

UNIVERSITÀ DEGLI STUDI DI MILANO DIPARTIMENTO DI FISICA

PhD First-Year Workshop

## EXPERIMENTAL AND MODELLING APPROACHES TO INVESTIGATE OPTICAL PROPERTIES OF ATMOSPHERIC AEROSOL

Sara Valentini

Atmospheric aerosol (PM): collection of solid and liquid particles suspended in the atmosphere



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Particle composition, size, shape





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Radiative forcing of climate between 1750 and 2011

Forcing agent



Figure 8.15 | Bar chart for RF (hatched) and ERF (solid) for the period 1750–2011, where the total ERF is derived from Figure 8.16. Uncertainties (5 to 95% confidence range) are given for RF (dotted lines) and ERF (solid lines).

IPCC, 2013

#### RF: net flux at tropopause

ERF: net flux at top of atmosphere

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Adapted from IPCC, 2013





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# Aerosol mixing state: impact on aerosol optical properties

Particle mixing state influences aerosol optical properties (e.g. shell of non-absorbing material can enhance absorption by an absorbing core)





Bond and Bergstrom, 2006

Fig. 1. Schematic of the effect of  $C_{\text{Clear}}$  and  $C_{\text{Brown}}$  shells on *BC* absorption. Lack and Cappa, 2010



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Fig. 1. Schematic of the effect of  $C_{\text{Clear}}$  and  $C_{\text{Brown}}$  shells on BCabsorption.





- Problems in models representativeness: assumptions on particle shape and mixing state
- Need to measure atmospheric aerosol optical properties without sample pre-treatment



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Visibility: the greatest distance at which a black object of suitable dimensions can be seen and recognized when observed against the horizon sky [*WMO*, 2008]









Air quality related value





**Objective definition?** 





#### PhD research activity - 1<sup>st</sup> Year



### PhD research activity - 1st Year

- Multi-wavelength measurement of absorption coefficient of atmospheric aerosol samples collected with different time resolution
  - Participation to the international collaborative project CARE (Carbonaceous Aerosol in Rome and Environs), for the determination of absorption and scattering coefficients of samples with high time resolution
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- Feasibility study on an experimental approach to retrieve the aerosol scattering coefficient of aerosol samples using the Polar Photometer developed by the Environmental Physics Research Group (currently used to obtain the aerosol absorption coefficient)





Laser sources for streaker sample analysis

Polar Photometer developed by the Environmental Physics group (PP\_UniMI)









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 Based on the measurement of the angular distribution of the radiation scattered by blank filter (before sampling) and loaded filter (after aerosol collection)





Laser sources for streaker sample analysis





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- Based on the measurement of the angular distribution of the radiation scattered by blank filter (before sampling) and loaded filter (after aerosol collection)
- Optimized for analyses of both low- and hightime resoultion samples collected on 47-mm filters and by streaker samplers







Laser sources for streaker sample analysis





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Loser sources for streaker sample analysis



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### PP\_UniMI: Two-layer radiative transfer model



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Large collaborative project (13 national and international partners)



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On-line and off-line analyses



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Data analysis still in progress...





• Multi-wavelength measurements of aerosol absorption coefficient  $(b_{ap})$  on daily and hourly  $PM_{2.5}$  samples (performed with PP\_UniMI)



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Need to tailor coefficients to make the algorithm site-specific!



Credits to M. Lazzarini, ARPA Lombardia





Main aerosol components:

- Ambient size distributions (measured)
- Densities, complex refractive indices (from literature)





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Tailored dry mass extinction efficiencies

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• Model check and improvement by comparison with parallel direct measurements of  $b_{ext}$  or visual range





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- Model check and improvement by comparison with parallel direct measurements of  $b_{ext}$  or visual range
- Implement the use of the algorithm in standard monitoring networks, in order to obtain visual range as an additional parameter




• Planned activities:



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  - Multi-wavelength determination of  $b_{ap}$  in samples collected in an area heavily impacted by anthropogenic sources (Terni), to be coupled with a detailed chemical characterization (collaboration with Università la Sapienza - Rome): outdoor and indoor (first time!) samples



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  - Possibility of a 6-month period at Vienna University:
    - Improve knowledge of on-line instrumentation measuring aerosol optical properties
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  - Development of a methodology to obtain aerosol scattering coefficients with PP\_UniMI for samples collected on suitable filter media



