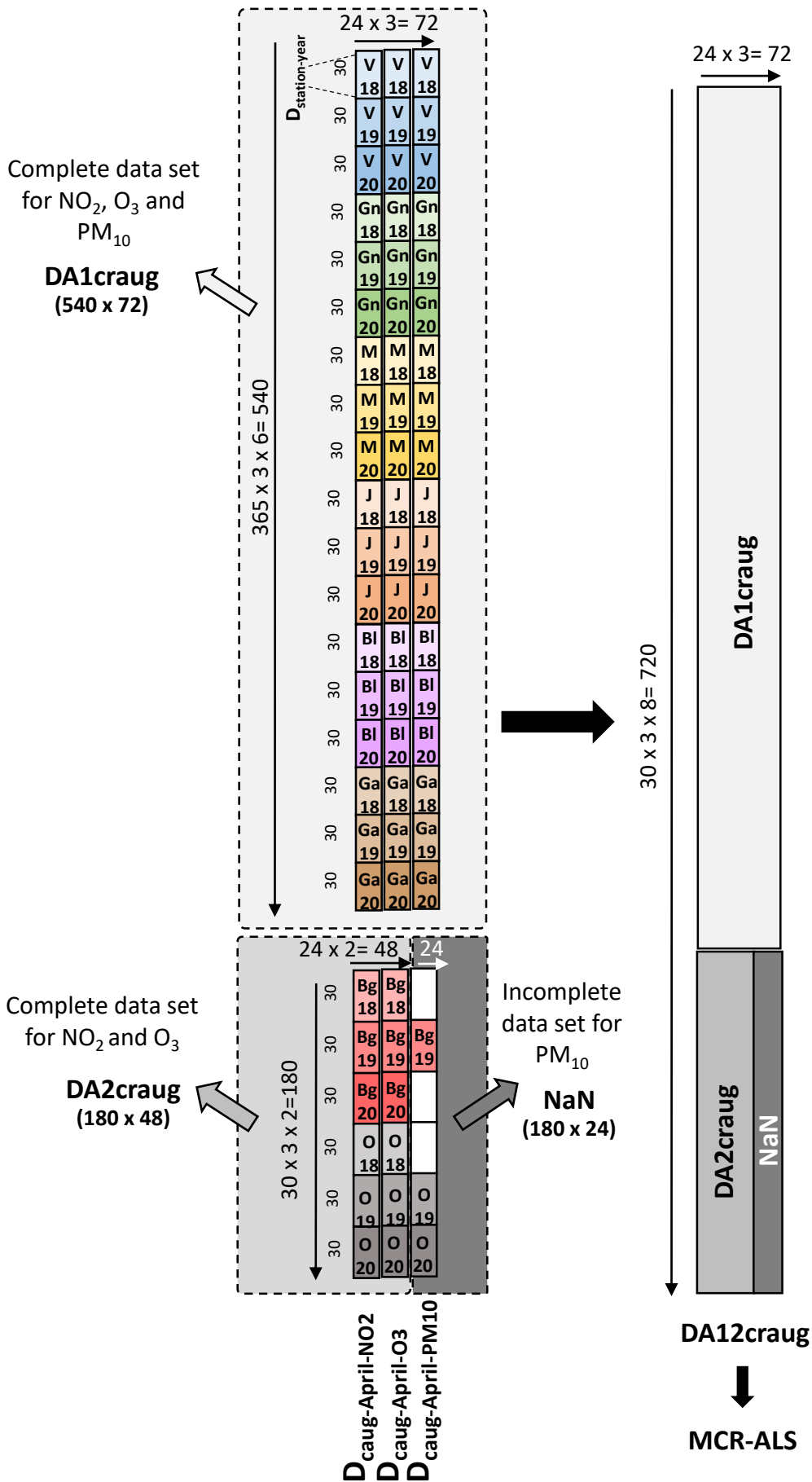
