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Supplement of

Porous aerosol in degassing plumes of Mt. Etna and Mt. Stromboli

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Application of the Dubovik's code

Detailed information on the Dubovik's code can be found in the works by Dubovik et al. (2006), Dubovik et al. (2011) and references therein. This supplement reproduces some basic information from those publications and underscores details of the code application in our work.

S1 Kernels for direct and inverse problems

Direct simulations and retrievals use the concept developed by Dubovik et al. (2006) and model the particles for each size bin as a mixture of spherical and non-spherical aerosol components. The nonspherical component was modeled by ensembles of randomly oriented spheroids (ellipsoids of revolution). Aerosol single-scattering properties are approximated utilizing look-up tables of scattering kernels.

The kernels were computed at grid points. The grid points are equidistant for the real part n of the refractive index, logarithmically equidistant for the spheroid axis ratio, i.e., aspect ratio ε, logarithmically equidistant for the imaginary part χ of the refractive index, and logarithmically equidistant for particle radius r bins. The size parameter $x = 2\pi r/\lambda$, where λ is the wavelength. We employed the code version where those parameters are varying in the following ranges.

$1.29 \leq n \leq 1.696$,

$0.0005 \leq \chi \leq 0.5$,

$0.3 \leq \varepsilon \leq 3.0$,

$0.012 \leq x \leq 626$.

The surface of particles can be smooth $\sigma = 0.0$ or severely rough $\sigma = 0.2$, where σ specifies the magnitude of roughness (Yang et al., 2013). According to Yang and Liou (1998), $\sigma = 0. - 0.005$, $\sigma = 0.005 - 0.05$ and, $\sigma = 0.05 - 0.2$ correspond to slight, moderate, and severe roughness in terms of smoothing corresponding phase functions.

An advanced version of the look-up tables of scattering kernels was used in the work by Kolokolova et al. (2015) were the rough spheroid model was employed to simulate polarization properties of cosmic dust.

S2 Input parameters for inverse problem

The Dubovik's package, we used to retrieve aerosol characteristics, has a large set of input parameters. The following lists present the main subset of the parameters, i.e., the characteristics that are necessary to explain the approach of our supervised retrievals.

The parameters related to the instrumentation and experimental data are the following:

- the number and the values of scattering angles at which the Polar Nephelometer (PN) measures angular scattering intensities (ASIs);
- the measured values of the angular scattering intensities;
- the wavelength of the PN laser beam.

The following parameters are assigned by an operator in the input file of the inversion code.

- the regularization parameter, i.e., the smoothness parameter;

- the number of the bins of the size distributions;
- the minimum particle size;
- the maximum particle size;

- the initial guess for the real part of the refractive index;
- the initial guess for the imaginary part of the refractive index;

- the surface roughness σ parameter.

S3 Output characteristics

The package, we used in this work, provides the following output characteristics as the retrieval results.

- the residuals;
- the retrieved value of the spherical/non-spherical partitioning ratio (SNR);
- the retrieved value of the real part of the refractive index;
- the retrieved value of the imaginary part of the refractive index;
- the retrieved values of the size distribution;
- the phase function computed for the retrieved size distribution and the refractive index;
- the value of the extinction coefficient computed for the retrieved size distribution and the refractive index.

S4 Supervised retrievals

We employed the code version where the surface roughness σ is not retrieved and has to be assigned. It can take only two values, i.e., either $\sigma = 0.0$ or $\sigma = 0.2$. Thus, supervised retrievals have to be performed at each value of the surface roughness σ individually.

The peculiarity of the inversion code, we used in this work, consists in the fact the executable file works with only one input file and it provides one corresponding output file. In other words, an operator has to prepare a set of input files and has to analyze the corresponding set of output files when the parameters mentioned in Section S2 are varied.

In the following, we try to explain our retrieval approach using programming terms or more specifically, **for**-loops of the C++ Language.

```
for (minimum particle size)
  {
   for (maximum particle size)
     {
       for (regularization parameter)
          {
```

```
        for (initial guess for the refractive index)
          {
              Retrievals
          }
        }
    }
}
```

where "**for** (parameter)" means that the parameter takes a set of values. The set of values is chosen/assigned by an operator. For example, the set of the regularization parameter is assigned within the range that contains the value corresponding to the "L-curve" method.

It can be seen, inverse problem is solved on the multidimensional grid of the input parameters. The accepted solution to the inverse problem corresponds to the minimum of the residuals. And, we notice that the minimum and the maximum sizes of particles belong to the set of assessed parameters in addition to the characteristics listed in Section S3.
Generally, the approach is time consuming. On the other hand, it assures that the obtained solution corresponds to the global minimum of an objective function in a generally non-linear case.

S5 Retrieval-quality assurance

The final step consists in the verification whether retrieval results are scarcely affected by small variations of the input parameters. The supervised retrievals are performed another time but on the smaller grid of the input parameter and the regularization parameter is varied within a tighter range.

```
for (regularization parameter)
  {
    for (initial guess for the refractive index)
      {
          Retrievals
      }
  }
```

The final solution is considered to be stable, i.e., of good quality, when the retrieved values of the refractive index and of the spherical/non-spherical partitioning ratio are the same; and the variations of the residuals and of the retrieved size distribution are small.

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