# S1. List of a-priori smoothing constraints

Table S1 provides the list of the a-priori smoothing constraints used for the optimum retrieval presented in Sect. S2, for each selected case from the EARLINET COVID-19 campaign, containing mostly anthropogenic particles (based on data and back-trajectory analysis).

**Table S1.** List of the a-priori smoothing constraints that provided the optimum solution for each of the cases studied herein, for each EARLINET station. OFD is the Order of Finite Difference, LM is the Lagrange multiplier, FM is the fine mode and CM is the coarse mode.

| EARLINET station ID | | Smoothness constraints | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Volume size distribution | | | Real part of refractive index | | | Imaginary part of refractive index | | | Concentration profile | | |
| | | OFD | LM FM | LM CM | OFD | LM FM | LM CM | OFD | LM FM | LM CM | OFD | LM FM | LM CM |
| AKY | 23 May, 2020 | 3 | 0.05 | 0.1 | 1 | 50 | 100 | 1 | 0.5 | 1 | 3 | 0.005 | 0.005 |
| | 25 May, 2020 | 3 | 0.005 | 0.05 | 1 | 30 | 30 | 1 | 0.1 | 0.1 | 3 | 0.05 | 0.05 |
| BAR | 18 May, 2020 | 3 | 0.005 | 0.005 | 1 | 30 | 40 | 1 | 1 | 1 | 3 | 0.002 | 0.002 |
| | 25 May, 2020 | 3 | 0.005 | 0.005 | 1 | 30 | 40 | 1 | 1 | 1 | 3 | 0.002 | 0.002 |
| COG | 4 May, 2020 | 3 | 2 | 0.5 | 1 | 1000 | 1000 | 1 | 10 | 10 | 3 | 0.00001 | 0.00001 |
| KUO | 23 May, 2020 | 3 | 150 | 0.5 | 1 | 500 | 500 | 1 | 1 | 0.3 | 3 | 0.0002 | 0.0002 |
| | 24 May, 2020 | 3 | 150 | 0.5 | 1 | 500 | 500 | 1 | 1 | 0.3 | 3 | 0.0002 | 0.0002 |
| SAL | 7 May, 2020 | 3 | 0.1 | 0.1 | 1 | 300 | 300 | 1 | 2 | 2 | 3 | 10 | 10 |
| | 27 May, 2020 | 3 | 0.1 | 0.1 | 1 | 300 | 300 | 1 | 2 | 2 | 3 | 10 | 10 |
| WAW | 22 May, 2020 | 3 | 3 | 1 | 1 | 2000 | 2000 | 1 | 1.5 | 1.5 | 3 | 0.0002 | 0.0002 |
| | 26 May, 2020 | 3 | 0.05 | 0.5 | 1 | 2000 | 2000 | 1 | 0.5 | 2.5 | 3 | 0.0002 | 0.0002 |
| PAY | 6 May, 2020 | 3 | 1 | 0.1 | 1 | 300 | 300 | 1 | 2 | 2 | 3 | 0.01 | 0.01 |
| | 20 May, 2020 | 3 | 1 | 0.1 | 1 | 300 | 300 | 1 | 2 | 2 | 3 | 0.01 | 0.01 |

# S2. Case studies

This section documents each selected case from the EARLINET COVID-19 campaign, expected to contain mostly anthropogenic particles (based on data and back-trajectory analysis).

For each case is reported:

- the comparison of the optimum retrievals against the SCC products (e.g. Fig. S1)
- the back-trajectory analysis from 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at the corresponding EARLINET station (e.g. Fig. S2)
- the time-height plot of the attenuated backscatter coefficient at 1064 nm, or 532 nm in case the 1064 nm channel was not available, and volume linear depolarization ratio at 532 nm, if available (e.g. Fig. S3)
- the AOD measurements at 500 nm from the CIMEL sunphotometer, and the product of fine mode fraction (e.g. Fig. S4)
- the calculation of the center-of mass for the retrieved concentration profile (e.g. Fig. S5)

The cases are presented in chronological order.

## S2.1 Belsk May 4, 2020
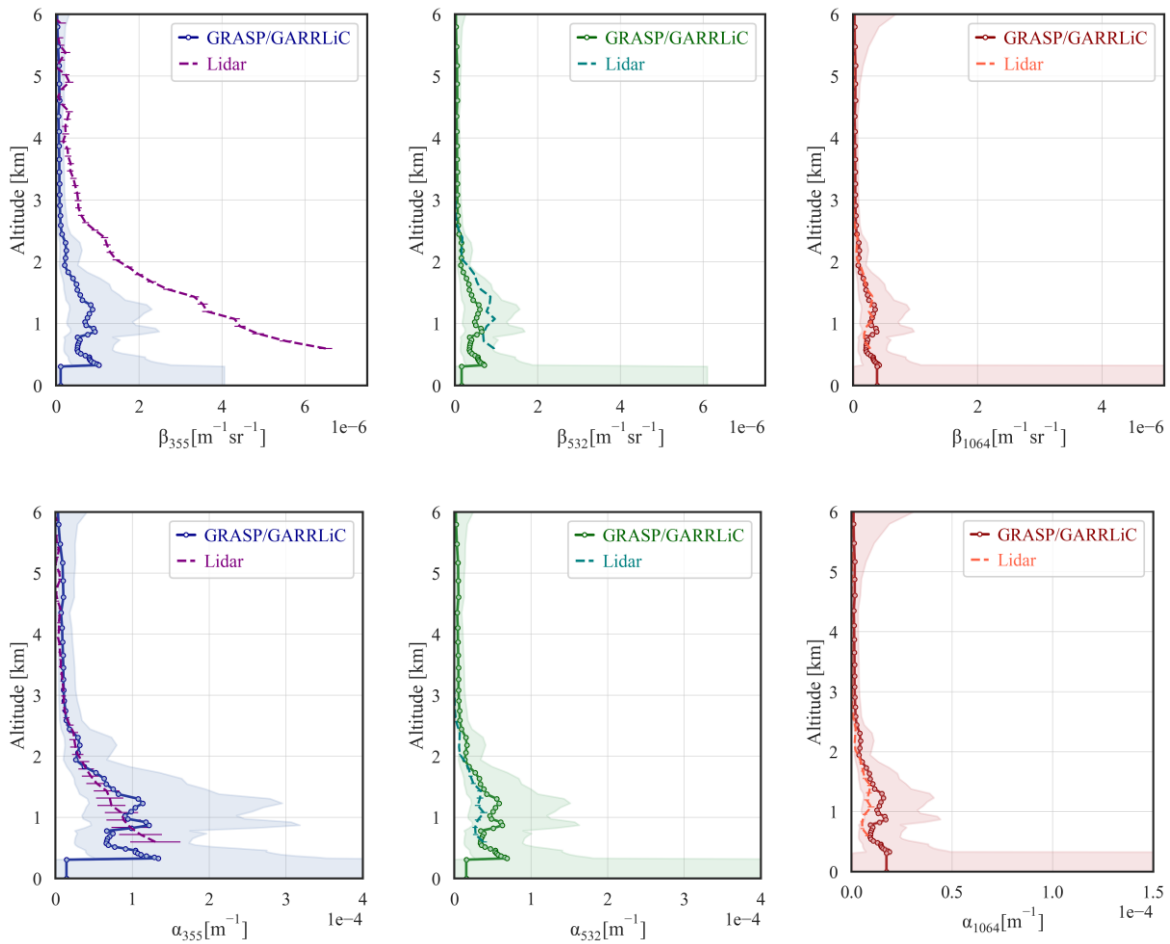
**Fig. S1)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left), 532 (middle) and 1064 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 06:15 and 07:15 UTC, 4 May, 2020.
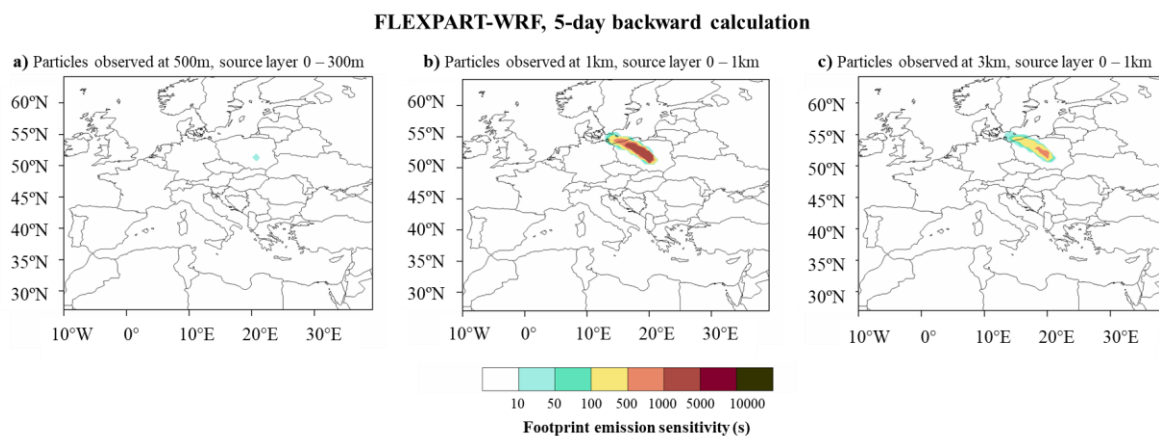
**Fig. S2)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Belsk station, on May 4, 2020, at 14:25 UTC at heights a) 500m, b) 1km and c) 3km.
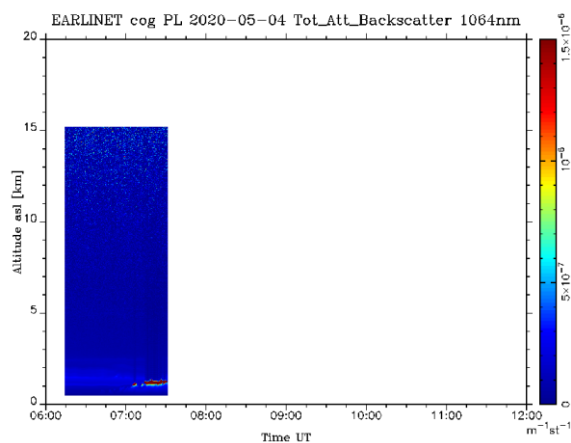
**Fig. S3)** Time-height plot of the attenuated backscatter coefficient at 1064 nm measured at Belsk station on May 4, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 06:15 - 06:45 UTC) the majority of the aerosol load is found below 3 km.
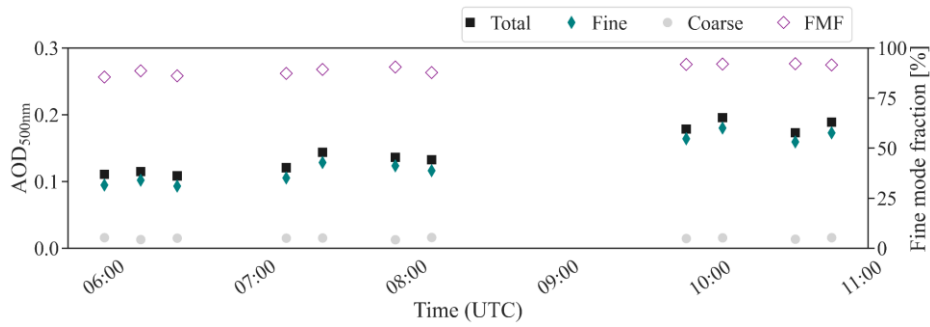
**Fig. S4)** Sun-photometer measurements from Belsk station on May 4, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 6:30 UTC, so as to overlap with lidar measurements.

**Fig. S5)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Belsk station on May 4, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