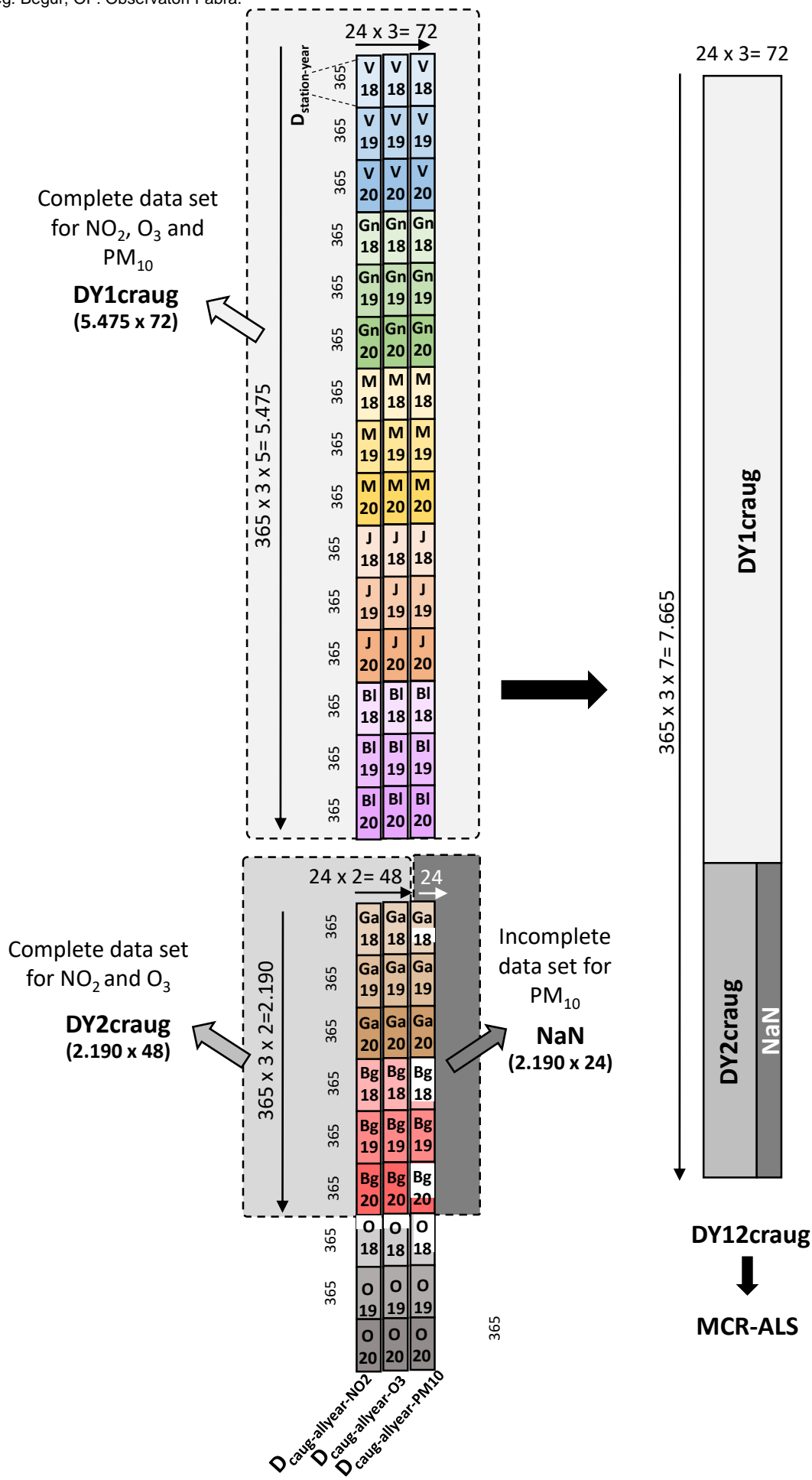


