

Spatial variations and source apportionment of the marine aerosol over the Atlantic Ocean

Shan Huang, Laurent Poulain, Manuela van Pinxteren, Dominik van Pinxteren, Zhijun Wu, Hartmut Herrmann, Alfred Wiedensohler

Leibniz-Institute for Tropospheric Research, Leipzig, Germany

email: huang@tropos.de



INTRODUCTION

The marine aerosol plays an important role in global climate regulation and marine biogenic system. Influenced by both continents and oceans, the chemical composition, spatial variation and the source of the marine aerosol are still poorly understood. This creates uncertainties in the estimation of hygroscopic growth and optical properties of aerosol particles.

In this study, physicochemical properties of aerosol particles in the marine boundary layer were measured by a comprehensive suite of on-line instrumentation on the Germany research vessel Polarstern during 4 cruises over the Atlantic Ocean mainly in spring or autumn (Fig. 1 and table below). Results of these cruises (CR) were presented here.

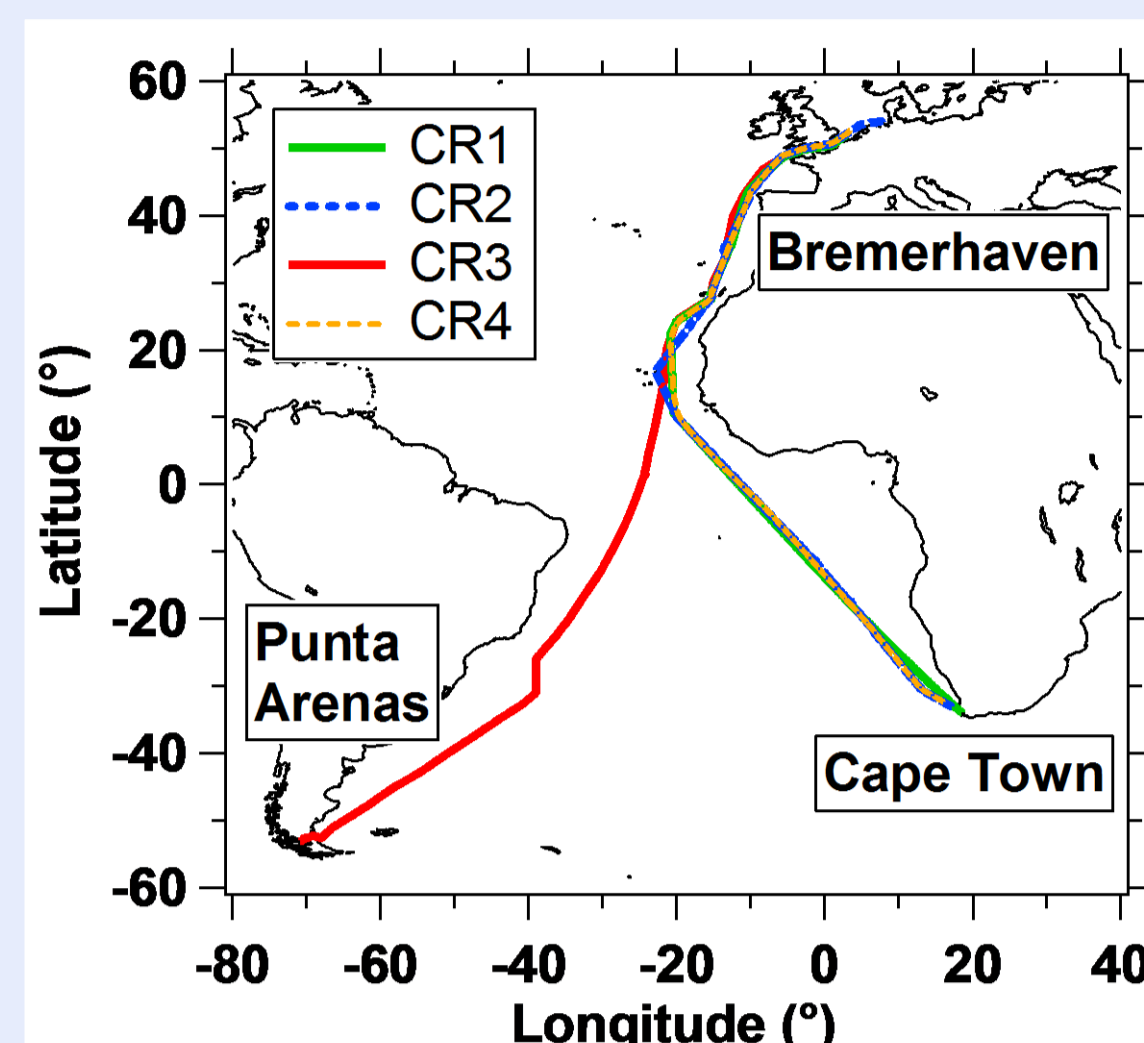


Fig. 1: Ship tracks in 4 cruises

Expedition	Port	Duration
CR 1 (ANT-XXVII/4)	Cape Town - Bremerhaven	20.04.2011 - 20.05.2011
CR 2 (ANT-XXVIII/1)	Bremerhaven - Cape Town	28.10.2011 - 01.12.2011
CR 3 (ANT-XXVIII/5)	Punta Arenas - Bremerhaven	10.04.2012 - 15.05.2012
CR 4 (ANT-XXIX/1)	Bremerhaven - Cape Town	27.10.2012 - 27.11.2012

Instrumentation:

Chemical composition:

- High Resolution Time of Flight Aerosol Mass Spectrometer (HR-ToF-AMS, main instrument for this study)
- Daily PM1 Digital filter sampler

Hygroscopic Growth

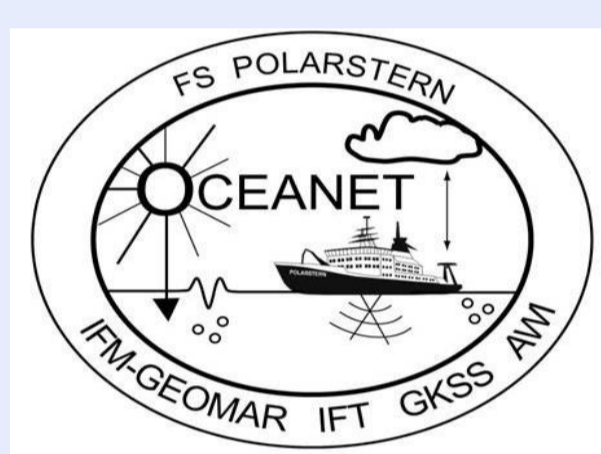
- Hygroscopicity Tandem Differential Mobility Analyzer (HTDMA)
- Differential Mobility Analyzer-Cloud Condensation Nuclei Counter (DMA-CCNC)

Particle Number Size Distribution:

- Aerodynamic Particle Size Spectrometer
- Mobility Particle Size Spectrometer

Optical properties:

- Integrating Nephelometer
- Multi Angle Absorption Photometer (MAAP)