# Thank you for your attention!

#### **Publications and presentations**

- Bernardoni, V., Elser, M., Valli, G., <u>Valentini, S.</u>, Bigi, A., Fermo, P., Piazzalunga, A., and Vecchi, R.; *Size-segregated aerosol in a hot-spot pollution urban area: Chemical composition and three-way source apportionment*; **Environmental Pollution** 231 (2017), 601-611
- <u>Valentini S.</u>, Bernardoni V., Massabò D., Prati P., Valli G. and Vecchi R.; *Tailored coefficients in the algorithm to assess reconstructed light extinction at urban sites: A comparison with the IMPROVE revised approach*; **Atmospheric Environment** (accepted)
- Vecchi R., Bernardoni V., Fermo P., Piazzalunga A., <u>Valentini S.</u>, and Valli G.; *Assessment of light extinction at a European polluted urban area during wintertime: Impact of PM1 composition and sources*; **Environmental Pollution** (under second review)
- Costabile F., et al.; First results of the "Carbonaceous aerosol in Rome and Environs (CARE)" experiment: beyond current standards for PM10; Atmosphere (submitted)
- <u>Valentini S</u>., Bernardoni V., Massabò D., Prati P., Valli G. and Vecchi R.; *Tailoring coefficients in IMPROVE algorithm to assess site-specific chemical light extinction*; poster presentation at **Congresso del Dipartimento di Fisica 2017**
- Forello A.C., Bernardoni V., Calzolai G., Chiari M., Lucarelli F., Massabò D., Nava S., Prati P., <u>Valentini S.</u>, Valli G., Vecchi R.; *High-time* resolved atmospheric aerosol characterization for source apportionment studies; poster presentation at Congresso del Dipartimento di Fisica 2017
- <u>Valentini S.</u>, Bernardoni V., Massabò D., Prati P., Valli G. and Vecchi R.; *Light extinction estimates using the IMPROVE algorithm: The relevance of site-specific coefficients*; poster presentation at European Aerosol Conference EAC 2017
- Bernardoni V., Forello A.C., Mariani F., Paroli B., Potenza M.A.C., Pullia A., Riccobono F., Sanvito T., <u>Valentini S.</u>, Valli G., Vecchi R.; *Innovative instrumentation for the study of atmospheric aerosol optical properties*; Proceedings in Physics - Congresso del Dipartimento di Fisica, Springer (submitted)





#### Radiative transfer model: 2-layer scheme





#### Radiative transfer model: Adding method





#### Radiative transfer model: Adding method





Particles [Hänel, 1987]

Scheme of radiative processes in a loaded filter

Tp

X TpBf+FpBf

X 8° B4

 $X (B_p^*)^2 (B_f^*)^2$ 

FpBf

X B<sub>p</sub>\*

X (B<sub>p</sub>\*)<sup>2</sup>B<sub>f</sub>\*

X (Bp\*)<sup>3</sup>(Bf\*)<sup>2</sup>

 $T_p$  = transmittance of the particle layer

 $F_p$  = fraction of parallel incident light scattered in the forward hemisphere by the particle layer

Incident (=1)

Bpf

X P<sub>p</sub>\*

X Bp\*Bf\*Pp\*

 $X (B_{p}^{*})^{2} (B_{f}^{*})^{2} P_{p}^{*}$ 

 $B_n$  = fraction of parallel incident light scattered in the backward hemisphere by the particle layer

 $P_p^*$  = fraction of light scattered by the filter and re-scattered by the particle layer towards the forward hemisphere

 $B_p^*$  = fraction of light scattered by the filter and re-scattered by the particle layer towards the backwards hemisphere





T<sub>p</sub>P<sub>f</sub>

F<sub>p</sub>P<sub>p</sub>\*

X B<sub>p</sub>\*P<sub>f</sub>\*

X (Bp\*)<sup>2</sup>Bf\*Pf\*

X (Bp\*)3(Bf\*)2Pf\*

Filter

P<sub>pf</sub>

### Radiative transfer model: **Two-stream approximation**



It can be decomposed in:  $I^{+}(\tau, \mu) = I(\tau, \mu) \in I^{-}(\tau, \mu) = I(\tau, -\mu)$  for  $0 \le \mu \le 1$ 

The two-stream approximation states that:  $I^+(\tau,\mu) = I^+(\tau) \in I^-(\tau,\mu) = I^-(\tau)$  (introduction of «effective» intensities, hemisphere-dependent)

Boundary conditions for an external source (parallel beam):

$$I^{-}(\tau = 0) = I_{0}$$
  
 $I^{+}(\tau = \tau^{*}) = 0$ 

scattering and absorbing atmosphere



Boundary conditions for an internal source (diffuse radiation):

$$I^{-}(\tau = 0) = 0$$
  
 $I^{+}(\tau = \tau^{*}) = 0$ 



### Radiative transfer model: Two-stream approximation



For diffuse radiation terms:

$$B_{p}^{*} = \frac{b(1 - T_{p}^{2\sqrt{B}})}{\sqrt{B} + a + (\sqrt{B} - a)T_{p}^{2\sqrt{B}}}$$
$$P_{p}^{*} = \frac{1}{2\sqrt{B}} \Big[ (\sqrt{B} - a + B_{p}^{*}b)T_{p}^{-\sqrt{B}} + (\sqrt{B} + a - B_{p}^{*}b)T_{p}^{\sqrt{B}} \Big]$$
$$a = 2 \Big[ 1 - \omega_{p}(1 - \beta_{p}^{*}) \Big], \ b = 2\omega_{p}\beta_{p}^{*}, \ B = a^{2} - b^{2}$$