## S2.2 Payerne 6 May, 2020
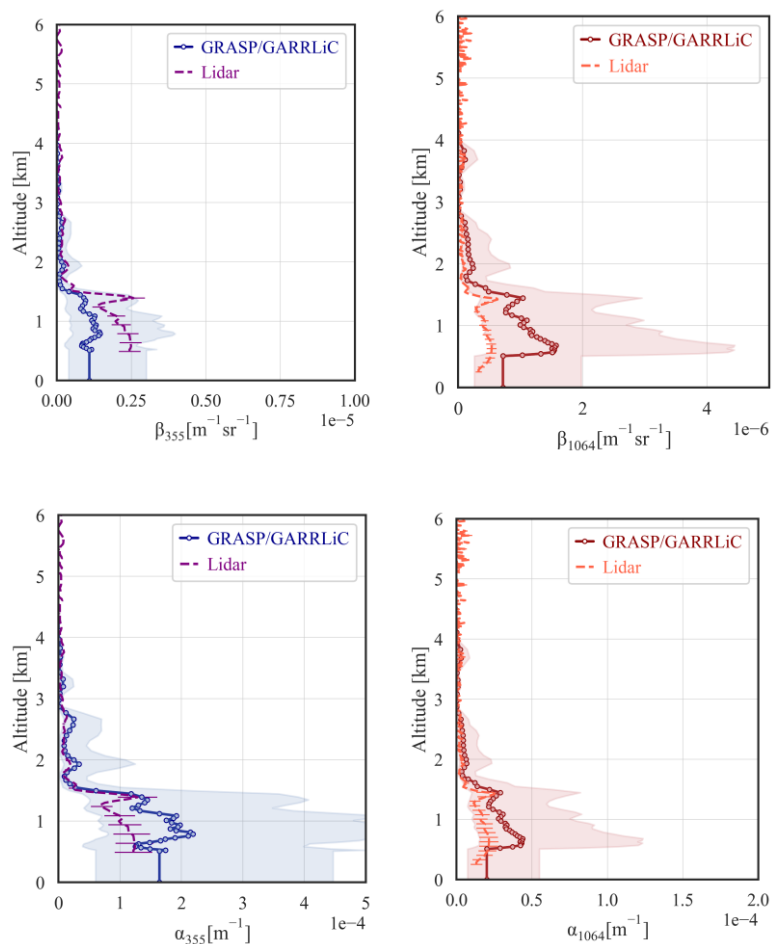
**Fig. S6)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left), and 1064 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 15:30 and 16:30 UTC, 6 May, 2020.
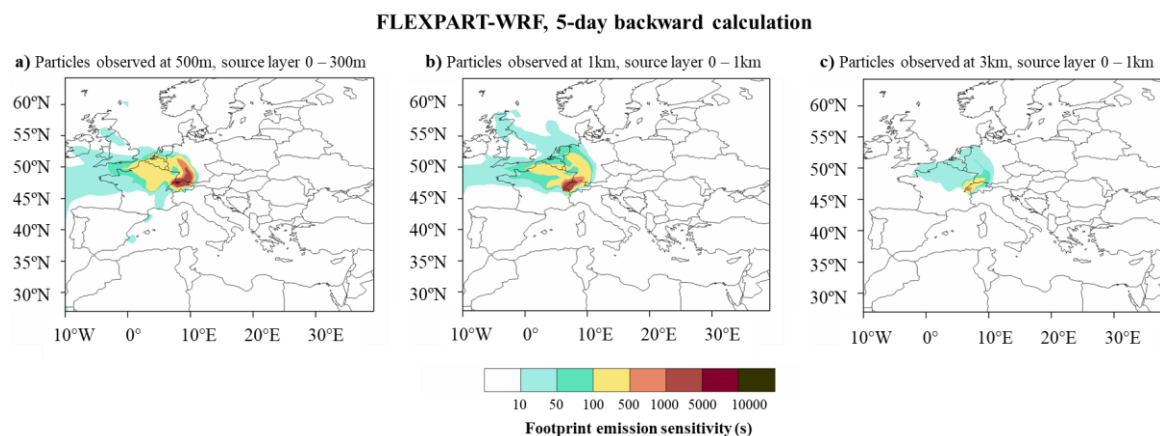
**Fig. S7)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Payerne station, on May 6, 2020, at 16:00 UTC at heights a) 500m, b) 1km and c) 3km.
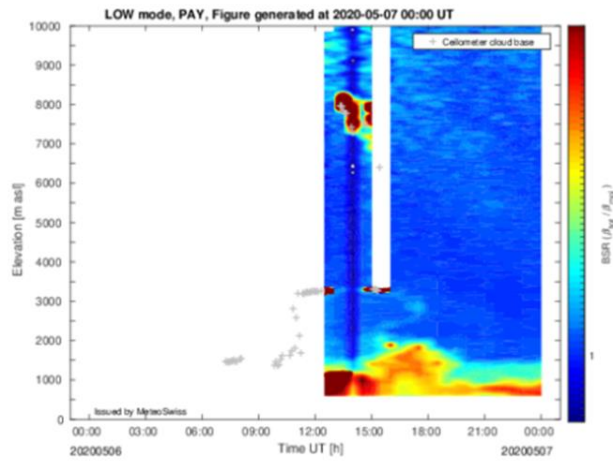
**Fig. S8)** Time-height plot of the attenuated backscatter ratio at 1064 nm measured at Payerne station on May 6, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 15:00 - 16:00 UTC) the majority of the aerosol load is found below 3 km.
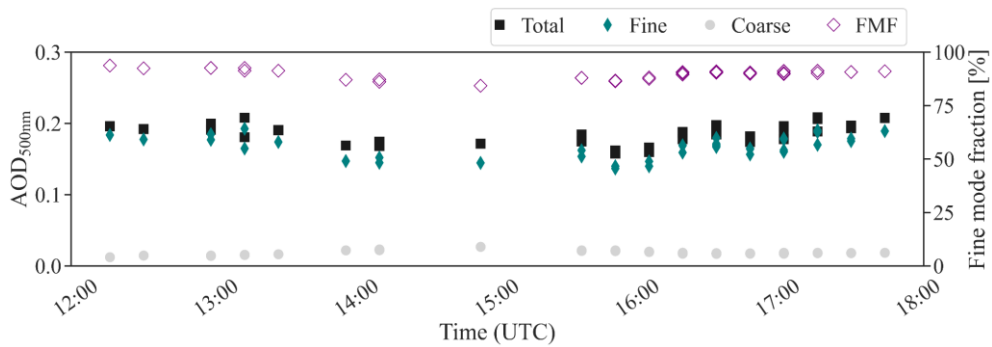
**Fig. S9)** Sun-photometer measurements from Payerne station on May 6, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 15:57 UTC, so as to overlap with lidar measurements.
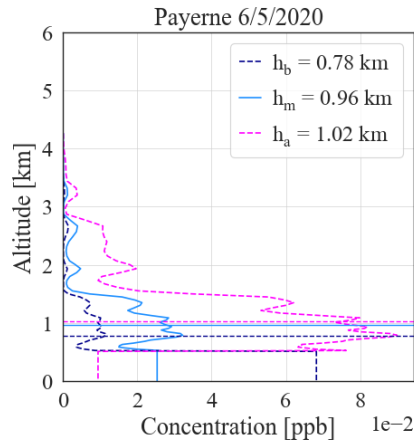
**Fig. S10)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Payerne station on May 6, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

## S2.3 Lecce 7 May, 2020
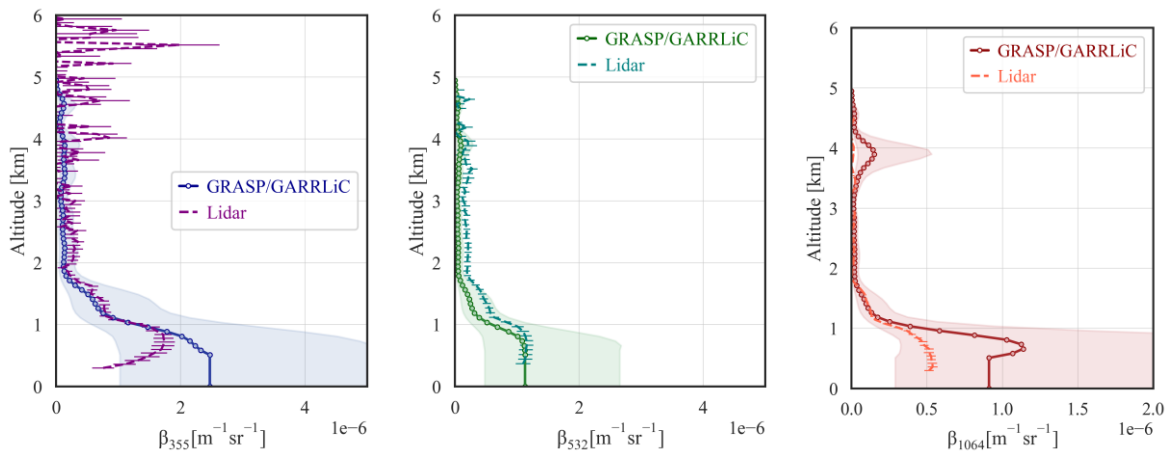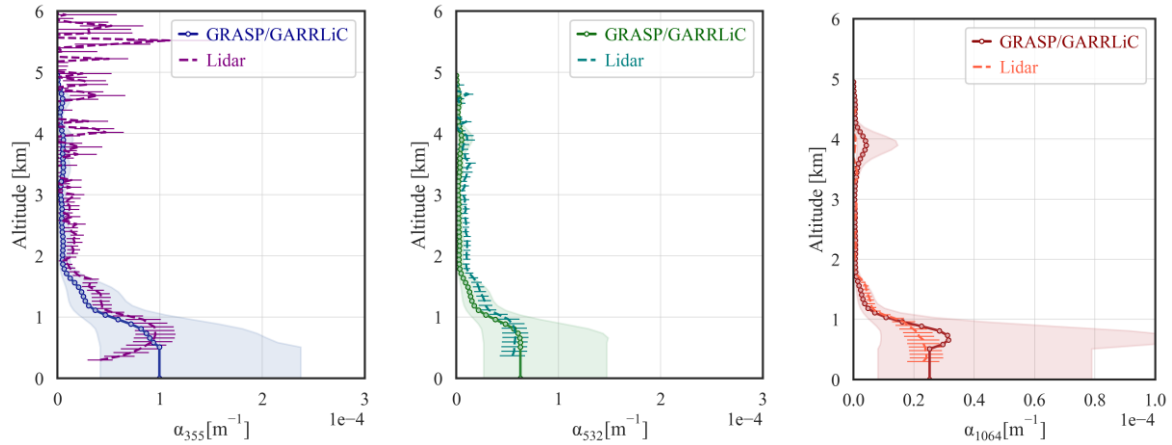
**Fig. S11)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left), 532 (middle) and 1064 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 12:27 and 13:27 UTC, 7 May, 2020.
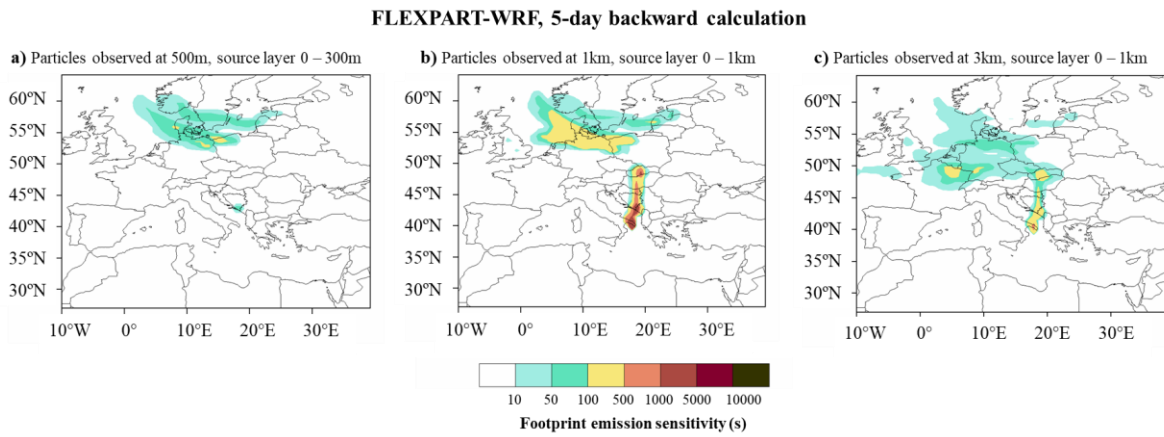
**Fig. S12)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Lecce station, on May 7, 2020, at 13:56 UTC at heights a) 500m, b) 1km and c) 3km.
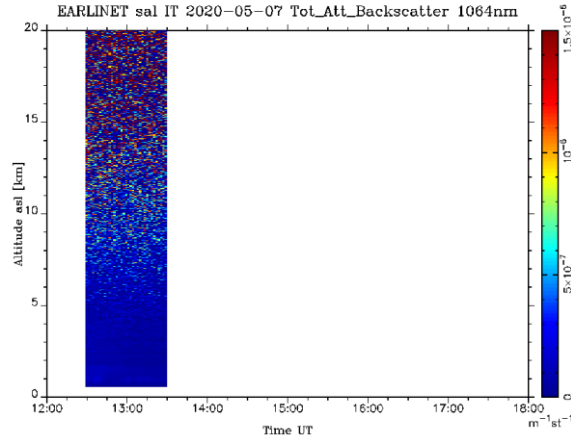
**Fig. S13)** Time-height plot of the attenuated backscatter coefficient at 1064 nm measured at Lecce station on May 7, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 12:27 - 13:27 UTC) the majority of the aerosol load is found below 2 km.
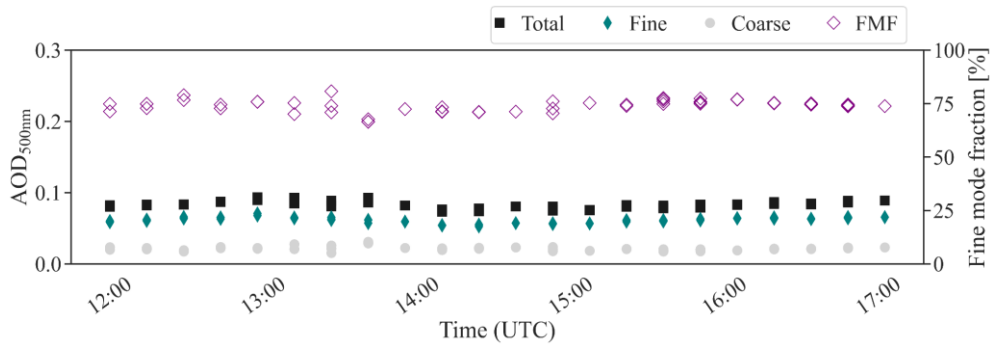
**Fig. S14)** Sun-photometer measurements from Lecce station on May 7, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 13:56 UTC, so as to be close in time with the corresponding lidar measurements.
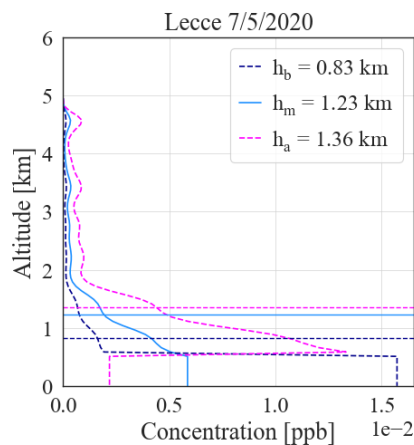
**Fig. S15)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Payerne station on May 7, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

