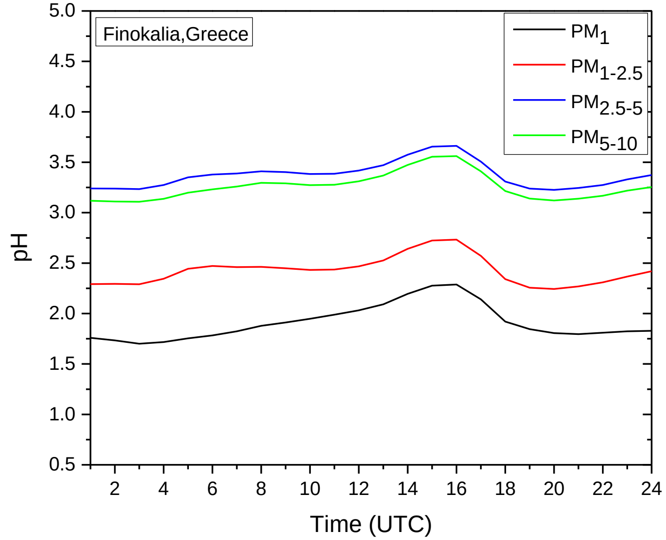
pH variation with height

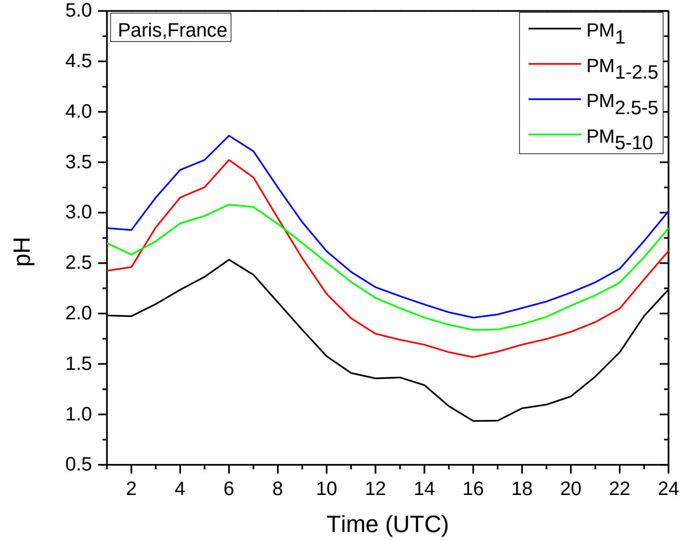


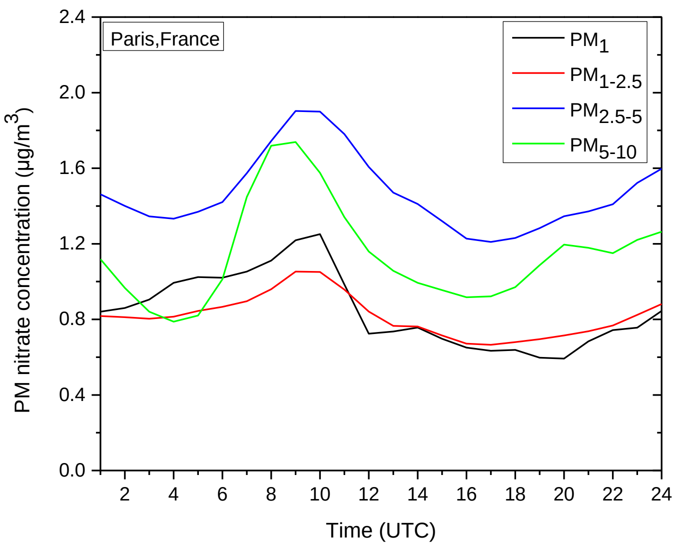
- Large changes of aerosol pH with height.
- pH decreases with altitude. This is primarily driven by a decrease in relative humidity
- Continental-marine contrast almost disappears above the boundary layer