SPATIAL VARIATION

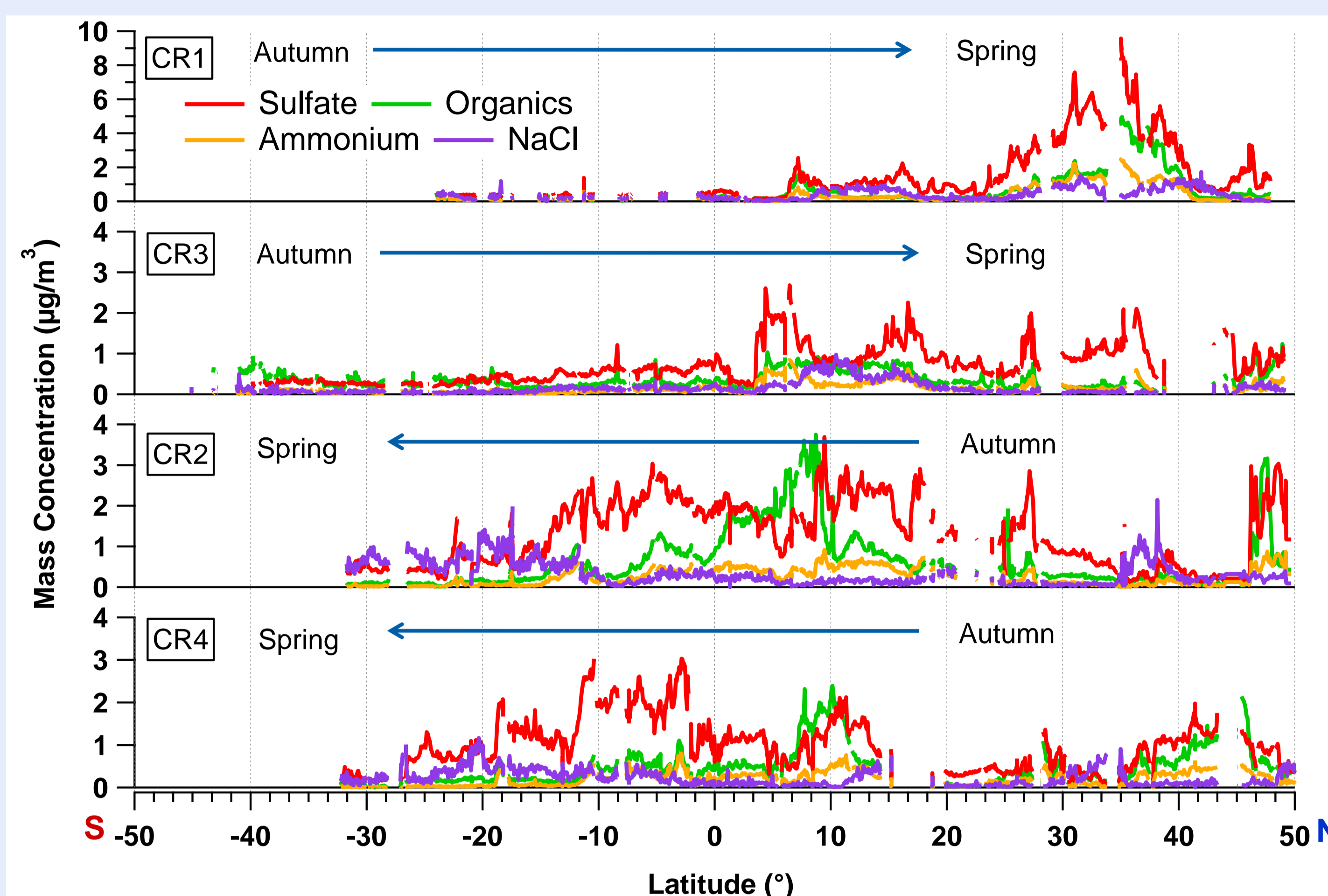


Fig. 2: Mass concentration of main species vs. latitude for the 4 cruises

The concentrations of four main species *sulfate*, *organics*, *ammonium* and *NaCl* were less than $4 \mu\text{g m}^{-3}$ in most period of the cruises, but relatively higher concentrations were observed when the ship was 1) close to the Europe (30° - 50°N), and 2) near the North Africa coasts (0° - 20°N), indicating the possible influence from land. NaCl concentrations were estimated based on HR-ToF-AMS and validated by results from filter measurement. (Gaps correspond to period with/of ship exhaust contamination and have been deleted).



