Supplement

Section 2 showed how plume transmittance can be computed from measured satellite radiance images with only plume temperature as additional input. In order to implement the VPR procedure in real life, the parameters of Eqs. (4), (5), (6), and (8) have to be calculated first. The preliminary analysis required to compute these parameters and their resulting values is briefly described below for some aerosol particle types and for two volcanoes (Mt. Etna, Sicily, Italy, and Eyjafjallajökull, Iceland).

As already noted, the relationships between radiance at sensor and plume transmittance depend mainly on aerosol optical properties, and to a lesser extent on local climatology and sensor response functions. A set of parameters is therefore required for each type of aerosol, volcanic area, and radiometer used. To compute this set of parameters a series of representative scenarios are simulated using the MODTRAN radiative transfer model. The parameters given in Tables (S1 to S7) derive from a series of 27648 scenarios computed for each aerosol type with the volcanic cloud modelled as a uniform layer of one kilometre in thickness and located at 4 different heights (4, 6, 8, and 10 km). The cloud contains spherical particles of 8 different radii (R_e, the applied range is indicated in the tables), with 6 optical depths at 550 nm (δ^*, in the range 0-1.25), seen under 12 vertical zenith angles (θ_z) within the foreseen MODIS range of 0-65 degrees (or air mass factor μ), embedded in 12 monthly mean atmospheres. From these simulations, the total transmittance $\tau \cdot \tau_p$, the up-welling L_u, and down-welling L_d radiances are available. Now, assuming under the volcanic cloud an ocean surface with emissivity $\varepsilon = 0.98$ and temperature T_s, which is the climatic monthly mean temperature of the ocean area representative of the volcano considered, it is possible to compute the radiance measured by the sensor with the following Eq. (S1):

$$L_p = [\varepsilon \cdot B(T_s) + (1 - \varepsilon) \cdot L_d] \cdot \tau \cdot \tau_p + L_u \qquad \text{(S1)}$$

From the radiance at the sensor (L_p and the plume transmittance τ_p) it is easy to compute the two linear regressions shown in Fig. 3b and their intersection point for each month and plume height. The final step is determination of the parameters of Eqs. (4), (5), and (6). These are computed from the linear fit of 48 values of B_{up}, B_{dn}, and τ_t (12 months and 4 heights) as function of B_p, the Planck function at the mean plume temperature T_p.

Two volcanoes, Mt. Etna and Eyjafjallajökull, were considered with the typical monthly mean climatological atmospheres (pressure, temperature, and relative humidity) and sea surface temperature computed for each specific area. For the Mt. Etna volcano, the upper-air atmospheric radio sounding measured at the WMO Trapani station, and the sea temperature from the NOAA in the area 14-18 East, 34-38 North were used (see Pugnaghi et al. 2013).

The monthly mean values used for the Eyjafjallajökull area (330-350 East, 58-62 North) were obtained from the NCEP database of the NOAA/ESRL Physical Sciences Division (http://www.esrl.noaa.gov/psd/data/reanalysis/reanalysis.shtml).

In Tables (S1 to S7) the parameters of Eqs. (4), (5), (6) are reported for some volcanic ash types of known complex refractive index. These are pumice volcanic particles from Volz (1973), which appear to work well for the Mt. Etna volcano, along with andesite (often used for Eyjafjallajökull) and obsidian, both from Pollack et al. (1973). The laboratory measurements of the refractive index of the ash collected by Peters (2013) during the recent Eyjafjallajökull eruption are also available and the VPR parameters were computed, but only for this volcano.

Three other kinds of particle were also taken into account: water droplets, ice, and sulphuric acid. The ice formation is a known phenomenon that occur also inside a volcanic cloud (Rose et al., 1994; Rose et al., 2004; Durant et al., 2008). Moreover, droplets of sulphuric acid can be detected in volcanic clouds after the beginning of an eruption because these droplets are produced by oxidation of sulphur dioxide in the presence of water. The refractive index of the sulphuric acid depends on its temperature and a reference temperature of 215 K was used here.

The parameters, band by band, reported in Tables (S1 to S7) are extremely similar for the Terra and Aqua satellites because their differences are only due to minor variations in the effective wavelengths used for the two MODIS radiometers. No major variations were identified for the different volcanic areas, while there are more obvious variations due to the different ash types and even greater variations were observed between ash and non-ash particles.

Table S8 reports the parameters required by Eq. (8) to compute B_s from the Planck emission B_p. A series of representative scenarios again had to be simulated. In this case a 1 km thick volcanic cloud was considered containing only sulphur dioxide (10 values in the range 1-10 g m^{-2}), located at the same 4 previous altitudes and embedded in the same previous 12 monthly mean atmospheres, with the plumes observed under 12 zenith angles (0-65 deg.). This produced 5760 simulated scenarios.

The computed coefficients shown in Table S8 are very similar both for the two volcanoes, and for the two MODIS spectrometers.