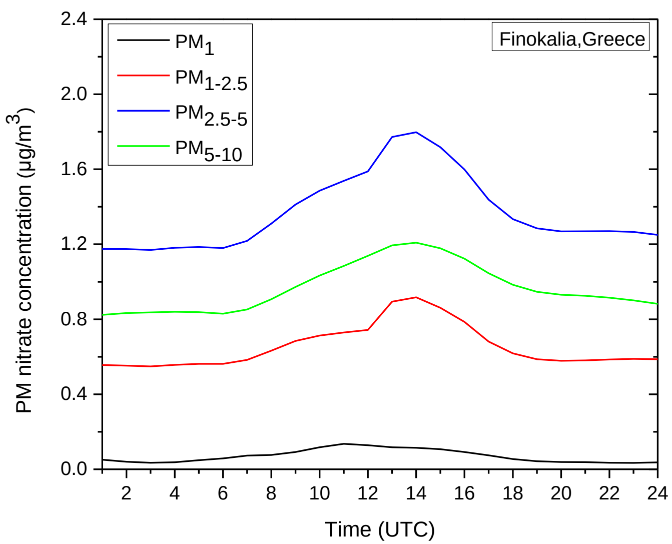
Zakoura et al., ACPD, 2020

Temporal evolution of pH – impact on inorganic nitrate



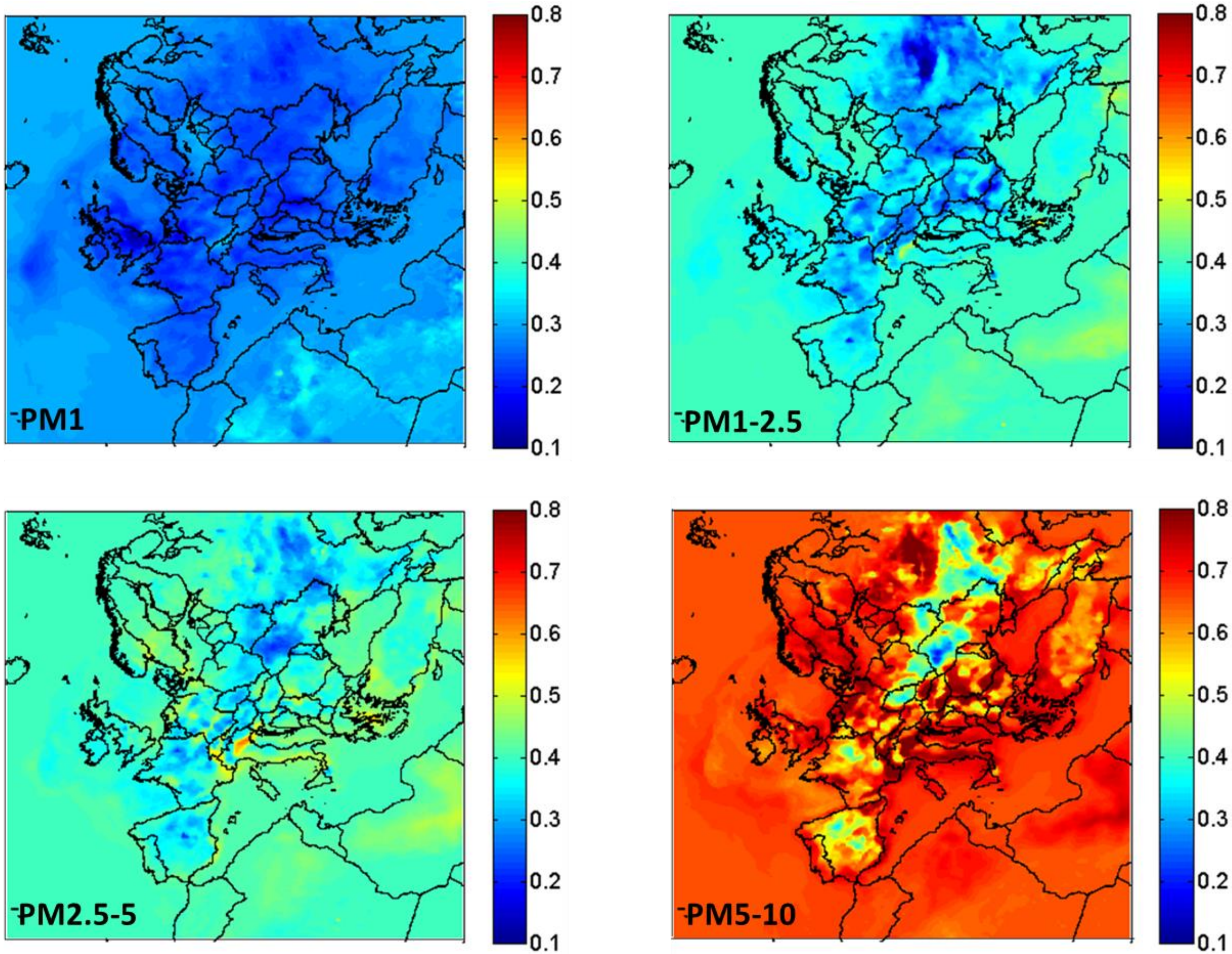






Zakoura et al., ACPD, 2020

The effect of other non-volatile cations in the pH of each size class: calcium from dust



- There is a small amount of Ca present in small PM1 – negligible impact on pH.
- PM1-2.5 sizes experience an influence that shifts pH up to 0.5 units.
- PM5-10 tends to have considerable effects – up to 1 pH unit.

[Zakoura et al., ACPD, 2020]

Conclusions

- pH increases with particle size by 1 to 4 units, with the highest pH values predicted in coastal areas of northern Europe.
- Difference between PM_1 and $PM_{1\text{-}2.5}$ in pH values, that can reach up to 1 unit over continental areas.
- Particles of all sizes more acidic higher in the atmosphere (0.5-2 units pH decrease).
- Highest pH and nitrate levels for the 2.5-5 μm range over Europe. pH is one of the main driving factors controlling its presence.
- pH increases when calcium is present, especially for $PM_{2.5\text{-}5}$ and $PM_{5\text{-}10}$, as calcium is mostly present in coarser particles.
- The pH changes are in the range that has an important impact on the solubility of metals (Fe, Cu) and other species (P) that can impact chemical processes, public health and biogeochemical cycles.

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https://doi.org/10.5194/acp-2019-1146

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Submitted as: research article

19 Feb 2020

Review status
This preprint is currently under review for the journal ACP.

Size-resolved aerosol pH over Europe during summer

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Received: 15 Dec 2019 – Accepted for review: 04 Feb 2020 – Discussion started: 19 Feb 2020