CASE STUDY OF CRUISE ANT-XXVIII/5

Air mass classification

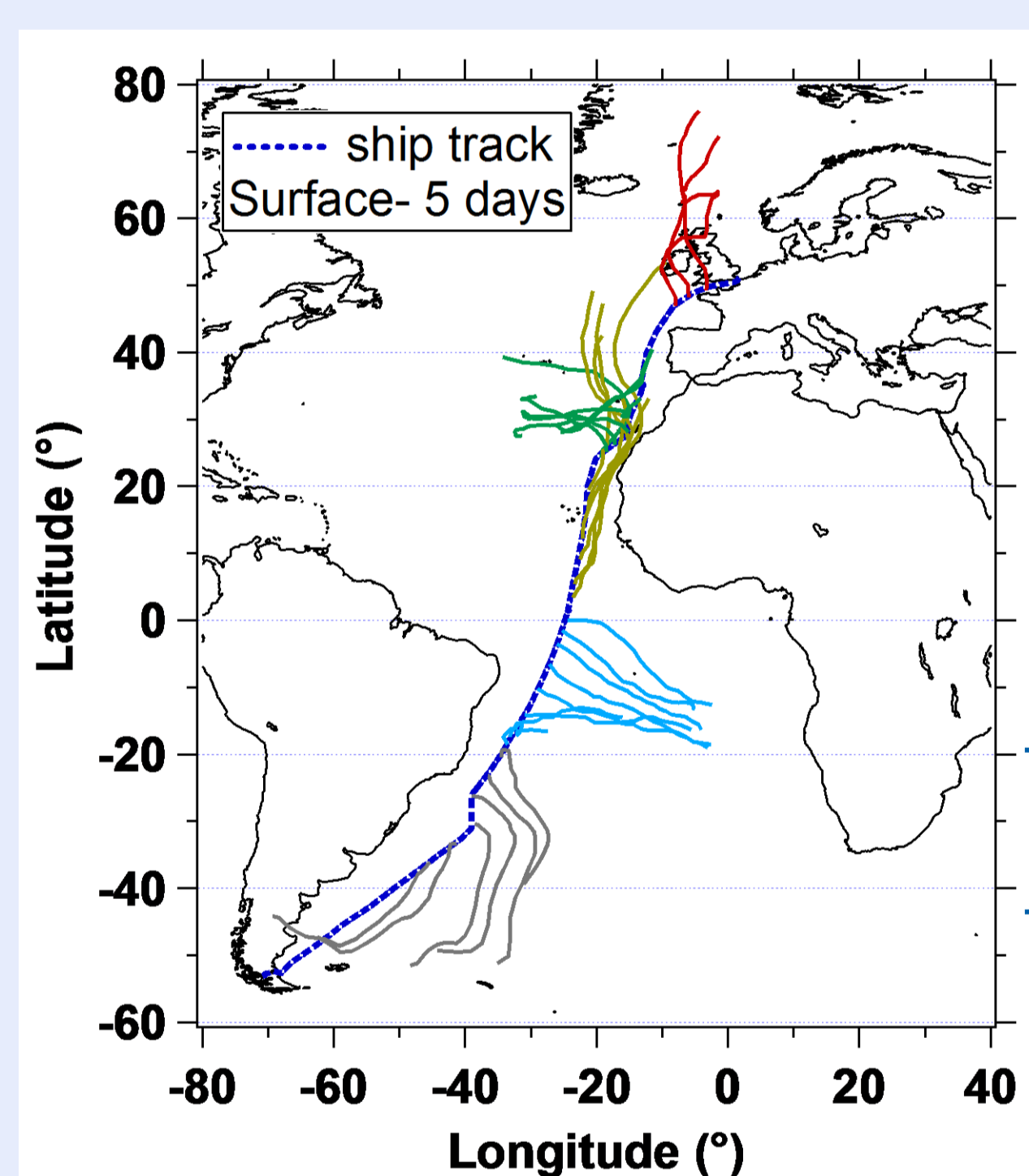


Fig. 3: Back trajectories along ship track

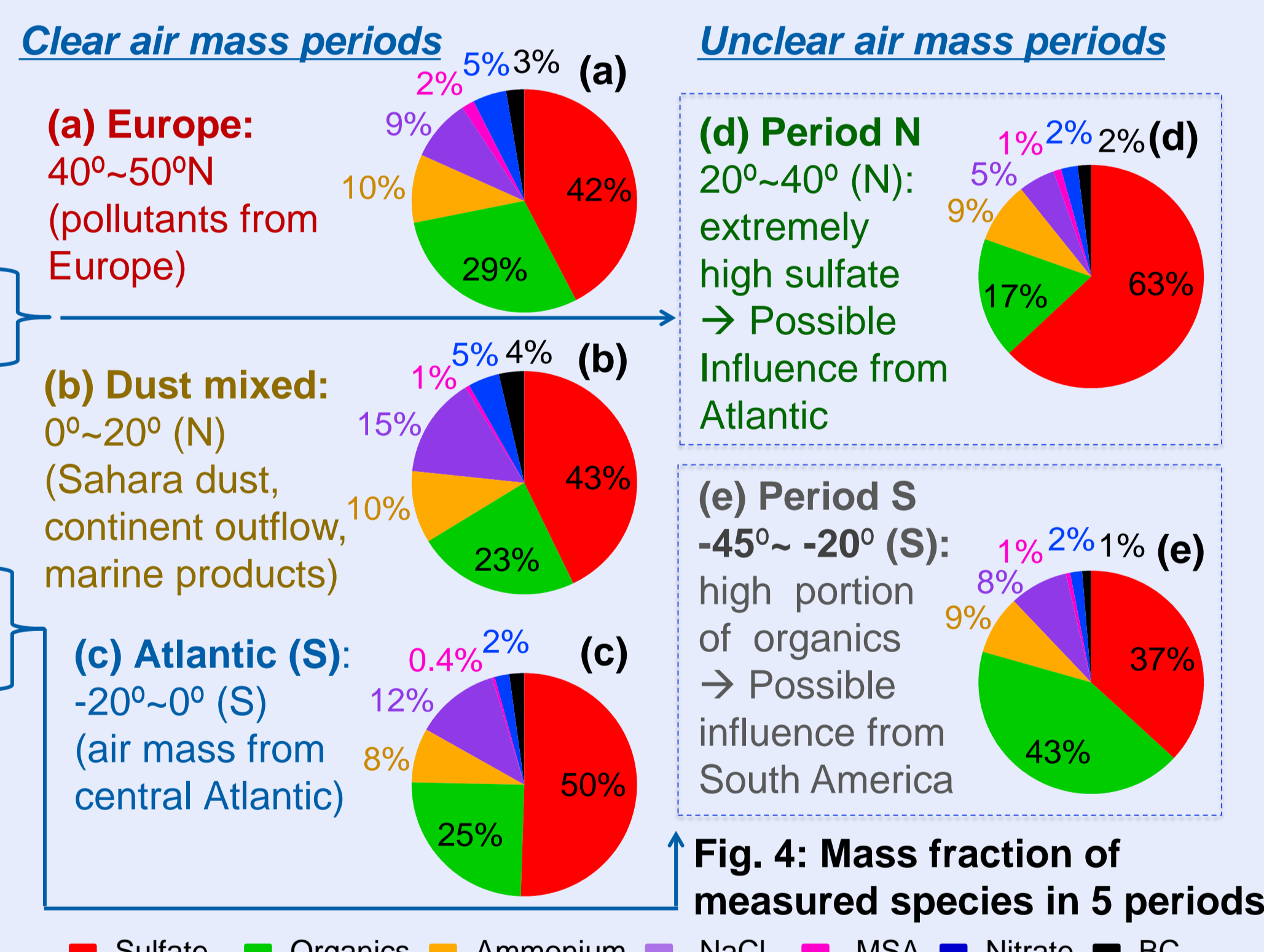


Fig. 4: Mass fraction of measured species in 5 periods

Quantification of MSA and NaCl

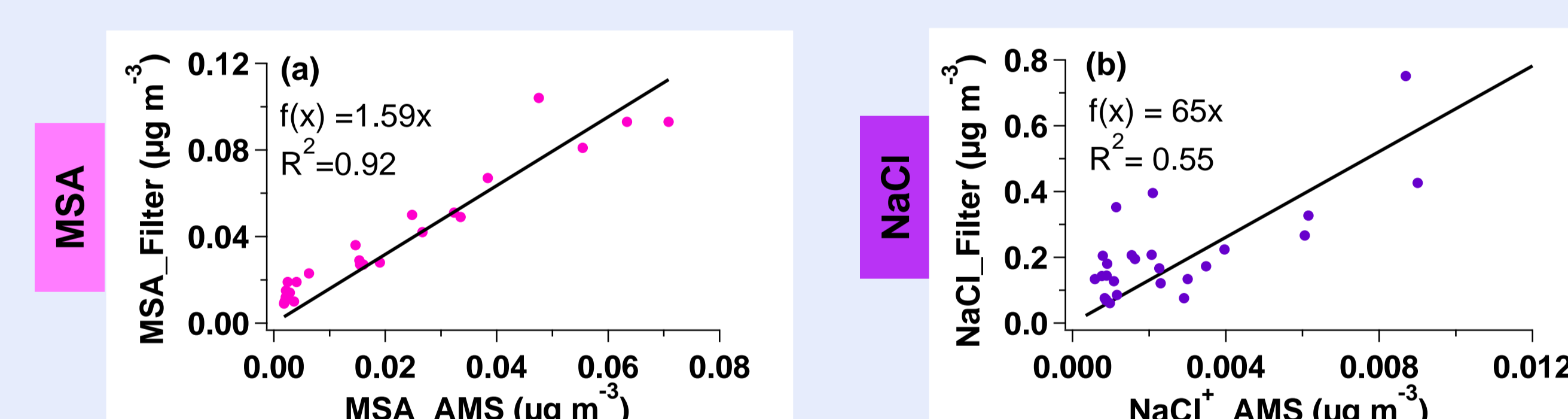


Fig. 5: The comparison between results from AMS and filters for (a) MSA and (b) NaCl

- **MSA** (methanesulfonic acid), the typical marine tracer, could be quantified by HR-ToF-AMS (Zorn et al., 2008, *Atmos. Chem. Phys.*).
- Good linear correlation between results from AMS and filter (PM1) measurements confirms the reliability of the method.
- The factor of 1.59 between the two instruments are probably caused by the two different analyzing methods.
- **NaCl** from submicrometer sea spray aerosol was quantified by HR-ToF-AMS by the method first reported by Ovadnevaite et al. (2012, *J. Geophys. Res.*).
- Comparing NaCl⁺ mass concentration from HRMS with results from the filter measurements, a scaling factor (65) is found, which is comparable to the result in Mace Head (51) mentioned above.