Table S1: Coefficients of Eqs. (4), (5), (6) for pumice (Volz, 1973) particles, radius range 0.8-10 μm.

Volcano	Satellite	Band (μm)	a_{up}	b_{up}	a_{dn}	b_{dn}	a_{tt}	b_{tt}	r^2
Mt. Etna	*Terra*	8.7	0.572	2.589	0.738	1.009	-0.0163	0.337	0.9952
		11	0.621	2.449	0.793	0.927	-0.0185	0.358	0.9970
		12	0.591	2.500	0.782	0.931	-0.0226	0.353	0.9962
	Aqua	8.7	0.570	2.611	0.735	1.001	-0.0169	0.336	0.9952
		11	0.620	2.445	0.793	0.927	-0.0185	0.358	0.9970
		12	0.591	2.502	0.782	0.932	-0.0226	0.353	0.9962
Eyjafjallajökull	*Terra*	8.7	0.560	2.155	0.682	1.159	-0.0183	0.327	0.9956
		11	0.604	2.153	0.730	1.202	-0.0214	0.347	0.9971
		12	0.574	2.225	0.719	1.199	-0.0255	0.338	0.9964
	Aqua	8.7	0.558	2.175	0.678	1.162	-0.0192	0.326	0.9956
		11	0.604	2.154	0.730	1.202	-0.0215	0.347	0.9971
		12	0.573	2.226	0.719	1.200	-0.0253	0.338	0.9964

Table S2: Coefficients of Eqs. (4), (5), (6) for andesite (Pollack et al. 1973) particles, radius range 0.8-10 μm.

Volcano	**Satellite**	**Band (μm)**	a_{up}	b_{up}	a_{dn}	b_{dn}	a_{tt}	b_{tt}	r^2
Mt. Etna	*Terra*	8.7	0.804	1.068	0.803	0.553	-0.0068	0.312	0.9975
		11	0.679	2.080	0.809	0.849	-0.0176	0.344	0.9968
		12	0.551	2.764	0.760	1.042	-0.0204	0.348	0.9956
	Aqua	8.7	0.800	1.113	0.798	0.568	-0.0059	0.300	0.9972
		11	0.678	2.088	0.809	0.850	-0.0176	0.345	0.9968
		12	0.550	2.768	0.760	1.044	-0.0203	0.348	0.9956
Eyjafjallajökull	*Terra*	8.7	0.802	0.898	0.771	0.659	-0.0083	0.304	0.9976
		11	0.664	1.836	0.751	1.109	-0.0187	0.322	0.9968
		12	0.533	2.453	0.695	1.301	-0.0233	0.337	0.9958
	Aqua	8.7	0.797	0.937	0.771	0.652	-0.0000	0.263	0.9974
		11	0.663	1.843	0.750	1.111	-0.0187	0.322	0.9968
		12	0.532	2.456	0.695	1.312	-0.0231	0.337	0.9958

Table S3: Coefficients of Eqs. (4), (5), (6) for obsidian (Pollack et al. 1973) particles, radius range 0.8-10 μm.

Volcano	**Satellite**	**Band (μm)**	$\mathbf{a_{up}}$	$\mathbf{b_{up}}$	$\mathbf{a_{dn}}$	$\mathbf{b_{dn}}$	$\mathbf{a_{tt}}$	$\mathbf{b_{tt}}$	$\mathbf{r^2}$
Mt. Etna	*Terra*	8.7	0.740	1.483	0.761	0.814	-0.0035	0.271	0.9925
		11	0.617	2.500	0.791	0.941	-0.0194	0.348	0.9968
		12	0.619	2.406	0.792	0.915	-0.194	0.343	0.9960
	Aqua	8.7	0.742	1.479	0.758	0.823	-0.0041	0.272	0.9924
		11	0.617	2.502	0.791	0.942	-0.0193	0.348	0.9968
		12	0.619	2.404	0.792	0.915	-0.0196	0.344	0.9960
Eyjafjallajökull	*Terra*	8.7	0.733	1.250	0.714	0.946	0.0034	0.227	0.9923
		11	0.601	2.194	0.730	1.204	-0.0215	0.331	0.9969
		12	0.602	2.143	0.733	1.162	-0.0215	0.328	0.9962
	Aqua	8.7	0.735	1.244	0.712	0.953	0.0041	0.222	0.9922
		11	0.601	2.196	0.730	1.205	-0.0214	0.331	0.9967
		12	0.602	2.141	0.734	1.161	-0.0216	0.329	0.9963

Table S4: Coefficients of Eqs. (4), (5), (6) for Eyja ash particles (Peters, 2013), radius range 0.8-10 μm.