## S2.4 Barcelona 18 May, 2020
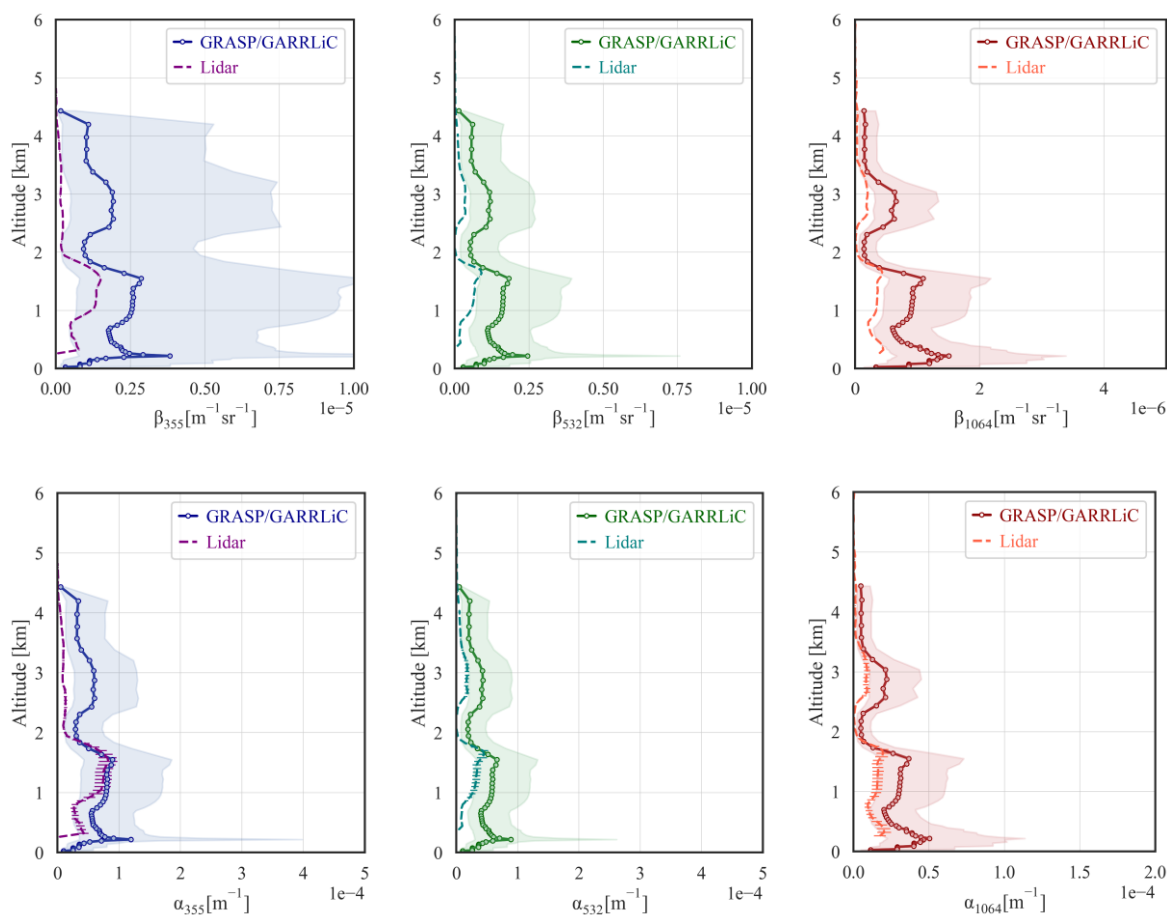
**Fig. S16)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left), 532 (middle) and 1064 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 08:25 and 09:05 UTC, 18 May, 2020.
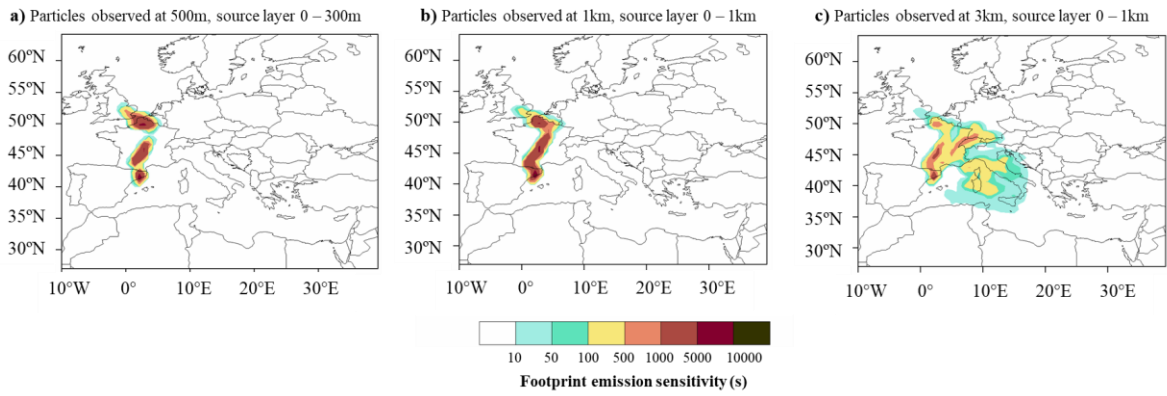
**Fig. S17)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Barcelona station, on May 18, 2020, at 08:50 UTC at heights a) 500m, b) 1km and c) 3km.
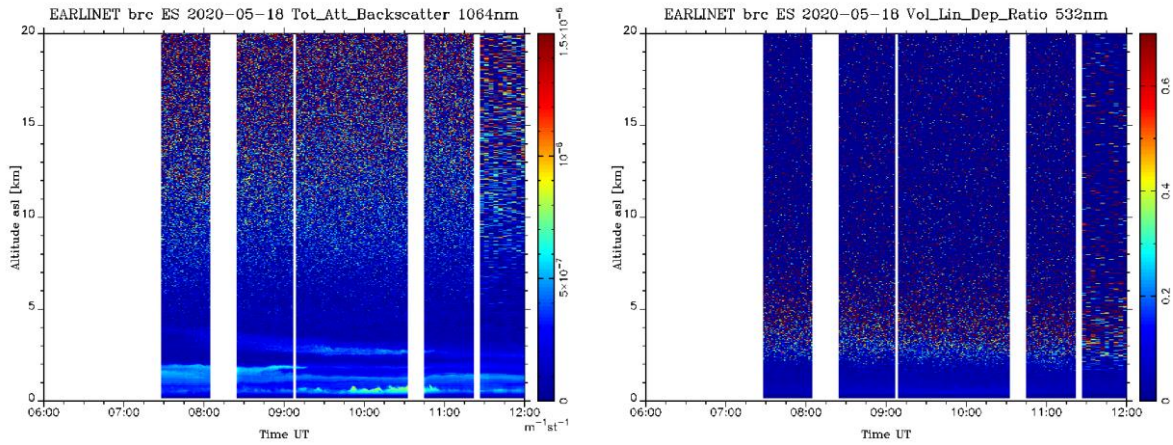
**Fig. S18)** Time-height plot of the attenuated backscatter coefficient at 1064 nm and volume linear depolarization ratio at 532 nm measured at Barcelona station on May 18, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 08:25 - 09:05 UTC) the majority of the aerosol load is found below 5 km.
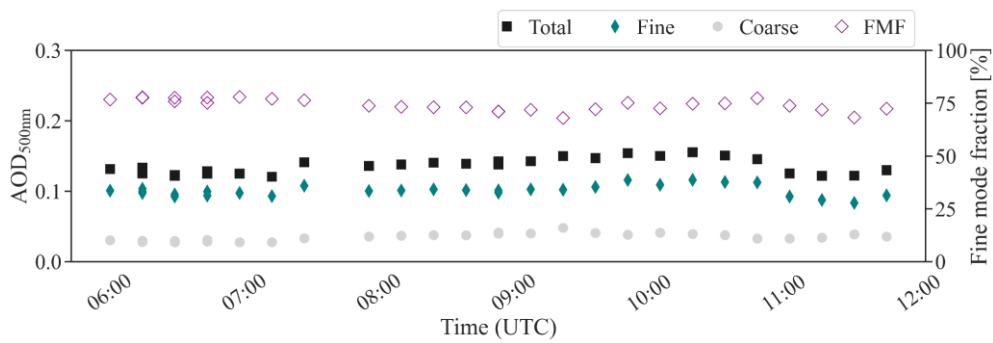
**Fig. S19)** Sun-photometer measurements from Barcelona station on May 18, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 8:50 UTC, so as to overlap with the lidar measurements.
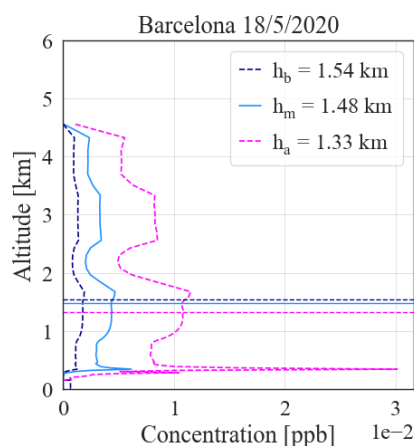
**Fig. S20)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Barcelona station on May 18, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

## S2.5 Payerne 20 May, 2020
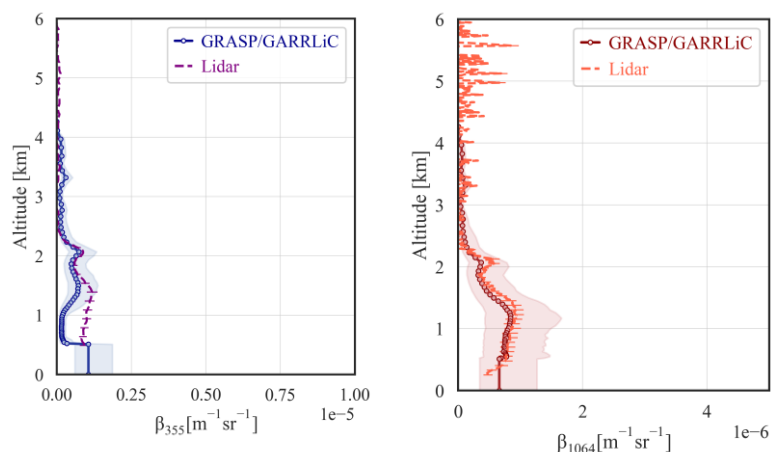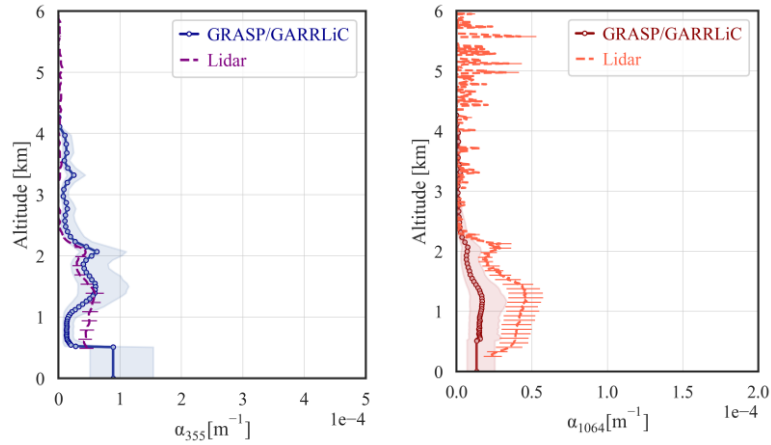
**Fig. S21)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left) and 1064 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 14:30 and 15:30 UTC, 20 May, 2020.
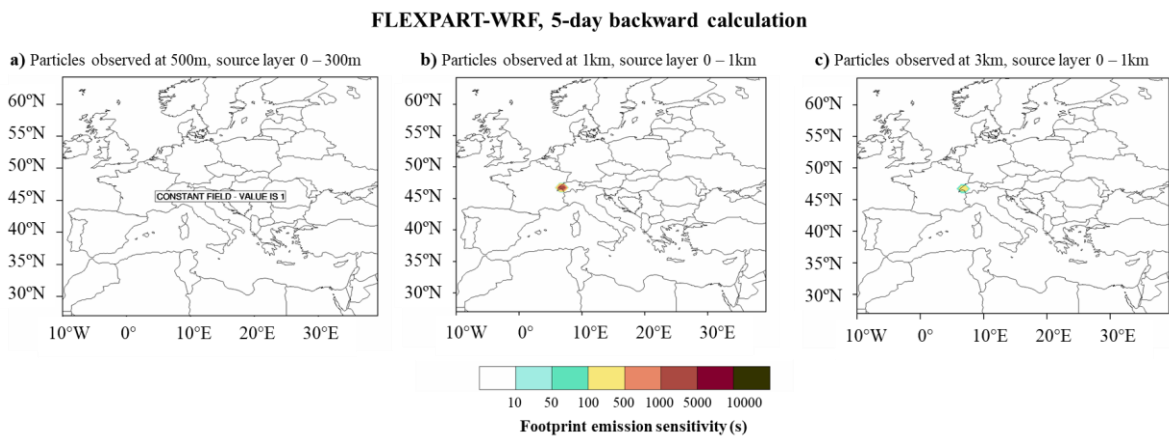
**Fig. S22)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Payerne station, on May 20, 2020, at 14:40 UTC at heights a) 500m, b) 1km and c) 3km.
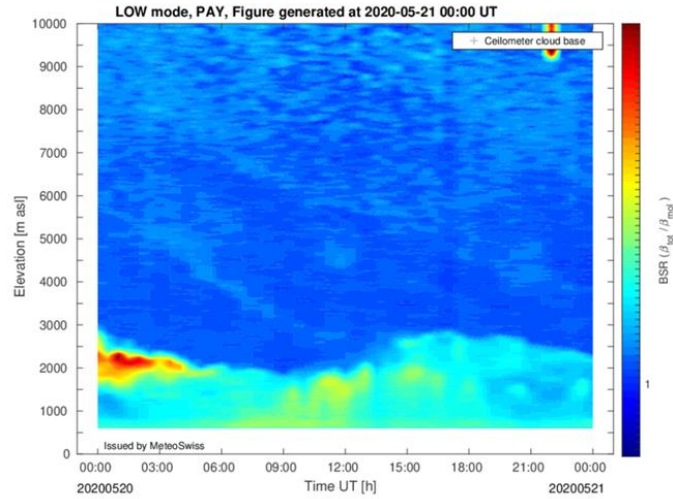
**Fig. S23)** Time-height plot of the attenuated backscatter ratio at 1064 nm measured at Payerne station on May 20, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 14:30 - 15:30 UTC) the majority of the aerosol load is found below 3 km.
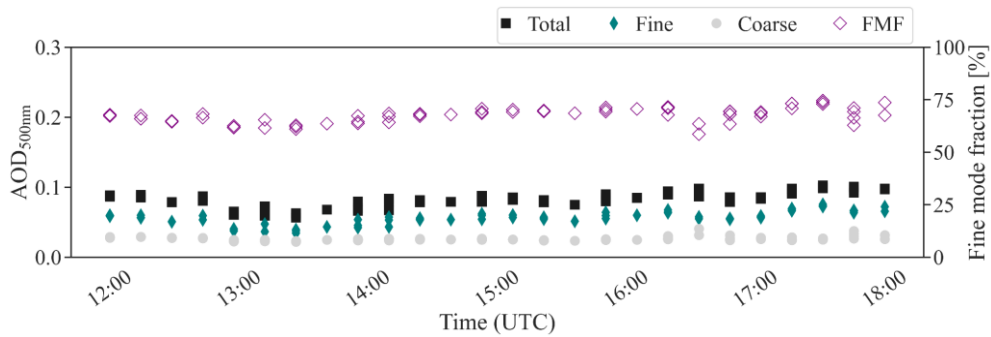
**Fig. S24)** Sun-photometer measurements from Payerne station on May 20, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 14:40 UTC, so as to overlap with lidar measurements.
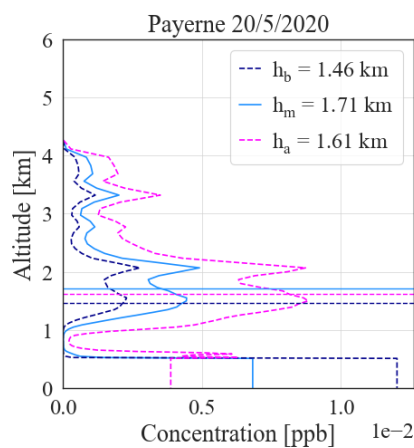
**Fig. S25)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Payerne station on May 20, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