Source apportionment based on PMF-AMS method

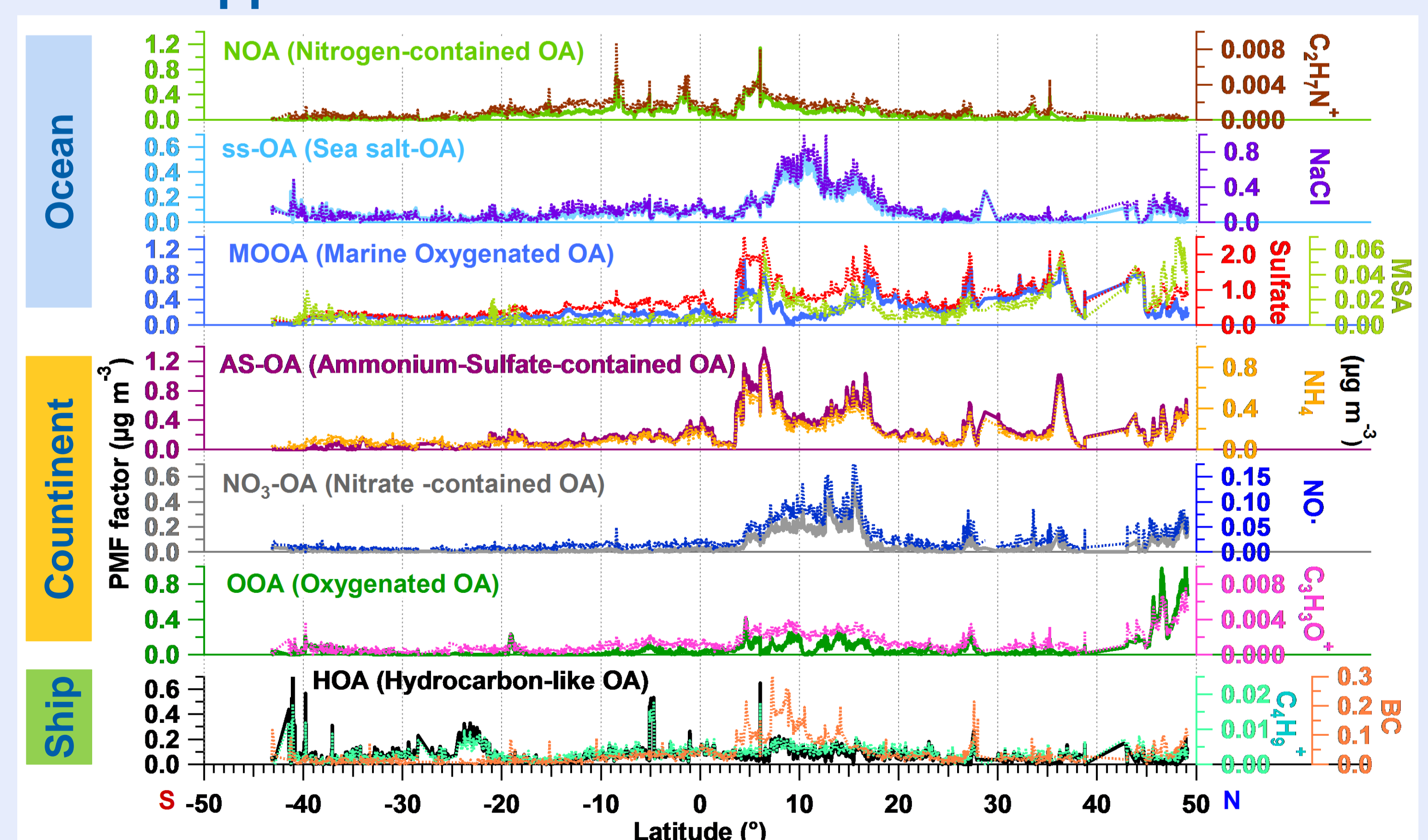


Fig. 6: The mass concentrations of OA (Organic Aerosol) components and corresponding tracers (right axes) vs. the latitude (PMF analysis based on organics + inorganics matrix)

3 OA components are probably influenced by Atlantic Ocean, the other 3 by continents and 1 represents the diffused ship exhaust. $\text{C}_2\text{H}_7\text{N}^+$ is typically from dimethylamine, which was found to be related with algae (Mueller et al., 2009, *Atmos. Chem. Phys.*), hence NOA is regarded as a marine factor.