Volcano	**Satellite**	**Band (μm)**	a_{up}	b_{up}	a_{dn}	b_{dn}	a_{tt}	b_{tt}	r^2
Eyjafjallajökull	*Terra*	8.7	0.769	1.079	0.746	0.818	0.0066	0.241	0.9955
		11	0.658	1.851	0.745	1.110	-0.0205	0.316	0.9967
		12	0.601	2.113	0.735	1.150	-0.0228	0.333	0.9964
	Aqua	8.7	0.769	1.083	0.743	0.824	0.0077	0.227	0.9953
		11	0.657	1.853	0.745	1.111	-0.0205	0.317	0.9967
		12	0.602	2.108	0.736	1.148	-0.0226	0.334	0.9964

Table S5: Coefficients of Eqs. (4), (5), (6) for water droplets, radius range 2-50 μm.

Volcano	Satellite	Band (μm)	a_{up}	b_{up}	a_{dn}	b_{dn}	a_{tt}	b_{tt}	r^2
Mt. Etna	*Terra*	8.7	0.615	2.486	0.751	1.016	-0.0160	0.355	0.9847
		11	0.867	0.946	0.920	0.288	-0.0184	0.415	0.9992
		12	0.888	0.744	0.927	0.260	-0.0127	0.378	0.9995
	Aqua	8.7	0.616	2.485	0.749	1.036	-0.0172	0.365	0.9848
		11	0.867	0.943	0.920	0.288	-0.0184	0.415	0.9992
		12	0.887	0.745	0.927	0.260	-0.0127	0.378	0.9995
Eyjafjallajökull	*Terra*	8.7	0.605	2.054	0.709	1.079	-0.0211	0.361	0.9850
		11	0.861	0.838	0.896	0.411	-0.0243	0.430	0.9991
		12	0.882	0.679	0.903	0.384	-0.0148	0.378	0.9995
	Aqua	8.7	0.607	2.053	0.707	1.097	-0.0231	0.375	0.9851
		11	0.862	0.836	0.896	0.410	-0.0242	0.430	0.9991
		12	0.882	0.679	0.903	0.385	-0.0148	0.378	0.9995

Table S6: Coefficients of Eqs. (4), (5), (6) for ice particles, radius range 2-50 μm.

Volcano	**Satellite**	**Band (μm)**	a_{up}	b_{up}	a_{dn}	b_{dn}	a_{tt}	b_{tt}	r^2
Mt. Etna	*Terra*	8.7	0.617	2.464	0.748	1.021	-0.0180	0.361	0.9847
		11	0.882	0.830	0.918	0.307	-0.0112	0.376	0.9995
		12	0.827	1.184	0.891	0.451	-0.0123	0.348	0.9988
	Aqua	8.7	0.615	2.484	0.749	1.014	-0.0177	0.361	0.9848
		11	0.882	0.837	0.917	0.309	-0.0112	0.375	0.9995
		12	0.826	0.826	0.891	0.452	-0.0124	0.349	0.9988
Eyjafjallajökull	*Terra*	8.7	0.607	2.037	0.706	1.087	-0.0241	0.369	0.9849
		11	0.876	0.749	0.892	0.436	-0.0136	0.379	0.9995
		12	0.818	1.065	0.856	0.617	-0.0133	0.339	0.9988
	Aqua	8.7	0.605	2.055	0.707	1.082	-0.0236	0.369	0.9851
		11	0.876	0.752	0.892	0.439	-0.0135	0.378	0.9995
		12	0.817	1.073	0.855	0.617	-0.0139	0.342	0.9988

Table S7: Coefficients of Eqs. (4), (5), (6) for sulphuric acid droplets, radius range 2-50 μm.

Volcano	**Satellite**	**Band (μm)**	a_{up}	b_{up}	a_{dn}	b_{dn}	a_{tt}	b_{tt}	r^2
Mt. Etna	*Terra*	8.7	0.707	1.847	0.787	0.727	-0.0132	0.336	0.9971
		11	0.719	2.007	0.831	0.764	-0.0153	0.354	0.9980
		12	0.644	2.298	0.780	0.976	-0.0266	0.397	0.9959
	Aqua	8.7	0.705	1.864	0.785	0.732	-0.0132	0.335	0.9971
		11	0.718	2.012	0.831	0.767	-0.0154	0.354	0.9980
		12	0.644	2.299	0.780	0.977	-0.0265	0.397	0.9959
Eyjafjallajökull	*Terra*	8.7	0.697	1.546	0.742	0.870	-0.0146	0.325	0.9973
		11	0.707	1.762	0.780	0.990	-0.0185	0.351	0.9980
		12	0.628	2.045	0.722	1.213	-0.0351	0.413	0.9958
	Aqua	8.7	0.696	1.560	0.740	0.877	-0.0146	0.324	0.9973
		11	0.706	1.766	0.779	0.993	-0.0186	0.351	0.9980
		12	0.628	2.045	0.722	1.213	-0.0349	0.413	0.9958

Table S8: Coefficients of Eq. (8), band at 8.7 μm.

Volcano	**Satellite**	**a_s**	**b_s**
Mt. Etna	*Terra*	0.9419	0.1120
	Aqua	0.9412	0.1101
Eyjafjallajökull	*Terra*	0.9492	0.0918
	Aqua	0.9477	0.0934