Abstract. The dependence of aerosol acidity on particle size, location and altitude over Europe during a summertime period is investigated using the hybrid version of aerosol dynamics in the chemical transport model PMCAMx. The pH changes more with particle size in northern and southern Europe owing to the enhanced presence of non-volatile cations (Na, Ca, K, Mg) in the larger particles. Differences of up to 1–4 pH units are predicted between sub- and super-micron particles, while the average pH of $PM_{1-2.5}$ can be as much as 1 unit higher than that of PM_1. Most aerosol water over continental Europe is associated with PM1, while $PM_{2.5-5}$ and PM_{5-10} dominate the water content in the marine and coastal areas due to the relatively higher levels of hygroscopic sea salt. Particles of all sizes become increasingly acidic with altitude (0.5–2 units pH decrease over 2.5 km) primarily because of the decrease in aerosol liquid water content (driven by humidity changes) with height. Inorganic nitrate is strongly affected by aerosol pH with the highest average nitrate levels predicted for the $PM_{2.5-5}$ range and over locations where the pH exceeds 3. Dust tends to increase aerosol water levels, aerosol pH and nitrate concentrations for all particle sizes. This effect of dust is quite sensitive to its calcium content. The size-dependent pH differences carry important implications for pH-sensitive processes in the aerosol.

How to cite: Zakoura, M., Kakavas, S., Nenes, A., and Pandis, S. N.: Size-resolved aerosol pH over Europe during summer, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-1146, in review, 2020.

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