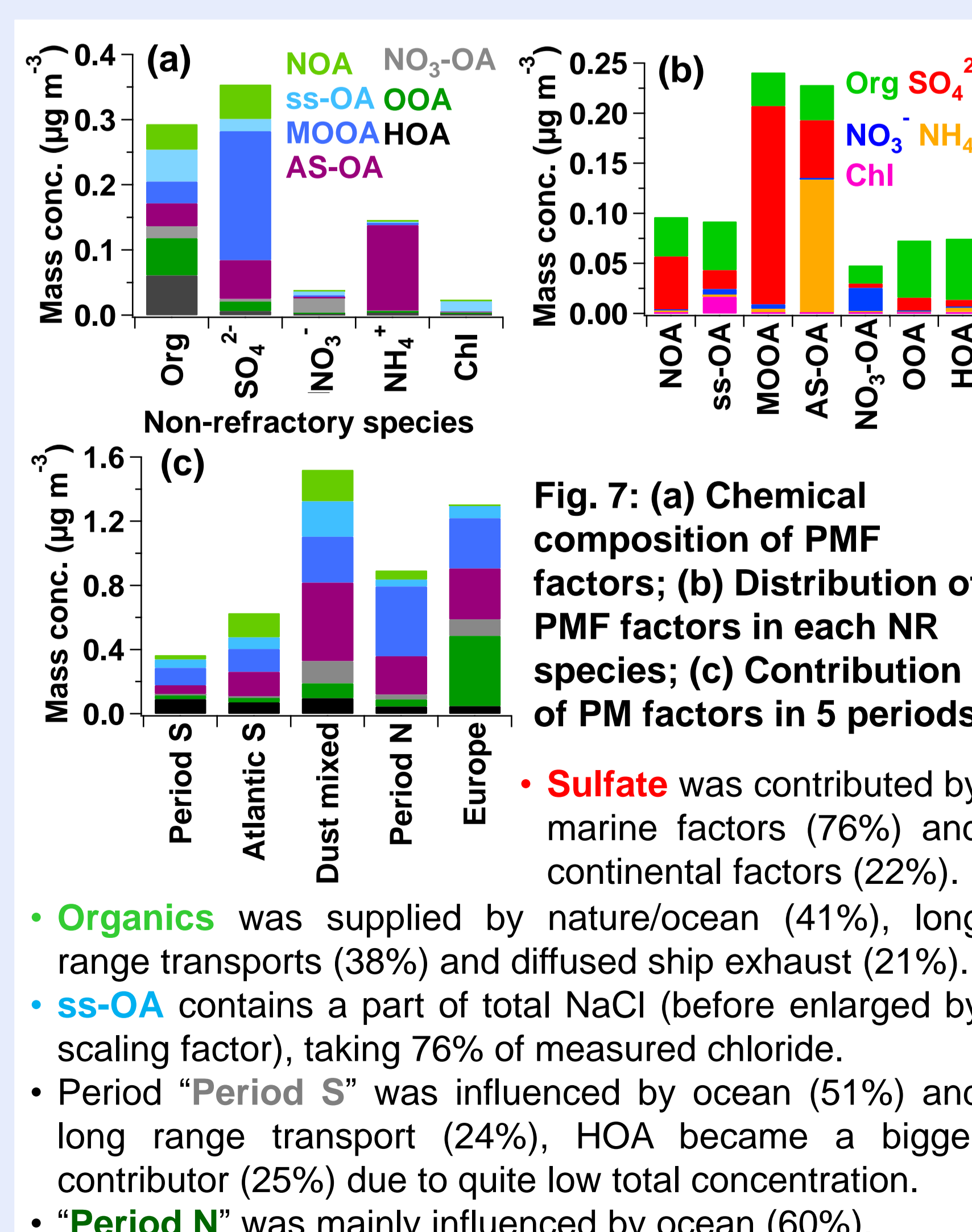


Fig. 7: (a) Chemical composition of PMF factors; (b) Distribution of PMF factors in each NR species; (c) Contribution of PM factors in 5 periods

- **Sulfate** was contributed by marine factors (76%) and continental factors (22%).
- **Organics** was supplied by nature/ocean (41%), long range transports (38%) and diffused ship exhaust (21%).
- **ss-OA** contains a part of total NaCl (before enlarged by scaling factor), taking 76% of measured chloride.
- Period "Period S" was influenced by ocean (51%) and long range transport (24%), HOA became a bigger contributor (25%) due to quite low total concentration.
- "Period N" was mainly influenced by ocean (60%).

SUMMARY

- Sulfate was the *dominant species* of PM1 non-refractory marine aerosol, followed by organics in all 4 cruises.
- During the Cruise ANT XXVIII/5 (CR3), *the main sulfate sources were coming from the ocean* (NOA, ss-OA and MOOA, 76%); only 22% of sulfate was related to long range transport and 2% to diffused ship exhausts.
- *Organics seems to be mainly influenced by ocean (41%)*, followed by long range transport sources (38%) and diffused ship exhaust (21%).
- On the base of air mass classification by back trajectories, PMF results showed more complex sources of each period:
 - Contributions of *marine sources* varied from 30% (Europe) to 60% (Period N, mainly influenced by ocean).
 - Most AS-OA (32%) was found during dust mixed period, indicating a *continent outflow*.
 - OOA took 34% of total mass in period "Europe", also implying *long range transport* from the land.