For parallel incident radiation terms:

$$B_{p} = \frac{c - \frac{p_{1}}{1 + \sqrt{B}} - \left(c - \frac{p_{1}}{1 - \sqrt{B}}\right) T_{p}^{2\sqrt{B}} - \frac{2p_{1}\sqrt{B}}{1 - B} T_{p}^{1 + \sqrt{B}}}{\sqrt{B} + a + (\sqrt{B} - a) T_{p}^{2\sqrt{B}}}$$

$$F_{p} = \frac{1}{2\sqrt{B}} \left[ \left(d + B_{p}b + \frac{p_{2}}{1 + \sqrt{B}}\right) T_{p}^{-\sqrt{B}} - \left(d + B_{p}b + \frac{p_{2}}{1 - \sqrt{B}}\right) T_{p}^{\sqrt{B}} \right] + \frac{p_{2}}{1 - B} T_{p}$$

$$c = \omega_{p}\beta_{p}, d = \omega_{p}(1 - \beta_{p}), p_{1} = c - ac - bd, p_{2} = -ad - bc - d$$

Where  $\beta_p$  and  $\beta_p^*$  are ratios of backscattered radiation to collimated and diffuse radiation, respectively, and are related to the asymmetry parameter g(assumption: g = 0.75 for atmospheric particles).

$$\beta_{p} = \frac{1}{2} \left[ 1 - g - \frac{4}{25} \left( 1 - \frac{|1 - 2g|}{8} - \frac{7}{8} (1 - 2g)^{2} \right) \right]$$
$$\beta_{p}^{*} = \frac{1}{2} \left[ 1 - \frac{g}{4} \left( 3 + g^{3 + 2g^{3}} \right) \right]$$
Intensity scattered at angle  $\theta$ 
$$g = \frac{1}{2} \frac{\int_{0}^{\pi} \cos \theta F(\theta) \sin \theta d\theta}{\int_{0}^{\pi} F(\theta) \sin \theta d\theta} = \frac{1}{2} \int_{0}^{\pi} \cos \theta F(\theta) \sin \theta d\theta$$
Phase function



### Radiative transfer model: $b_{ap}$ calculation

Given the assumption on g (thus  $\beta_p$  and  $\beta_p^*$  known), only 2 unknown quantities remain in the expression obtained by the adding method:  $\omega_p$  and the transmittance of particle layer  $T_p$ 

## Modelling approaches for the evaluation of $b_{ext}$





### Modelling approaches for the evaluation of $b_{ext}$ : visibility estimates

Example:

Application of the tailored approach to reconstruct light extinction for 1-week period (Milan, 2015)



Comparison of estimated visual range (VR) with visibility measured at Linate airport

Good agreement, BUT slope < 1! (non-idealities in measurements performed at airports)

- Possible reduction of the Koschmieder constant  $(-\ln 0.02 = 3.912)$  (already observed...)
- Need to compare the model results with directly measured  $b_{ext}$  or VR



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#### Modelling approaches for the evaluation of $b_{ext}$ : visibility estimates

#### Example:

Application of the tailored approach to reconstruct light extinction at an urban site

b <sub>ext</sub> (in Mm⁻¹)	Total	Amm. Sulphate	Amm. Nitrate	ОМ	<b>b</b> <sub>ap</sub>	Fine Soil	Coarse Mass	Rayleigh Scattering	NO2	Visual Range (km)
mean	287.2	24.1	108.9	77.1	28.2	0.8	21.1	12.0	14.9	18.8
std. dev.	158.1	20.0	88.1	44.9	15.2	0.5	11.6	0.2	4.4	13.2
min	45.0	1.6	1.4	12.4	5.5	0.3	1.9	11.5	4.9	4.3
max	919.9	111.5	510.9	214.4	75.1	4.8	64.8	12.5	26.1	86.9
average percentage		8.5%	34.1%	27.0%	10.5%	0.4%	7.3%	5.7%	6.4%	

#### Light extinction apportioned by sources



Range of VR during this period: 4.3-86.9 km Similar to values reported by literature studies: 10 km: polluted atmosphere 80-100 km: clear air

Possibility of tackling visibility impairment acting directly on PM source emissions