**Supplementary Figure S1.** Data arrangement for MCR-ALS simultaneous analysis of incomplete multiblock data sets. Individual data matrices of  $\text{NO}_2$ ,  $\text{O}_3$  and  $\text{PM}_{10}$  for the month of April were rearranged as explained in Section 2.7. to conduct their simultaneous MCR-ALS analysis. Three new matrices were generated:  $\text{DA1}_{\text{craug}}$  (540,72),  $\text{DA2}_{\text{craug}}$  (180,48) and  $\text{NaN}$  (180,24). The concatenation of these three matrices resulted in  $\text{DA12}_{\text{craug}}$  (5.475,24). VH: Vall d'Hebron, Grn: Granollers, Mn: Manlleu, Jun: Juneda, Bell: Bellver, Gra: Gràcia, Beg: Begur, OF: Observatori Fabra.

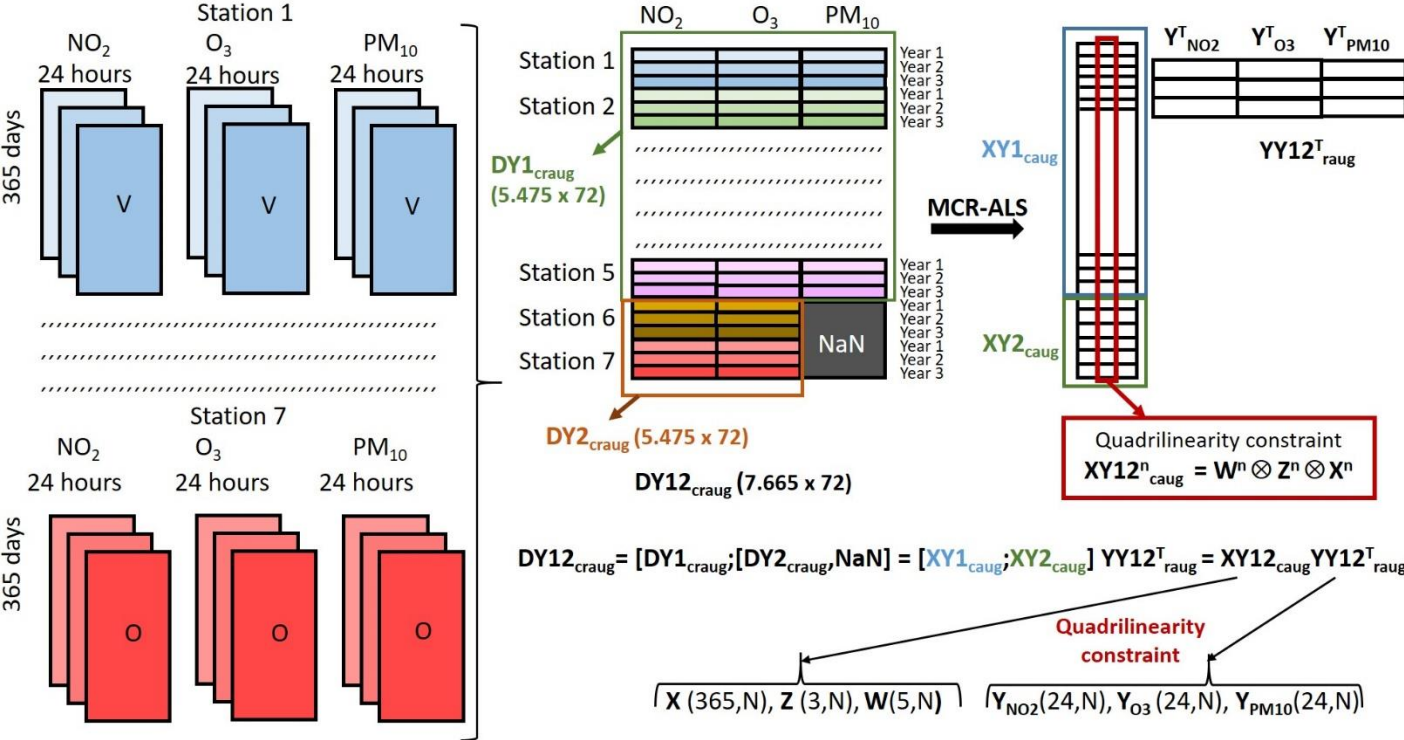


**Supplementary Figure S2.** Data arrangement for MCR-ALS simultaneous analysis of incomplete multiblock data sets. Individual data matrices of NO<sub>2</sub>, O<sub>3</sub> and PM<sub>10</sub> for the entire year were rearranged as explained in Section 2.7. to conduct their simultaneous MCR-ALS analysis. Three new matrices were generated: **DY1<sub>craug</sub>** (5,475,72), **DY2<sub>craug</sub>** (2,190,48) and **NaN** (2,190,24). The concatenation of these three matrices resulted in **DY12<sub>craug</sub>** (7,665,72). VH: Vall d'Hebron, Grn: Granollers, Mn: Manlleu, Jun: Juneda, Bell: Bellver, Gra: Gràcia, Beg: Begur, OF: Observatori Fabra.