## S2.6 Warsaw 22 May, 2020
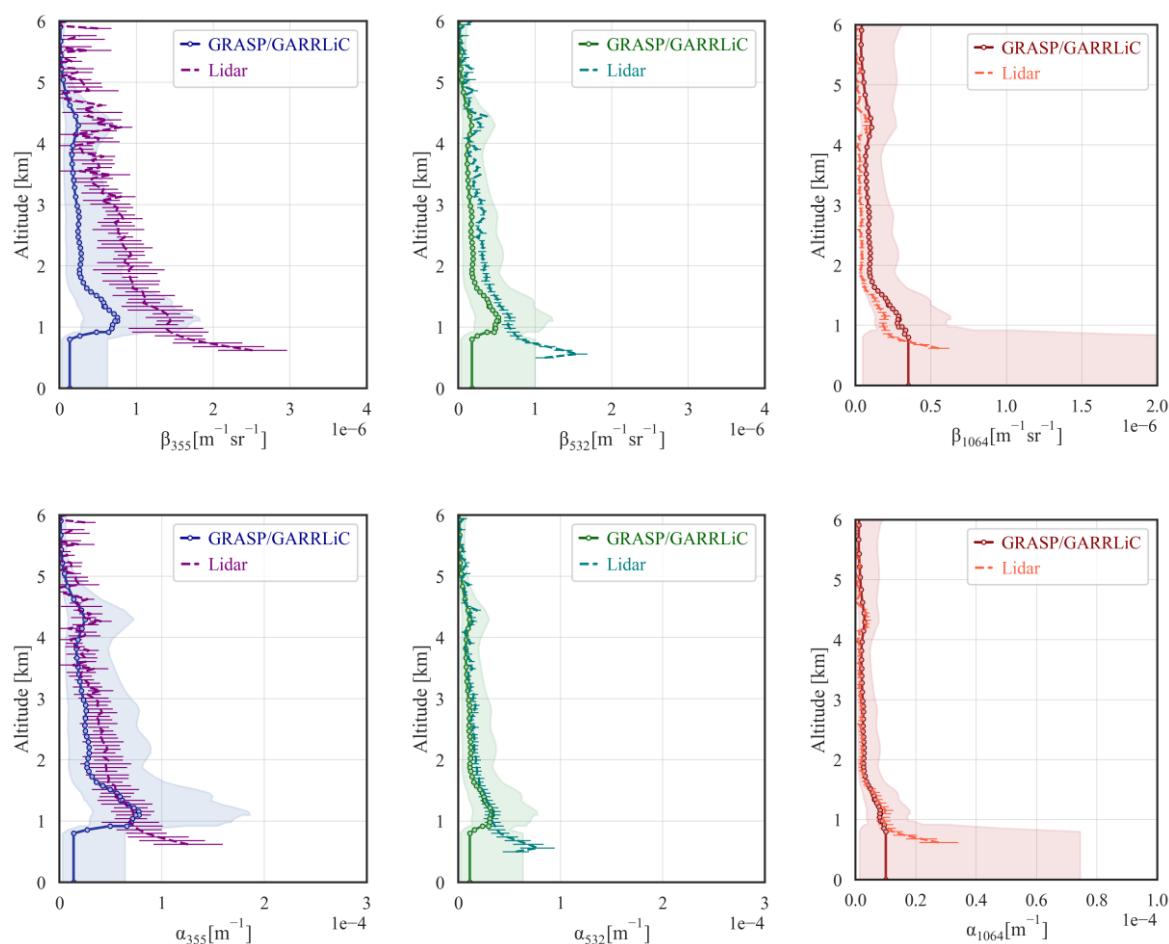
**Fig. S26)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left), 532 (middle) and 1064 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 06:00 and 06:55 UTC, 22 May, 2020.
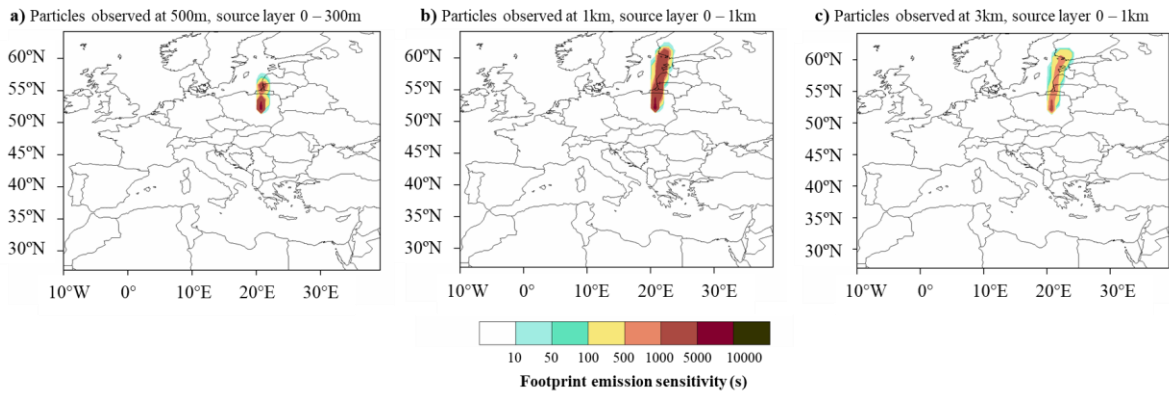
**Fig. S27)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Warsaw station, on May 22, 2020, at 06:16 UTC at heights a) 500m, b) 1km and c) 3km.
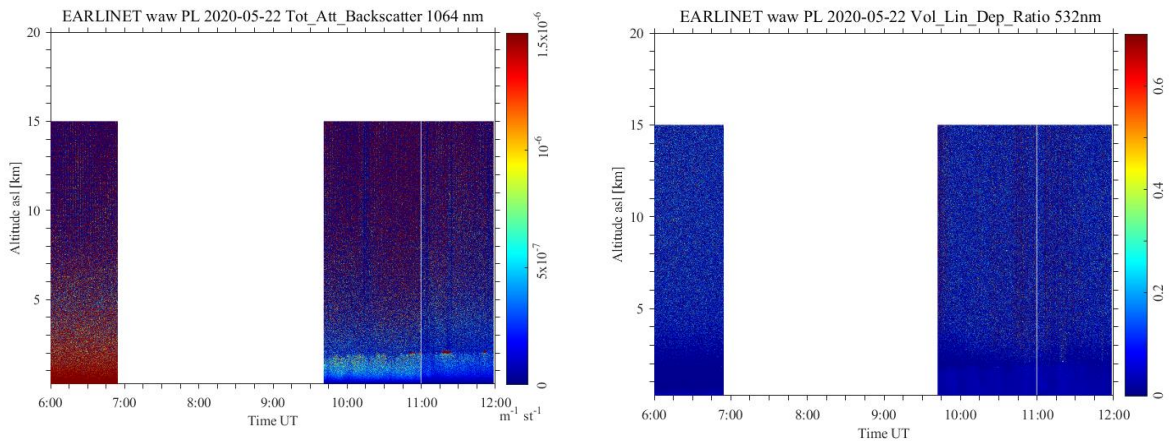
**Fig. S28)** Time-height plot of the attenuated backscatter coefficient at 1064 nm and volume linear depolarization ratio at 532 nm measured at Warsaw station on May 22, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 06:00 - 06:55 UTC) the majority of the aerosol load is found below 3 km.
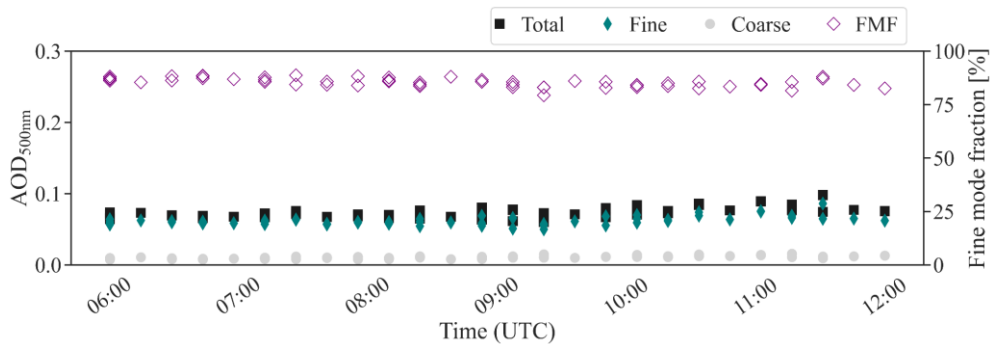
**Fig. S29)** Sun-photometer measurements from Warsaw station on May 22, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 6:16 UTC, so as to overlap with lidar measurements.
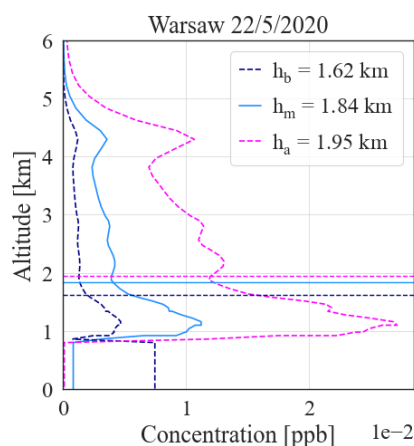
**Fig. S30)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Warsaw station on May 22, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

## S2.7 Antikythera 23 May, 2020
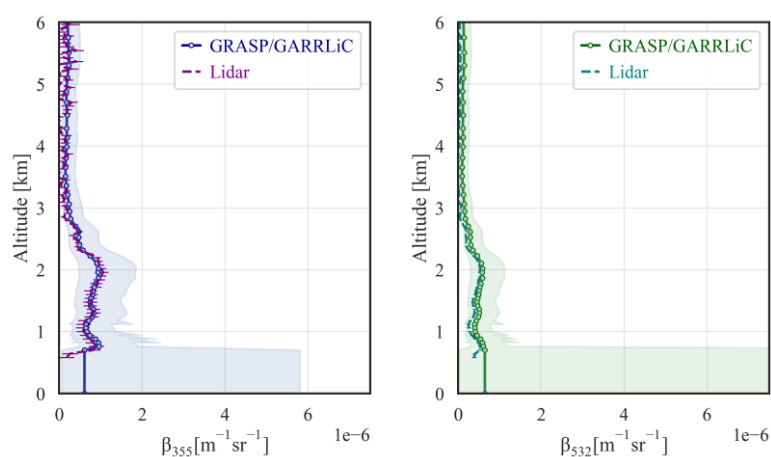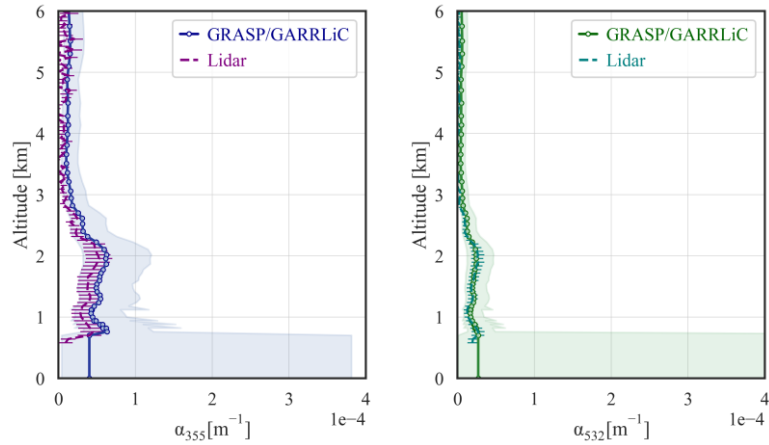
**Fig. S31)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left), and 532 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 04:00 and 06:00 UTC, 23 May, 2020.
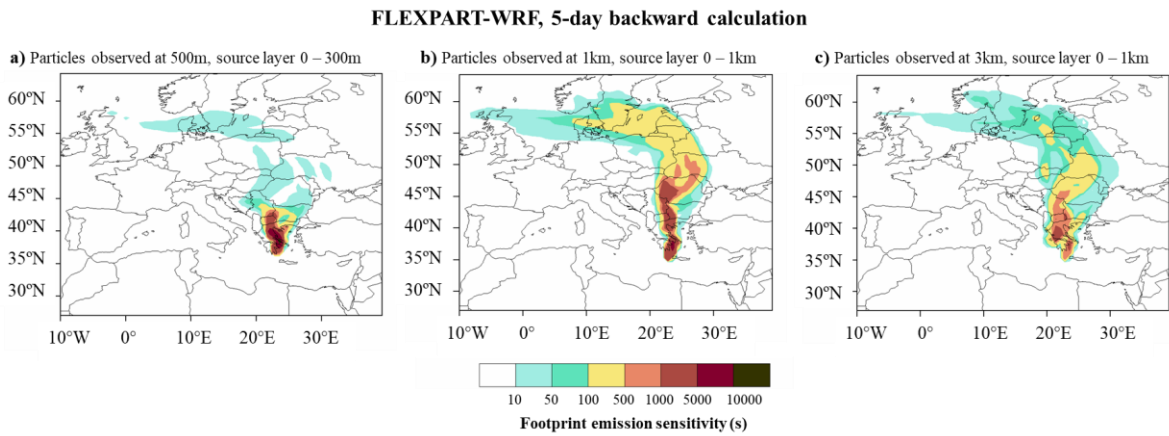
**Fig. S32)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Antikythera station, on May 23, 2020, at 05:04 UTC at heights a) 500m, b) 1km and c) 3km.
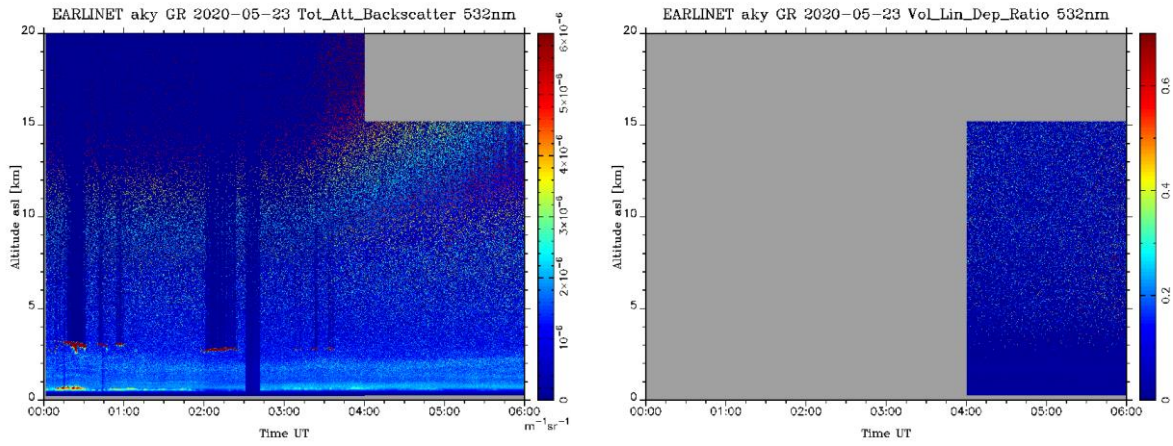
**Fig. S33)** Time-height plot of the attenuated backscatter coefficient at 1064 nm and volume linear depolarization ratio at 532 nm measured at Antikythera station on May 23, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 04:00 - 06:00 UTC) the majority of the aerosol load is found below 3 km.
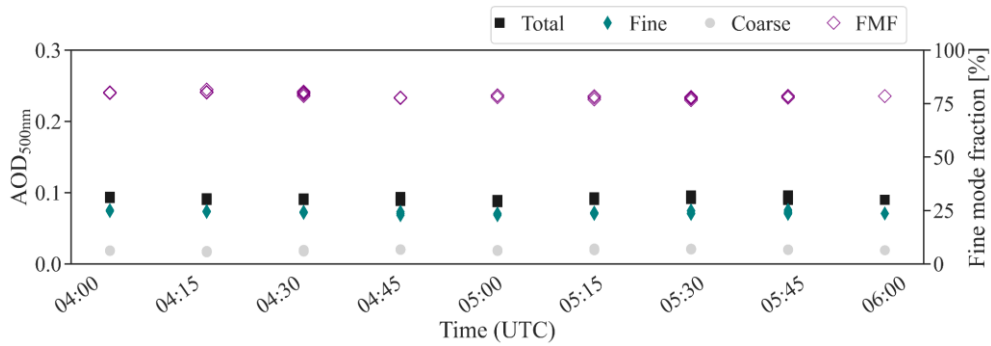
**Fig. S34)** Sun-photometer measurements from Antikythera station on May 23, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 05:04 UTC, so as to overlap with lidar measurements.

**Fig. S35)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Antikythera station on May 23, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

## S2.8 Kuopio 23 May, 2020
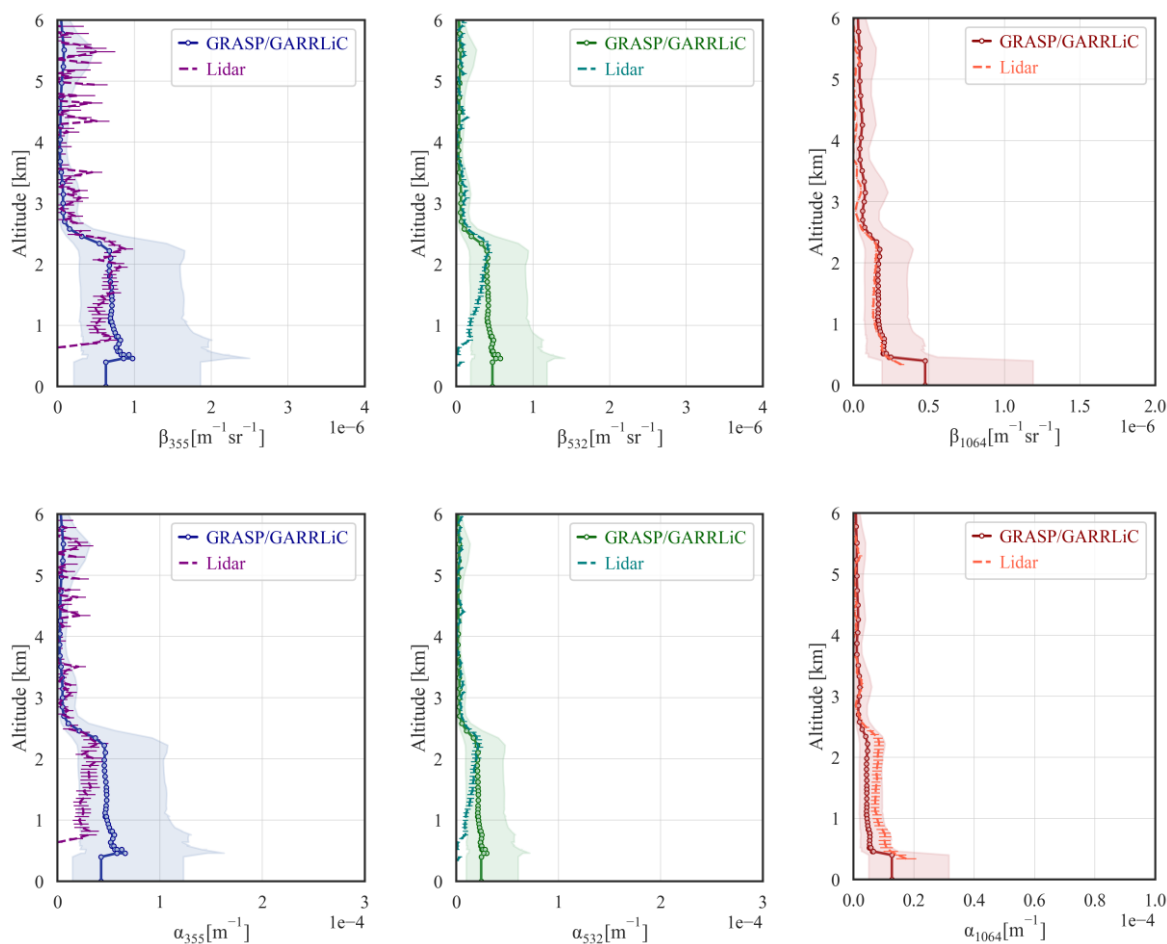
**Fig. S36)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left), 532 (middle) and 1064 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 14:00 and 16:00 UTC, 23 May, 2020.

**FLEXPART-WRF, 5-day backward calculation**
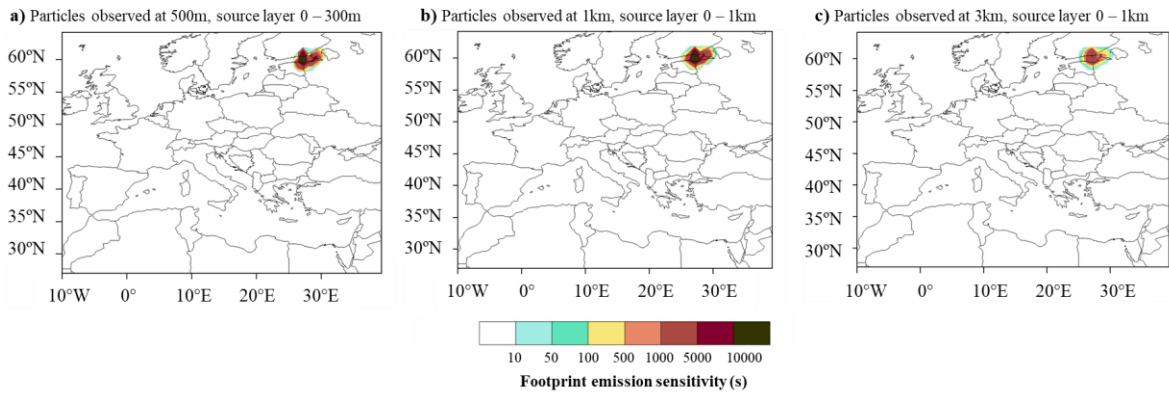
**Fig. S37)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Kuopio station, on May 23, 2020, at 14:32 UTC at heights a) 500m, b) 1km and c) 3km.
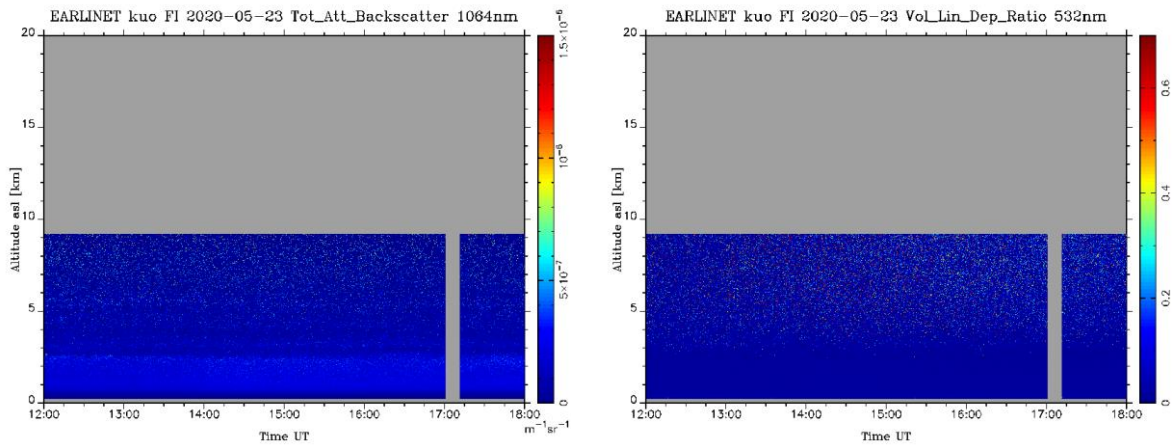
**Fig. S38)** Time-height plot of the attenuated backscatter coefficient at 1064 nm and volume linear depolarization ratio at 532 nm measured at Kuopio station on May 23, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 14:00 - 16:00 UTC) the majority of the aerosol load is found below 3 km.
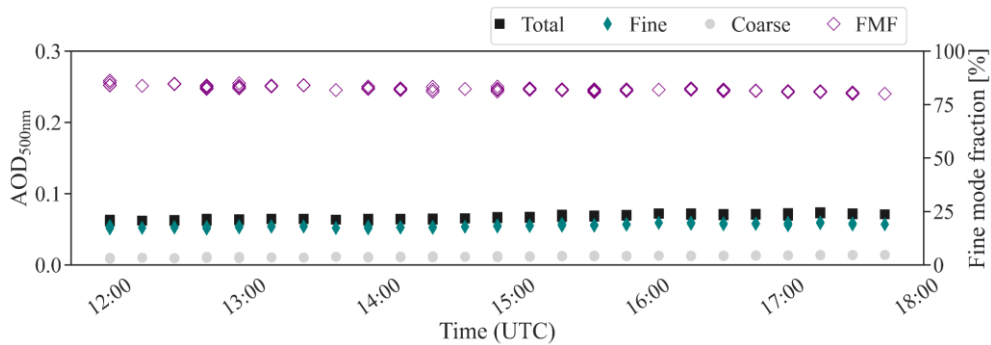
**Fig. S39)** Sun-photometer measurements from Kuopio station on May 23, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 14:32 UTC, so as to overlap with lidar measurements.