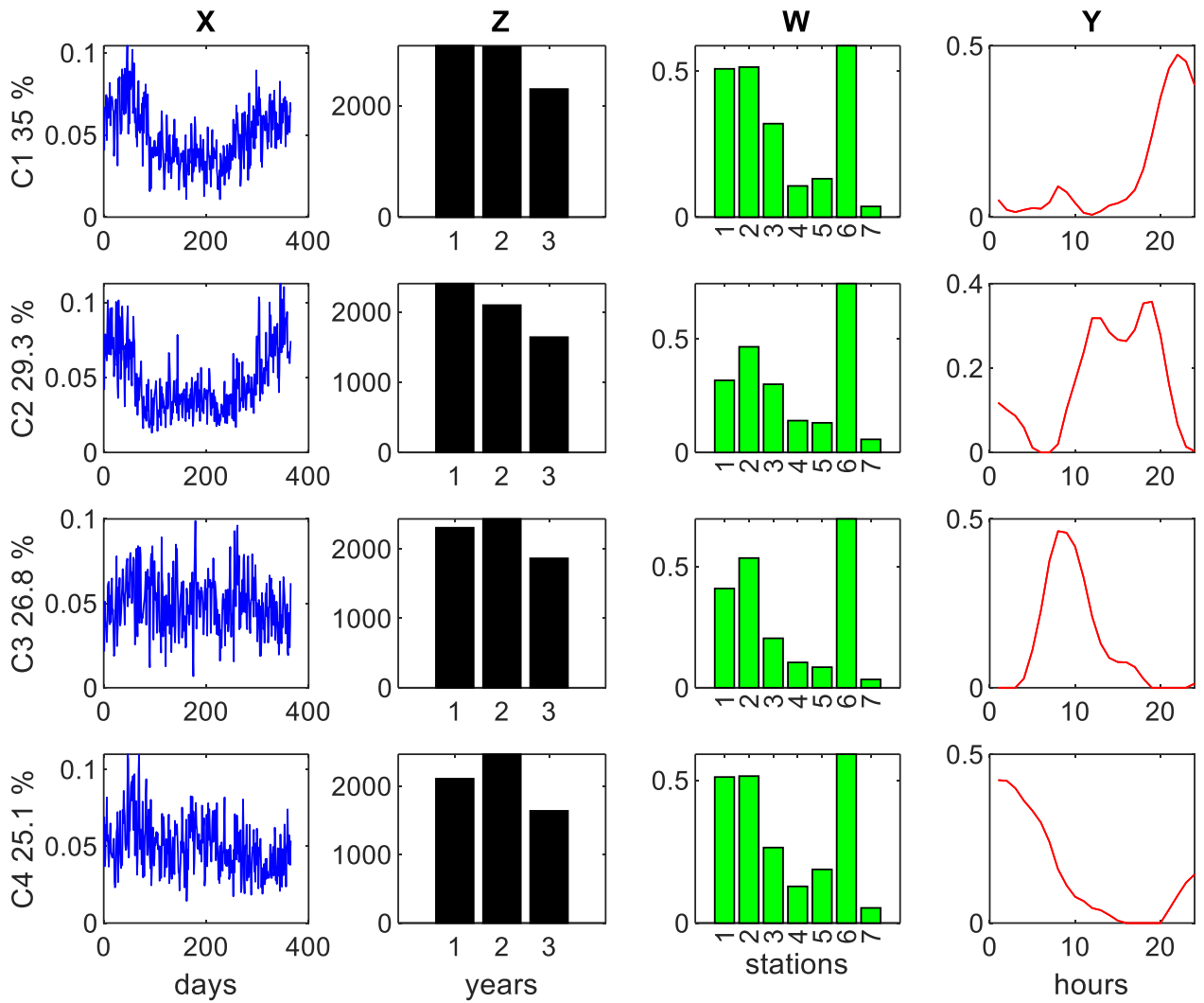


**Supplementary Figure S3.** MCR-ALS with the quadrilinearity constraint for the simultaneous analysis of the three contaminants in the incomplete multiblock data set of the entire year. See Equation 9 and their explanation in the manuscript.