**Fig. S40)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Kuopio station on May 23, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

## S2.9 Kuopio 24 May, 2020
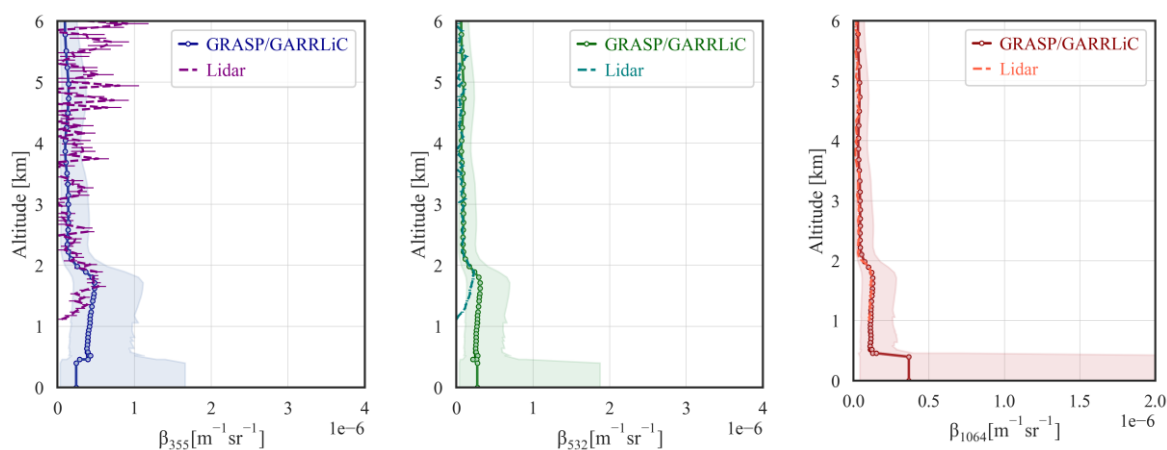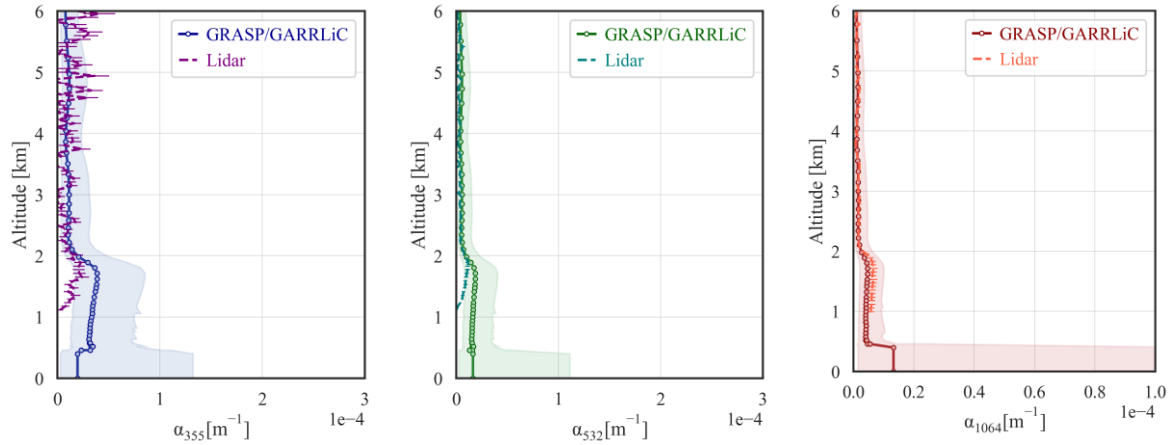
**Fig. S41)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left), 532 (middle) and 1064 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 08:00 and 10:00 UTC, 24 May, 2020.
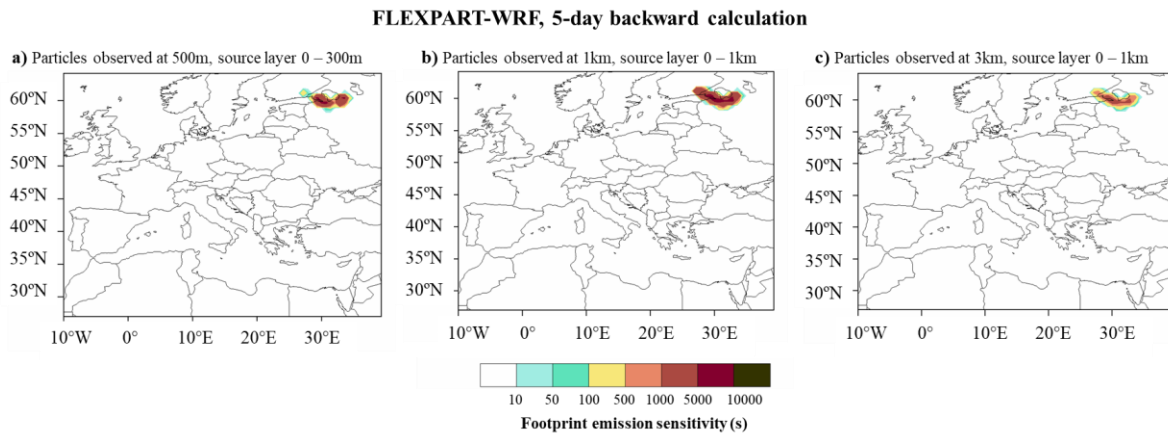
**Fig. S42)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Kuopio station, on May 24, 2020, at 08:18 UTC at heights a) 500m, b) 1km and c) 3km.
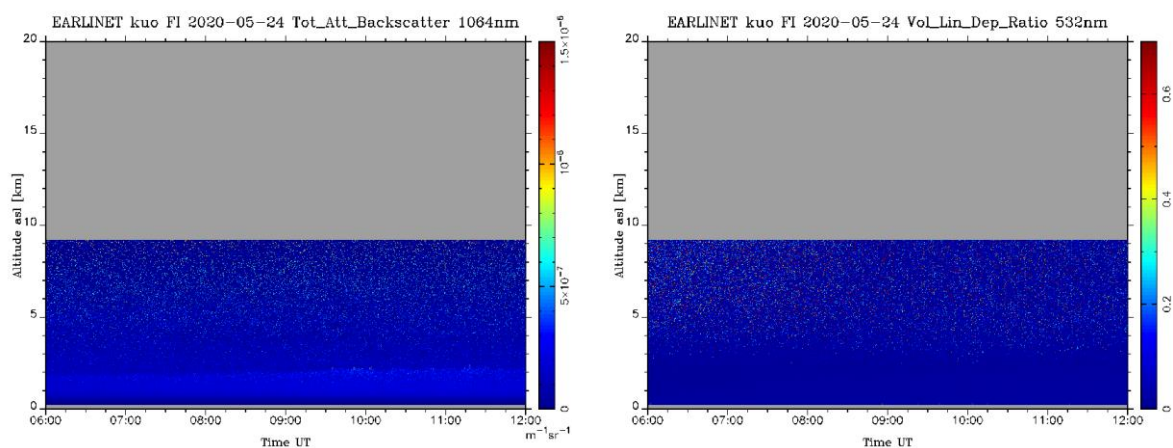
**Fig. S43)** Time-height plot of the attenuated backscatter coefficient at 1064 nm and volume linear depolarization ratio at 532 nm measured at Kuopio station on May 24, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 08:00 - 10:00 UTC) the majority of the aerosol load is found below 2 km.
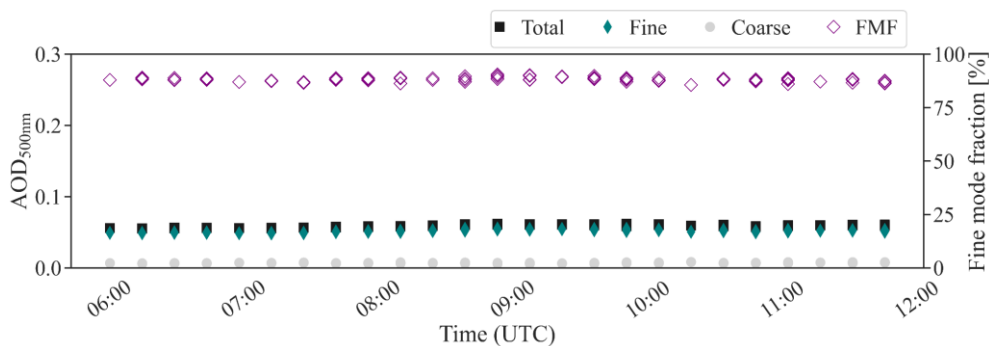
**Fig. S44)** Sun-photometer measurements from Kuopio station on May 24, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 08:18 UTC, so as to overlap with lidar measurements.
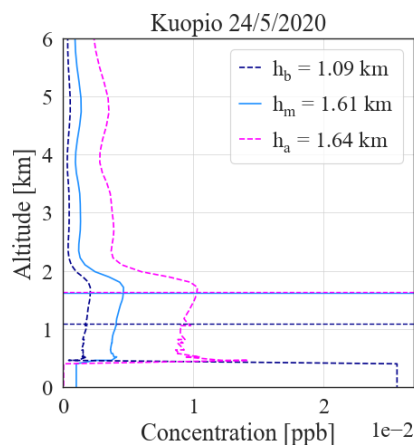
**Fig. S45)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Kuopio station on May 24, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

## S2.10 Antikythera 25 May, 2020
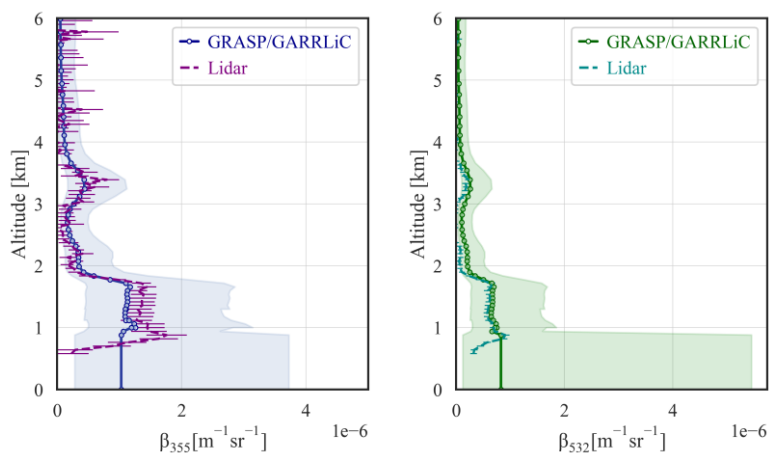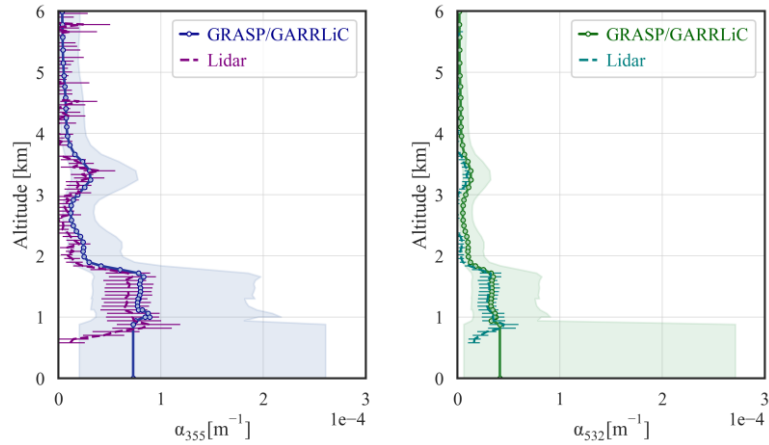
**Fig. S46)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left) and 532 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines).For the latter, lidar signals were averaged between 13:15 and 15:00 UTC, 25 May, 2020.
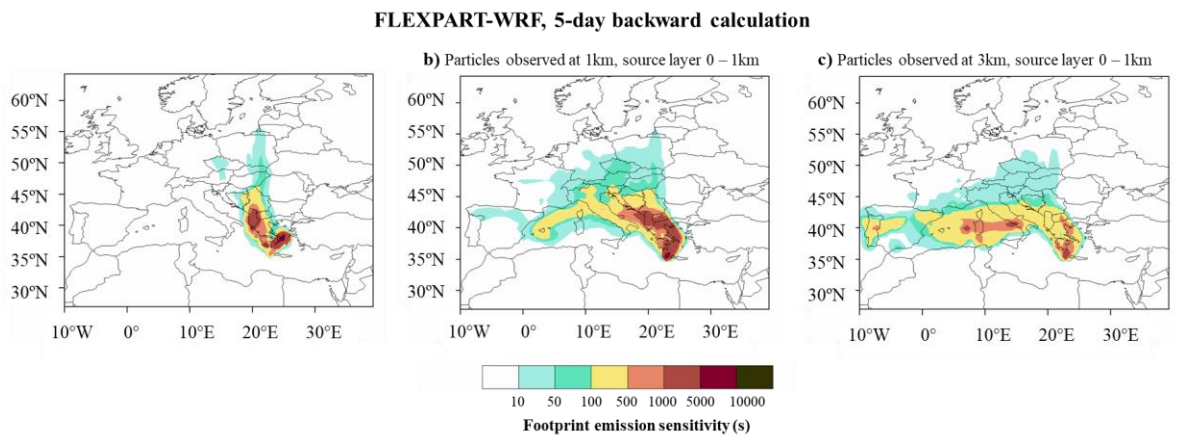
**Fig. S47)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Antikythera station, on May 25, 2020, at 14:25 UTC at heights a) 500m, b) 1km and c) 3km.
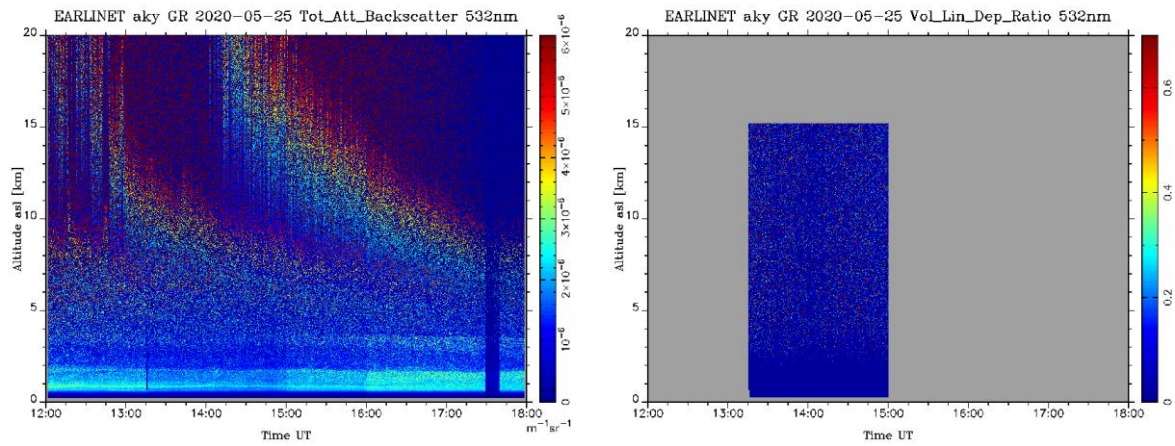
**Fig. S48)** Time-height plot of the attenuated backscatter coefficient at 1064 nm and volume linear depolarization ratio at 532 nm measured at Antikythera station on May 25, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 13:15 - 15:00 UTC) the majority of the aerosol load is found below 2 km while there is also a thin layer between 3 and 4 km.
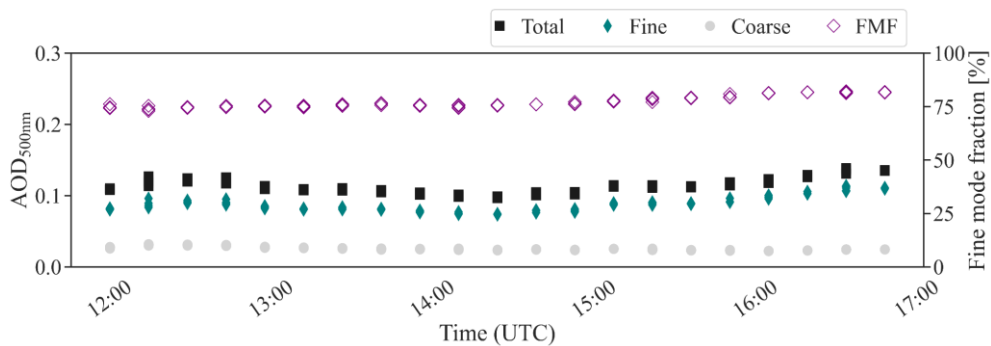
**Fig. S49)** Sun-photometer measurements from Antikythera station on May 25, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 14:25 UTC, so as to overlap with lidar measurements.

**Fig. S50)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Antikythera station on May 25, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

## S2.11 Barcelona 25 May, 2020
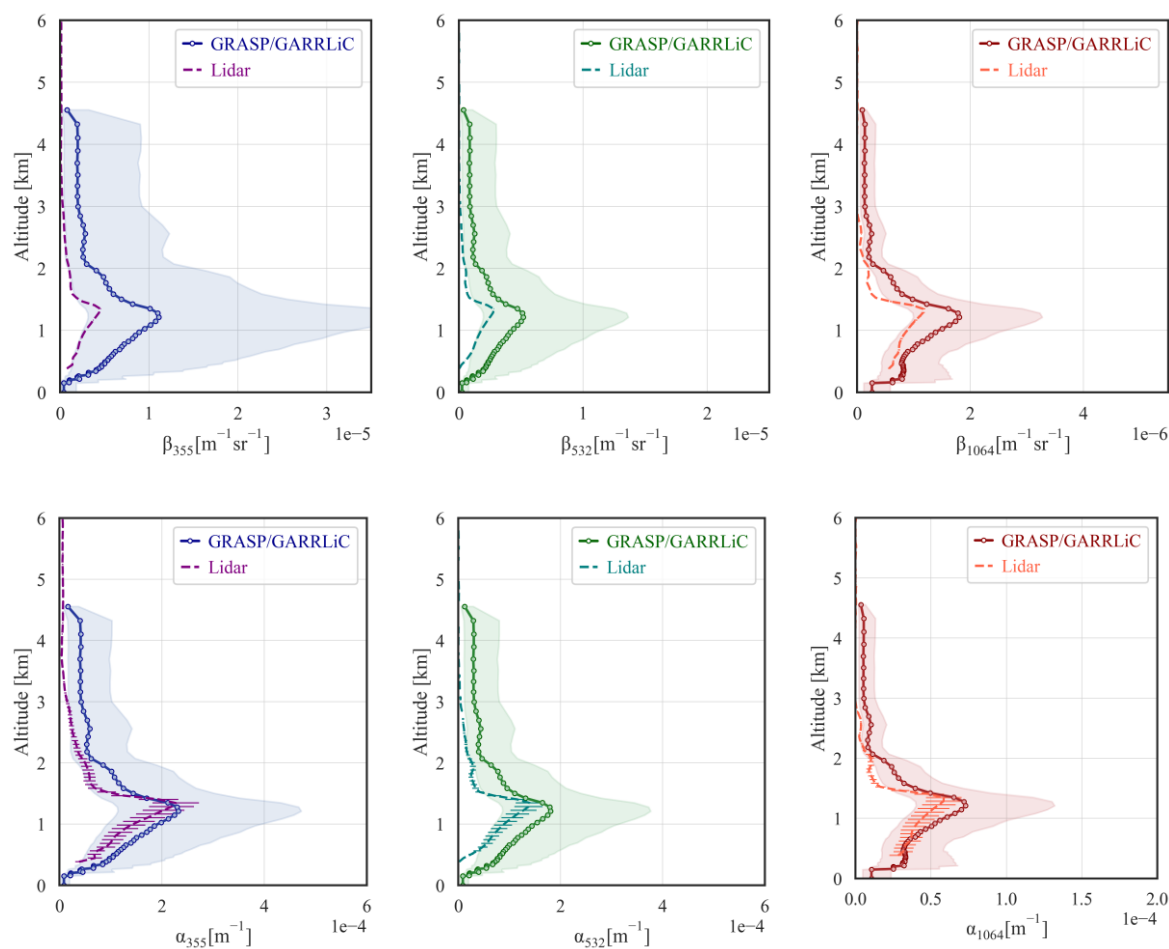
**Fig. S51)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left), 532 (middle) and 1064 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 17:09 and 19:09 UTC, 25 May, 2020.

**FLEXPART-WRF, 5-day backward calculation**
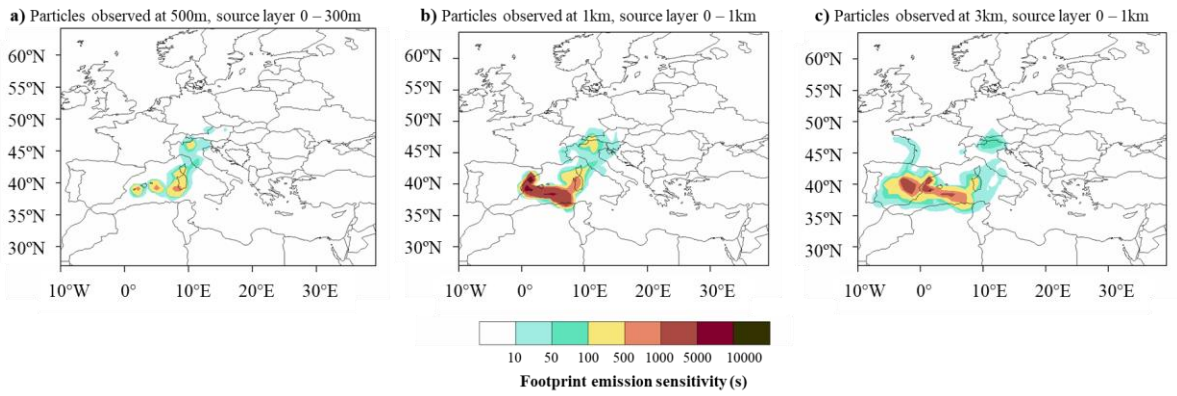
**Fig. S52)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Barcelona station, on May 25, 2020, at 15:50 UTC at heights a) 500m, b) 1km and c) 3km.
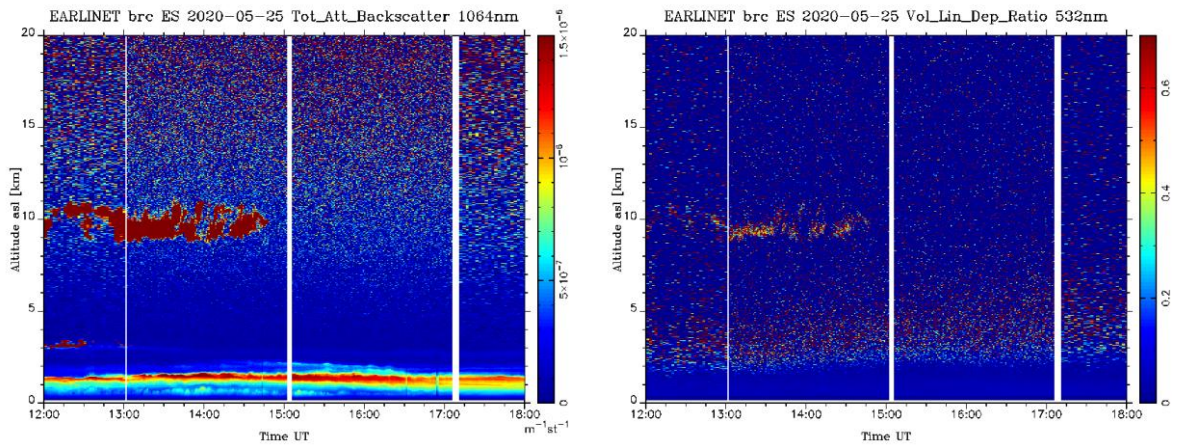
**Fig. S53)** Time-height plot of the attenuated backscatter coefficient at 1064 nm and volume linear depolarization ratio at 532 nm measured at Barcelona station on May 25, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 17:09 - 19:09 UTC) the majority of the aerosol load is found below 3 km.
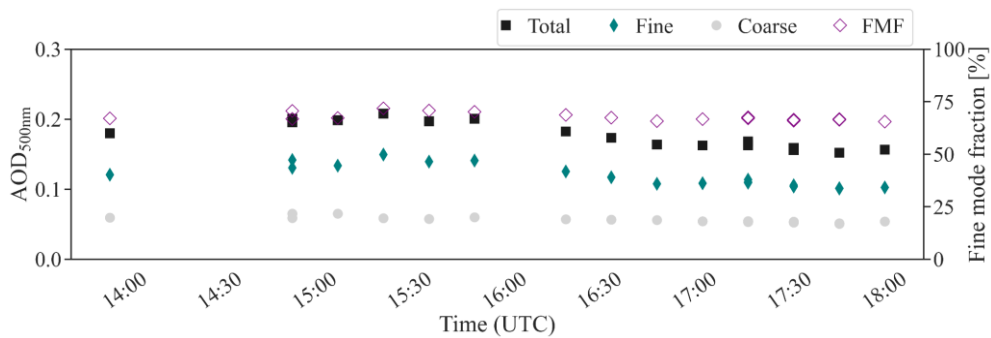
**Fig. S54)** Sun-photometer measurements from Barcelona station on May 25, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 15:50 UTC, so as to be close in time with the corresponding lidar measurements.

**Fig. S55)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Barcelona station on May 25, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