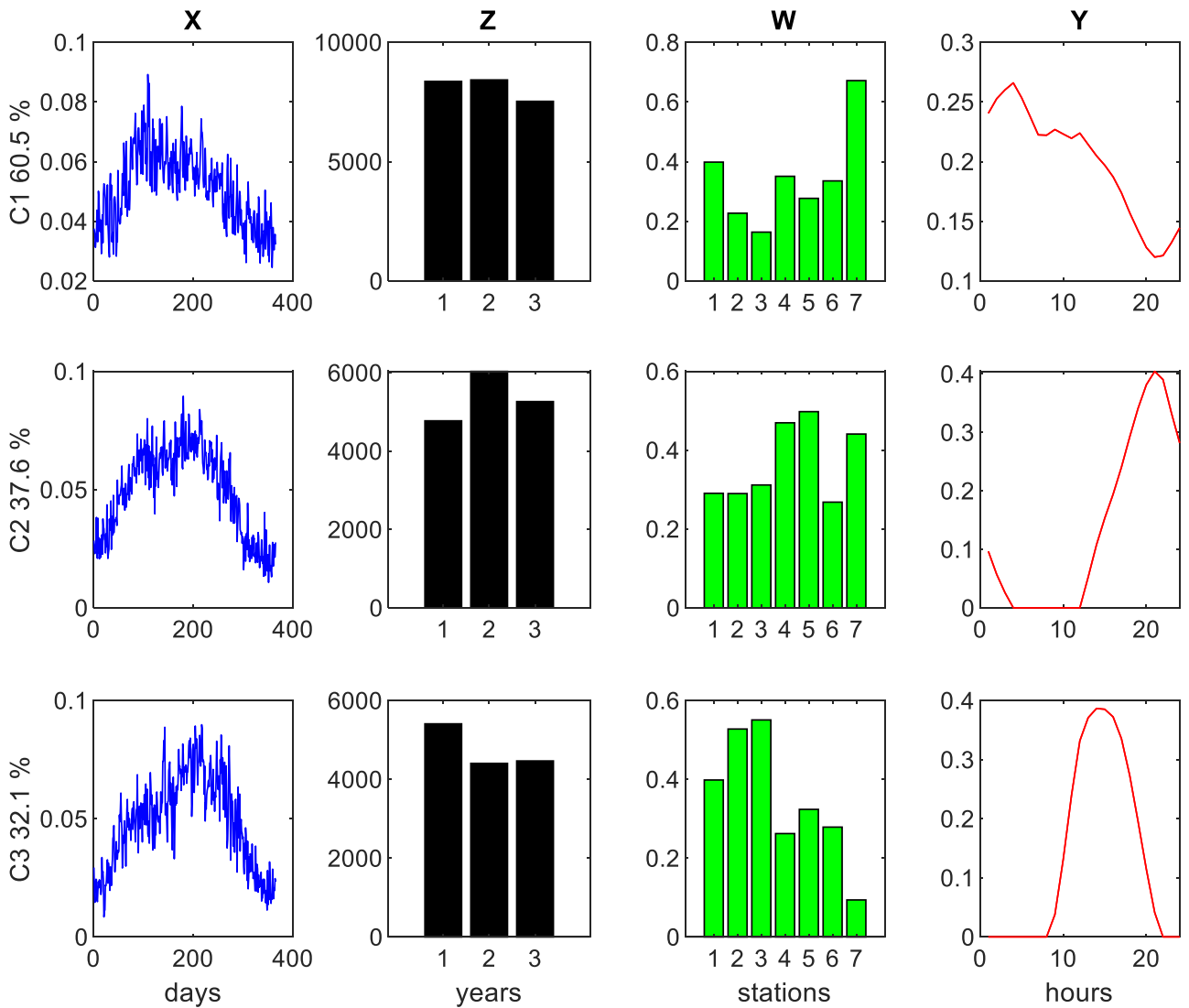
**MCR-ALS simultaneous analysis of incomplete multiblock data with quadrilinearity constraint for the entire year**



**Supplementary Figure S4.** MCR-ALS analysis of NO<sub>2</sub> concentrations in the column-wise super-augmented data matrix  $D_{\text{caug-allyear-NO}_2}$  (Equation 3) using non-negativity and quadrilinearity constraints. Profiles of the four different data modes are given in different colors: **(X)** in blue days of the year; **(Z)** in black year 1=2018, 2=2019 and 3= 2020; **(W)** in green stations 1: Vall d’Hebron, 2: Granollers, 3: Manlleu, 4: Juneda, 5: Bellver, 6: Gràcia, 7: Begur, and **(Y)** in red, hours of the day.



**Supplementary Figure S5.** MCR-ALS analysis of  $O_3$  concentrations in the column-wise super-augmented data matrix  $D_{\text{caug-allyear-}O_3}$  (Equation 3) using non-negativity and quadrilinearity constraints. Profiles of the four different data modes are given in different colors: (**X**) in blue days of the year; (**Z**) in black year 1=2018, 2=2019 and 3= 2020; (**W**) in green stations 1: Vall d’Hebron, 2: Granollers, 3: Manlleu, 4: Juneda, 5: Bellver, 6: Gràcia, 7: Begur, and (**Y**) in red, hours of the day.



**Supplementary Figure S6.** MCR-ALS analysis of  $PM_{10}$  concentrations in the column-wise super-augmented data matrix  $D_{\text{caug-allyear-}PM_{10}}$  (Equation 3) using non-negativity and quadrilinearity constraints. Profiles of the four different data modes are given in different colors: **(X)** in blue days of the year; **(Z)** in black year 1=2018, 2=2019 and 3= 2020; **(W)** in green stations 1: Vall d’Hebron, 2: Granollers, 3: Manlleu, 4: Juneda, 5: Bellver, and **(Y)** in red, hours of the day.