# S2.12 Warsaw 26 May, 2020
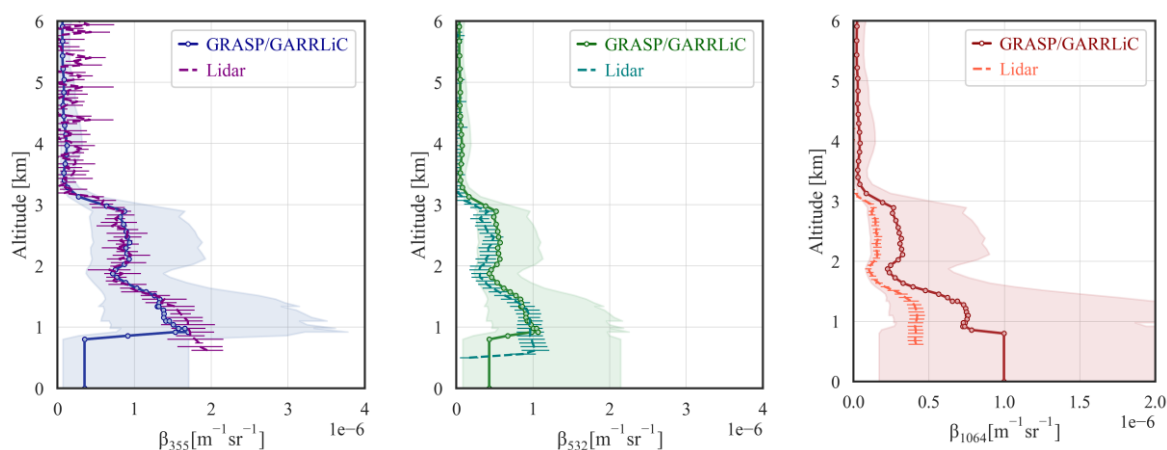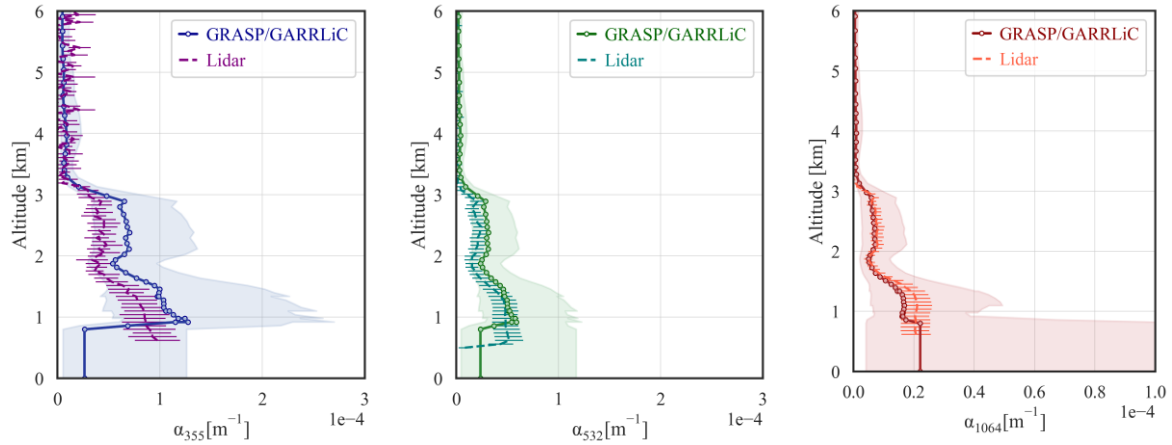
**Fig. S56)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left), 532 (middle) and 1064 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 16:20 and 17:05 UTC, 26 May, 2020.
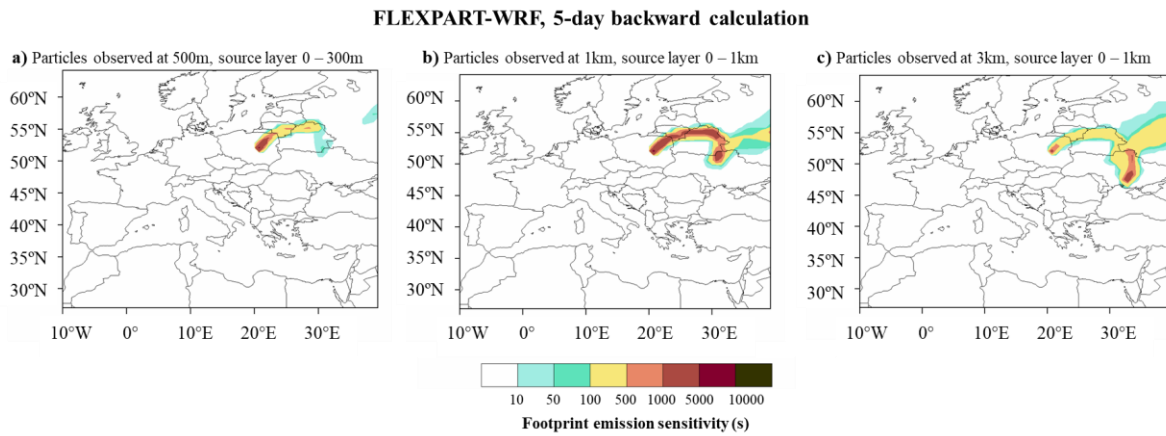
**Fig. S57)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Warsaw station, on May 26, 2020, at 16:48 UTC at heights a) 500m, b) 1km and c) 3km.
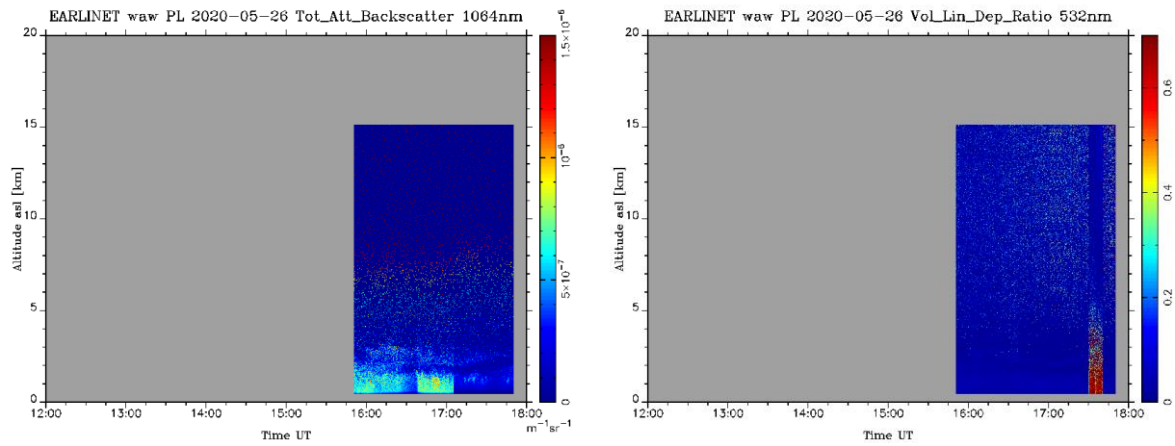
**Fig. S58)** Time-height plot of the attenuated backscatter coefficient at 1064 nm and volume linear depolarization ratio at 532 nm measured at Warsaw station on May 26, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 16:20 - 17:05 UTC) the majority of the aerosol load is found below 3 km.
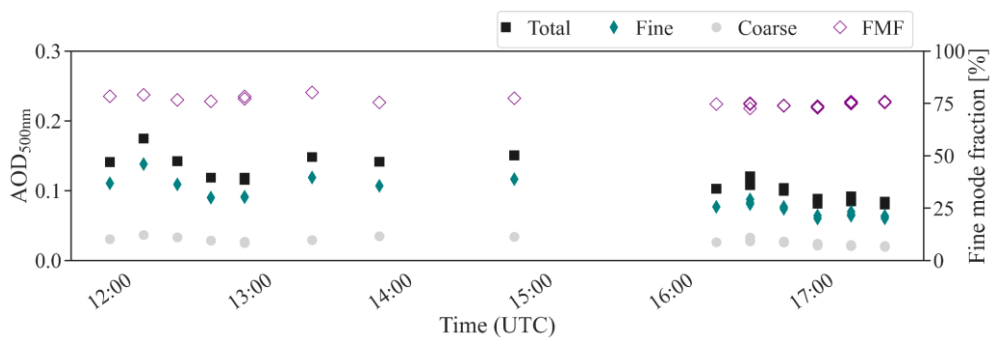
**Fig. S59)** Sun-photometer measurements from Warsaw station on May 26, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 16:48 UTC, so as to overlap with lidar measurements.
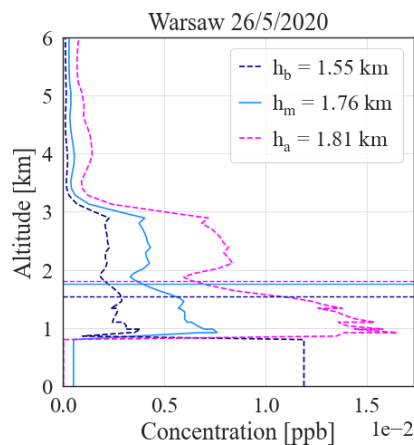
**Fig. S60)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Warsaw station on May 26, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.

## S2.13 Lecce 27 May, 2020
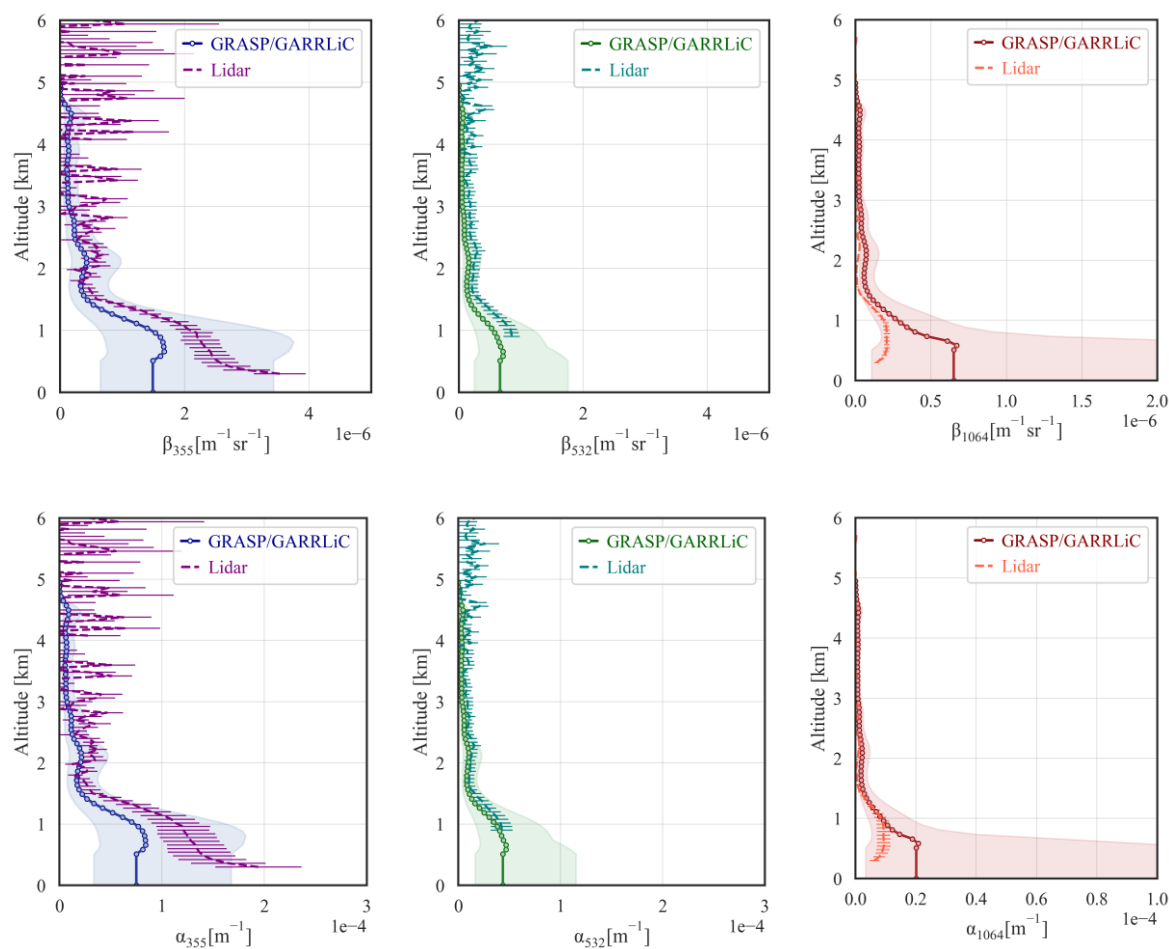
**Fig. S61)** Evaluation of the backscatter (upper row) and extinction coefficient profiles (lower row) at 355 (left), 532 (middle) and 1064 nm (right). The products derived from GRASP/GARRLiC (solid lines) are compared to the SCC derived products (dashed lines). For the latter, lidar signals were averaged between 13:50 and 15:00 UTC, 27 May, 2020.

**FLEXPART-WRF, 5-day backward calculation**
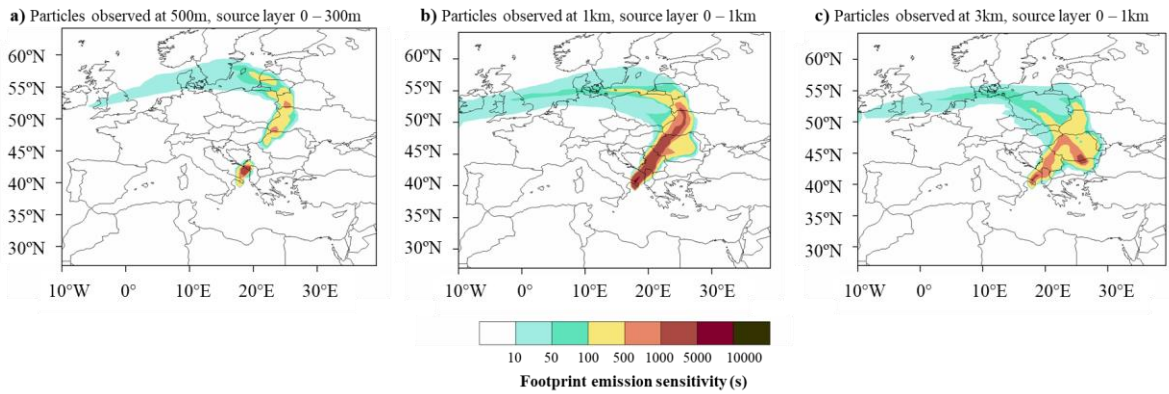
**Fig. S62)** 5-day backward source-receptor FLEXPART-WRF simulations for the particles arriving at Lecce station, on May 27, 2020, at 13:57 UTC at heights a) 500m, b) 1km and c) 3km.
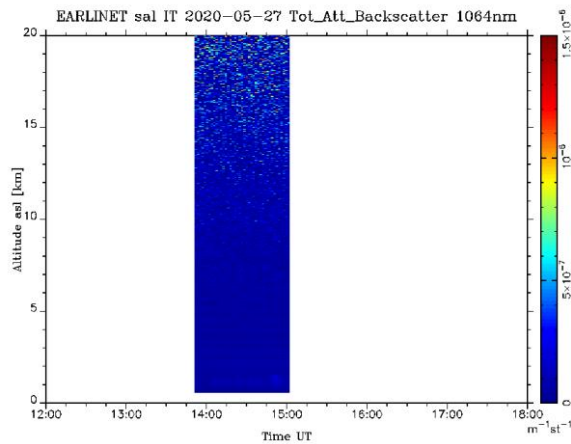
**Fig. S63)** Time-height plot of the attenuated backscatter coefficient at 1064 nm measured at Lecce station on May 27, 2020. At the time window used for GRASP/GARRLiC retrievals (i.e., 13:50 - 15:00 UTC) the majority of the aerosol load is found below 2 km while there is also a very low, in particles' concentration, layer just above 2 km.
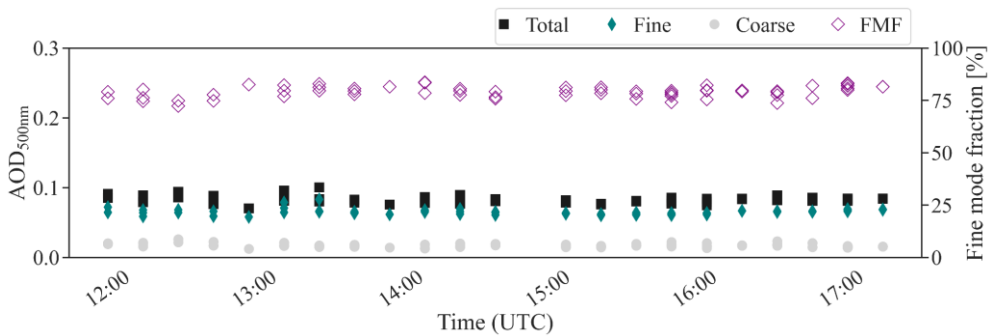
**Fig. S64)** Sun-photometer measurements from Lecce station on May 27, 2020: a) AOD at 500 nm (total; black squares, fine; green diamonds, and coarse; gray circles) and b) fine mode fraction (purple diamonds). Radiances and AOD values used for GRASP/GARRLiC retrievals were derived at 13:57 UTC, so as to overlap with lidar measurements.

**Fig. S65)** The anthropogenic component concentration profile, as derived from GRASP/GARRLiC for Belsk station on May 4, 2020. The light blue line ($h_m$) corresponds to the retrieval without accounting for the retrieval uncertainty. Due to the retrieval uncertainty, we construct the profiles "$h_a$" (light pink) and "$h_b$" (dark blue) as follows: "$h_a$" is the concentration profile taking into account the maximum value above the full overlap height and the minimum value below the full overlap height. "$h_b$" is the concentration profile taking into account the minimum value above the full overlap height and the maximum value below the full overlap height. The corresponding centre of mass height for each profile is shown in the legend in km.